High-power terahertz generation at megahertz repetition rates using few-cycle pulses

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High-power terahertz generation at megahertz repetition rates using few-cycle pulses

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May 2021
Supervisor: Professor Peter Uhd Jepsen
Co-supervisor: Associate Professor Edmund John Railton Kelleher
To my grandfather

Albert Friedrich Gerhard Buchmann
Acknowledgements

At the end of the year 2017 I had a choice to make - accept an offer as product quality manager for Europe with one of the world’s largest electronics producers, or accepting a 3-year PhD contract at DTU. As these two choices were fundamentally different - industrial vs. academic - it was not an easy choice, but after almost three and a half years at DTU I am convinced that I did not make the wrong decision. Science has since long been a passion of mine. I was lucky to receive a broad education resulting in three degrees in Physics and Geology prior to starting my PhD. Through this and from constant discussions with colleagues from different fields I realised how interconnected science is, and must be. Having a thorough and detailed understanding of scientific topics and being able to explain them in layman terms to non-scientists has been a key-takeaway from my time at DTU - especially in a time where many people believe their access to the internet equates gaining knowledge over years of research and studying. For this reason, I have written this thesis in a way that allows interested readers from other fields to understand the entire concept behind my work. None of this would have been possible though without the support of a whole range of people and institutions, which I wish to express my gratitude to in no particular order.

I have great appreciation for my principle supervisor Prof. Peter Uhd Jepsen and his support throughout the years. This naturally includes the frequent personal and scientific discussions, but also his ability as a skilled leader of the Ultrafast Infrared and Terahertz Science group, allowing for several bright minds to work together closely and in a collaborative manner. Additionally, I must stress my gratitude towards his great flexibility that allowed me to continue my PhD path as intended despite the global situation resulting from the COVID-19 pandemic.

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Larsen, they were also involved in DTU Lys which allowed for several great events and beer-tapping fun which I enjoyed thoroughly. Also, our table-football games in the kitchen were fiercely competitive and would act to provide energy and motivation after lunch to continue working on my PhD.

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All these people, and many more that I have failed to mention, were part of this thesis and my life for the last three years and therefore deserve credit for all they have done to help me personally and professionally. The connections I have made during this tenure will hopefully last for a long time, and I am looking forward to meeting any of all these people in the future.
Abstract

Terahertz (THz) radiation can be used in an abundance of applications ranging from fundamental science to in-situ quality control of industrial processes. The electric field strengths of strong THz-radiation allows for studying fundamental properties of matter, and ultrafast THz-spectroscopy is a promising tool for material analysis and security applications. Semi-transparent to many common objects that are opaque at optical frequencies, THz-radiation is used in non-invasive imaging applications. On top of this, THz-imaging offers the additional benefit over high-frequency methods like X-Rays by having very low photon-energies and thus being non-damaging to the materials exposed to it. The largest disadvantage of THz-radiation is its strong absorption by water molecules, which are present in many materials of interest (e.g. biological samples), and omnipresent in the atmosphere as humidity. Hence, for the practicality of the above mentioned applications, a strong THz-source is required. Currently, most high-efficiency, broadband THz-generation methods require strong pump-light energy, which is typically provided with large lasers operating at low repetition rates of a few hertz or kilohertz at most. Regrettfully, this low repetition rate drastically affects measurement time or data acquisition speed. As a consequence, the signal-to-noise ratio tends to be low when fast measurements are required.

To circumvent both of these issues, this thesis provides a feasible approach to generate high-power THz-radiation at MHz repetition rates by using a compact fibre-laser system and a simple external pulse compression method. The combination produces high peak power laser pulses which are applied to drive optical rectification (OR) in a highly efficient organic crystal to produce THz-radiation with a comparatively large efficiency. The output of a near-infrared femtosecond ytterbium-doped fibre-laser is passed through a polarisation maintaining large mode area photonic crystal fibre (LMA-PCF) inducing spectral broadening to a full-width half-maximum (FWHM) bandwidth of ~100 nm from an initial FWHM of 8 nm. The strongly polarised output is sent through a pair of SF11-glass prisms, compressing the spectrally broadened pulse from 250 fs to 22 fs at FWHM pulse duration. Ignoring fibre coupling loss, this method provides an almost tenfold increase in peak power to 13.8 MW at a repetition rate of 10 MHz. Further scaling of the repetition rate (and thus the average power) proved possible, as the damage mechanism were solely peak power dependent at >2 MW.

The 22 fs beam is focussed onto the organic crystal HMQ-TMS, which provides a highly efficient option for THz-generation through OR in a collinear setup, reducing complexity compared to other efficient methods for THz-generation, while also providing a far larger spectral bandwidth. It spans from below 1 THz to over 6 THz at an average power of 1.38 mW with peak electric field strengths exceeding 6 kV·cm⁻¹ in standard atmospheric conditions for fairly large THz-spot sizes.
of 369 µm in radius at $1/e^2$-intensity. Thus, the field strengths can easily exceed 20 kV·cm$^{-1}$ through tighter focussing and by operating in dry-air environments. The optical to THz-generation efficiency of $5.5 \cdot 10^{-4}$ is more than an order of magnitude larger than what is typically achieved with inorganic crystals at similar pump parameters.

The compression setup and THz-source described in this thesis is easily implemented and shows long-term stability. Thus, the work presented as part of this PhD thesis can greatly benefit a multitude of applications and propel THz-technology meaningfully out of the laboratories and into the commercial realm, while also providing a powerful tool for fundamental scientific applications.
Terahertz (THz) -stråling kan bruges i et væld af af applikationer lige fra grundlæggende videnskab til kvalitetskontrol på stedet af fabriksprocesser. De elektriske feltstyrker ved stærk THz-stråling gør det muligt at studere grundlæggende egenskaber ved stof, og ultrahurtigt THz-spektroskopi er et lovende værktøj til materialeanalyse og kan anvendes indenfor sikkerhed. Mange almindelige objekter der er uigennemsigtige for optiske frekvenser, er semi-transparente for THz-stråling, og det kan derfor bruges i ikke-invasive billedbehandlingsapplikationer. Oven i dette tilbyder THz-billeddannelse en yderligere fordel i forhold til højfrekvente metoder som røntgenstråler ved at have meget lave foton-energer og således ikke beskadige de materialer, der udsættes for det. Den største ulempe ved THz-stråling er dens stærke absorption af vandmolekyler, som er til stede i mange materialer og prøver, der skal undersøges, og allestedsnærværende i atmosfæren som fugtighed. Derfor kræves en stærk THz-kilde for at tillade, at de ovennævnte applikationer er effektive. I øjeblikket kræver de fleste højeffektive THz genereringsmetoder stærk pumpelysenergi, som typisk er forsynet med store lasers, der fungerer ved lave gentagelseshastigheder. Denne lave gentagelseshastighed påvirker drastisk målingstider, dataopsamling og som en konsekvens signal-støjforholdet, når der kræves hurtige målinger.

For at omgå begge disse problemer giver denne afhandling mulig tilgang til at generere THz-stråling med høj effekt ved MHz-gentagelseshastigheder ved hjælp af et kompakt fiberlaser-system og en simpel ekstern pulskomprimeringsmetode til at tilvejebringe laserpulser med høj spidsseffekt, der er tilstrækkelige til at køre "optical rectification" (OR) i en meget effektiv organisk krytal til frembringe af THz-stråling med forholdsvis stor effektivitet. Strålen fra en nær-infrarød femtosekund ytterbium doteret fiberlaser ledes gennem en polarisationsbevarende fotonisk krytal fiber (LMA-PCF) med et stor mode-område, hvilket inducerer spektral udvidelse fra en fuld bredde halv maksimum (FWHM) båndbredde på 8 nm til en FWHM båndbredde på ~ 100 nm. Det rent polariserede output sendes gennem et par prismer ud af SF-11 glas for at komprimere den spektralt udvidede puls fra 250 fs til 22 fs i FWHM-pulsvarighed. Under hensyntagen til effekttab efter fiberen giver denne metode en næsten ti gange stigning i spidsseffekt til 13,8 MW ved en gentagelseshastighed på 10 MHz. Yderligere skalering af gentagelseshastigheden (og dermed den gennemsnitlige effekt) var mulig, da skadesmekanismen udelukkende var afhængig af spidsseffekt.

22 fs-strålen fokuseres derefter på den organiske krytal HMQ-TMS, som giver en yderst effektiv mulighed for THz-generation gennem OR i en opsætning hvor laserstrålerne deler den optiske vej, hvilket reducerer kompleksiteten sammenlignet med andre effektive metoder til THz-generation, samtlig med at den giver en langt større spektral båndbredde. Båndbredden spaner fra
under 1 THz til over 6 THz med en gennemsnitlig effekt på 1,38 mW med maksimale elektriske feltstyrker, der overstiger 6 kV·cm⁻¹ under standard atmosfæriske forhold for forholdsvis store THz-spotstørrelser på 360 µm i radius ved 1/e²-intensitet og kan således let overstige 20 kV·cm⁻¹ ved strammere fokusering og ved at arbejde i et tørt luftmiljø. Effektiviteten af THz generationen fra optiske bølgelængder er på 5,5·10⁻⁴, hvilket er mere end en størrelsesorden større end hvad der typisk opnås med uorganiske krystaller ved lignende pumpeparametre.

En sådan THz-kilde er let implementeret og viser langvarig stabilitet. Således kan det arbejde, der præsenteres som en del af denne ph.d.-afhandling, være til stor gavn for en lang række applikationer til at drive THz-teknologi meningsfuldt ud af laboratorierne og ind i det kommercielle område, samtidig med at det giver et stærkt værktøj til grundlæggende videnskabelige anwendelser.
Preface

This PhD-thesis, titled *High-power terahertz generation at megahertz repetition rates using few-cycle pulses*, is the summary of my work as a PhD-student at the Department of Photonics Engineering of the Technical University of Denmark. Starting date of the PhD period was the 01. February 2018, running through to the 13. May 2021. The employment was interrupted by an unpaid leave period of two months due to global pandemic related delays with the external research planned at Kyoto University, Japan. Additionally, an extension of six weeks was granted upon request. Throughout the entire period the main supervisor was Peter Uhd Jepsen, with Morten Bache as a co-supervisor until his departure from DTU Fotonik, after which Edmund John Railton Kelleher became co-supervisor in 2019. All three supervisors were from DTU Fotonik.

All experimental results presented in this thesis were obtained at DTU Fotonik, with results obtained at Kyoto University being part of ongoing analysis that was not finished in time for the thesis deadline. Simulations and calculations on pulse compression were also carried out at DTU, while simulations and calculations regarding the THz-radiation were mainly conducted by Dr. Mojca Jazbinsek at the Institute of Computational Physics, Zurich University of Applied Sciences, 8400 Winterthur, Switzerland. Organic crystal design, growth, and preparation was conducted by Prof. O-Pil Kwon from the Department of Molecular Science and Technology, Ajou University, 443-749 Suwon, South Korea in collaboration with Prof. Fabian Rotermund from the Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), 34141 Daejeon, South Korea. High-quality spintronic samples were provided by Prof. Tobias Kampfrath from the Department of Physical Chemistry, Fritz Haber Institute, 14195 Berlin, Germany. All other optics were purchased commercial products or were manufactured at the in-house DTU workshop.

During the PhD, I have received funding twice from *Otto Mønsteds Fond* for conference participation abroad in South Korea and my external research stay at Kyoto University, Japan. Additionally, *P.A. Fiskers Fond* granted me with funds for my external research stay at Kyoto University, Japan. The PhD project was otherwise fully funded by DTU Fotonik.

Tobias Olaf Buchmann
Kongens Lyngby, May 13th, 2021
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CHAPTER 1

Introduction

The Nobel Prize in Physics 2018 was awarded to Arthur Ashkin, Gérard Mourou and Donna Strickland for their groundbreaking inventions in the field of laser physics. Arthur Ashkin’s work on “[
...
] optical tweezers and their application to biological systems [...]” not only sheds light on this very fascinating technology, but also highlights the reach of optical physics across different scientific communities. With the help of such laser-based applications, scientists in chemistry, biology, material sciences and many more fields are now able to investigate the science relevant to them on timescales previously unimaginable - namely in the realm of femtoseconds. A femtosecond (fs) is only a tiny fraction of a second, precisely a one millionth of a billionth of a second which reads as $0.000000000000001$, or as $10^{-15}$. To put this into perspective, a femtosecond corresponds to a second similar to the way a second corresponds to the time passed since a large asteroid led to the extinction of most dinosaurs. It is an incredibly short timescale that allows to observe physical and chemical reactions in real-time, such as molecular bonding, charge transport or phonon propagation. Very much like “the horse in motion” from 1878 by Eadweard Muybridge shown in fig. 1.1, in order to resolve a process in time, one requires a probing frequency - or “camera shutter” - that is at least double the frequency of the process to be observed (also termed the Nyquist criterion).

Figure 1.1. “The horse in motion”¹, by Eadweard Muybridge, 1878.

¹Image taken from https://en.wikipedia.org/wiki/The_Horse_in_Motion - accessed on 14.02.2021
Another great property of ultrashort timescales in combination with laser pulses is the fact that the contained energy is distributed only over an incremental short amount of time. To put this into perspective, a standard nuclear power plant produces an average of 1 gigawatt of power, sufficient to meet the energy needs of three million average German households. A typical laser pulse from a commercial system with a pulse energy of 1 mJ contained within a pulse duration of 100 fs corresponds to a peak power over the duration of the laser pulse of 10 GW. This is ten times the power of the nuclear power plant. This temporal confinement of power into a set of light pulses allows to explore several interesting physical processes such as nonlinear optical phenomena, opening up yet another highly important fundamental and also industrial research path. However, with great power comes great responsibility - especially towards the optical components in one’s system which often cannot handle elevated levels of peak power without degradation or outright destruction. The simple yet genius solution to this problem is why Gérard Mourou and Donna Strickland received their share for the Nobel Prize in Physics 2018, namely for “[...] their method of generating high-intensity, ultrashort optical pulses [...]”^3, better known as chirped pulse amplification. High peak power and chirp (a temporal property of a laser pulse), play an important role in this thesis, and thus my work would not have been possible without the contribution of countless researchers in the field of optics - with or without Nobel prizes - which contributed to the development of high-power ultrashort laser sources. Their combined efforts have played a vital role in a better understanding of our universe and everyday processes, but has by no means reached the end of its possibilities.

Another important factor is the range of frequencies between those that are currently easily accessible and those that are not. A substantial lack of suitable and matured sources limits applications in the frequency range termed the “terahertz-gap”, situated between infrared (IR) radiation below 30 terahertz (THz) and high radio frequencies at several gigahertz, such as the emerging 5G-technology. Within this hard-to-reach frequency band lie several interesting physical properties of importance to a wide range of multidisciplinary sciences, motivating the enhanced emphasis on research in this area since approximately the turn of the millennium. The reason for the lack of adequate THz-sources are manifold, but one of the leading difficulties is the typically low conversion efficiency when generating THz-radiation through the use of optical driving lasers. Supported by the demand for high-power lasers elsewhere, this has led to rapid development of powerful femtosecond lasers bypassing the low efficiencies by simply applying larger power and intensity.

The development of these sources will be briefly outlined in the following chapter before returning to THz-radiation and the great potential of combining the captivating fields of femtosecond physics and THz-generation in chapter 1.2. The introduction is followed by the main part of the thesis which first describes high-power fibre-lasers and external pulse compression in technical detail in part I, followed by its application to generate THz-radiation using an organic crystal in part II. After this, a discussion on the issue of humidity related absorption in THz-applications is presented in chapter 6, and the thesis closes with the conclusion.

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^2https://www.ndr.de/nachrichten/info/Watt-Das-leisten-die-Anlagen-im-Vergleich,watt250.html - accessed on 15.02.2021
1.1 Femtosecond lasers

Starting in 1960, the race to develop Light Amplification by Stimulated Emission of Radiation (LASER) was incredibly intense and close, with Theodor Maiman’s solid-state ruby laser [1], He-Ne gas laser [2], semi-conductor laser [3], and several more variants being discovered and realised [4]. As part of this research field, Snitzer et al. demonstrated clean mode-propagation in optical waveguides in 1959 [5] and realised the first fibre-laser in 1961 [6], followed up by the first fibre-laser amplifier in 1964 [7]. Despite its apparent advantages of compactness and easily directed laser output, the initial success of other laser-forms and the limited power options of the fibre-lasers made the technology take a back seat in the laser world. This changed about two decades later in 1985, when Mears et al. [8] doped fibre cores with neodymium, allowing for very low pump-power thresholds of just 0.1 mW to achieve lasing, and taking another large step by introducing erbium-doped fibres offering very high gain [9]. The corresponding group around David N. Payne also suggested various potential applications, including spectroscopy and wavelength division multiplexing for telecommunication due to the tunability of such laser sources. In the wake of this discovery, the suitability of erbium amplifiers for telecommunications was cemented by their low cross-talk and broad bandwidth required for wavelength division multiplexing. Together with the gain bandwidth of erbium, this was an important contributing factor to shift the typical operating wavelength for telecommunications applications to the so called “C-band” between 1530–1565 nm at which is sits today. It was also a University of Southampton based group that (re-)discovered the potential of ytterbium as a promising dopant for fibre-lasers [10] in 1995 due to its useful gain and emission bandwidths in the near infrared. These properties have made ytterbium one of the main dopants for near-IR high-power femtosecond fibre-based laser systems commonly used today. Initial issues about beam quality and thermal heating have subsequently been circumvented through continued development and engineering efforts, ultimately resulting in fibre-lasers using photonic crystal fibres or fibre-rods as the main amplification stage. This development has allowed contemporary diode-pumped ytterbium fibre-lasers to typically reach wall-plug efficiencies of $\sim30\%$ [4], with record efficiencies of over 50 $\%$ [11] - a huge step compared to the 2 $\%$ which were considered state-of-the-art in 1988 [12].

After establishing reliable laser systems, the focus shifted towards emitting shorter pulse durations. Ultrashort laser pulses in the pico- or femtosecond realm are typically achieved through a method termed mode-locking, where a large amount of light modes propagating in a cavity are phase-locked to each other by using active or passive mode-locking techniques. One example is Kerr-lens mode locking for spatial control through an aperture. Depending on the gain bandwidth, laser cavities can support few to many thousand modes, as long as the cavity length is an integer multiple of half a wavelength. The idea behind mode-locking is to suppress transmission of low intensity light and allow it for high intensity light, which occurs when the multiple modes are phase matched. This can be achieved actively or passively, with passive modulators having faster reactions times and thus lead to shorter output pulses, for example semiconductor saturable absorbers. They act as a high absorption device for low intensities, but once the absorption reaches saturation, they can be seen as a transmission device. This way, the cavity and modulator will initially select a strong enough set of random photon bursts from the gain
medium to propagate in the cavity, undergoing another round trip through the gain medium and being amplified even further, strengthening its position in time vs. other photons until steady state operation is reached. This set of photons now travels periodically through the cavity and has the involved modes phase-locked to each other, with the pulse duration being shorter for more modes being in phase. The pulse duration is also dependent on the recovery time of the absorber - the shorter, the better. As one cavity end mirror is partially transmissive, a set of pulses with a repetition rate corresponding to the round-trip time of the cavity is emitted. Initially, dye-lasers were used to produce ultrashort pulses [13], but in the early 1980’s, solid-state systems based on titanium-sapphire crystals replaced the dye lasers as they offered a more stable and practical solution to generate femtosecond pulses [14] and are still a widely used laser system today. Since then, other options to generate femtosecond pulses have emerged such as fibre-lasers or semiconductor lasers.

For a long time, the laser was initially a “solution looking for a problem”, but those problems quickly found themselves. Interest grew enormously across several fields, spanning from material processing, defence applications, dermatology, ophthalmology, communications, entertainment, and many more. This interest was a strong driving force for the rapid development of lasers, both continuous wave (cw) and pulsed versions, with pulse durations covering nano- to femtoseconds, depending on the application.

At DTU Fotonik, several research groups are working on different topics, not only on ultrafast lasers, e.g. telecommunications and optical sensing devices. As part of the interdisciplinary approach to connect the scientific research of the various research groups, femtosecond lasers were used to solve some difficulties that two of my colleagues came across. To give the reader an impression of the applicability of femtosecond lasers, these experimental excursions during the PhD-duration will be discussed henceforth. In both applications, high-power, ultrafast lasers were used to ablate material of optical fibres in a desired pattern with a fairly straightforward setup. Image a) in fig. 1.2 shows the microscopic side-view of an optical fibre with a conical hole drilled into the facet on the right hand side with a diameter of 40 µm. What is shown here is a two-mode step-index fibre which is intended to be spliced to a photonic lantern allowing for spatial division multiplexing [15] - a technology that allows to increase existing data-transmission rates by the number of spatial modes that can be separated. Due to the lantern’s end facet being far smaller than the core diameter of the step-index fibre, splicing the two together turned out to be rather difficult. The required force to push the two fibres together during the splice would displace the smaller fibre from the initially intended position. By drilling a conical hole into the facet, the smaller fibre would thus be wedged into position, allowing for the splicing process to proceed as intended⁴. Here, femtosecond lasers offer an option to act as a drill on such small spatial dimensions with zero mechanical force applied to the involved objects, increasing drilling precision. By altering the characteristics of the laser beam, namely the power, illumination period, and spot size, the depth and size of the cone can be custom modelled. Following a similar approach, image b) in fig. 1.2 shows the result of micromachining the side of a hollow-core fibre used in optical sensing, with the dark spots in the centre of the fibre being laser-drilled holes. In this specific

⁴Results on the transmission effects of the machining and thus the feasibility of this method are yet to be published
case, any gas filling the inside of the hollow-core fibre will alter the transmission properties, allowing to be cross-referenced to existing databases and thus sensing specific chemical compounds. For practical reasons, the sensing-active hollow-core optical fibre is spliced to a solid-core fibre, which means that there are no opportunities for gas to enter or exit the hollow-core fibre from the end-facets. The laser-induced holes on the side of the hollow-core fibre allow interaction with any outside gas and thereby making them applicable to localised sensing applications. Furthermore, the transmission properties of the stretch of fibre that has undergone this treatment is not significantly reduced, allowing for the high-quality optical sensing application. In further experiments, even large removal of cladding material showed no significant decrease in guidance properties, which is why the initial process of laser ablation was changed to focussed ion-beam milling for the final application [16], allowing for larger holes and better transition of fluids or gases into the fibre core.

Figure 1.2. a) Microscope side-view of an optical fibre after laser-drilling a hole into the fibre facet, visible as a black groove. b) Microscope image showing the top-view of a fibre after drilling holes (black dots) into the cladding. The positional displacement of the holes result from stage misalignment. c) Close-up side-view of larger holes in the same fibre.
1.2 Terahertz radiation

Definitions of the onset and end of the "THz-gap" in the electromagnetic spectrum differ, but can generally be attributed to the frequency range between 0.1-30 THz, despite the lower boundary overlapping with existing microwave technology and the upper boundary in the mid-IR competing with existing optical techniques, especially at 30 THz, which corresponds to a wavelength of roughly 10 µm - very close to the emission wavelength of CO₂-lasers. Nonetheless, a strong focus lies on the range between 0.3-10 THz (or a wavelength of 1-0.03 mm), where a plethora of applications have been envisioned, with some of them already having matured to commercial grade, while others still remain in the realm of science-fiction. The historical difficulty of generating and detecting this "THz-gap" lies with the inherently low efficiencies in generation. It also demands powerful driving sources, accompanied with downsides such as system size, power consumption, power-handling and heat-management, whereas the detection is inhibited by the lack of suitable (semi-conductor-) materials. The typical techniques used in either optical or radio frequency detection cannot simply be extrapolated to the "THz-gap". Extending optical semi-conductor technology for detection and generation will not work due to the very low band gap required for the photon energies in the THz-frequency range. Thermal background radiation at room-temperature would also increase the noise on such materials, making it difficult to have "THz-only" detection without strong cooling of the detectors. Existing infrared detection already has issues with exactly this problem, despite the photon energy being more than an order of magnitude larger than that of THz. Extrapolating existing radio-frequency techniques such as LC-circuits to the THz-region also proves to be a difficult task, as the high frequency oscillations of THz-radiation is often too fast for such circuits and electronic switching speeds. Adding to these detrimental issues, THz-radiation also suffers from strong absorption by water-molecules present in the atmosphere. The corresponding power-loss necessitates highly sensitive detectors that ideally act across a large frequency range and has been a major restraint for THz-radiation to be used in telecommunication applications exceeding a few meters of free-space propagation distance.

These difficulties may have all but terminated the interest in THz-technology, but luckily several scientists across the world took up the challenge to make it work, particularly by using ultrafast lasers for generation. As a consequence though, with the early focus on efficient and reliable THz-generation and detection, the THz-community was more of a scientific niche rather than an enabler of technology. Much of the fundamental work in this regard was conducted by David H. Auston and coworkers. They demonstrated photoconductive antennas in combination with ultrashort lasers to generate and detect THz-radiation in 1988 [17] - a concept they demonstrated for millimetre wave generation a few year earlier in 1983 [18]. Shortly after, Grischkowsky et al. performed terahertz time-domain spectroscopy (TDS) on water vapour [19] and other materials [20], showing the great potential for chemical and molecular spectroscopy. With these convincing demonstrations of the practical applicability of THz-TDS based on ultrashort laser sources, THz-radiation became of interest to a wider audience. This resulted in several practical applications in e.g. telecommunications, molecular spectroscopy and chemical sensing, security and imaging, as well as medicinal diagnostics. As THz-frequencies can transmit through many materials, unlike optical frequencies, non-invasive imaging and monitoring applications helped drive the
continued growth of this technology. Especially since THz-frequency photons are not energetic enough to cause any of the harmful effects in the way that high energy X-rays do. The downside vs. X-ray imaging is the reduced spatial resolution, but in several uses X-ray spatial resolution is not necessary, for example in security-scanners at airports. This real-time imaging technique is also applied in medical applications to detect skin-cancer [21] and shows potential for monitoring layer-thicknesses in several industries where non-destructive measurement methods are currently unavailable. Other than these applications, THz-radiation still plays an important role in basic scientific research, especially due to the strong electric field strengths that can be achieved at these low frequencies, applied for example in THz-scanning tunnelling microscopy [22]. Further examples and historic overview will be given in part II of this thesis starting with chapter 4, but the reader is also referred to reference [23] which gives an overview of current THz-technology as of 2017. What is clear though, is that with time THz-applications will become more widespread as their potential and benefits are obvious. In order to accelerate this process, fundamental research in THz-generation and detection is still required and therefore several institutes worldwide are working to find new ways or to improve existing techniques.

The Ultrafast Infrared and Terahertz Science group at the Technical University of Denmark (DTU) is part of this global effort and has provided me with an outstanding workplace to conduct my PhD. The above listed applications each have an ideal source and detector, but many of them that have not reached commercial or at least widespread scientific breakthrough would greatly benefit from a high-efficiency, high repetition rate, and broad bandwidth THz-source with moderately strong field strengths. Precisely this interest has motivated the research outlined in this thesis, which tries to provide for a stable THz-source that can satisfy the desire for these parameters. Hence, a promising technique to acquire such a source, relying on the transformation of highly energetic laser pulses at optical frequencies to THz-frequencies, was applied and optimised over the last three years. The results described in this thesis hopefully allow fellow research groups to apply similar techniques at their respective facilities. The resulting increase in THz-related research will then ideally excel this exciting technology well beyond scientific and isolated industrial applications in the same way that optical lasers and diodes have transformed our world in the past half century.

This thesis is split into two main parts which both can be read and understood individually. The first part introduces the concept of external pulse compression and the experimental results obtained from applying them to a high-power fibre-laser in chapters 2 and 3. Following this, the second part focuses on THz-generation techniques and detection in chapter 4 as well as the experimental results (chapter 5) achieved by pumping an organic crystal with the compressed output of the laser from part one. It finishes with a detailed analysis of humidity-related absorption of THz-radiation in air in chapter 6.
Part I

Nonlinear pulse compression
2.1 Optical fibres and fibre-lasers

The desire to guide and control light is perhaps as old as the very first artificial mirror, but it did require a few thousand years before documented scientific concepts became advanced enough to understand the physics of guiding light. Most important was the description of refraction and reflection of light in the 19th century by Colladon and Babinet [24] (despite much older similar findings in ancient Greece and the middle-east). After another 100 years of experimental process in optics, the first modern-type optical fibre was demonstrated and patented [25] - just a few years after the invention of the laser. The principle used at the time - total internal reflection (TIR) - is still the most common method for optical wave-guiding used in telecommunications. TIR is easily understood by following Snell’s law, which describes the propagation of a ray when passing from one medium with refractive index \( n_1 \) into another with refractive index \( n_2 \). This incident angle on the layer boundary is \( \Theta_1 \), defined with respect to the boundary surface normal, with \( \Theta_2 \) the angle after the boundary. The relation of these parameters is shown in eq. (2.1),

\[
\frac{n_2}{n_1} = \frac{\sin(\Theta_1)}{\sin(\Theta_2)}. \tag{2.1}
\]

For the case that a light ray initially travels within the higher refractive index material with \( n_1 \) and is then incident upon the boundary layer with the lower refractive index material with \( n_2 \) - i.e. \( n_2 < n_1 \) - at an angle \( \Theta_1 \), the propagation angle required for \( \Theta_1 \) in order to achieve a given output angle \( \Theta_2 \) within medium \( n_2 \) can be calculated by rearranging eq. (2.1) to:

\[
\Theta_1 = \arcsin \left( \frac{\sin(\Theta_2) n_2}{n_1} \right). \tag{2.2}
\]

Now the requirement can be set that \( \Theta_2 \) is perpendicular to the normal, i.e. \( 90^\circ \), which will set the sinus-term in eq. (2.2) to 1. The input angle \( \Theta_1 \) that is calculated from the remaining \( \frac{n_2}{n_1} \) term is also called the critical angle. Any input angle larger than the critical angle will lead to total internal reflection of the beam at the boundary layer, so that the light-ray is confined within the first medium, for example in a circular fibre-geometry. This is the basic principle behind step-index optical fibres, which are commonly used for single-mode operation, and hence termed single-mode fibres (SMF). Typically, fused silica - commonly known as glass - is used to manufacture these fibres, where the core region is doped with certain elements to increase the refractive index with respect to the un-doped outer cladding layer. Another option is to engineer a gradual decrease
of the refractive index, leading to graded-index fibres, commonly used for multi-mode operation. To address requirements that standard step-index fibres cannot satisfy, several other waveguides have been developed. These include hollow-core fibres operating on anti-resonant guidance or photonic band gap guidance [26] (in both cases the core refractive index is in fact lower than that of the cladding), and also more exotic twist-induced guiding which requires no classic core-cladding structure [27].

For the purpose of this thesis, the most important additional fibre-design is the photonic-crystal-fibre (PCF), demonstrated by J.C. Knight et al. at the University of Southampton in 1996 [28], which allows single-mode operation at large core sizes [29]. This is important because the intensity within a fibre core scales inversely quadratic with increasing core size, and in a standard SMF the core size of only a few µm limits the operational average power one can propagate through it. Contrary to this, PCFs allow for large throughput powers (while maintaining single-mode guidance) due to the larger core size and mode-field diameter (MFD) of the guided beam. To understand why this is so, the physical principles of PCFs will be elaborated on in more detail. Figure 2.1a) shows the front-facet of the aforementioned PCF with the core region surrounded by a series or regular spaced black dots. In fig. 2.1b) the far-field pattern of the guided light is shown, exhibiting a hexagonal pattern as some of the light squeezes through the gaps between the holes (both images in fig. 2.1 are taken from [30]).

The black dots are in fact air-filled capillaries, or "holes", which have the refractive index of air (∼1). The air-encircled solid-core material will have a higher refractive index, leading to a quasi-step-index structure similar to a SMF. The resulting large refractive index difference allows smaller critical angles to suffice for (modified) TIR, which is in turn beneficial for guiding optical rays.

In both cases, SMF and PCF, the modes guided by a waveguide are the electric-fields that can be determined by solving the resulting wave-equation derived from Maxwell’s equations after applying typical physical restraints and simplifications for optical fibres. Additionally, in the general case of an optical fibre, low-loss and spherical independence of the refractive index both
in the core and cladding can be assumed, which simplifies the wave-equation to the Helmholtz-equation,
\[ \nabla E(r, \omega) + n^2(\omega) \frac{\omega^2}{c^2} E(r, \omega) = 0, \] (2.3)
with the radial electric field \( E(r, \omega) \) in the frequency domain, frequency dependent refractive index \( n(\omega) \), angular frequency \( \omega \), and the speed of light in vacuum \( c \). Equation (2.3), converted to cylindrical coordinates, can be solved with the help of Bessel functions. A single-mode guiding fibre therefore is defined by a single solution for eq. (2.3). Since optical fibres are dependent on their specific parameters such as refractive indices and core radius \( r_c \), an additional parameter termed the \( V \)-number is introduced. It is obtained by setting the Bessel function for the cladding region to 0 - i.e. the mode is only supported in the core and does not radiate and decay into the cladding - and solving the remaining equation. This leads to the expression for the \( V \)-number:
\[ V = \frac{2\pi f r_c}{c} \sqrt{n_c^2 - n_{cl}^2} = \frac{2\pi f r_c}{c} NA, \] (2.4)
with \( f = \frac{\omega}{2\pi} \) the optical frequency in [Hz], \( r_c \) the core radius in [m], \( n_{cl} \) the cladding refractive index, and \( n_c \) the core refractive index. The square-root term is also the definition of the numerical aperture \( NA \) of an optical fibre. The \( V \)-number allows for determining the (integer) number of modes a given fibre can guide (through \( V^2/2 \) for step-index fibres and large \( V \) [31]). For single-mode guidance, only the fundamental mode is allowed to propagate through the fibre, meaning that the next higher mode (LP\(_{11}\)) must not be able to propagate. This requirement allows for calculating the necessary \( V \)-number so that the cut-off frequency for a given waveguide lies below the desired single-mode frequency by using eq. (2.5). This yields an upper limit of \( V \approx 2.405 \) until which single-mode guidance occurs at a given frequency. For larger \( V \)-numbers, the next order mode can propagate and the fibre is a multi-mode fibre. Since the polarisation direction counts separately, the integer "number of modes" for single-mode guidance is in fact two. Hence, according to eq. (2.4), to maintain a low \( V \)-number for a given frequency, one requires either a smaller index difference between core and cladding (imposing restrictions on the accepted angles for TIR according to eq. (2.2)), and/or one must maintain a small core-radius \( r_c \). For example, a typical SMF (e.g. Thorlabs SM980G80\(^1\)) will have \( n_c = 1.46058 \) and \( n_{cl} = 1.45068 \). When setting the condition of \( V < 2.405 \) for a given frequency \( f = 305.91 \) THz (equivalent to a wavelength of 980 nm), the core radius must be no larger than \( \sim 3.3 \) µm in order to exhibit single-mode guidance in a SMF.

In contrast to this, the \( V \)-number for a PCF does not depend on the core radius but rather on the spacing between the air-holes surrounding the core [29,32], termed the pitch \( \Lambda \), leading to the expression
\[ V = \frac{2\pi f \Lambda}{c} \sqrt{n_c^2 - n_{2\text{air}}^2} = \frac{2\pi \Lambda}{\lambda} NA. \] (2.5)
The equation shows that for a given \( NA \), the \( V \)-number depends on the ratio between the pitch and the wavelength of interest. In fact, with an effective index approximation for the cladding structure, the \( NA \) can also be modified by changing the ratio of the pitch and the hole-size (i.e.

\( ^1 \)https://www.thorlabs.com/thorproduct.cfm?partnumber=SM980G80 - accessed 15.03.2021
the air filling fraction). Due to the changes in geometry and guidance, the SMF value for single-mode operation of $V < 2.405$ no longer holds. Detailed work by Mortensen et al. has derived a value for multi-mode cut-off in PCFs at $V = \pi$ [32]. Since the pitch $\Lambda$ is a manufacturing parameter and the core-radius is omitted from eq. (2.5), one can manufacture PCFs with large core-sizes while still maintaining single-mode guidance at desired frequencies. To better understand the guidance, one can imagine the holes as bars and the pitch as the distance between the bars, similar to a prison cell. Any mode within this prison cell that can slip through the bars will escape into the cladding, while the larger fundamental mode is trapped inside the core.

The manufacturing freedom-of-choice is also where the real advantage of PCFs lies, since it can specifically be tuned to guide wavelengths at desired characteristics by varying the pitch and the hole-size. By altering these parameters one can tune the cut-off frequency for single-mode guidance and the zero-dispersion wavelength, which offers a great advantage over classical SMFs. The meaning of these properties will be explained in detail in the following chapter 2.2 along with other critical fibre parameters. Further tuneability is achieved by adding specific dopants to the core, allowing to change for example the nonlinear refractive index, and by adding so termed “stress-rods”, which change the structural birefringence in such a way that polarisation maintaining (PM) propagation is enabled. Such a large mode area (LMA), PM-PCF is the fibre used for spectral broadening in this thesis, the LMA-40-PM-PCF (fibre under development), and is depicted in fig. 2.2, where the stress-rods are identified as the darker vertically parallel shaded structures on either side of the core.

![Figure 2.2](image-url)

Figure 2.2. a) SEM-image of the core-region of the LMA-40-PM-PCF showing the hole structure up close. The stress-rods are visible as vertically shaded areas beside the core. b) Microscope image of the LMA-40 front facet. The polarisation maintaining stress rods are the dark areas next to the core.
2.2 Fibre properties

In the previous chapter, fibre-specific parameters such as the zero-dispersion wavelength, were briefly touched upon but not given a detailed explanation. This chapter will give a more thorough overview of the most important fibre properties, as listed below:

- Dispersion and the nonlinear refractive index
- Fibre losses and the numerical aperture
- Nonlinear fibre parameters

2.2.1 Dispersion

A well-known phenomenon of dispersion is the breaking of white light into its spectral colours when passing through a prism, termed angular dispersion. The reason for this effect is that the different frequencies of light bundled together in white light experience different refractive indices within the prism, and thus different output angles according to eq. (2.1). In a simplified picture on an atomic level, the electrons will respond to a frequency-dependent electromagnetic field applied to them. This response is governed by several factors, but the impact of the incident electromagnetic field is determined by its strength and its (angular)\(^2\) frequency \(\omega\), which is thereby changed for light at different frequencies. To a first degree, the overall response of a material can be expressed through its refractive index \(n\), which as a consequence is therefore frequency dependent, i.e. \(n(\omega)\). This frequency dependence is termed chromatic dispersion, and has a profound impact on the group- and phase velocity of light within a given medium, changing the propagation velocity according to

\[
v(\omega) = \frac{c}{n(\omega)}.
\]

The typical frequencies of visible or infrared light are far from medium resonance frequencies. This has led to the empirical expression of the Sellmeier equation\(^{[33,34]}\), allowing to model the refractive index as a function of frequency (or rather the vacuum wavelength \(\lambda\)) according to

\[
n(\lambda) = \sqrt{1 + \sum_i A_i \lambda^2 \lambda^2 - B_i}.
\] (2.6)

Equation (2.6) is valid for materials with negligible absorption at the wavelength region of interest and depends on the experimentally determined and material specific coefficients \(A_i\) and \(B_i\). With the help of the Sellmeier equation one can determine the group velocity \(v_g\) and group refractive index \(n_g\) through

\[
\frac{1}{v_g(\omega)} = \frac{n_g(\omega)}{c} = \frac{n(\omega) + \omega \frac{\partial n(\omega)}{\partial \omega}}{c},
\] (2.7)

which governs the velocity of the envelope of a pulse propagating in a medium. In fibre-optics, the mode-propagation constant \(\beta\), defined as the change of the wavenumber \(k\) with frequency

\(^{2}\)For brevity, the term "angular" is often dropped in this thesis, but \(\omega\) always refers to the angular frequency, while \(f\) refers to normal frequency.
\[ \beta_i = \frac{\partial^i}{\partial \omega^i} \] has become a useful way to describe the effects of dispersion by mathematically expanding it into a Taylor-series at the centre-frequency \( \omega_0 \) of a spectrum according to [35]:

\[ \beta(\omega) = \beta_0 + \beta_1(\omega - \omega_0) + \frac{1}{2}\beta_2(\omega - \omega_0)^2 + \frac{1}{3!}\beta_3(\omega - \omega_0)^3 + \cdots . \] (2.8)

It contains important parameters for the understanding of propagation in fibre-optics. The first term, \( \beta_0 \), describes a simple overall phase shift within the envelope. The second term, \( \beta_1 \) in units of \([s/m]\), is the same as \( \frac{1}{v_g(\omega)} \) in eq. (2.7) (i.e. the inverse group velocity) and describes the resulting time-delay of an optical pulse envelope propagating through a specific medium. This is relevant for example in telecommunications applications to determine (part of) the latency of a signal sent between two points. Given the small lengths of fibres used and the general irrelevance of arrival time in this thesis, \( \beta_1 \) plays no significant role per se. The next higher-order term is \( \beta_2 \) in units of \([s^2/m]\), the third term in the Taylor series, which is also termed the Group Velocity Dispersion (GVD), and perhaps the most important fibre-parameter for this thesis. It describes, as the name says, the change of the group velocity, and can be determined through the refractive index according to:

\[ \beta_2 = \frac{1}{c} \left( 2 \frac{\partial n}{\partial \omega} + \omega \frac{\partial^2 n}{\partial \omega^2} \right). \] (2.9)

For fused silica - the most common optical fibre material - the GVD crosses zero at 1273 nm, which determines the zero-dispersion wavelength \( \lambda_{ZDW} \), as shown in blue in fig. 2.3. For wavelengths shorter than \( \lambda_{ZDW} \), the group velocity decreases with shorter wavelengths (due to an increasing group index), leading to a positive GVD parameter. This means that longer wavelengths will experience a lower refractive index and thus travel faster than the shorter wavelengths in the same medium. Historically, this has been termed as normal dispersion, while anomalous dispersion exhibits the opposite behaviour, with group velocity decreasing with longer wavelengths, slowing down longer wavelengths with respect to shorter ones. Anomalous dispersion is detrimental for physics involving solitons, however, for the scope of this thesis, the relevant dispersion regime lies in the normal region, meaning that solitons and other effects arising from anomalous dispersion do not play any role in the further analysis.

As mentioned before, the \( \lambda_{ZDW} \) of PCFs can be tuned largely at will, and thus PCFs find plenty of applications in telecommunications where operation at \( \lambda_{ZDW} \) is desired to maintain the temporal structure of a pulse. The reason for this is that the spectral components of a pulse propagating far away from \( \lambda_{ZDW} \) will experience different group velocity, and thus the pulse will be more strongly stretched in time. Despite this effect of pulse broadening being a problem for telecommunications, it does offer several potential benefits for other physical applications, such as external pulse compression, which will be explained in detail in chapter 3.3 and 2.4.

When operating with ultrashort pulses, the bandwidths tend to be large, so that higher-order dispersion parameters from the Taylor expansion also need to be taken into account, such as \( \beta_3 \) in eq. (2.8), also termed Third-Order-Dispersion (TOD), shown in red for fused silica in fig. 2.3. The effects of such higher-order dispersion will be discussed in more detail in chapter 3.4.
2.2 Fibre properties

Figure 2.3. GVD of fused silica (blue) as a function of wavelength. For wavelengths below the zero-dispersion wavelength at $\sim 1273$ nm, $\beta_2$ is positive, i.e. normally dispersive, meaning that shorter wavelengths travel slower than longer wavelengths. The third order dispersion (red) increases strongly for longer wavelengths.

As indicated, the dispersion parameter is highly important to understand the development and propagation of optical pulses in a waveguide such as an optical fibre. In general, the dispersion of a waveguide is made up out of several distinct contributions, listed and explained in regard to large core PCFs as follows:

- Modal dispersion:

  Each solution to the wave-equation is a modal distribution of the electric field that can propagate through a waveguide. This is more profound in step-index multi-mode fibres than in graded-index fibres, and can be figuratively explained by considering the range of permitted angles for TIR. Some rays will propagate a more direct path through the fibre at shallow angles, while others propagating at a steeper angle will essentially travel a farther total distance through the fibre, delaying their arrival and separating them from the faster modes. Since the fibres used in this thesis are single-mode PCFs, modal dispersion is not present and therefore can be neglected.

- Material dispersion:

  Each material has its specific dispersion profile which determines the frequency-dependent refractive index. In the case of bulk material, this is by far the most dominant contributing effect. For waveguides it still plays the most important role and can be tailored to suit specific needs, for instance through careful doping with other elements. This is often applied to optical fibres, where the core material, typically fused silica, is doped with specific elements such as germanium or fluoride in order to change the refractive index according to
application requirements. Material dispersion generally controls the dispersion parameter for silica-based LMA-PCFs, such as the fibre used in this thesis.

- **Waveguide dispersion:**
  The shape of a waveguide also has an effect on the dispersion, since the propagated field experiences a group index that is not entirely defined by the core, but to an extent also by the cladding. This is due to the radial distribution of the core-light, which slightly protrudes into the cladding. It is most important for very small core radii that approach the dimension of the guided wavelength. In contrast to this, as the waveguide mode field diameter becomes larger, the effects of the waveguide dispersion reduce, leaving material dispersion to dominate. To illustrate this, fig. 2.4 shows the GVD for two LMA-PCFs with core diameters of 10 and 20 µm respectively, as well as that for bulk fused silica. For an increased core size there is an approach to the curve of the pure fused-silica material values, meaning that waveguide dispersion becomes less relevant for large core PCFs, as also stated by Seidel et al. [36]. The fibre used in this thesis has a large enough core size to assume an insignificant impact of waveguide dispersion.

- **Polarisation mode dispersion:**
  A linearly polarised field launched into an optical fibre will couple to its orthogonal polarisation state. This is due to the nature of real-life applications not exactly matching theoretical predictions. A real-world centrosymmetric material will show deviations from the theoretical symmetry due to impurities, manufacturing imperfections, and induced micro-stress. All of these induce some level of birefringence, allowing for polarisation crosstalk to take place. As a result of the changes in guidance for different polarisation modes the pulse broadens during propagation. A polarisation maintaining fibre prevents this by specifically inducing a desired birefringence in the fibre. This prevents crosstalk to amplify due to a periodic change of the phase applied to the undesired polarisation state, leading to its cancellation. Such birefringence can be induced through stress-rods as seen in fig. 2.2. With the LMA-40 being a polarisation maintaining fibre, polarisation mode dispersion can be disregarded for the purpose of this thesis.

- **Nonlinear dispersion:**
  At sufficiently high optical intensities $I \sim 10^{20} \text{ W} \cdot \text{m}^{-2}$ [37], a given material’s refractive index will be noticeably altered by its nonlinear index $n_{nl}$ according to [35]:
  \[
  n(\omega, I) = n(\omega) + n_{nl}(\omega)I. \tag{2.10}
  \]
  The reason for this is the induced dipole-moment (polarisation) not behaving linearly with the applied electric field (hence: nonlinear), since the field strengths are sufficient enough to distort and control the electron movements in a non-harmonic fashion. In the case of extreme field strengths, ionisation of the atoms is also possible, which is relevant for a particular THz-generation technique in a gaseous medium described in chapter 4.2, while such intensities in a solid-core fibre lead to laser induced damage. As the material properties are changed on an atomic scale due to the strong displacement of the electrons, the propagation parameters also differ compared to the low-intensity, harmonic regime. The effect
induced by the electric field therefore changes the refractive index and thus the group index, leading to additional dispersion effects. For optical fibres, where the core materials used have inversion symmetry (so their second-order susceptibility becomes zero\(^3\)), the nonlinear index \(n_{nl}\) (assuming a linear polarised field) is related to the third-order susceptibility \(\chi^{(3)}\) according to [35]:

\[
n_{nl} = \frac{3}{8n} \text{Re} \left( \chi_{xxx}^{(3)} \right),
\]

(2.11)

with \(\text{Re}\) indicating the real part of a complex number. The resulting index changes allow for several nonlinear effects, the most thesis-relevant of which are explained in chapter 2.3. The third-order susceptibility is a material specific property, and following the above argumentation, the only two relevant dispersion contributions for the PCFs used in this thesis are thus resulting from the core-material.

The main effect of the dispersion on femtosecond pulses comes from the GVD defined in eq. (2.9) which has a strong impact on the pulse duration. For a pulse with limited bandwidth, i.e. when the refractive index does not change significantly over the frequency range, the group velocity can be assumed as constant over the entire spectral bandwidth. As a consequence, its dispersion (GVD) is also assumed as constant, i.e. having no impact on the pulse. For pico- and especially femtosecond pulses, this assumption no longer holds, since the refractive index change across the optical bandwidth is large enough to have an impact, leading to lower frequencies travelling faster than higher frequencies in the same medium (normal dispersion regime). This will lead to one of three effects depending on the initial chirp (i.e. the distribution of the pulse’s frequency components over time) conditions of the pulse:

- **Unchirped pulse**: All frequency components are overlapping in time.
- **Positively chirped pulse**: Lower frequency components are leading ahead of higher frequency components in time.
- **Negatively chirped pulse**: Lower frequency components are trailing higher frequency components in time.

In the case of initially unchirped pulses, GVD in the normal dispersion regime results in temporal stretching of the pulse, as higher frequency components experience a larger refractive index and thus travel more slowly than lower frequency components. The output pulse duration at full-width half-maximum (FWHM) \(\tau_{out}\) on an initially unchirped (Gaussian) pulse of duration \(\tau_{in}\) that propagates through a material of length \(z\) can be calculated through [39]:

\[
\tau_{out} = \tau_{in} \sqrt{1 + \frac{(4\ln(2)\beta_2(\omega_0)z)^2}{\tau_{in}^4}}.
\]

(2.12)

\(^3\)This assumption is generally valid. However, at extreme intensities, second-order effects can be observed as explained in [38].
The result of the GVD multiplied with the length of a material is the group-delay dispersion (GDD) - a specific amount of dispersion for any given material of a certain thickness, with its units in \( [s^2] \). Similar mathematical relations as in eq. (2.12) are also derived for pre-chirped Gaussian pulses and other pulse shapes in [35], while including higher-order dispersion parameters. The applications of eq. (2.12) will be addressed in chapter 2.4.

A positively chirped pulse will accumulate additional positive chirp when propagating through a normally dispersive medium, which in turn will result in an increased temporal stretch of the pulse. For negatively chirped pulses propagating in a normally dispersive medium, the pulse will initially compress to an unchirped state (assuming a linear chirp profile), after which it will follow the positive chirp accumulation described above. This phenomenon is often used to increase the efficiency of nonlinear actions, since it allows the pulse’s intensity to remain high over a longer propagation path before dropping due to the temporal elongating of the pulse [40]. GVD in a long enough section of optical fibre will therefore eventually lead to a (linearly) stretched output pulse for any (linearly) chirped input state. The temporal stretching process is reversible when applying the exact amount of opposite sign GDD acquired by the pulse.

Figure 2.4. GVD of the PCFs LMA10 (blue) and LMA20 (red), and of fused silica (green). The dispersion parameter \( \beta_2 \) of large core PCFs based on fused silica will behave increasingly like that of bulk fused silica for increasing core size\(^4\).

2.2 Fibre properties

2.2.2 Fibre losses

The accumulated contribution of factors reducing the optical power within an optical fibre is
collectivised in the attenuation coefficient $\alpha$, which reduces the initial power $P_0$ after a given
propagation length $L$ (or material thickness) to $P(L)$ according to:

$$P(L) = P_0 e^{-\alpha L}.$$ (2.13)

This exponential behaviour for the intensity-loss of light through a medium is commonly known
as the Beer-Lambert law and is the result of century-spanning efforts by scientists Bouguer, Lam-
bert, and Beer [41]. The absorption coefficient, also called attenuation coefficient, has units of
$m^{-1}$ and typically is frequency dependent. The power diminishing factors are dominated by
material absorption (especially at resonance frequencies of specific atoms and molecules) and
Rayleigh scattering, with confinement losses following up in the case of optical fibres. It is there-
fore to no surprise that the common telecommunication wavelengths lie at the lowest loss area
for fused silica, and that the loss ultimately determines the operating frequencies used with opti-
cal fibres. For the most common fibre material, fused silica, the low-loss window is framed by the
electronic resonances at ultraviolet wavelengths below $\sim 0.2 \mu m$ and vibrational resonances at
infrared wavelengths exceeding $2 \mu m$ [42]. Rayleigh scattering results from density variations in
the core material and sets a fundamental intrinsic loss level, even at frequencies not coinciding
with resonances [35].

The "effective length" $L_{\text{eff}}$ of a fibre is determined by the attenuation coefficient and is com-
monly used in analysis and equations when working with long or high-loss optical fibres, according
to [35]:

$$L_{\text{eff}} = \frac{1 - e^{-\alpha L}}{\alpha}.$$ (2.14)

For low-loss or short fibres, the effective length very closely matches the real fibre length $L$. In this
thesis, the longest length of PCF used experimentally was 0.25 m, and typical PCF-attenuation
coefficients lie below $1.1513 \times 10^{-3} m^{-1}$ ($< 5 \text{ dB-km}^{-1}$) at $\sim 1030 \text{ nm}$ [43]. At such short distances,
the loss corresponds to 0.000125 dB (or $\sim 0.029 \%$), which can be considered negligible and hence
further analysis assumes loss-less propagation within the PCF once the light has been coupled to
the core. Another important factor regarding the loss in fibres is known as bending-loss. This loss-
mechanism originates from increased confinement loss due to bending of a fibre, which leads
to increased coupling from the core to the cladding, as well as the consequently induced stress
leading to polarisation mode losses. The PCF used in this thesis has a fairly stiff nature, but can be
coiled with a large enough radius. However, as the fibres used are fairly short (<20 cm), they are
not bent or coiled, which eliminates any source of bending-loss.

The above statements eliminate the most relevant loss-sources for fibre-propagation, but there
are still other loss-mechanisms that need to be considered, most importantly coupling loss

\[^5\]This value is also specified by suppliers of LMA-PCFs, e.g. https://www.nktphotronics.com/lasers-fibers/product/large-mode-area-photonic-crystal-fibers/ , or https://www.thorlabs.com/newgrouppage9.cfm?objectgroup_id=1902, accessed 12.03.2021. Conversion from dB·m$^{-1}$ to $m^{-1}$ through $\alpha_{\text{dB}} = 4.343\alpha$ [35]
and micro-stress loss. Micro-stress is induced e.g. by clamping the fibre in place to maintain its position, which asserts pressure onto the fibre at the point of clamping, marginally changing the fibre properties at that point. This change is large enough though to enhance confinement loss and thus mode-coupling into the cladding, and also acts as a fibre-fuse initiation point (discussed in more detail in chapter 3.5). The largest loss-component with most optical fibres is related to medium interfaces such as air-fibre or fibre-fibre at coupling junctions or splices. Due to the refractive index change from air to glass, reflection losses at the interface are not avoidable. For fused silica, with a refractive index of \( n_2 = 1.45 \) at 1035 nm [44] and P-polarised light, the reflected portion of light coming from air at \( n_1 = 1.00027 \) can be calculated with the help of the Fresnel equations as

\[
R_P = \frac{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\alpha_i) \right)^2} \cos(\alpha_i)}{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\alpha_i) \right)^2} \cos(\alpha_i) + n_2 \cos(\alpha_i)}.
\] (2.15)

The above values yield a result of 3.37 % for fused silica that is reflected at normal incidence \( \alpha_i = 0 \). Inside the scope of this thesis, coupling loss from free-space into the fibre results in the largest loss contribution, with the specific values being discussed in chapter 3.4. The reason for this can be understood when recalling the condition for TIR from chapter 2.1. In a simple ray-angle picture, any ray that enters the fibre at an angle larger than the critical angle (or acceptance angle) is not totally internally reflected and is subsequently lost into the cladding. The acceptance angle for the PCF used in this thesis can be determined analogously to a SMF through the numerical aperture \( NA \) (though the \( NA \) of PCFs in general is not universally defined), or by measuring the divergence angle \( \theta \) of the beam after the fibre output in the far-field. The divergence angle in this thesis is defined as the half-angle of the total beam-cone divergence. This leads to the case that if the \( NA \) is small, the acceptance angle is also narrow. Thus, as a general rule for launching from free-space into optical fibres, the divergence angle (in radians) of the beam should not exceed the \( NA \) of the optical fibre.

For a better understanding, the MFD of an optical fibre is introduced. It is defined in analogy to the beam diameter of a Gaussian beam in free-space where the intensity of the beam drops to \( 1/e^2 \), and holds for single-mode fibres. To achieve optimal coupling one therefore should try to match the Gaussian mode profile at the focussed beam-waist of the input beam with the propagation MFD of the fibre-core. In an ideal situation, only the propagation mode is excited at the input facet, with the MFD matching that of the fibre so that the beam is immediately guided as intended and loss due to cladding-coupling is minimised. In reality however, this is sometimes not as easy to achieve as one might think, and substantial coupling losses can occur. This is exacerbated by the fact that the \( NA \) and diffraction limited spot size \( r_{min} \) of a Gaussian beam counteract each other, with the \( NA \) increasing for smaller spot sizes, according to:

\[
NA = M^2 \frac{\lambda}{\pi r_{min}},
\] (2.16)

with \( M^2 \) being the beam quality factor, often called \( M \)-squared. By defining the \( NA \) of a free-space beam as \( NA = n \sin(\theta) = \theta \) (due to small angle approximation), a preset fundamental
limit for the $NA$ to spot size ratio is thus given by the properties of the source laser beam. Any alteration of the divergence angle $\theta$, for example with a telescope to increase the beam-size in order to achieve a tighter focal spot with a lens, will thus also change the $NA$ accordingly. A simple example illustrates the issue at hand. By assuming an $NA$ of the PCF to be $\sim 0.02$ (which is close to that of the PCF used in this thesis), and applying the condition that the $NA$ of the input beam should not be larger than that, it becomes clear from calculation with eq. (2.16) that $r_{\text{min}}$ is to be no smaller than $\sim 16 \, \mu m$. In continuation with the above argumentation, the maximum divergence angle of a laser-source (or input beam) for optimal coupling to such a PCF can thus also be calculated to be $\theta_{\text{max}} = 0.02 \, \text{rad}$, or $1.146 ^\circ$. If the perfectly Gaussian free-space beam exceeds these parameters, additional coupling loss is unavoidable, while any difference of the real laser beam mode compared to a perfect Gaussian (i.e. an increased $M^2 > 1$) exacerbates the issue.
2.2.3 Relevant fibre parameters

As showcased in eq. (2.10), at large intensities the refractive index is markedly affected by its nonlinear index contribution through the Kerr-effect. To describe these effects more clearly, especially in fibre-optics, important parameters will be introduced that allow for a better understanding of the processes. The nonlinear refractive index \( n_{\text{nl}} \) and the dispersion parameter \( \beta \) (or GVD) have already been introduced in chapter 2.2, and therefore will not be repeated here, with the remaining parameters being the:

- **Nonlinear coefficient \( \gamma \):**
  
  The nonlinear coefficient, also termed nonlinear parameter, is defined as
  \[
  \gamma(\omega_0) = \frac{n_{\text{nl}}(\omega_0)\omega_0}{cA_{\text{eff}}} ,
  \]  
  in units of \([\text{W} \cdot \text{m}]^{-1}\), with \( A_{\text{eff}} \) being the effective mode area. Clearly, \( \gamma \) can be increased either by selecting a suitable substrate with a high nonlinear index \( n_{\text{nl}} \), or by reducing the effective area, e.g. through a smaller core. Much attention is given to chalcogenide glasses to replace fused silica and to access the mid-IR due to their orders of magnitude larger nonlinear index and high transparency [45, 46].

- **Effective mode area \( A_{\text{eff}} \):**
  
  For more complex modes, the illuminated area can not simply be approximated as a circular distribution with the core radius, especially since the mode partly extends into the cladding as explained above. Instead, the effective mode area is defined through the modal distribution \( F(x, y) \) as [35]
  \[
  A_{\text{eff}} = \frac{\left( \int \int_{-\infty}^{\infty} |F(x, y)|^2 \, dx \, dy \right)^2}{\int \int_{-\infty}^{\infty} |F(x, y)|^4 \, dx \, dy} .
  \]  
  For the fundamental mode in single-mode fibres, the effective area is defined through the MFD as \( A_{\text{eff}} = \pi \left( \frac{\text{MFD}}{2} \right)^2 \).

- **Nonlinear length \( L_{\text{NL}} \):**
  
  The nonlinear length gives an estimate of fibre length after which nonlinearities become relevant to the optical pulse and need to be considered for pulse propagation. It is defined through \( \gamma \) and the peak power of the pulse at the fibre-input \( P_{\text{peak}} \) as [35]
  \[
  L_{\text{NL}} = \frac{1}{\gamma P_{\text{peak}}} .
  \]
  For example, a 1 ps pulse from a fibre-laser at 0.1 W and a repetition rate of 20 MHz would exert a peak power of roughly 5.5 kW. With \( \gamma \sim 100 \cdot 10^{-4} \text{ [W} \cdot \text{m}]^{-1} \). This results in a nonlinear length of \( \sim 0.02 \text{ m} \), meaning that with fibre-lengths larger than 2 cm, nonlinear effects need to be taken into account. Naturally, for telecommunication purposes, the input-laser and fibre parameters are very different, leading to nonlinear lengths of several km.
• Dispersion Length $L_D$:

In analogy to the nonlinear length, the dispersion length defines a length of fibre after which dispersive effects need to be considered, such as GVD leading to pulse-broadening. For a temporally sech$^2$-shaped pulse with a FWHM duration of $T_{\text{FWHM}}$, the power independent dispersion length is defined as

$$L_D = \frac{1}{(2\ln(1 + \sqrt{2}))^2} \frac{T_{\text{FWHM}}^2}{|\beta_2|}.$$  \hspace{1cm} (2.20)

Here, the textbook definition $\frac{T_0^2}{|\beta_2|}$ from [35] with the pulse duration $T_0$ defined at $1/e$-intensity has been altered by converting the textbook pulse width $T_0$ to the pulse-width at FWHM using the relation $T_{\text{FWHM}} = T_0 \cdot 2\ln(1 + \sqrt{2})$ for temporally sech$^2$-shaped pulses [35]. Continuing with the above example, for a fused silica fibre with a GVD of $\sim 16 \cdot 10^{-27} \text{s}^2\text{mm}^{-1}$ at 1064 nm centre wavelength, the dispersion length for a 1 ps pulse would be 20 m. In this case, any operation with a fibre length significantly shorter than 20 m needs to take nonlinear effects into account, while dispersive effects become relevant at lengths of 20 m or more. For a 50 fs pulse in the case above, the dispersion length reduces to only 5 cm.

When working with fibres of adequate length and the high intensities typically encountered in ultrafast nonlinear physics, the dispersion and nonlinear lengths become comparable to the fibre length, leading to interesting phenomena that can be exploited, such as optical solitons (in the anomalous dispersion regime) or effects that allow for external pulse compression.
2.3 Relevant nonlinear effects

This section will elaborate in detail on the relevant nonlinear effects encountered in the pulse-compression experiments for this thesis. Due to fibre geometry and initial pulse and operating conditions, these nonlinearities can be summed up in the following list:

- Optical Kerr effect: Self-phase modulation and self-focussing
- Stimulated Raman scattering
- Optical wave breaking

The two main contributions in optical fibres arising from the optical Kerr effect are self-phase modulation (SPM) and self-focussing (SF). In the case of SF, when a high intensity pulse with centre-wavelength $\lambda_0$ propagates in a medium of refractive index $n$ and nonlinear index $n_{nl}$, the resulting change of refractive index according to eq. (2.10) also becomes a spatially varied effect due to the intensity distribution across the beam. Since the intensity profile of the fundamental mode (or Gaussian distribution) is highest at the centre and decays radially, the resulting Kerr-effect in this case is thus also strongest in the centre of the pulse, and leads to a radially decaying gradient of the refractive index change. At sufficiently high intensities, the resulting effects on the pulse due to the refractive index change become stronger than any geometrical or other dispersive effects, which leads to a lens-like structure, spatially focussing the pulse towards its centre. This effect, self-focussing, is then self-sustaining and accelerating in magnitude, as increasing portions of the beam intensity are focussed towards its centre, exacerbating the refractive index change and increasing the focussing strength of the "lens". The phenomenon continues until the beam reaches a point of such intense focussing and intensity that optically induced damage occurs (in solids) and the beam collapses. In case of a waveguide like an optical fibre, this effect is (in part) connected to the fibre-fuse effect which can destroy immense lengths of fibres and will be discussed in detail in chapter 3.5. For obvious reasons, this kind of catastrophic self-focussing should be avoided in daily operation. Luckily, self-focussing requires a certain critical peak power threshold $P_{\text{crit}}$ to instigate the effect, which is irrelevant of the initial beam-width, and can be determined by [47]:

$$P_{\text{crit}} = \frac{0.148 \lambda^2}{n n_{nl}}.$$  \hspace{1cm} (2.21)

For fused silica at a centre wavelength of 1035 nm, the resulting critical peak power for SF lies at $\sim 4$ MW for linearly polarised light and at $\sim 6$ MW for circular polarisation [48].

Just as the intensity of a pulse varies radially in space causing SF, the temporal shape and intensity variation of a pulse also affects the refractive index due to the Kerr-effect. The resulting phenomenon is termed self phase modulation and generally leads to spectral broadening\(^6\). This can be explained through the intensity-dependent refractive index change leading to a temporal variation of the pulse’s phase $\phi$, which affects the instantaneous (angular) frequency $\omega$

---

\(^6\)Spectral narrowing initially occurs for negatively chirped pulses in media with a positive nonlinear index
2.3 Relevant nonlinear effects

Through $\omega = \frac{1}{2\pi} \frac{\partial \phi}{\partial t}$. The difference $\Delta \omega$ between the instantaneous frequency $\omega$ and the centre frequency $\omega_0$ thus varies over the temporal profile of the pulse. For a better understanding it is represented in fig. 2.5 (red line), with an emphasis to be directed to the sign of $\Delta \omega$. At the leading edge (rising intensity) of the pulse, the SPM-induced frequency shift is negative, leading to a red-shift (i.e. the generation of lower frequencies). Across the centre of the pulse, the intensity variation is small, and thus a constant (read: linear) change of the instantaneous frequency shift with a zero-crossing at the peak of the pulse (or at the centre frequency) occurs. As the intensity of the pulse decays, the SPM-induced frequency shift is positive, leading to a blue-shift (i.e. generation of higher frequencies). Thus, as the pulse propagates in the fibre, new frequency components are constantly added as long as the intensity is large enough for the Kerr-effect to take place. This phenomenon thus broadens the spectrum of the pulse.

![Figure 2.5](image)

**Figure 2.5.** Intensity (blue) and SPM-induced frequency shift (red) of a temporally $\text{sech}^2$-shaped pulse. With increasing intensity on the leading edge, the instantaneous frequency shift is negative, i.e. it results in a red-shift. Across the centre of the pulse, the instantaneous frequency shift profile is close to linear, i.e. the pulse develops a linear chirp here (quadratic phase). As the intensity drops on the trailing edge, the instantaneous frequency shift is positive, leading to a blue-shift.

Stimulated Raman scattering (SRS) is a nonlinear process that offers great advantages in applications such as microscopy, material science, or tuneable laser-sources [49, 50]. It also can be a highly undesirable effect, limiting performance for example with pulse compression through phase distortion or in fibre amplifiers by shifting sufficient amounts of pump power out of the desired amplification frequency window. Discovered by C.V. Raman in 1928 [51], it has been studied extensively due to its several useful applications. Generally speaking, SRS exploits the vibrations of molecules in a medium which are excited by the incident pump beam. The material reacts to the electric field with a short delay, which leads to a transfer of energy from the pump photon to the lattice structure. This difference results in an optical phonon in the material (molecular vibration).
while the remaining photon energy continues as a lower-energy photon at a longer wavelength, termed the Stokes-wave. The total number of photons in a loss-less material is thereby maintained. The phonons are specific to each material and result in a characteristic red-shift of a fraction of the pump light to lower frequencies. For sufficiently high pump-powers and long propagation distance, the resulting Stokes-beam can itself act as another pump, feeding a second Stokes-beam. This process is called cascaded Raman scattering, used for example in cascaded Raman lasers. For amorphous materials, especially for fused silica, the excited molecular vibrations spread over a certain frequency-range, resulting in a spectrally broad Raman-gain with its maximum close to 13.2 THz of the pump frequency. This is distinctly different to crystalline structures, where only specific frequencies are amplified, and offers a great spectroscopic tool in combination with optical fibres. To quantify the strength of the effect, the Raman gain coefficient $g_R$ was introduced, with units of $[W^{-1}\cdot m]$. Several studies have since been conducted to accurately determine this coefficient for fused silica, finding that it scales inversely with the pump frequency. For practical applications, a signal beam that lies within the Raman-gain bandwidth is co-launched with the pump, leading to Stimulated Raman Scattering, while in the absence of such a signal beam spontaneous amplification of noise (spontaneous Raman scattering) leads to a build-up of the Stokes-wave, which then acts as a signal to be further amplified through the same process as stimulated Raman scattering. For this reason, the abbreviation SRS used in this thesis refers to either initial process in the theory, while in the practical terms of the experiments all amplification results from spontaneous Raman scattering as no signal-beam is co-launched with the pump.

The most profound impact of SRS in this thesis is the resulting phase contribution it exerts onto the pulse. Since SRS is a nonlinear third-order effect, it induces higher-order phase on the pulse which complicates pulse compression methods. Precisely this impact on the pulse shape has been studied carefully in the late 1980’s [52–54]. All three studies found that SRS leads to an asymmetry in the final spectra, which results from depletion of the leading edge of the input (SPM broadened) pulse towards the Stokes wave. The subsequent steepening of the pump leading edge together with walk-off effects and cross-phase modulation of the two pulses lead to an array of effects that have a profound impact on the linearity of the chirp across the majority of the pulse. This asymmetry-induced nonlinear chirp differs from that which could be compensated for by a quadratic compressor (e.g. gratings or prisms), meaning that the compressed pulse has a significant amount of residual energy showing up as temporal wings i.e. uncompensated higher-order phase contributions. Furthermore, after a given threshold, any increase in pump power results in an increase in the Stokes wave, while the pump-power remained constant, termed “clamping” by the authors [52].

To mitigate this effect without elaborate and high-loss higher-order compression techniques presented in [52, 55], one can estimate a threshold for a given fibre that indicates the point at which SRS becomes strong enough to negatively affect the pulse. Previous work on this topic recommended a conversion of less than 5% [54, 56]. This threshold, and its modification for LMA-fibres is described in chapter 3.3 on page 42. For SRS, at an input pulse centre wavelength of 1035 nm, the Stokes wave is red-shifted by 49 nm (a frequency-offset of 13.2 THz resulting from the peak Raman gain in fused silica). Subject to positive GVD and a significant group-velocity mismatch, the Stokes pulse travels faster with respect to the main pump pulse, further depleting the
leading-edge of the input pulse in time, followed by the emergence of a prominent red-shifted spectral component at longer wavelengths. This leads to an asymmetry of the SPM-induced spectra across the pump pulse, as well as distorting the linearity of the chirp due to cross-phase-modulation interaction between the pump and Raman pulse \[53\]. The growth and amplification of the Stokes pulse is ultimately limited by the walk-off length

\[
L_w = \frac{T_{\text{FWHM}}}{2 \ln \left(1 + \sqrt{2}\right) \left| v_{gP}^{-1} - v_{gs}^{-1} \right|}
\]

of the two pulses that describes the length-scale over which the two pulses can interact (pump \(p\) and stokes \(s\) group velocity \(v_g\), input pulse duration \(T_{\text{FWHM}}\) \[35\]. For long walk-off lengths, SRS generates a potentially substantial pre-pulse, destroying the desired square temporal pulse shape, which ultimately has a negative effect on the quadratic compressibility and should therefore be avoided or has to be tackled through loss-heavy spectral windowing \[57\].

![Figure 2.6](attachment:image.png)

**Figure 2.6.** An unchirped ideal sech\(^2\)-shaped pulse (dashed black) and it’s computed, square-shaped output considering GVD, SPM, OWB and SRS (blue: SRS off, red: SRS on). The effects of SRS on the pulse shape are visible as ripples across the leading edge, while OWB causes the oscillations at the pulse edges.

As GVD broadens a pulse to a temporally rectangular shape within the fibre, the intensity change at the beginning and end of the pulse becomes strong, almost step-like. The resulting SPM-induced chirp at these points is nonlinear in contrast to that induced by GVD, resulting in a nonlinear superposition of the two. As new frequency components are produced through SPM, especially at the pulse edges where chirp is nonlinear, they overtake (are overtaken) by the initial frequencies of the pulse at the leading (trailing) edge. In the normal dispersive regime, this overlap results in interference between the involved frequencies, resulting in temporal oscillations at the temporal edges of the pulse (see fig. 2.6), which is termed optical wave breaking (OWB).
2.3 Relevant nonlinear effects

OWB also produces new frequencies through four-wave mixing at the spectral edges of the pulse. Since the low-frequency components of this are clouded by the SRS build-up, the most noticeable manifestation can be seen on the high-frequency end of the spectrum, see fig. 3.4.
2.4 External pulse compression

External pulse compression relies on the compensation of any chirp a pulse has acquired in a previous step. The acquired chirp may be obtained for different reasons, e.g. from dispersion accumulated by a short pulse in a given experiment, from deliberately induced dispersion in combination with spectral broadening (see fig. 2.7), or from applications where chirp is added and then removed in order to allow for chirped pulse amplification - the Nobel prize winning idea mentioned in the introduction.

![Simplified schematic of external pulse compression. A pulse is spectrally broadened (e.g. in a PCF) with the new frequency components being out of phase, i.e. stretched in time. This chirp is then compensated for, e.g. with a prism compressor, so that all frequency components are in phase. The pulse is now at its transform-limit.](image)

**Figure 2.7.** Simplified schematic of external pulse compression. A pulse is spectrally broadened (e.g. in a PCF) with the new frequency components being out of phase, i.e. stretched in time. This chirp is then compensated for, e.g. with a prism compressor, so that all frequency components are in phase. The pulse is now at its transform-limit.

In the most simple case, the dispersion is of second order (i.e. quadratic phase), being group-delay dispersion (GDD) that manifests itself with a linear chirp. Several materials exist that offer opposite dispersion values allowing for precise control of the sign-switched GDD applied to the pulse, which can be understood by slowing down the leading frequency components compared to the trailing ones that can thus “catch up”. Once all dispersion is compensated for, meaning that all frequency components arrive simultaneously in time and the phase is constant across the pulse, it is transform-limited, i.e. at its shortest possible duration. In this case, the pulse-envelope resembles a Gaussian or similar clean pulse-shape. For practical applications, Frequency-Resolved-Optical-Gating (FROG) measurements allow for accurate recording of the phase of a pulse, which can then be tackled through various compensation methods.

These most commonly include chirped mirrors, which have a given GDD per reflection and thus are slightly tuneable by alignment to allow for more or fewer bounces off of them. They also are anti-reflection (AR)-coated, meaning that loss is minimised with reflectivities above 99.9 %, but often come with an increased price tag, especially when ordering custom designs. A more economical and variable option is given with gratings or prisms which utilise a combination of geometric dispersion together with material specific GDD. Other options consist of using gas-filled
hollow-core fibres that allow for precise tuning of the GDD by varying the density of the gas, or by specifically designed solid-core fibres [58–60]. In this thesis, the method used for external pulse compression relies on spectral broadening and the development of a linear chirp across the larger part of the pulse, which is then compensated for by using a prism-compressor setup. Such a setup requires four prisms, but symmetry allows for two prisms and a reflection surface, with single-prism options also available by using two special mirrors. Nonetheless, since these variations simply make the setup more compact, the fundamental maths behind the four-prism setup remains the same. The GDD induced by a prism-compressor can be calculated based on [55] with the following expression:

\[
GDD = \frac{\lambda^3}{2\pi c^2} \left[ 4L \left( \left( \frac{d^2n}{d\lambda^2} + \left( 2n - \frac{1}{n^3} \right) \frac{dn}{d\lambda} \right)^2 \sin(\beta) - 2 \left( \frac{dn}{d\lambda} \right)^2 \cos(\beta) \right) \right] (2.23)
\]

\[
\cdots + 4 \left( \frac{d^2n}{d\lambda^2} \right) (2D) \right]. (2.24)
\]

Here, \( L \) is the prism spacing between their respective apexes, \( n \) the refractive index of the prism, \( D \) the diameter of the beam at \( 1/e^2 \) intensity at the first prism input, \( \lambda \) the centre wavelength, and \( \beta \) the angle of the dispersed beam after the first prism. The GDD can be tuned by varying the distance between the prisms, since all parameters except \( L \) are set by the beam diameter, Brewster’s angle arrangement, and the chosen prism material. Ideally, the distance is chosen in such a way that the resulting GDD of the prisms matches the GDD of the pulse after spectral broadening, but with opposite sign (assuming no higher-order phase distortion). Given the fact that the time-bandwidth product of the output of the fibre used in this thesis is too large to be measured by the FROG available, the GDD was instead calculated through eq. (2.12). It allows for calculating the induced GDD of a given material that a known input pulse has propagated through. Hence, eq. (2.12) can be re-written to be expressed using experimentally observable quantities, such as the output pulse duration \( \tau_{out} \) and the frequency bandwidth \( \Delta f \) defined through the centre-wavelength \( \lambda_0 \), the speed of light in vacuum \( c \) and the measured wavelength bandwidth \( \Delta \lambda \) according to \( \Delta f = \frac{\Delta \lambda c}{\lambda_0^2} \). With the addition of a pulse-shape dependent time-bandwidth product \( b \), which is 0.441 for a Gaussian and 0.315 for a sech^2-shaped pulse, the GDD is calculated through:

\[
GDD = \frac{1}{4\ln(2)} \left( \frac{b \tau_{out}}{\Delta f} \right)^2 - \left( \frac{b}{\Delta f} \right)^4. (2.25)
\]

The drawback of using prism compressors are the higher losses compared to individually designed AR-coated chirped mirrors and the inability to individually address higher-order dispersion. This is related to the dominant contribution of the GDD in the prisms, which is set and tuned by changing the distance of the prisms to an optimum length that accommodates for the second order dispersion one wishes to remove. This fixed length carries a residual TOD value, which cannot be selectively tuned without changing the second order dispersion compensation achieved in the previous step. This residual higher-order dispersion often encountered in high intensity applications sets a limit to the pulse quality that can be achieved with a quadratic compressor like the four-prism setup. It can, however, be addressed by spectral windowing or by using a grating-compressor, termed a Grism compressor [61, 62] (a combination of prisms and gratings). Through
2.4 External pulse compression

this, the third-order dispersion (TOD - cubic phase term) can be addressed individually since the TOD of prisms and gratings is of opposite sign, meaning that one can tune the separation distance of the prisms in parallel to the grating parameters so that the pulse’s GDD and TOD are compensated for. On the downside, gratings typically have a low transmission or reflection, so substantial power loss might occur which might be larger than the additional benefit to the peak power obtained from having a cleaner temporal shape compared to a simple prism compressor.

When using a prism compressor, a certain tolerance to the mismatch between achieved compression and theoretical optimum is necessary, which can be reduced to some extent by the choice of an optimal prism-material with a suitable intrinsic TOD value. Furthermore, a prism compressor is more economical and allows for a less complex setup, which translates to a more practical approach for the compression. To sum this up verbally: **better is the enemy of good** - a too complex and elaborate compression setup will require more resources and time for a potential gain that is not necessary for the applications envisioned in this thesis.

To better understand the process of spectral broadening in a normally dispersive fibre and its effect on the temporal structure of an experimentally recorded input pulse, fig. 2.8 is introduced. It shows the output pulse after propagating an experimentally recorded input pulse through 13.5 cm of fused silica at 1.41 MW peak power and 250 fs FWHM as well as its phase and chirp profile. Especially when comparing to the SPM-induced chirp profile in fig. 2.5 on page 25, the greater linearity of the chirp across the pulse duration becomes apparent. This is the result of the interplay between SPM and GVD. The modulations at the pulse edges from OWB are also clearly visible and the onset of SRS affecting the pump pulse becomes apparent. This is the result of the underlining principle of this kind of external pulse compression essentially requires the GVD-imposed chirp to develop in a linear fashion over the newly generated SPM-induced frequency components, while simultaneously producing a square-shaped temporal profile [35, 63, 64]. The frequency modulation from SPM leads to a linear frequency chirp across the region of maximum intensity, i.e. the centre of the pulse. In an optical fibre, the GVD has a defining role in the evolution of the pulse-shape throughout the spectral broadening process, and is particularly pronounced for femtosecond pulses because of the broader initial input bandwidth [65]. For normally dispersive fibres, the positive GVD temporally broadens the pulse envelope, adding to the accumulation of positive chirp. Importantly, this dispersive broadening acts to further linearise the SPM-induced chirp profile, which means that the final, spectrally broadened pulse, has a linear chirp that can be addressed through quadratic compression, and results in a compressed pulse with higher peak power, broader bandwidth, and shorter pulse duration than the initial pump-pulse, motivating the entire process.

A higher peak power will result in a stronger impact of nonlinear processes and such a broadband femtosecond source would have multiple potential applications. The high bandwidth would for example allow for a widely tuneable optical parametric amplification setup to generate far-IR frequency components, while the corresponding ultrashort pulse durations can be readily used in pump-probe experiments requiring high temporal resolution. The concept of increasing
2.4 External pulse compression

Figure 2.8. Simulated temporal output intensity (blue), chirp (red), and phase (green) of an experimentally measured input pulse after propagating a 13.5 cm stretch of PCF at standard experimental settings (1.4 MW, 250 fs, 10 MHz). The distortion of the generally linear chirp profile across the leading edge results from the higher-order phase originating from SRS. This simulation includes the RIN of the laser.

The peak power of a laser source through such an external compression is nothing new, but the application in combination with a high-power, high repetition rate source promises to allow for far more diverse applications in the laboratory. Many processes requiring high peak intensities could hereby be scaled up, increasing conversion efficiency as the active area of a process is increased. One particular application that will be addressed in this thesis is the generation of THz-radiation through a process termed optical rectification, covered in detail in chapter 4.2.1. This process would immensely benefit from a high peak power source as described above.

These applications - pulse compression and THz-generation at MHz repetition rates - are the two main elements of this thesis. In the following chapters, the devices used for the pulse compression experiments are introduced and the experimental setup is elaborated on. Experimental results and further calculations for the PCF and power settings follow up on this. They are then discussed and put into context with contemporary work with similar setups. So far, all relevant physical parameters and processes that are required to understand the following experimental procedure have been introduced, which is why the following chapters will have less theoretical content.
CHAPTER 3

Compression to 22 fs

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3.1 Driving laser analysis

The driving laser source used for the work conducted in this thesis was a ytterbium based master oscillator power amplifier by NKT photonics (Yb:fibre-laser). It is a prototype-like predecessor to the now commercially available NKT aeroPULSE FS-series. The master fibre oscillator emits its output into an amplifier rod, which is pumped by several diodes, enhancing the throughput power of the laser pulses. The laser emits a vertically linear polarised beam with >23 dB polarisation extinction ratio at a centre wavelength of 1.035 µm and a spectral bandwidth of 8 nm, as shown in fig. 3.8. Internal dispersion control, directly accessible through the control software, allows the temporally sech²-shaped output pulses to be adjusted from ~250 fs to ~1 ps duration (FWHM). The transform-limit for a 8 nm pulse centred at 1035 nm lies at ~140 fs. At such short pulse durations, however, fibre-based lasers have their limitations due to higher-order phase contributions accompanying various nonlinearities. Together with the limited gain bandwidths these are the main reasons why the transform-limit of 140.7 fs FWHM is not reached.

To control dispersion, an element with opposite-sign GVD to the GVD imposed by the laser is introduced - often with an optional control of the amount of GDD to be compensated for. Classical second-order phase compensation with tuneable free-space solutions generally require long path-lengths leading to constructional limitations. All-fibre options on the other hand can include tuneable fibre-Bragg gratings installed in the compact setup of the fibre-laser. This is often implemented to compensate for GVD, but leaves TOD unaddressed. To compensate for limitations from these acquired higher-order phase contributions, complex and usually free-space optical setups are required, mitigating a main benefit of fibre-lasers (being not free-space). This uncompensated phase contribution also limits the potential compressibility using external pulse compression with second-order dispersion control. Ruefully, TOD can not be avoided in solid fibre-lasers at such

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1The exact design is intellectual property of NKT Photonics
3.1 Driving laser analysis

large powers and short pulse durations. These limitations, geometrical as well as higher-order phase contributions, therefore set a boundary on the achievable pulse duration within classic fibre-lasers. Nonetheless, other designs incorporating hollow-core fibres for GVD control (replacing gratings) circumvent the geometrical issue, but add higher TOD contribution [66].

Any pulsed-laser characterisation requires the measurement of the pulse duration, especially when working with ultrashort pulses. In order to measure femtosecond pulse duration, a technique termed auto-correlation is applied rather than classical "stop-watch" methods. The reasoning behind this is that no electrical switching process is fast enough to resolve the temporal structure of femtosecond pulses. Instead, the pulse itself is used to analyse its own temporal duration. This is possible due to nonlinear processes such as second-harmonic generation (SHG) or two-photon absorption, which exhibit a nonlinear response with increased intensity. The initial pulse is split into two parts which undergo an equivalent beam path to each other and are then spatially recombined, either collinearly or in a focal point, depending on the type of autocorrelation used. One beam path is periodically lengthened and shortened, which results in a periodic temporal overlap and detachment of the two pulses, resulting in a nonlinear response according to the degree of the overlap. This low-frequency periodic movement can be calibrated with the speed of light in air to a femtosecond timescale by allocating a certain real time-step derived from the spatial movement to a measured time-difference on an oscilloscope. The result allows for femtosecond time resolution despite the used detectors and electronic materials having response-times far longer than femtoseconds. As a last step, depending on the specific type of autocorrelation used, further mathematical analysis and deconvolution according to the temporal pulse-shape have to be applied.

For temporal pulse measurements applied in this thesis, a self-built interferometric, two-photon absorption version is used [67] in combination with the MOSAIC-software provided by the Department of Physics and Astronomy at the University of New Mexico. The software allows for real-time analysis of an interferometric autocorrelator (IAC) trace yielding the FWHM and some further information on the chirp of the trace [68,69]. The AC-method allows for pulse duration analysis, but falls short in accuracy once the temporal pulse-shapes become more complex, as the resulting AC-traces of different input pulse shapes may look the same. To fully analyse a pulse in time and phase, a modified version of autocorrelator called FROG is used. Here, the resulting nonlinear signal is not only recorded temporally, but also spectrally, which allows for a full spectrogram recording of the pulse revealing the distribution of the spectral components over time as well as the phase of the pulse.

Figure 3.1a) shows the shortest possible laser-output recorded as an IAC trace and a zoomed-in image depicting the cycle oscillations. The ideal IAC-ratio of the peak to background of 8:1 is not achieved despite increased alignment efforts. Figure 3.1b) contains the FROG retrieved field, phase, and the FROG spectrogram. It shows a linear residual temporal phase across the centre of the pulse that becomes nonlinear at the pulse edges, meaning that across the centre of the pulse most of the GDD is compensated for, while higher-order residual phase is still present at the edges. The detailed temporal analysis of ultrashort pulses is a science in itself, with several more options for pulse-characterisation on ultrafast timescales, but a thorough explanation of
the topic is outside the scope of this thesis. Further information can be found in textbooks such as [63], with the relevant take-away from this brief introduction being that IAC allows for initial monitoring of the temporal duration of a pulse as well as an indication of the chirp [68, 69]. A more detailed understanding of the temporal and phase structure is acquired through FROG, especially for externally compressed ultrashort pulses.

![Figure 3.1](image)

**Figure 3.1.** a) normalised IAC measurement (blue) of the 250 fs pulse from the fibre-laser. The cycle length is 3.45 fs (inset). FWHM values extracted from IAC are equal to the width of a sech²-curve (red, offset by +1 to match the background signal). b) Reconstructed FROG image (inset), extracted temporal intensity (blue) with an ideal sech²-shaped fit (dashed black), and phase (dotted red).

The output repetition rate of the laser, *i.e.* the number of pulses per second, can be set at distinct intervals over the range of 2-20 MHz. These values correspond to integer divisions of the internal oscillator’s fundamental frequency, allowing for pulse energies up to 22 µJ at a maximum output power of 44 W. The output power of the laser as a function of the software set drive-current is depicted in fig. 3.2a), which shows the typical exponential behaviour for laser gain, a nonlinear process.

The spatial properties of the output beam are characterised by an M² value <1.2 and a beam divergence of θ =0.8 mrad. The M² value is a measure of the quality of a laser-beam, with a value of 1.0 being equivalent to a perfect Gaussian beam. An exemplary image of the intensity distribution of the laser beam is shown in fig. 3.2b), which highlights the clean Gaussian
mode profile. By measuring the beam diameter over a set of fixed distances, the divergence was experimentally determined to lie at $\theta \sim 0.008$ rad, which relates to an $NA$ of 0.008 for the laser output beam.

Figure 3.2. a) Output power of the laser as function of the drive-current setting. b) Spatial beam intensity of the laser output showing a close to Gaussian profile.

In order to have for the focussed spot size to match the MFD of the PCF while also keeping the focussing angle as low as possible to allow for adequate acceptance angles, the beam was enlarged directly behind the laser output by a factor of five to $\sim 9.4$ mm in diameter. Due to divergence, this lead to a spot size at the position of the focussing lens of 11.73 mm. The initial enlargement also aimed to protect subsequent optics from damage due to the high intensities resulting from a smaller spot size. In general, a beam can be enlarged or reduced in size using a telescope setup. The two variants are called either the Galilean or Keplerian versions, which both operate on the same principle using two lenses. The first lens with focal length $f_1$ will focus or diverge the beam, while the second lens with suitable focal length $f_2$ is placed at a given distance $d = f_1 + f_2$ to collimate the resulting reduced or enlarged beam for further propagation, with the ratio of the focal lengths of the lenses determining the change to the beam-size $\Delta D$ through:

$$\Delta D = -\frac{f_2}{f_1}.$$  \hspace{1cm} (3.1)

For stable and repeatable operation, the final settings of the laser were set to a 10 MHz repetition rate, a pulse duration of 250 fs, and an operating power of 13.3 W. Precise control of the
power was achieved with a polarising beamsplitter set behind a $\lambda/2$ waveplate, which allows for modulation of the P-polarised (i.e. parallel to the plane of the laboratory table\textsuperscript{2}) transmitted beam-power in a sinusoidal fashion from $\sim 0 \%$ to $\sim 100 \%$ of the laser operating power. Since all experiments conducted were solely pulse-energy dependent, the repetition rate was set to 10 MHz instead of 20 MHz for ease of operation (i.e. less average power and thermal drift). The overall laser output power was not set to its maximum of 44 W, because the power required for the compression setup was limited by the damage threshold of the fibre and there was no need for the excess laser-output.

\textsuperscript{2}The S-polarised output of the laser is rotated by 90° by an optical isolator.
3.2 Setup overview

Figure 3.3. Pulse compression setup. The laser output is expanded and then focussed into the PCF. The spectrally broadened pulse is collimated and sent through the prism compressor. A pick-off mirror steers the compressed pulse towards the applications.

The setup used for the compression-part of this thesis is shown in fig. 3.3. The output of the fibre-laser is sent through an isolator (EOT PAVOS Ultra High Power Optical Isolator, over 33 dB feedback suppression) to prevent damage to the laser from potential back-reflections. The isolator was ordered with custom design parameters being an operating temperature of 25 °C, a 5 mm aperture, a 1035 nm centre-wavelength, and a rotation of polarisation from vertical to horizontal. The beam then passes a periscope assembly to change the propagation beam-height to match the experiment, using high-power withstanding dielectric mirrors (Thorlabs NB1-K14) with a reflection of >99.8 % at the laser centre wavelength. This is followed by a Galilean telescope-arrangement, consisting of two achromatic doublets with focal lengths of -50 mm and 250 mm (Thorlabs ACN127-050-B, Thorlabs AC254-250-B-ML), exerting a five-fold beam expansion to a beam diameter of ~9.4 mm at 1/e² intensity. This enlargement allows for a better match between the focussed beam properties and the MFD of the PCF. As a positive side effect, the divergence of the initial beam is also reduced by a factor of five to 1.6 mrad. The following variable power attenuator consists of a λ/2 waveplate (Thorlabs WPH05M-1030) designed at a centre-wavelength of 1030 nm and a polarising beamsplitter cube (Thorlabs CCM5-PBS203/M) with an extinction ratio of transmitted P-polarised light to S-polarised light of 30 dB. The thickness of the substrate does not measurably affect the pulse duration through dispersion. The cleanly

³The isolator and periscope are not shown in the setup figure.
3.2 Setup overview

P-polarised beam is then guided towards the focussing lens for the PCF by two anti-reflection coated broadband dielectric mirrors (Thorlabs BB1-EO3), having a reflection of \( \sim 99.00 \% \) each at the centre wavelength of 1035 nm. The focussing lens used is an air-spaced doublet with a focal length of 200 mm (Thorlabs ACA254-200-1064) that allows for a focal spot size of 27 \( \mu \)m and a Rayleigh length of 0.46 mm, given an input spot size of 11.73 mm in diameter at 1035 nm central wavelength with an \( M^2 \)-value of 1.2. The lens was chosen specifically to deliver the desired focal spot size conditions as close as possible, since the fibre-launching process is critical to the experiment. The difference from the ideal 32 \( \mu \)m MFD spot size resulting from calculations with an assumed spot-diameter of 10 mm rather than the 11.7 mm. The lens choice was also affected by the lens options available for purchase. At the beam-waist, the wavefront can be assumed with minimal divergence, and the Rayleigh length gives a measure of the distance over which the wave-front maintains the focussed beam-width to an order of \( \sqrt{2} \) (a doubling of the mode area) on either side of the focus.

Ideally, the fibre-facet meets the wavefront at the narrowest point of the beam (where divergence is minimal), while a longer Rayleigh length will allow for easier alignment. The 13.5 cm long PCF has a core diameter of 40 \( \mu \)m, an outer cladding diameter of 450 \( \mu \)m, and with a high temperature acrylate coating the final diameter is 540 \( \mu \)m. It is placed on a three axis manual stage (Thorlabs MB1616D/M) allowing for precise alignment in the X, Y, and Z directions with a precision of 1 \( \mu \)m and is fixed in position with a V-grooved fibre-clamp (Thorlabs HFF001). Due to constraints from the cleaving process, as well as to avoid potential damage to the fibre (explained in detail in chapter 3.5), the PCF was stripped from its acrylate coating up to 2 cm behind the facet. At the fibre output, the pulse has undergone significant SPM and has broadened to almost 100 nm in bandwidth (FWHM), which all originate from the same "focal point" (the end-facet), and standard lenses would lead to a sub-optimal collimation performance due to chromatic aberration. For this reason, the collimating lens used to collect the diverging beam after the fibre exit is an achromatic lens with a focal length of 50 mm (Thorlabs AC127-050-B-ML), placed on a manual translation stage to adjust for optimal collimation in the far field. As a result, the lens was not placed precisely at the focal length, but rather adjusted in such a way that the resulting beam offered a suitable and low divergence spot size at several metres of propagation distance, e.g. at the point of THz-generation or for pulse-measurement at the autocorrelator. By measuring the spot size of the non-collimated output of the fibre at different distances from the end facet, the divergence, and thus the \( NA \) of the fibre, was measured to be 0.022, almost identical to the manufacturers estimation of 0.02.

Just behind the collimating lens, a Wollaston prism (Thorlabs WP10) with extinction ratio of 10,000:1 is placed temporarily. A Wollaston prism splits unpolarised light into two orthogonal components (P- and S-polarisation) and separates them by an angle of propagation. Since the input beam to the fibre is strongly P-polarised and the PCF is polarisation maintaining, the P-polarisation is preserved in case the stress rods of the PCF are aligned either parallel or orthogonal to the horizontal plane (i.e. the P-polarisation). Any other orientation of the PCF will lead to some level of splitting between S- and P-polarised light. With help of the Wollaston prism, one can separate the two components and minimise the intensity of the S-polarisation by rotating the PCF to achieve the desired orthogonal alignment. This offers an easily implemented option to correctly
align the PCF according to the polarisation of light in the core. Subsequently, it allows for a clean power measurement of the output core-light and thereby the calculation of the overall coupling efficiency. Once this alignment process is finished, the Wollaston prism is removed, the beam collimated and then guided by a dielectric mirror (EO3) towards the prism compressor stage with a total power of 4 W and a pulse energy of 0.4 µJ.

The first uncoated equilateral SF11-glass prism has side-length dimensions of 15 mm and 15 mm height, with a 60° apex angle (Edmund Optics #47-278). It is inserted just enough to avoid any visible beam-clipping from the prism apex, which translates to a beam insertion of 1 mm. The output angle of the refracted beam is tuned by adjusting the rotation-mount on which the prism is placed in such a way that the minimum divergence angle is reached (equivalent to Brewster’s angle). High accuracy for this is achieved by monitoring the beam displacement at over 100 cm of beam path behind the prism. Brewster’s angle, which has minimal reflection losses for P-polarised light, is a function of the refractive indices involved (and thus the wavelength), which cannot perfectly match the broadband pulse across its entire spectral width. For narrow-band applications, custom cut apex angles to match Brewster’s angle can be ordered, however for this experiment, the optimal settings for the centre wavelength of 1035 nm of Brewster’s angle at 60.322° and apex angle of 59.356° are very close to the off-the-shelf 60° apex angled SF11 prisms. As the pulse enters and exits the prism, its spectral components are split horizontally at different propagation angles. Due to the distance between the prisms of 480 mm, this results in a significant horizontal spatial broadening of the pulse, requiring the second uncoated equilateral SF11-prism to be larger than the first. Its dimensions are 30 mm in height and a 30 mm side length, also with a 60° apex angle (Edmund Optics #47-276). The prism is angled equally to the first prism, with the exiting beam matching the propagation angle of the input beam to the first prism, and insertion is managed while monitoring the FROG-signal to achieve optimal compression, estimated to be approximately 5 mm from the apex. On the downside, due to the different propagation angles of the spectral components, the divergence from Brewster’s angle for each frequency is now more profound compared to the first prism. For example, the refracted beam at 1035 nm leaves the first prism at an angle of 62.4°, which is then also the incidence angle on the second prism, differing by 2° from the ideal Brewster’s angle of 60.3°. This accumulates to an average power loss at each prism of ~2%, and a total reflection loss for all four prisms of 283 mW, or 7.4 %. Due to the blue frequency components experiencing a shorter beam path through the SF11 glass than the red components, they temporally “catch up”, leading to a shorter pulse duration by de-chirping the pulse.

To avoid further GDD induced by dielectric mirrors, the following beam path uses protected silver mirrors (Thorlabs PF10-03-P01), due to their extremely low GDD contribution across the entire bandwidth of approximately 3-5 fs², with the disadvantage being a lower reflection of ~97.00 %. The exit-beam of the second prism is sent back through the prisms on the same beam path after reflection off a silver mirror. The mirror is slightly vertically angled to allow for an offset of the back-propagating pulse over the initial one. Another silver mirror (the pick-off mirror) is placed above the input beam in front of the first prisms to steer the back-propagating, compressed beam towards any desired applications.
3.2 Setup overview

The total power-loss (from prisms and mirrors) compared to the exit power of the fibre at 4 W to the beam position after the pick-off mirror at 3.45 W is 13.7 %. Considering only the compression part of this thesis, the direct applications are the pulse characterisation by monitoring through IAC and FROG, which are very sensitive and require low powers. The required attenuation is achieved with a series of beamsplitters that pick off a small amount of power from the main beam and direct it with silver mirrors towards the analysis instrument. The GDD from the reflection of beamsplitters on the pulse duration is of negligible impact, with values in the low single digit fs\(^2\)-regime, whilst transmission through optical substrate is avoided as far as reasonably possible. The unavoidable transmission objects in the IAC-setup consist of two ultra-thin beamsplitters (Thorlabs UFBS5050), one for each arm of the autocorrelator. While the focus is achieved using a silver paraboloidal mirror rather than a transmission lens, offering the further benefit of no chromatic aberration and thus no temporal pulse elongation in the focus. In fact, the UFBS5050 beamsplitters used actually induce negative GDD of \(~\sim 40\) fs\(^2\) (reflection and transmission combined), thereby coincidentally compensating for the positive GDD of the eight silver-mirror reflections encountered after the pick-off mirror of \(~\sim 24\) fs\(^2\). The dispersion values are taken from data-sheets provided by the manufacturer, which might differ from batch-to-batch production and individual units.

For the FROG measurements, a commercial system (MesaPhotonics FROG Scan) based on SHG in a BBO with a cross-correlation method is used. The beam has to propagate through a \(\lambda/2\)-waveplate for polarisation matching of the SHG-crystal, adding 60 fs\(^2\) and also not offering equal retardance across the entire bandwidth. Following this, one arm is reflected off the internal beamsplitter, while the other transmits it (inducing different amounts of GDD to each arm). Lastly, the beam is focussed into a 100 \(\mu\)m BBO crystal with a transmission lens (higher GDD dielectric mirrors are used for beam steering). Exact materials used within the FROG are known only to the manufacturer, meaning that a precise theoretical estimate of the induced GDD is not possible, but assumed to be on the order of an additional 40-60 fs\(^2\), leading to a total of approximately 100 fs\(^2\). This matches very well with the experiments, as the IAC measured duration of 22 fs is elongated in theory to 25 fs when GDD of 100 fs\(^2\) is applied - exactly the duration measured with the FROG. The FROG is thus not as well suited for ultrafast analysis as the IAC setup, which results in a slight discrepancy between the acquired results from the FROG-temporal width vs. that of the IAC of a few femtoseconds (or \(~\sim 10\) %). The pulse durations reported in this thesis are those measured through IAC unless mentioned otherwise, while the FROG is mainly used to monitor the phase and temporal structure of the pulse.

Other than these crucial tools, an optical spectrum analyser (ANDO: AQ6317B) and a thermal power meter are used to monitor the spectral bandwidth and power. This concludes the overview of the setup used for the compression experiments and remains the same for all applications described in this thesis. Subsequent setup figures list the entire pulse-compression setup as a boxed source.
3.3 The “golden ratio”

The first experimental approach with the setup consisted merely of recording the output spectrum as a function of fibre-length and peak power monitored behind the collimating lens in order to gain a deeper understanding of the spectral development. Figure 3.4 shows the characteristic development of spectral broadening through SPM in a normally dispersive fibre for an increasing set of peak powers at a fibre length of 13.5 cm. At 0.734 MW the spectrum shows a clean development of the typical modulations resulting from SPM, with no noticeable features at the spectral edges in the low dB range. As the peak power is increased to 1.151 MW the spectral effects of additional nonlinearities become visible as spectral content below 1000 nm (from OWB) and a skewed shift to the red after 1100 nm (from SRS). Both features grow stronger at 1.367 MW and the SRS shows a noisy behaviour, while the main part of the pulse still shows clean SPM oscillations. Finally, at 1.877 MW, the cleanliness of the SPM oscillations over the centre part of the spectrum is disturbed and SRS develops a new peak beyond 1150 nm by draining power from the nearby leading edge frequencies.

The peak power values reported in this thesis is calculated according to the adjusted peak power formula for temporally sech\(^2\)-shaped pulses [70]:

\[
P_{\text{peak}} = 0.88 \frac{P_{\text{avg}}}{f_{\text{rep}} \tau_{\text{FWHM}}},
\]

where \(P_{\text{avg}}\) is the average power, \(f_{\text{rep}}\) is the repetition rate, and \(\tau_{\text{FWHM}}\) is the FWHM pulse duration. Briefly, it can be seen that above a certain peak power threshold (or equivalently after sufficiently long propagation, see fig 3.5), additional features to the spectrum appear and the initial symmetrical development of the spectrum is disrupted. For a better understanding of the effects on the chirp and thus the compressibility, the output temporal spectrum, field and chirp for fibre lengths of 0 mm, 9 mm, 53.5 mm, and 135 mm were computed when pumping the fibre with 2 MW of peak power.

As seen in fig. 3.5 column a), the bandwidth resulting from pumping at a high peak power of 2 MW also shows significant spectral components at the spectral edges which are not an artefact of SPM. They significantly distort the chirp profile of the pulse and cause the development of strong oscillations across the previously linear curve for the temporal profile in fig. 3.5 column b). Following the simple understanding that a pulse with a large spectral bandwidth correlates to a short temporal duration is valid in theory, so this additional bandwidth should be desired. However, in practical terms, compressors are usually limited to individual tuning of the second (and sometimes third) order dispersion compensation, which requires a pulse with quadratic phase, i.e. a linear chirp, for clean compression. Given that these very high intensity related spectral features are a result of nonlinear effects that contribute higher-order phase and the fact that in the experimental setup used in this work can individually tune only for second order dispersion (GVD), the resulting pulse compression quality is compromised [53,71]. This can further be understood by viewing the resulting chirp from the simulation of fig. 3.5b), which shows a nonlinear chirp profile for the long fibre length, particularly on the leading edge of the pulse which is depleted by SRS and is a hindrance for a quadratic compressor setup.
Figure 3.4. Output spectrum of a 13.5 cm long PCF pumped with increasing peak power (top to bottom). At low peak power, SPM dominates the spectrum, while OWB and SRS contribute stronger as the peak power is increased. For high peak powers, the symmetry of the broadening is broken due to SRS.

The dominant effect that causes the spectral intensity bump at 941 nm results from OWB, which also adds to the spectral intensity at 1139 nm. Ultimately, OWB is connected to SPM and GVD - the two driving forces for fibre-based pulse broadening and compression - and is unavoidable. As the pulse propagates through the fibre, newly developed SPM frequency components at the red (blue) edge of the spectrum travel faster (slower) than the existing spectral boundary components due to GVD. The new frequency components pass the pre-existing ones, causing
interference between the two frequencies involved. This leads to a temporal oscillatory fine structures at the edges of the pulse, seen clearly in fig. 2.6. These spectral side-lobes can be thought of as a four-wave mixing process [35]. The resulting chirp profile induced by SPM and OWB at the edges of the pulse is not linear due to higher-order phase contributions from these processes. Hence, this additional bandwidth cannot be utilised to generate shorter pulse durations by using a quadratic compressor, but instead will reduce the temporal quality of the compressed pulse. However, as mentioned, OWB is inherently linked to SPM and GVD and thus unavoidable. Furthermore, since the OWB contributions are situated at the edges of the temporal and spectral pulse, the effect it has on the main part of the pulse width is minuscule, and thus OWB is a tolerable phenomenon.

The same can not be said for the other main contribution occurring at prolonged distance and/or higher peak power, being stimulated Raman scattering (SRS). SRS develops independently of the other nonlinear effects and generates a distinctly red-shifted pulse that depletes the low-frequency side of the SPM-broadened pulse as the pulse propagates. This breaks the symmetry and draws out the low-frequency end of the spectrum, as can be seen in fig. 3.5a), second row. Here, the onset of Raman is visible as a small bump at 272 THz, matching the Raman frequency gain of 13.2 THz (∼50 nm) in fused silica from the nearest SPM modulation at 285 THz (∼1052 nm). As the fibre-length is increased, this peak gains spectral power, and fig. 3.5a) third row shows that together with the SPM, the SRS-peak has also shifted. The new peak is at 249 THz, ∼13 THz from the steep spectral SPM-feature rising at 261 THz from. With even longer propagation, simulated in row four of fig. 3.5a), the SRS starts depleting and asymmetrically broadening the SPM-structure. It develops a very distinct far-shifted peak at ∼231 THz. An interesting observation in fig. 3.5a), third row, is that the SRS has not yet depleted the steep low-frequency spectral edge of the SPM bandwidth, but as the pulse propagates further it drains that part of the spectrum towards longer wavelengths.

The red-shifted SRS pulse in a normally dispersive fibre will travel faster than the main pulse and ultimately split from it, with the length-scale of this being termed the walk-off length introduced in eq. (2.22) on page 27. It describes the length-scale over which the two pulses can interact and after which the two pulses are temporally separated. For high intensity pulses, SPM and GVD will broaden the pulse spectrally and temporally, elongating the time or fibre length the SRS pulse requires to leave the main pulse, as it is initially encompassed by the growing main pulse, leading to prolonged interaction. In this case, the SRS pulse affects the central part of the main pulse, adding higher-order phase contribution to it.
Figure 3.5. Column a) shows the spectral behaviour of a 2 MW input pulse at four distinct fibre lengths. SPM develops alongside OWB and SRS (peak below 250 THz), broadening the input spectrum. Column b) shows the corresponding temporal profile (blue) of the pulse normalised to the input, and the acquired chirp (red) becoming strongly nonlinear on the leading edge due to SRS. The y-axis of the last image of the right column is shifted to allow for better readability.

Since SRS initially is an independent process (unlike OWB), one can try to minimise its impact on pulse compression by choosing a suitable launch-power and fibre length. This ratio leads to an optimal peak power for a given fibre length, giving this chapter its title. To accurately calculate these parameters, two equations are essential.
• The optimal fibre length for SPM and GVD induced pulse modification and subsequent quadratic compression

• The threshold at which significant SRS contribution to the pulse occurs.

The method of pulse compression used in this thesis has been thoroughly studied by several authors (e.g. [72, 73]), who have provided empirically or numerically derived equations to estimate the optimal fibre length for given input parameters. A concise expression for the optimum fibre length introduced by G.P. Agrawal in Ref. [73] is based on extensive numerical analysis and reads:

\[ L_{\text{opt}} \approx \sqrt{6L_D L_{\text{NL}}}, \quad (3.3) \]

where \( L_D \) is the dispersion length and \( L_{\text{NL}} \) the nonlinear length as introduced earlier in eq. (2.19) and eq. (2.20). In his book, G.P. Agrawal points out that eq. (3.3) is valid when considering the effects of GVD and SPM, with a too short fibre not having yet developed the rectangular temporal shape (and linear chirp) desired, while too long fibres reduce the effect of SPM and simply add dispersion. In other words, the optimal fibre length derived using eq. (3.3), does not account for additional nonlinearities such as SRS, which set a physical limit on what quadratic compressors can achieve. It is therefore only valid for peak powers below the Raman threshold.

In the fibre-community, the typical equation taken to estimate the Raman threshold is an approximation of the original formula provided by R.G. Smith in 1972 [74], which assumes long and small core fibres and a Stokes to pump power ratio of 1:1 (i.e. 50 % conversion). However, as Jauregui et al. point out [75], the relation given by Smith can not simply be applied to other fibre structures, such as in high-power fibre amplifiers or - more importantly - PCFs [76]. This is because the “classic” relation is independent of the effective area. So while it holds true for small fibre cores, where the area is comparable across many fibres, it becomes inaccurate for LMA fibres. Out of this reasoning, Jauregui et al. propose a modified simplification [76] of the original equation in [74], which yields a Raman threshold power \( P_{\text{th}} \) of:

\[ P_{\text{th}} = \frac{20.3 + \ln \psi + \ln \left( \frac{A_{\text{eff}}}{g_R L_{\text{eff}}} \right)}{g_R L_{\text{eff}}} A_{\text{eff}}. \quad (3.4) \]

Here, \( \psi \) is the ratio between the Raman and the pump power at the fibre output, \( A_{\text{eff}} \) the effective area, \( g_R \) the material and frequency specific Raman gain coefficient, and \( L_{\text{eff}} \) the effective length which in this case is equal to the fibre length (see eq. (2.14) in chapter 2.2.2). The thresholds calculated through the method in [74] tend to be 50 % lower than those received using the effective area respecting eq. (3.4) for LMA fibres. For the parameter \( \psi \), which defines the ratio of power that is converted from the pump to the Raman pulse, results in [52] from 1988 (before the invention of PCFs) show an upper limit of 11 % at which significant pulse reshaping takes effect, marking a threshold after which Raman amplification strongly increases. However, more recent results argue for a more modest value of 1 % [75] from a point of fibre-amplifiers where SRS is to be

\[ \text{In the cited proceedings of SPIE article, the equation shows } 20.3 - \ln \beta \text{ instead of } \cdots + \ln \beta, \text{ with } \beta \text{ being } \psi \text{ in this thesis. After correspondence with the author of the article, this has been verified as an error resulting from a previous definition of } \beta \text{ being larger than } 1. \]
avoided as much as possible and is therefore the \( \psi \)-value adopted in this thesis. Since eq. (3.4) cannot be easily re-written into an expression for the fibre length, a set of power threshold values calculated from their corresponding lengths was inversely plotted and fitted with an exponential function in the form of \( L = aP_{th}^{-c} \). This provided an analytical expression of fibre length as a function of peak power.

It has to be noted, that the above equations and relations are derived for cw-operation, and thus the applicability to pulsed operation has to be demonstrated as a final step before starting calculations. G.P. Agrawal [35] states that one can apply the cw-criterion as long as the effective length \( L_{eff} \) is taken to be \( \sqrt{\pi} \)-times larger than the walk-off length \( L_W \):

\[
L_{eff} = \sqrt{\pi} L_W \approx \frac{T_{\text{FWHM}}}{|v_{\text{gp}} - v_{\text{gs}}|}.
\]

(3.5)

Entering the group velocity values for the centre wavelength of 1035 nm, the first Raman peak at 1084 nm and a pulse duration of 250 fs results in a walk-off length of 8.5 cm, which, when multiplied with \( \sqrt{\pi} \) as in eq. (3.5), yields 15.1 cm. The optimal fibre length recorded in this thesis lies below this effective length, and thus the cw-based relations used above to estimate peak power and fibre length are assumed valid for pulsed operation. Considering that this model ignores the bandwidth and temporal broadening of the pump (and stokes) pulse, it furthermore acts as a lower boundary on the applicable lengths for the cw-criterion, as a broader pump pulse will overlap for longer time with the Raman pulse leading to a longer walk-off length.

Lastly, intrapulse Raman scattering can occur within the same pulse by amplification of low-frequency components acting as the Stokes wave through high-frequency parts that act as the pump-beam. For this to take place, the bandwidth must be large enough to cover the Raman-gain bandwidth. Furthermore, the involved frequencies must have not reached walk-off lengths that would terminate the process. For normally dispersive fibres and initially unchirped pump pulses that fulfil these requirements, the centre of the frequency shifts only 0.5 THz (~1.8 nm) [35] to longer wavelengths. For input bandwidths that require SPM to achieve the necessary width for intrapulse Raman scattering (e.g. broadening to over 50 nm), the reduced peak power after SPM-broadening further reduces this effect. Given the parameters of the pump pulse and fibre in this thesis, intrapulse Raman scattering therefore does not exhibit a relevant impact.

For the optimal fibre length according to eq. (3.3), the relevant parameters affecting the result are the material specific nonlinear coefficient \( \gamma \) and \( \beta_2 \), as well as the pulse dependent parameters of the peak power \( P_{\text{Peak}} \) and the input pulse duration \( T_{\text{FWHM}} \). For materials with lower \( \gamma \) and \( \beta_2 \), and for longer \( T_{\text{FWHM}} \), the optimal fibre length increases, while it decreases for larger \( P_{\text{Peak}} \). The fibre parameters of the LMA-40-PM-PCF can be treated equal to those of fused silica (as explained in chapter 2.2.1), the nonlinear index \( n_{nl} \) of which determines \( \gamma \) together with the effective area \( A_{\text{eff}} \) and the wavelength \( \lambda \).

Several studies on the \( n_{nl} \)-value of fused silica have been published at wavelengths of 1064 nm and 1053 nm (though not explicitly at 1035 nm). The results span over a range of values between
1.9·10^{-20} \text{ m}^2\text{ W}^{-1} \text{ and } 3.3·10^{-20} \text{ m}^2\text{ W}^{-1} [44,77–82]. More recent publications from 2015 and 2019 report a value of 2.23·10^{-20} \text{ m}^2\text{ W}^{-1} [83] and 2.19·10^{-20} \text{ m}^2\text{ W}^{-1} [84] both at 1030 nm. Using a code from NKT-employee Etienne Genier [85], the FROG-recorded field of the 250 fs pulse is propagated through GNLSE, including the relative intensity noise (RIN) of the laser and using the material and dispersion properties of fused silica with the effective area of the LMA-40. By running the code using the wide array of the above mentioned values for \( n_{nl} \), the best matching value for \( n_{nl} \) can be obtained by comparing the computed spectral output to an experimental one. The most accurate fit was obtained using the value reported by [79] of \( n_{nl} = 2.74·10^{-20} \text{ m}^2\text{ W}^{-1} \) at 1053 nm. With the value for \( n_{nl} \) fixed and \( \beta_2 = 18.606·10^{-27} \text{ s}^2\text{ m}^{-1} \) for fused silica taken from [44], the peak power dependent length according to eq. (3.3) can be calculated.

For the Raman threshold in eq. (3.4), the critical parameter is the Raman gain coefficient \( g_R \). Similar to \( n_{nl} \), several reports on \( g_R \) have been published at 1064 nm. They range from 0.74·10^{-20} \text{ m} \cdot \text{ W}^{-1} \text{ to } 0.92·10^{-20} \text{ m} \cdot \text{ W}^{-1} [86–88]. Since \( g_R \) has a strong impact on the Raman threshold, the highest value leading to the lowest Raman threshold is used. Additionally, scaling models [86,89] are used for a legitimate estimate of the Raman-gain coefficient at the centre wavelength of 1035 nm, resulting in the final value of \( g_{R,\text{high}} = 0.99·10^{-20} \text{ m} \cdot \text{ W}^{-1} \).

![Figure 3.6](image-url)

**Figure 3.6.** Optimal fibre length for quadratic compression using the classic formula which ignores higher-order effects (blue). The onset of undesired SRS is set at a 1 % threshold (red). The intersection lies very close to the experimentally used peak power of 1.41 MW (vertical dashed line) and a fibre length of 13.5 cm (horizontal dashed line).

With these values, two graphs are generated as shown in fig. 3.6. For low peak powers, the Raman threshold length lies far above the optimal fibre length derived through the classical approach in eq. (3.3). However, as the peak power increases, the LMA-Raman threshold length
crosses that of the optimal fibre length at around 1.5 MW, leading to difficulties when using a quadratic compressor. Therefore, launched peak powers above this intersection and up to the damage threshold of the fibre at 2.1 MW (explained in more detail in chapter 3.5) will not lead to the desired clean compressibility of the output pulse, despite a broader generated bandwidth. Instead, for the LMA-40-PM-PCF the optimal length and peak power combination from fig. 3.6 seems to be at 14.5 cm and 1.49 MW. Empirically confirming these initial theoretical estimations are the final settings used for compression of 13.5 cm fibre length and 1.41 MW peak power, which lie very close to the calculation.

The above analysis shows that choosing the highest possible power does not necessarily lead to the cleanest and shortest compression when using a solid-core PCF and a quadratic compressor, as additional factors need to be considered. This concludes the theoretical framework necessary to understand the pulse-compression experiments. The experimental outcome based on these calculations will be discussed in the next chapter.
3.4 Compression results

This chapter will cover the experimental results regarding pulse compression and their analysis. First is the PCF analysis, then the compression results from SF10 and SF11 prisms and overall system efficiency, the noise of the system, and lastly the spatial beam profile. For the source laser properties, please refer to chapter 3.

The first crucial step is coupling the beam into the fibre. By using the 200 mm focal length lens together with a beam diameter of 11.7 mm, the focussed beam waist is calculated to be 27 µm. This acceptably matches the MFD of the PCF of 32 µm, with the similarity of these two parameters being important for efficient coupling. With a Rayleigh length of 0.46 mm, the lateral accuracy of the three-axis fibre-stage is sufficient to accurately place the fibre facet into the beam waist. Nonetheless, launching the fundamental mode in the fibre with optimal coupling efficiency requires careful and precise alignment. The fibre-facet has to face the beam at an absolute zero-degree incidence angle, so that x-y movement will not shift the facet out of the Rayleigh length. This alignment was performed by hand, reducing accuracy. With the fibre facet in place and coupling optimised, the polarisation maintenance of the PCF as a function of peak power was recorded. This also yielded the coupling efficiency of the system, as any light coupled to the core will retain its polarisation state at the exit, while any light that might propagate in the cladding will undergo a polarisation change. For this, the ratio of the P-polarised output of the fibre to the input beam power is recorded as the input power is changed through the variable attenuator. As shown in fig. 3.7, the ratio achieved lies between 46 % and 50 %, with a tendency to lower values at increased input power. With an increasing power, the intensity at the edges of the Gaussian beam also increase. This enhances the effect caused by the mismatch of the intensity edges between the hexagonal intensity profile around the core and the Gaussian input mode, while the centre of the beam is equally well guided at higher power.

Earlier publications on smaller, non-polarisation maintaining PCFs (a 10 µm core [90] PCF, and a PCF with MFD of 16 µm [91]) at similar wavelengths (1064 nm, 1030 nm) show a better coupling efficiency of 60 %. More recent publications demonstrate 80 % [92] and 90 % at a MFD of 26 µm [64], both at 1030 nm (though the exact experimental method and coupling efficiency definition are not given). These efficiencies lie within the range of simulated coupling efficiencies shown by Nahar et al. [93], where free-space coupling into at LMA-35 PCF was calculated\(^5\), though at 1550 nm. The publication emphasises the importance of tilt-angle, e.g. the offset from a perpendicular beam path to the fibre-facet, which quickly reduces coupling efficiency with an increasing offset angle. For example, a 0.2° offset results in a -5 dB loss (or only ∼30 % coupling efficiency). The exact matching of the Gaussian free-space diameter with the MFD of the fibre, as well as the lateral displacement from the core play a smaller, yet still relevant role. The tilt-angle is connected to the acceptance angle, and due to the low $NA$ of LMA-PCFs, this exacerbates the coupling losses when mismatched. The optimally computed and measured coupling loss in the publication by Nahar et al. [93] lied at ∼-2.15 dB, corresponding to a coupling efficiency of

\(^5\)The coupling efficiency was defined as the ratio of output to input power of a fibre, and thus includes the entry facet, propagation absorption, and exit facet losses.
3.4 Compression results

Figure 3.7. Coupling efficiency for the LMA-40-PM-PCF vs. input peak power. An average efficiency between 45-50 % is achieved, but could be improved.

61 %. By using additional field lenses, it was improved to -1 dB (70 % coupling efficiency). A subsequent publication by the same author addressed the optimisation in detail using several lenses to increase coupling efficiency [94]. The improved value (ignoring the lenses’ Fresnel reflections) reaches a theoretical minimal coupling loss for the LMA-35 of -0.5 dB (or 89 % coupling efficiency). It also suggests an interesting approach through surface manipulation of the collapsed end-cap of the fibre to remove the need of the optimisation lenses altogether [94].

In practice, the focussed beam suffers from aberration as well as mode mismatch between the Gaussian free-space mode and the fibre-mode, which are the two main loss-contributions apart from Fresnel reflections. Compared to the just mentioned coupling efficiencies with similar LMA-PCFs, the result achieved here seems to be at the lower end of potential efficiency. The difference can be further explained through the polarisation maintaining nature of the fibre coupled with the lack of a high-precision rotation option (for the experiments, the fibre was rotated manually by hand until acceptable polarisation output was recorded after the Wollaston prism). This results in some loss due to polarisation-alignment mismatch, adding to the general alignment issues of the stage and the fibre position. The beam characteristics of the source laser (especially the mode-size-mismatch and the $M^2$ value of 1.2 differing from a perfect Gaussian shape by 20 %) together with additional Fresnel loss of 3.37 % from measuring the power behind a Wollaston prism further explain the discrepancy. The high efficiencies reported above do not elaborate in detail on the methods used. Presumably, total power was measured at the exit of the fibre, possibly including potential cladding contributions that are filtered out in the setup presented for this thesis. Lastly, with the low propagation loss of the fibre, the coupling efficiency remains equal for all fibre lengths tested (39 mm to 185 mm).
3.4 Compression results

Figure 3.8. Input (red) and output (blue) spectrum after the LMA-40-PCF at input values of 1.41 MW and 250 fs. A tenfold increase of bandwidth is achieved. The -10 dB spectral feature below 960 nm results from OWB, while the centre of the spectrum is dominated by typical SPM oscillations.

With the PCF set in place for optimal coupling and P-polarised output, the impact of peak power on the output bandwidth was recorded and produced results as expected from theory. With increasing peak power, the generated bandwidth also extends for a given fibre length, or with increasing fibre length for a given peak power (see fig. 3.4). Taking the final set peak power of 1.41 MW, the bandwidth extends over 181 nm at -20 dB, centred at 1035 nm, as shown in fig. 3.8, or ∼94 nm at -3 dB (equivalent to FWHM).

A FROG scan of the uncompressed output pulse was not possible, as the time-bandwidth product was too large in the uncompressed state for the settings and algorithm to catch both the full spectral bandwidth as well as the entire pulse duration. Hence, to estimate the rough spacing between the prisms, the pulse was monitored through the IAC while positioning the second prism at set distances and recording the pulse duration (no adjustments to prism insertion were applied at this point). Plotting the pulse durations vs. the distance of the prisms will show a curve with a single minimum, indicating the (rough) optimal prism separation distance to compensate for the acquired GDD in the fibre. Once at this position, the insertion of the second prism was altered for fine-tuning the GDD compensation for the shortest possible pulse duration. Initial experiments were conducted with SF10-prisms, resulting in a GDD compensation of ∼6646 fs² and a TOD compensation of ∼13364 fs³.

Figures 3.9a) and 3.10a) show the FROG results for the best compression achieved with SF10 prisms, which was also the pulse used in the first publication of this PhD [95]. The temporal structure of the pulse indicates that it contains a still significant amount of higher-order dispersion that has not completely been compensated for. Also, the large separation distance of ∼830 mm
between the prisms and their relatively small size lead to spatial clipping of the beam at the far prism. To circumvent both problems, SF11-glass prisms were introduced to the setup. Since its GVD is stronger than that of SF10, the prism separation required for compensating an equal amount of GDD as with the SF10 prisms could be reduced to ∼480 mm, while at the same time compensating stronger for TOD of -15505 fs$^3$. Furthermore, the larger size of the SF11-prisms allowed for clipping free operation and thus a better power performance. The resulting improved pulse duration and FROG-trace are shown in fig. 3.9b and fig. 3.10b. By calculating the GDD and TOD resulting from the PCF (2512 fs$^2$ and 5618 fs$^3$) and adding it to the prism values, one can determine the residual phase implemented by the fibre-laser itself. This results in a GDD of 4134 fs$^2$ and a TOD of 9887 fs$^3$ acquired by the pulse before the PCF, i.e. the laser oscillator and amplifier rod.

**Figure 3.9.** Comparison of the reconstructed FROG traces of the a) SF10 and b) SF11 prism compressed pulses. A 20 THz offset corresponds to ∼72 nm. The SF11 result shows more spectral intensity along the temporal centre of the pulse.

The fitted curves in fig. 3.10 show an ideal sech$^2$-shaped pulse over the central part of the pulse with the SF10-prism compression yielding 30 fs, and the SF11 compression 22 fs (the values are in fact taken from analysis of the IAC traces in fig. 3.11, since the FROG induces more GDD during the measurement resulting in a slightly lengthened pulse duration). Taking the area of the time trace intensity, i.e. the pulse energy (power integrated over time), and comparing it to the area of the sech$^2$-fit over the central peak as seen in fig. 3.10, reveals the amount of energy contained in the centre-peak. For the SF10 prism compression this value lies at 69 % (though the
fit is not ideal), while for the SF11 prism it lies at 60%. Assuming an equal total pulse energy, the temporal widths of 22 fs (SF11) and 30 fs (SF10), and the just mentioned central-peak content, the SF11 pulse shows a peak power advantage of over 17% compared to that of the SF10 pulse, despite the lower power content in the central peak. In fact, due to the lower loss with the larger SF11 prisms, the actual total pulse energy is 0.345 $\mu$J compared to the 0.25 $\mu$J of the SF10 compression. This leads to a peak power advantage of the SF11 prisms of 64% over the SF10 compression, with peak power values of the center-pulse of 8.28 MW (SF11) and 5.06 MW (SF10). The residual FROG error for the SF11 compression is 0.0177 and 0.0090 for the SF10, where the FROG error is the root mean square difference between the measured and retrieved traces divided by the number of points in the trace.

![Figure 3.10](#)

**Figure 3.10.** Temporal traces obtained through FROG after compression with a) SF10 and b) SF11 prisms. The SF11 compressed pulse shows a cleaner profile that is more closely resembled by a sech²-shaped pulse (dashed black) and has less developed side-lobes compared to the SF10 result. The insets show a zoomed-in area of the phase, with the SF10-trace having an undesirably steeper gradient than the SF11-trace. The temporal FWHM are 30 fs (SF10) and 22 fs (SF11).

As a result of this strong performance increase of the SF11-prism and also the slightly cleaner temporal structure, the SF10 prisms were disregarded for future setups and experiments. It should be noted again, that this compression setup is a simple quadratic compressor, and thus the prism distance and insertion is dominated by the GDD one wants to compensate for. Consequently, any higher-order phase compensated by the prisms is a fixed value. This leads to the temporal structures at the leading and trailing edges of the pulses, acting as pre- and post-pulses of lower
intensity. Nonetheless, the phase across the centre peak of the pulse is almost linear at a value of 0, with the SF11 showing a less steep residual phase, meaning that overall phase compensation across the centre-peak is improved. Since the SHG-FROG used here has time-ambiguity, it cannot be said which side of the pulse is the leading or trailing edge (regrettfully, experiments to determine this by placing pieces of glass into the beam path were not conducted). Cleaner pulses can be obtained with more elaborate compression setups [55] involving individually tuneable cubic phase compensation.

![Graphs showing SF10 and SF11 compression results](image)

**Figure 3.11.** Comparison of a) SF10 and b) SF11 prism compression results recorded as IAC traces. The SF11 result is narrower and the side-lobes are less developed, which both are indications for cleaner compression. The insets show the few-cycle centre-peak from -60 fs to 60 fs.

The impact of the satellite pulses becomes more clear when viewing the IAC traces in fig. 3.11, rather than in fig. 3.10. Any side-pulses lead to structuring of the IAC trace next to the main pulse in a symmetric way. The stronger these side-lobes show in the IAC trace, the higher their impact is on the pulse, as they contain more energy. In conclusion, this means that a pulse with low side-lobes will therefore have a less structured pulse with reduced energy per side-lobe. This is important, as a strong pre-pulse can already induce nonlinear-effects in a material that will then interfere with the following main pulse. The SF10 IAC trace shows such a stronger side-lobe influence compared to the SF11 IAC trace. Furthermore, the SF11 IAC trace has a better peak to background ratio (an ideal IAC has a ratio of 8 : 1) and shows a better symmetry behaviour. Both observations indicate a better alignment and cleaner pulse structure.
Overall, the system power loss lies at \(\sim 60\%\), with 86\% of that resulting from the fibre coupling process. Comparing the peak power launched into the fibre to the peak power achieved from the entire pulse after compression, there is an almost tenfold increase from 1.41 MW to 13.8 MW, clearly justifying the external pulse compression. Even when taking only the peak power across the centre peak of the compressed pulse of 8.28 MW, a significant peak power increase that compensates for the loss of power is demonstrated.

Since the side-lobes are assumed to be resulting from non-compensated higher-order phase contributions, a theoretical verification of this can be achieved by adding a set amount of higher-order dispersion to the recorded complex FROG trace, ideally removing the side-lobes. Through eq. (3.6) one can simulate this by altering different values of each component of the Taylor expansion in the exponent of the additional phase, which is mathematically provided by adding a phase component to the complex field in the frequency domain and then applying FT to convert back to the time-domain.

\[
E'(\omega) = E(\omega) e^{-i \left( \beta_0 + \beta_1 (\omega - \omega_0) + \frac{1}{2!} \beta_2 (\omega - \omega_0)^2 + \frac{1}{3!} \beta_3 (\omega - \omega_0)^3 + \frac{1}{4!} \beta_4 (\omega - \omega_0)^4 + \ldots \right) L}.
\] (3.6)

The frequency dependent propagation constants \(\beta_i\) for the simulation are taken from a known material’s dispersion profile, e.g. fused silica, and by changing the simulated length \(L\) in the exponent, the additional phase can be altered. Opposite signage for the values of \(\beta_i\) needs to be taken, since the idea is to compensate for higher-order phase contributions, not to add more. The first two components can be set to zero, as they do not change the pulse shape (see eq. (2.8) on page 14). Furthermore, one can set any element of the expansion to zero to see the effect of only one contribution, e.g. only the TOD. If the side-lobes are indeed a result of one specific higher-order dispersion, this method should allow to remove them from the trace. The obtained optimal result using eq. (3.6) is shown in fig. 3.12. Disappointingly, the pulse shape still exhibits side-lobes.

In theory, the phase can also be modelled with a high-order polynomial fitted to the data recorded by FROG (see the phase in fig. 3.10). Multiplying the phase with the inverse of the fitted function would set the phase to a constant value. However, finding a suitable material that applies a phase contribution equal to the inverse of the recorded phase is not trivial, and even the fitting process can be very complicated. This stems in part from phase-noise being amplified by nonlinear effects such as SRS or four-wave-mixing.

The theoretical approach of verifying the assumed origin for the side-lobes shows that the limiting factor for the pulse compression achieved here results from higher-order residual phase and amplified phase noise in the system. These factors can not be adequately addressed by using the dispersion profile of fused silica (other material’s dispersion profiles were not applied). An additional aspect are several equal-frequency components at different points in time, as seen in fig. 3.9. By changing the phase of a particular frequency component, one applies this phase shift to the multiple equal-frequency appearances across the pulse duration. The difference in phase of these equal-frequency components will therefore remain the same. Multiple equal-frequencies can result from several different nonlinear effects, for example through OWB, SPM,
Figure 3.12. The measured 22 fs pulse’s temporal intensity profile (blue) compared to the same pulse after simulating additional phase compensation (assuming the opposite dispersion of fused silica). The computed trace shows slightly less energy in the side-lobes, but a certain amount cannot be removed.

SRS-shifted frequency or four-wave mixing. Each method generates frequency components individually and thus also with an individual phase that is not necessarily equal to that of the other generation methods. Hence, it becomes clear that the pulse modified through the LMA-PCF and high peak powers in this thesis is far more complex than initially assumed and thus cannot be compressed to an ideal textbook-shaped pulse.

To determine the pulse-to-pulse stability of the pump laser and possible introduction of intensity noise from the compression stage, spectrally resolved RIN measurements using a large bandwidth photodiode (Thorlabs: DET08CFC, 5 GHz) and a fast (4 GHz) real-time oscilloscope (Teledyne: LeCroy-HD09404) were conducted \[96, 97\]. At first, the average voltage response of the photodetector was recorded with the oscilloscope at different input-power values from 3 µW to 100 µW in order to determine the linear-response regime of the detector. The power was measured with a photodiode power sensor (Thorlabs S122C). The resulting linearity curve in fig. 3.13 shows that the linear response regime is valid for input powers below 20 µW, corresponding to an average voltage response between 50-450 µV. Subsequently, for each measurement to lie within the linear response regime of the photodetector, the power was adjusted to receive a voltage response in this band.

A total of 10005 pulses per measurement were recorded as a voltage versus time trace with the peak of the individual pulses in the oscilloscope reading being proportional to the integrated energy of the pulse. At every 25 nm of the spectra, both before and after the compression stage, 10 nm full-width half-maximum (FWHM) band pass filters were used for direct narrow-band spectral comparison. The nine resulting spectrally limited RIN measurements from 950 to 1150 nm
3.4 Compression results

Figure 3.13. RIN measurements. a) Linearity curve fit (red) to the oscilloscope’s voltage response (blue rings) and b) spectrally resolved RIN (blue) after spectral broadening in the LMA40-PCF (output spectrum in red). The RIN increases towards longer wavelengths due to SRS.

are shown as blue circles in fig. 3.13b). By plotting a histogram of the recorded voltage-response and fitting a Gaussian distribution, the RIN of the filtered spectra is determined by dividing the standard deviation of the data through its mean value. The overall source laser RIN was measured to be 1.10 %, whereas the uncompressed output at the exit of the fibre shows an (unweighted) overall RIN of 1.74 %. This suggests only a relatively small noise contribution from the nonlinear spectral broadening process. After passing through the prisms, the compressed output has an average RIN across its full bandwidth of 1.79 %. The very slight increase over the uncompressed output may be a result from additional noise-contributing nonlinear effects resulting from the now compressed pulse passing through the (fourth) prism material at full power. Also, effects within the optical fibre used to couple the light to the detector could have an impact due to the shorter pulse duration and higher peak power of the pulses. Since the measurements are taken over several thousand pulses, statistical error is minimised, while a systematic error of the photodetector cannot be ruled out. This is why attention is given to the uncompressed output pulse’s RIN for further analysis.

Starting from the low wavelength end at 950 nm in fig. 3.13 it can be seen that the spectral intensity arising from OWB has a similar RIN as the central part of the SPM pulse. Since OWB is inherently connected to SPM, the RIN therefore is expected to lie in a similar range. The next measurement at 975 nm shows a larger value, perhaps resulting from the lower signal-intensity at
3.4 Compression results

the spectral valley present here, which is the remaining spectrum at the edge of the SPM that is
not transferred to the OWB peak at 950 nm. The RIN for the central part of the SPM curve up to
1075 nm continues to remain low, with the exception at 1025 nm also attributed to the signal-valley
present at that wavelength due to destructive interference of the SPM. When approaching the
longer, red-shifted wavelengths beyond 1075 nm, the frequency content is now also affected by
SRS, with a clear increase of the RIN towards longer wavelengths. Following this analysis, fig. 3.13
demonstrates that the central part of the pulse with the majority of the spectral intensity has a
fairly low RIN. Thus, by filtering out the peripheral red- and blue-shifted frequency components
(i.e., bandwidth outside the range 1.000–1.075 \(\mu m\)) before compression, a reduced RIN value of
\(\sim 1.3\%\) is obtained. This pulse compression method can therefore be considered as a low-noise
process. If required, such filtering is fairly easily achieved in a prism-based setup by placing e.g.
knife-edge beam blocks in front of the second prism where the pulse is spread out horizontally
along its frequency components.

![Figure 3.14. Spatial intensity profile of the beam taken at the collimated fibre output in row a) and after the compression stage in row b). The Gaussian mode profile is preserved (red fit to yellow data).](image)

Lastly, the beam-quality of the spatial mode is assessed and shown in fig. 3.14. The collimated
output from the fibre shows a clean Gaussian profile, as does the profile after the compression
stage, though the latter has a slight intensity distortion. Due to the sensitivity of the IR-camera
and the short-pulse duration, the beam had to be heavily attenuated before measurement. This,
together with the not perfectly angle-matched prisms, might explain the lower quality of the
compressed pulse, though the Gaussian form is still clearly dominant. The images were taken at
unspecific positions in the set-up, so the resulting diameters shown have no specific relevance for
the set-up.
3.5 Solid-core limitations

Solid-core fibres have comparatively strong mechanical features over hollow-core options, are more economical and easier to manufacture, and offer a less complex environment for nonlinear processes as they do not require gas cells. On the downside, the peak power that can be propagated through solid-core fibres is ultimately limited by self-focussing, the process being explained in chapter 2.3, page 24. In a report on LMA PCF fibres, Seidel et al. have shown the potential peak power capacity to increase with larger core diameters [36], while peak irradiance decreases. Interestingly, the peak powers reported are well below the self-focussing threshold for fused silica at the given pump-wavelength (≈4 MW [48]), with a maximum peak power of just below 2 MW for the LMA-35 PCF (which has a core diameter of 35 µm). In the report, the authors attribute the damage to be accelerated by "(sub-)critical self-focusing" [36], but fail to provide any deeper analysis on the damage. The images provided are only input-facet images rather than also side-view images that could show damage-propagation effects. For the PCF used in this thesis with its core diameter of 40 µm, the damage threshold was found to be at 2.11 MW, much in line with the reports from Seidel et al. The method used to document this was to gradually increase the peak power and monitoring the polarised output power of the fibre-core through the Wollaston prism until a drop in power was observed. The last stable power reading before the damage occurrence was noted down as an upper limit. Additionally, a cracking-sound and bright light at the fibre entrance would occur when the damage threshold was reached.

The self-focussing threshold is independent of the initial beam-area and provides an upper damage limit for an ideal fibre. Experiments on focussing the pump laser into bulk fused quartz at distinct powers confirmed this threshold, showing damage occurring at peak powers between 4.4-4.9 MW. SF and the resulting beam collapse is an effect that requires a certain propagation distance, which reduces for higher power. It additionally needs to counteract the divergence of the beam, which explains the observed damage threshold above the theoretical value of 4 MW for this wavelength. Despite this slight discrepancy for the damage observed in bulk fused silica, the difference of over 2 MW in peak power between recorded damage threshold in the fibre and that of pure fused silica hints at a different damage mechanism. The most common and destructive damage method in optical fibres is termed the fibre-fuse effect. It will be described in more detail henceforth, based on an excellent summary by Raman Kashyap [98] who reported on the effect in 1987.

Generally speaking, a fibre fuse is a catastrophic, backwards propagating damage mechanism occurring in solid-core optical fibres, and is connected to temperature dependent absorption rather than self-focussing [99]. It expresses itself as a bright plasma plume propagating towards the input-laser source at several metres per second, destroying the guidance properties of the fibre along its path. Self-focussing (and to some extent thermal lensing), on the other hand, leads to a beam-collapse with high intensities allowing for laser-induced breakdown of the material. Its resulting damage pattern is localised to this point of propagation and therefore self-focussing does not explain the continuous propagation of the plasma within the fibre. This

---

6Defined as peak power in the fibre after which damage occurred.
3.5 Solid-core limitations

is emphasised by the fact that the plasma observed during fibre-fuse reaches temperatures of several thousand degrees Celsius, well beyond the break-down and melting point of SiO$_2$. This leads to a strong decrease of the refractive index that would terminate any further self-focussing effects. Hence, self-focussing can potentially contribute to the process in the initial stages of fibre-fuse, but is not the direct cause.

Instead, the absorption profile of SiO$_2$ provides the explanation, as shown in figure 7 in reference [98]. It increases exponentially after a specific temperature is reached that is well above operating temperatures for standard telecommunications applications. Once this boundary threshold is overcame, the heat gain through absorption outweighs the heat loss through conduction, convection, and radiation. This means it is self-feeding, as increasing absorption will lead to higher temperature and even higher absorption, and thereby also self-sustaining when given enough propagation material available for absorption. The rapid heat increase leads to the formation of a plasma plume that radially extends, i.e. also towards the source laser. This moves the point of maximum absorption in that direction and forces the plasma to travel along the fibre towards the source. The trailing end of the plasma is thereby starved of additional power, cools down, and detaches from the main plasma plume in a periodic fashion and distinct shape, leading to a “bullet train” as seen in fig. 3.15, with molecular oxygen trapped inside [98]. The shape of the damage bubbles becomes shorter and more asymmetric (i.e. bullet shaped) with shorter pulse duration due to rougher “surface-tension” effects at such short durations and high intensities. The process eventually terminates once the plasma runs out of fibre or if the fibre diameter is strongly and abruptly increased [100]. In the latter case, the feeding laser mode is also increased, leading to a smaller power fraction being absorbed by the plasma.

Interestingly, the initiation point of the fibre-fuse can be triggered at will. In general, it requires a localised increased absorption, which corresponds with guiding-loss. For example, strong bending of a fibre and the resulting bending loss can act as such a fuse-point, while other types of micro-stress lead to the same effect. Furthermore, external heat application, for example through radiation or a heat source, can also act as starting points. In the case of the setup mentioned for the compression experiments, the fibre-fuse point was repeatedly starting a few millimetres behind the input facet, where the (polymer-stripped) fibre was clamped down by the fibre-clamp (for un-stripped fibres, the coating would catch fire at far lower peak powers). Albeit that the clamping pressure was set to a minimum, the slight force induces micro-stress in the fibre, increasing the guidance loss at that point. Hence, at the given peak power of 2.1 MW (or 525 GW·cm$^{-2}$ in the fibre-core), the absorption in the core was stronger than the heat-loss processes, and the self-feeding absorption increase started, leading to the fuse-initiation and subsequent propagation towards the fibre entry facet. The long plasma-plume is shown in fig. 3.15b) at the beginning of the fibre, where further propagation is no longer possible. Along its previous path to the right, the typical bullet shaped detachments from the starting point are clearly visible in the microscope image$^7$. The damage threshold and pattern reported here was reoccurring for any fibre that experienced this violent fate. Therefore, a peak power of 2.1 MW sets the limit for the fibres used in this experimental setup.

$^7$Figure 3.15b) shows only the last segment of the destroyed fibre. The plasma bullets continue in the fibre core for several millimetres
3.5 Solid-core limitations

Figure 3.15. Microscope images of the LMA-40-PM-PCF after a fibre-fuse. a) Front facet with visible damage in the centre of the core between the stress rods. b) Characteristic bullet shaped pattern of plasma plumes detaching from the main propagation plasma which blasted material out of the entry fact.

In an approach to reduce the high fibre-loss in the initial segment of the fibre, the facets of several fibres were collapsed, as seen in fig. 3.16a) - some only at the entry facet, and some also at the exit-facet. The idea behind this is that the input angle of the incoming light from air to the collapsed fused-silica fibre-facet is reduced before entering the fibre-core, leading to a better coupling efficiency. For comparison, the LMA-40 was also spliced to a standard silicon fibre (fig. 3.16b) to increase the $NA$ of the input-facet (and thus the acceptance angle). This also was aimed at helping with handling issues of the collapsing method. The fibres with collapsed input- and end-facets led to improved coupling efficiencies with an average value of 67%. Fibres only collapsed at the input-facet showed an average coupling efficiency of 58%. It must be noted that these measurements did not use the Wollaston prism method to exclude the p-polarised core-light as described earlier, but instead were taken at a larger distance away from the end-facet. The assumption here being that the cladding light will diverge more strongly and therefore not be recorded by the power meter. This also helps to explain the higher coupling efficiency for the double-collapsed fibres, since the exit-light will experience a more narrow divergence between cladding and core light. This probably lead to inflated results, since the end-facet should no effect on the input-facet coupling efficiency.

The main reason why the collapsed fibres were not used in the final configuration was the difficult reproducibility of equally collapsed facets. The filament and machine used for collapsing the holes is typically applied to splice two fibres. In the collapsing configuration (e.g. with only one fibre), the initial position of the fibre was difficult to reproduce, as fibres were loaded manually onto the motorised stage. Despite equal filament power and process duration, the resulting outcome would largely differ from fibre to fibre. Some fibres were exposed too much to the filament-arc, leading to rounded end-facets that refract the light in an undesirable way, while others were too far away from the arc to collapse the holes. Combined with a limited supply of the LMA-40 fibre, a “trial and error” approach to sort out a few promising splices was not taken. In contrast to the collapsed fibres, the fibres spliced to a standard silicon fibre showed much lower coupling efficiency of only 29%, despite the larger $NA$ of the input-facet, attributed to the large MFD mismatch when the light propagates from the silicon into the PCF core (a tapered fibre-solution was not experimented with). Lastly, due to the abundant power available from the laser combined with the inherent limit of the solid-core fibre in terms of damage, the 50% coupling efficiency of bare fibres was sufficient, and no long term degradation due to dust or dirt entering the air-holes was observed either (negating the protection aspect of collapsing holes). This is not to say that
3.5 Solid-core limitations

Figure 3.16. Microscope images of the LMA-40-PM-PCF. a) The collapsed fibre facet (compare to fig. 2.2). b) The LMA-40 (right) spliced to a silicon-fibre (left). The opaque centre area is the spliced region. c) Side-view of a cleaved LMA-40. d) Side-view of a collapsed fibre with the air-holes being terminated shortly before the end of the fibre.

collapsing the end-facet(s) of PCFs does not yield improved coupling, but within the scope of this thesis and the limited technical knowledge and support on splicing and collapsing LMA-fibres, a thorough and detailed study on this topic was not conducted.

This concludes the analysis of the experimental data obtained with respect to the LMA-40-PM-PCF and the compression of 250 fs pulses. The following chapters will introduce applications of such short pulses, specifically through optical rectification to generate THz-radiation.
Part II

Terahertz radiation
CHAPTER 4
THz-generation, detection, and applications

Part of the content in the following chapter(s) is reproduced from [T. O. Buchmann et al. “High-power few-cycle THz-generation at MHz repetition rates in an organic crystal”, APL Photonics 5(10), 106103 (2020)], with the permission of AIP Publishing and from [T. O. Buchmann et al. “MHz-repetition-rate, sub-mW, multi-octave THz-wave generation in HMQ-TMS”, Optics Express 28(7), 9631 (2020)], with permission of the Optical Society of America

4.1 Applications and THz-TDS

Over the past two to three decades, THz-applications have seen increased attention. For example, many airports world-wide have implemented millimetre scanners that operate with low frequency THz-radiation, which is just one practical implementation of THz-radiation for imaging purposes. In laboratories, the most common application stems from THz-TDS. This technique allows for detailed material analysis, but the strong field strengths that can be produced have also led to the development of new fields, such as THz-scanning tunnelling microscopy (THz-STM [22]). In telecommunication applications, THz-generation and technology is a sought-after technology for higher data-transmission networks, though there are still several issues for free-space transmission due to atmospheric influences. Accompanying this field are various suitable THz-waveguides, which are still at an early development stage [101]. However, promising fast optical modulation using THz-waveguides is currently being researched [102], which might prove to be vital in areas without atmospheric limitations, e.g. in space. Satellite-to-satellite communication using THz-radiation is an interesting candidate for large data-transmission bandwidths [103]. The list of potential applications can be continued, for example with layer-thickness analysis through time-of-flight measurements or medical applications. A recently published and profound analysis covering the industries from “[...] polymers, paint and coatings, pharmaceuticals, electronics, petrochemicals, gas sensing, and paper and wood [...]” can be found in [104]. The most thesis-relevant and established technique is that of THz-TDS, which will be explained in detail in this chapter, while a more thorough overview of potential THz- and THz-TDS applications can be found elsewhere [105–107]. As a side-note it should be stated that the discussion regarding THz-radiation in this thesis assumes pulsed rather than cw sources.
THz-TDS is a powerful tool for the investigation of samples (solid, liquid, or gaseous), reported on in 1989 by measuring the absorption and dispersion of water-vapour in the low THz-frequency regime [19]. The technique requires ultrafast laser pulses that are shorter than the THz-pulse that is to be measured, giving incentive for the external pulse compression in the first part of this thesis. The main advantage of THz-TDS, other than accessing the THz-frequency range, is the fact that it is a coherent detection scheme for mapping the electric field of the THz-pulse in time. When applying a Fourier transform (FT)\(^1\) to the recorded time-domain signal \( \mathcal{E}(t) \) to transfer it into the frequency domain \( \mathcal{E}(\omega) \), one obtains the THz-pulse amplitude component \( A(\omega) \) and the phase component \( \exp(i\phi(\omega)) \) with respect to frequency, i.e. the full description of the complex electrical field in the frequency domain:

\[
\frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \mathcal{E}(t) e^{-i\omega t} dt = \mathcal{E}(\omega) = A(\omega) e^{i\phi(\omega)}. \tag{4.1}
\]

From the reference \( r \) and sample \( s \) traces recorded with THz-TDS one can extract the phase-change \( \Delta \phi = \phi_s - \phi_r \) experienced by a THz-pulse that travels through the sample material. By dividing the two traces through each other, this phase-change can then be used for further analysis. It varies depending on the exact setup and sample structure one is investigating, as the system response is governed by various transmission, reflection, and absorption effects. A well written tutorial on this is given by Schmuttenmaer et al. [108], which the reader is referred to for more information. The division yields:

\[
\frac{E_s}{E_r} = \frac{A_s(\omega)}{A_r(\omega)} e^{i(\Delta \phi(\omega))}. \tag{4.2}
\]

With the help of THz-TDS, one can therefore access several interesting material parameters, such as:

- The refractive index in the THz-regime, given that the thickness of the sample is accurately known.
- The thickness of a sample, given that the THz-refractive index is known.
- The extinction coefficient and thereby the frequency dependent absorption.
- The permittivity of a sample [109].
- The conductivity of a sample such as graphene [110].

A commonly-used THz-TDS approach to estimate the refractive index \( n \) and absorption coefficient \( \alpha \) of a known-thickness sample is practiced through equations (4.3) and (4.4), assuming a reference trace taken in air. The most important assumption is the negligible phase change at the Fresnel entrance and exit interfaces of the sample (i.e. no complex refractive index for the

\(^1\)When working with FT and THz-TDS, one needs to pay careful attention to the sign-convention and normalisation factor used in the FT and by numerical programs.
transmission coefficients). With this, the transmission coefficients become real-valued, allowing for the equations:

\[ n(f) = n_{\text{air}}(f) + \frac{1}{d} \frac{\Delta \phi}{2\pi f}, \]  
\[ \alpha(f) = -\frac{2}{d} \ln \left( \frac{E_s}{E_r} \left( 1 + n(f) \right) \right). \]

The frequency dependent refractive index \( n(f) \) here depends on the recorded phase change and sample thickness. The absorption coefficient is dominated by the field transmission term \( \frac{E_s}{E_r} \), as the refractive index fractional term is close to unity for refractive indices between \( n = 1 \) and \( n = 3 \). For eq. (4.4) to be valid, it must be assumed that the phase acquired by the THz-pulse during the transmission of the bulk part of the sample is orders of magnitude larger than the phase change acquired from the Fresnel transmission coefficients in and out of the sample. This is often the case e.g. when analysing optical materials or chemical substances such as narcotics or explosives [108, 111]. Such samples have low reflection and sufficient thickness. This assumption would be invalid, however, for very thin samples with etalon effects or materials with strong reflection such as metals, where the phase change is dominated at the Fresnel interface (and where transmission measurements would not be feasible to conduct). After calculating the refractive index and from it the absorption coefficient, one can then calculate the extinction coefficient \( \kappa \), e.g. the imaginary part of the refractive index, through eq. (4.5):

\[ \kappa(f) = \alpha(f) \frac{c}{4\pi f}. \]

After this, one must verify the initial assumption of \( \kappa \) being much smaller than the real part of the refractive index \( n \) by comparing the two parameters. If the difference is large in favour of real part, then the initial assumption is justified.

Despite being a useful and fairly straightforward method to determine material properties, there are some drawbacks to the THz-TDS method. These mainly include experimental parameters and system stability, since the analysis requires identical THz-pulses throughout the entire measurement process. For small changes to the system, such as thermal drift from the source laser or alterations to the environment (e.g. the relative humidity), the resulting THz-pulse mismatch may result in statistical error that is acceptable for many applications. However, especially long-duration \(^2\) or thin-sample measurements suffer greatly from changes to the THz-pulse. This results in an increased error on the acquired data, which is why THz-TDS should be conducted in the shortest possible time window. Additionally, statistical error should be reduced by acquiring and averaging the data of multiple measurements. Consistent with theme of this thesis, both aforementioned issues can be addressed by using MHz sources, as they inherently mitigate source fluctuations due to the high repetition rate and provide thousands of pulses in the time span of the lock-in integration. Also, faster scan speeds than those conducted with low repetition rate sources are possible, reducing precious measurement time. Such a MHz THz-source is inherently

\(^2\) Several minutes for a single scan can be necessary due to long lock-in integration times to reduce noise and/or for very long scan ranges.
linked to high-power MHz laser sources, such as the fibre-laser applied in this thesis (see chapter 3.1).

Another drawback is the real-world presence of humidity, as dry air is typically only available in controlled laboratory environments. The strong water absorption in air makes experimental propagation distances above a few tens of centimetres or metres unfeasible (depending on the application) [105], while detection in reflection geometry often further reduces the amount of signal that can be detected. Lastly, the water absorption bands might also coincide with desired frequency information for a specific material, which then cannot be extracted.
4.2 Generation of THz-radiation

What has slowed the development of THz-technology over a long time was, and to some extent is, the absence of suitable, low-cost, and stable THz-sources alongside a lack of detectors. Over the decades, a few distinct physical techniques have emerged as feasible and reliable generation methods. They will be briefly introduced hereafter, with the method of optical rectification being analysed in more detail. An elaborate overview and more detailed introduction to several generation methods, with an emphasis on tilted-pulse front techniques, can be found in a recently published review by Kampfrath et al. [112].

4.2.1 Optical rectification

For a weak electric field $<10^5 \text{ V} \cdot \text{m}^{-1}$ [113] incident on an atom or molecule, the reactionary electron movement (and with it the induced polarisation $P$) behaves as a harmonic oscillator, confined to a symmetric potential. Therefore, no nonlinear optical processes are observable. This changes when the electric field becomes sufficiently large to distort the potential and allows for a nonlinear motion of the electrons leading to a nonlinear induced polarisation. These field strengths can, for example, be achieved by focussing a short laser pulse onto a small area of a given material. The field threshold for this to take place is not clearly defined and varies over several orders of magnitude depending on what is considered as the onset of a nonlinear reaction. One estimation method requires it to be on the order of the lowest nonlinear expansion term of the susceptibility so that it becomes comparable to the linear term in magnitude, e.g. at applied fields on the order of the characteristic atomic field strength of approximately $10^{11} \text{ V} \cdot \text{m}^{-1}$ [37]. Another estimate can be derived from the value of the nonlinear coefficient, which requires a sufficiently high intensity to significantly change the overall refractive index. As the field relates to the square root of the intensity divided by the speed of light and the vacuum permittivity, $\sqrt{I/(\epsilon_0 c)}$, this leads to required field strengths of $>10^{10} \text{ V} \cdot \text{m}^{-1}$ for most commonly used materials. For non-centrosymmetric material structures, strongest nonlinearity is of second order, also called a $\chi^2$-process, due to the dominant effect of the second order susceptibility $\chi^2$ in the expanded term for the induced polarisation with the input electric field $E$:

$$P = E\chi^{(1)} + E^2\chi^{(2)} + E^3\chi^{(3)} + \ldots$$

(4.6)

The resulting electron movements in the molecular structure can be split into a linear and a nonlinear component, where the nonlinear component can be separated into an oscillatory AC and a constant offset - the DC part, which in is turn responsible for optical rectification. The incoming AC field is thus in part rectified (hence the name) to a DC contribution. Due to the nature of ultrashort pulses, this DC-component rises and falls rapidly in time (in accordance with the temporal pulse-shape), which allows for the emission of low-frequency THz-radiation. The resulting bandwidth is related inversely to the optical pulse duration. The OR-induced nonlinear polarisation $P^{(2)}$ can be written as [114]:

$$P^{(2)}(\omega_{\text{THz}}) = \sum_{j,k} \epsilon_0 \chi_{ijk}^{(2)}(\omega_{\text{THz}}, \omega + \omega_{\text{THz}}, -\omega) E_j(\omega + \omega_{\text{THz}}) E_k^* \omega, \quad (4.7)$$
4.2 Generation of THz-radiation

with the Cartesian components expressed through the indices $i, j, k$ and the vacuum permittivity $\epsilon_0$. Another way to view the process is to see OR as difference frequency generation within the bandwidth of an optical pulse itself. Naturally, the effect is dependent on a set of parameters which can be optimised to obtain the strongest possible THz-radiation, namely:

- The input-light polarisation and relative crystal orientation:

  From an intuitive point of view, the polarisation of the incoming light should be oriented to the crystal in such a way that it allows for maximum induced polarisation. The susceptibility of crystal structures can be expressed using tensors, which can take on fairly simple form depending on symmetries within the crystal structure. A commonly used crystal for THz-generation is zinc telluride (ZnTe) as its characteristic zincblende structure creates a tensor with only three non-zero elements. This allows for rather straightforward calculations with an angle dependent field. An expression for the maximum THz-intensity obtainable can be derived as a function of the angle $\theta$ between the polarisation of the electric field and a specific axis of the crystal. A more detailed derivation by Yun-Shik Lee [114] results in the expression (equation (3.70) in [114]):

$$I_{\text{THz}}(\theta) = \frac{3}{4} I_{\text{THz}}^{\text{max}} \sin^2(\theta) \left( 4 - 3 \sin^2(\theta) \right).$$  \hspace{1cm} (4.8)

The respective maxima of eq. (4.8) are obtained at values $\theta = 54.74^\circ$ and $\theta = 125.26^\circ$ (and at values $+180^\circ$), which induce the strongest polarisation and correspond to the direction of the chemical bonds between the Te and Zn atoms (experimental proof shown for example in [115]). These angles are equivalent for all zincblende structured crystals, such as gallium phosphide (GaP), but similar relations can be derived for other crystal structures. As the emitted THz-field direction lies parallel with the induced nonlinear polarisation, the THz-radiation emitted by OR has the same linear polarisation as the pump light for the given optimal angles stated above. In practice this rotation is easily achieved, as one can either monitor the emitted THz-power, or simply view the intensity of the SHG-signal that accompanies the process, as its angle dependence is equal.

- The refractive indices for the optical pump and the rectified frequencies:

  Unlike the angle dependence, which can be optimised in the laboratory setup, the material properties regarding the refractive index are far more important as they intrinsically limit the choice of suitable materials. In an ideal crystal void of dispersion, the refractive index would be equal across the entire frequency range. This means, that the generated THz-frequencies will travel at the same speed and in phase with the optical pump pulse, leading to enhanced amplification of the THz-pulse as the two propagate through the length of the crystal. In practice however, these ideal conditions are not met, which leads to non-equal propagation velocities due to the difference in refractive index. When propagated in such a medium, the THz-pulse will eventually be out of phase with the optical pump, so that newly generated THz-components will destructively interfere with the previ-
4.2 Generation of THz-radiation

Previously generated ones, reducing the overall THz-power\(^3\). This occurs after a phase shift of \(\pi\), with the coherence length \(l_c\) being the distance before such a phase shift is reached, calculated through:

\[
l_c = \frac{\lambda_{\text{THz}}}{2(n_{\text{THz}} - n_g)},
\]

(4.9)

with the refractive index \(n_{\text{THz}}\) at the THz-wavelength and the group index of the pump wavelength \(n_g\) [117]. To increase the coherence length, the mismatch between the refractive indices needs to be minimised. This is desirable because for thicker generation crystals, the THz-output is typically improved due to the longer interaction length. For a well phase-matched material, the thickness is ultimately limited by the temporal dispersion of the pump pulse and the corresponding peak power decrease. However, there is trade-off between THz-power from thicker crystals and the generated frequency bandwidth, as the refractive index is not constant across the optical or THz-pulse bandwidth. Generally, high THz-frequency content is lost with thicker crystals due to the index mismatch of most generation crystals increasing for higher THz-frequencies. To illustrate the issue at hand, fig. 4.1 shows the coherence length for GaP, with \(n_g\) and \(n_{\text{THz}}\) taken from [118, 119].

![Figure 4.1](image)

**Figure 4.1.** Colourmap showing the coherence length of GaP as a function of pump wavelength and generated THz-frequency. The optimal phase matching occurs at pump-wavelengths below 1 \(\mu\)m.

For longer pump wavelengths the mismatch in fig. 4.1 increases and the coherence length to generate the THz-frequencies decreases. With a pump source emitting light at, for example, 1550 nm, this necessitates very thin crystals for generation of frequency content.

\(^3\)The same issues exist with other frequency generation methods such as SHG, where a smart solution to the problem was the development of periodically poled lithium niobate (PPLN) that allows for quasi-phase matching over a long propagation distance. PPLN has also been used to generate tuneable cw THz-radiation [116].
4.2 Generation of THz-radiation

beyond 1 THz. This in turn limits the conversion efficiency due to the reduced interaction length. The data shows that GaP is well suited for pump wavelengths centred at approximately 1 µm, which is readily available using ytterbium-based lasers like the fibre-laser used in this thesis.

- The electro-optic coefficient:
  Another material specific value is the electro-optic (EO) coefficient \( r \) in units of \([\text{pm} \cdot \text{V}^{-1}]\), which is related to the susceptibility tensor elements \( \chi^{(2)} \) through \( r = -\frac{4\pi}{n_0^2 n_e^2} \chi^{(2)} \), with the ordinary \( n_0^2 \) and extraordinary \( n_e^2 \) refractive index \([106]\). Due to the equal units, the electro-optic coefficient \( r \) is often confused with the nonlinear optical coefficient \( d \) - the two being linearly proportional, though different in magnitude \((r < d)\). For clarity, the electro-optic coefficient \( r \) is the coefficient used in this thesis. It essentially quantifies the response strength for the induced polarisation and thus the emitted THz-radiation. As the THz-field strength is proportional to the induced polarisation, which depends linearly on the susceptibility as shown in eq. (4.7), it, by conversion, also depends linearly on the electro-optic coefficient (with the THz-power depending quadratically on \( r \)). Hence, a material with a larger value for \( r \) is preferable. The \( r \)-values for ZnTe and GaP lie between 1-3 pm-V\(^{-1}\) \([120, 121]\), while lithium niobate shows a value of above 6 pm-V\(^{-1}\) \([122, 123]\), and various organic crystals have significantly stronger electro-optic coefficients at approximately 50 pm-V\(^{-1}\) \([117]\). It strongly depends on the hyperpolarisability (i.e. the ability to induce a large electric dipole moment) of a molecule. Organic crystals offer the advantage of tailoring the electron donors and acceptors (e.g. the chemical bonds) within the molecule in such a way that optimises the hyperpolarisability, explaining their larger values. Additionally, the response time of the charge-distribution should lie in the femtosecond timescale and be easily displaced by an incoming electric field \([124]\).

- Absorption loss:
  Lastly, a strong limitation to achievable bandwidth and THz-power is absorption. When the optical pump light is absorbed at high repetition rates, it leads to thermal build-up and sets a thermal damage threshold on a crystal. This limits the fluence that can be shone onto the crystal and thereby the achievable THz-power and field strengths. For low repetition rates, a single-shot of sufficient intensity can also damage a material through non-thermal damage mechanisms. These include electron-avalanche and material breakdown resulting from the strong fields involved, though such laser-induced damage thresholds typically are higher than the thermal breakdown limit. The absorption of the generated THz-radiation on the other hand is typically converted into material specific phonons and sets a limitation on achievable bandwidth for OR in suitable materials. Phonons are lattice-structure vibrations at optical frequencies, where each material has specific phonons depending on the crystal structure and elements involved. The field of light of a specific frequency will couple to the polarisation induced by the displacement of atoms and molecules from the phonon, which leads to said THz-absorption and amplification of the phonon. This prohibits the transmission of these THz-frequencies out of the crystal, often acting as an upper cut-off frequency. As phonons can also be viewed as a heat-transport mechanism, their effects can be reduced by cooling the optical crystals. Heat-management can be improved
by bonding the generation crystals to substrates that alleviate the thermal accumulation, though such methods might add further complexity and additional issues. To give a few examples, the lowest transverse-optical (TO) phonon frequency in ZnTe lies at 5.3 THz [125] and at 11 THz for GaP [126]. They strongly absorb the THz-radiation at these frequencies. The absorption strength decays towards lower frequencies, leading to a reduced signal strength as one approaches frequencies closer to the TO frequency. However, through more complex second order phonon interaction, several side-bands at lower frequencies also act as THz-frequency blocks with an effective bandwidth rather than strict delta function like absorption.

A commonly used crystal for OR is lithium niobate, proposed for THz-generation in 2002 [127] and subsequently pumped with low [128] and high [129] repetition rate sources producing impressive results. More recently, it was used to produce 66 mW of THz-power when pumped at 112 W of optical power at 1 µm [130], though its achievable THz-frequency is limited and it requires a tilted-pulse front setup due to phase-matching conditions of the crystal. The most easily implemented OR setup is a collinear setup where the crystal is at zero-incidence to the pump light. This alignment is used for example with ZnTe, GaP, BNA or organic crystals. In fact, one of the first “THz” generation processes with a collinear setup was achieved in 1971 with lithium niobate, though the generated frequencies were below 0.5 THz and the pump laser had a pulse duration of 2000 fs [131]. This again shows the tremendous development on pump laser and THz-generation (and detection) that has taken place over the past few decades. As part of this thesis, which emphasises a low-complexity approach, collinear OR is the generation method implemented, and thus direct comparisons to other methods for benchmark purposes are omitted.

4.2.2 Other generation methods

In this chapter, generation methods other than OR will be introduced with limited technical detail compared to the previous chapter on OR. The three methods in question are, in order of appearance, Photoconductive switching, Laser driven gas ionisation, and Spintronics. Other methods for THz-generation, such as Quantum-cascade-lasers [132], were not encountered experimentally as part of this thesis and thus are not included here.

Photoconductive switching

Photoconductive switching can largely be attributed to the work of D. H. Auston, who pioneered the idea and the science behind it throughout his career, coining the term “Auston-switch” as a synonym for the photoconductive switch [18, 133, 134]. In principle, a suitable semiconductor is placed within an electrical circuit in such a way that it acts as an insulator under normal conditions. Once photons are incident on the semiconductor, the free carriers produced by the photo-effect turn the semiconductor into a conducting element of the circuit. It therefore essentially acts as a switch controlled through illumination, hence the name photoconductive switching. The switching time is controlled by the duration of the input-light generating the free carriers.
and the carrier lifetime before recombination. When an external bias-field is applied across the switch, the free charge carriers are accelerated. The hereby induced current modulation occurs on sub-picosecond timescales, which in turn leads to the emission of THz-radiation [114, 135].

The layout of this technology allows for a wide range of modulations, controlled through the semi-conductor material band gap, the distance of the switch and the break-down voltage, as well as the manipulation of the carrier lifetime by varying the concentration of defects. As the emitted THz-radiation originates from a spot smaller than the THz-wavelength, it is highly divergent and a slightly more complex collimating lens is required to guide the THz-radiation coherently towards a desired direction. Typical commercial products offer THz-pulse trains at 100 MHz repetition rate over a 2 THz (6 THz) bandwidth at -20 dB (-80 dB) [136]. The average THz-power of such system of 30 µW results from a system efficiency of 0.15 %, similar to what standard antennas achieve (~0.1 %) [137]. Advanced photoconductive THz-antennas, enhanced by plasmonic nanocavities, have offered impressive, high-power performance delivering up to 4 mW of average power at 76 MHz repetition rates at just Watt-level excitation (efficiency ~0.5 %). Their bandwidth spans over 2 THz at -20 dB [138], but due to the necessary tight confinement of the pump light, further power-scaling based on the fabrication of large-area devices could prove challenging. Outstanding conversion efficiencies of 7.5 % have been demonstrated using similar nanoscale devices, though at a lower average THz-power of 105 µW [139]. With the technology already at a commercial level, photoconductive switches offer a great advantage over other THz-generation techniques. The science is fairly well understood, and the devices can be tailor-made to suit a specific application, have a small-system footprint, and offer the long-term potential of implementation into integrated circuits [140]. The pump light is typically delivered by fibre, so the system is robust and compact, allowing for mobile and low-maintenance applications. Overall, the system offers plenty of potential for future industrial applications with low-frequency THz-applications and moderate power levels. The scaling towards large bandwidths exceeding 6.5 THz (though recorded at -80 dB [141]), with high average powers and/or strong field strengths is where photoconductive systems fall short. For example, in 2010 the record field strength reported was only 36 kV·cm⁻¹ [142]. Most applications requiring larger parameters are currently confined to industrial R&D or university research, which leads to the introduction of the next generation method, laser driven gas ionisation.

Laser driven gas ionisation

The generation and detection of THz-radiation by focussing ultrashort pulses with sufficient energy to turn a gas into a plasma was demonstrated by Hamster et al. in 1993 [143, 144]. The high-energy optical pulse that is focussed into a gas, e.g. air, ionises the gas-atoms, creating a plasma with a large number of free carriers. These carriers are accelerated by the ponderomotive force of the optical light field, leading to a photocurrent that emits THz-radiation. In order for the emitted radiation to reach a reasonable intensity, as well as a desired directional output, an asymmetric field needs to be induced that helps to generate a nonlinear current. Typically, this is achieved by frequency-doubling the fundamental pump-light and having the two frequencies co-propagate towards the same focus, called a two-colour setup or the "AC-bias
4.2 Generation of THz-radiation

The THz-field strength is linearly or even quadratically proportional to the field of the second-harmonic [114], depending on the optical pulse energy. However, it depends sinusoidally on the phase difference between the second harmonic and fundamental wave, e.g. it is at a maximum for a phase difference of $\pm \pi/2$ [146]. Efficiency can further be increased by a multi-colour setup [147], promising orders of magnitude improvements - in theory - when using three vs. two colours [148]. The generated THz-radiation depends on several factors, for instance the temporal overlap of the two frequencies, their phase difference, the relative intensities of each one, the operating gas, the pump pulse duration and more. This leads to high tuneability and optimisation on the one hand, but to a complex setup on the other. Nonetheless, this method has proven to provide the strongest THz-electric field strengths and good conversion efficiencies. For example - using a two-colour scheme - a field strength in excess of $100 \text{ MV} \cdot \text{cm}^{-1}$ with a conversion efficiency of 2.36 % was reported in 2020 at a repetition rate of 20 Hz, corresponding to magnetic field strengths above 33 T [149]. However, the driving laser requirements in the millijoule energy range currently prevent intense THz-plasma sources in the MHz regime. The fastest repetition rate achieved with this method lies at 100 kHz with an average power of 40 mW and a conversion efficiency of $4.2 \cdot 10^{-4}$, though this combines several laser-channels to maintain the required pulse energy [146].

The other great advantage of this generation method is the possibility to produce very large bandwidths, ranging from single-digit THz-frequencies up to 200 THz, and thus almost the entire IR-spectrum [150, 151]. This is a result of the free movement of the electrons and the absence of any plasmons as encountered in solid-state materials. Additionally, there is no inherent damage-threshold as with other generation methods, so the pump-intensity can be scaled at will, since the source-gas essentially is unlimited and constantly replenished. With such a plethora of advantages, it is hard to find many downsides to this generation method. However, the main drawback for these methods is the high-energy requirement on the source laser system, while the complex free-space setup adds another strain to potential users. Connected to the high-pulse energy typically extracted from regenerative amplifiers is a low repetition rate. It limits the data-acquisition speed or reduces the SNR in scientific experiments or applications which are envisioned by industrial users. A third and perhaps scientifically more exciting generation method is introduced in the next paragraph. It might well be a suitable candidate in the future to bridge these disadvantages if sufficient research is conducted.

Spintronics

Spintronics is the term coined for a THz-generation method that not only depends on the manipulation of the electron charge to generate a radiating transient charge current (as with gas-ionisation). Instead, it additionally exploits the spin properties of electrons through transient spin-to-charge transfer in magnetic multi-layer thin-films, pioneered by T. Kampfrath’s group at the Fritz Haber Institute of the Max Planck Society in Berlin [152, 153]. The operating principle also requires the generation of free charge carriers (electrons to be precise), usually achieved by illumination with a femtosecond pulse. The carriers are excited in a ferromagnetic (FM) layer with a perpendicular magnetic field applied to it, which results in a strongly spin-polarised set of mobile
4.2 Generation of THz-radiation

electrons, e.g. a spin-current. Adjacent to the FM layer is a non-ferromagnetic (NM) layer into which the spin-current can flow due to diffusion [152]. At the interface of the two layers, spin-orbit coupling leads to a conversion of the spin-current to a perpendicular charge-current when the spin-current electrons are deflected at opposite-sign angles depending on their respective spin-orientation. Due to the FM-layer being magnetised, the spin-polarisation results in a separation of the electrons in a majority and minority spin density. As it consists of a larger amount of electrons, the majority spin density dominates. This imbalance translates to an uneven distribution of charge-flow in one direction in the NM layer, effectively generating a charge-current. The process is known as the inverse spin Hall effect, experimentally demonstrated in 1984 [154]. Similar to the photoconductive antennas, this short-lived charge-current then leads to the emission of THz-radiation, which is perpendicularly polarised to the FM magnetisation and can be modulated by changing the magnetisation, potentially allowing for on-chip THz-applications [155].

Figure 4.2. Comparison of the recorded THz-field for two nominally equal spintronic samples consisting of a W/CoFeB/Pt tri-layer with thicknesses of 2.0/1.8/2.0 nm. Pump-conditions on both samples were equivalent, though the resulting red trace shows only 10 % the field strength of the blue trace. This difference is expected to result from the manufacturing quality, as the samples were provided by two different sources.

Spintronic THz-generation has been demonstrated to produced broadband THz-pulses covering up to 20 THz at -10 dB, with field strengths exceeding 300 kV·cm⁻¹ [156], and up to 30 THz at -20 dB [153]. The conversion efficiencies of the publications cited above lie in a range between that achieved through optical rectification in organic and inorganic crystals at ∼1·10⁻⁶, with average powers reported in the tens of µW-range. This makes spintronic emitters a worthy competitor for THz-generation. For more detailed information, a comprehensive review of the state of the art on spintronic development from 2021 makes for an interesting read [157]. The relatively low conversion efficiency and output power should not be seen as a weakness though, since one great advantage of these emitters is the scalability, and the research itself is not even a decade
4.2 Generation of THz-radiation

old. Given a sufficiently high fluence (remaining below damage threshold), the THz-generation process can be area-scaled at will, since the FM-NM layers can be economically produced in large dimensions. Adding a third NM layer by “sandwiching” the FM layer allows for increased efficiency as electrons diffusing into both direction can be utilised. The generated THz-radiation adds constructively due to the very thin dimensions of only a few nanometres for each layer, much smaller than the THz-wavelength. Furthermore, a reflective layer for the pump-light can be placed on the back-side, repeating the THz-generation process within a time-frame short enough so that the “second” THz-pulse is indistinguishable from the first.

Perhaps the most important advantage is the general pump-wavelength independence [158] (from 900-1500 nm). This allows the spintronic emitters to be used with most femtosecond lasers, as the only requirement on them is to lift the electrons in the FM layer to the conduction band. Lastly, spintronic emitters can be used in free-space setups or be combined with standard antennas in fibre-coupled systems [159], improving the THz-yield compared to photoconductive antennas. The disadvantages of spintronics are the relatively few research groups working on the still young topic. As a consequence, a detailed study on the damage threshold is missing, and there are limited facilities to produce suitable high quality samples. The last point is important, as the production quality has a strong effect on the THz-generation efficiency, as shown in fig. 4.2. It shows the recorded THz-output of two nominally equal tri-layer samples (W/CoFeB/Pt) at equal pump conditions, though with a significant difference in THz-output.
4.3 Detection of THz-radiation

In analogy to the previous chapter on THz-generation, THz-detection also offers several choices, which each have their own unique application spectrum, advantages, and disadvantages. The main detection method used in this thesis was through electro-optic sampling (EOS), which is why this method will be introduced in more detail. Alternative detection methods are briefly discussed afterwards. Generally speaking, the THz-frequencies are difficult to detect, as established techniques for optical frequencies or radio-frequencies are not applicable. This is due to the low photon-energy which makes typical photo-detection methods difficult to implement, while the THz-frequencies are too large for the low-frequency detection methods of radio-frequencies.

4.3.1 Electro-optic sampling

A useful detection method for THz-radiation, especially in laboratories, involves the second-order $\chi^{(2)}$ effect of non-centrosymmetric crystals such as ZnTe or GaP (EO crystals). In simple terms, a THz-field modifies the properties of a medium and the effect of this can be inscribed onto a probe beam passing through the modified crystal area. This process will be explained in more detail in this section and outlines the general detection method that is termed “EOS” for the remainder of this thesis.

Utilising the linear electro-optic effect (or Pockels effect) to coherently detect THz-radiation was demonstrated in 1995 [160]. Since then, several approaches to optimise and understand the process were taken. For non-centrosymmetric materials (e.g. materials where the $\chi^{(2)}$ is non-zero), an applied electric field changes the birefringence of the medium proportionally to the field strength. Similar to eq. (4.7), the second-order nonlinear polarisation through the Pockels effect is given by [114]:

$$P_i^{(2)}(\omega) = 2 \sum_{j,k} \epsilon_0 \chi^{(2)}_{ijk}(\omega, \omega, 0) E_j(\omega) E_k(0). \quad (4.10)$$

Considering a lossless medium, the two susceptibility tensors of OR and the Pockels effect are nominally the same, i.e. $\chi^{(2)}_{ijk}(0, \omega, -\omega) = \chi^{(2)}_{ijk}(\omega, \omega, 0)$, meaning that the tensor elements used in the description for OR can be used equally here. The electro-optic coefficients for OR can thus be used to determine the strength of the Pockels effect. Equation (4.10) shows that the effect depends linearly on the electric field of a given frequency, which manifests itself physically as a change in refractive index proportionally to the applied electric field. This is, for example, used in electro-optic modulators or as modifiable waveplates. Disregarding phase-matching conditions for now, i.e. a dispersionless medium with equal refractive indices across all frequencies, and by adding an optical probe pulse to propagate collinearly to the THz-beam, coherent detection of the THz-field is possible.

In the absence of the THz-field, the linearly polarised probe beam will propagate through the EO-crystal unaltered\(^4\), except for the time-delay caused by the refractive index. After passing the

\(^4\)In practice, crystal defects and production quality typically lead to a slight birefringence of the crystal.
EO crystal, the probe beam is sent through a quarter-waveplate, which is oriented in such a way that it produces a perfectly circular polarisation. In sequence, a Wollaston prism splits the circularly polarised light $I_0$ into two orthogonal components $I_x$ and $I_y$, which are of equal intensity $I_0/2$ due to the circular polarisation of the input pulse. These two beams are then detected with a balanced photodiode subtracting the response of the two signals from another, leading to a signal reading of $I_x - I_y = 0$. When the THz-pulse is present in the EO-crystal, a birefringence is induced in the EO-crystal due to the Pockels effect. This changes the phase-delay $\Delta \phi$ in the probe pulse as it experiences the THz-induced difference of refractive index along the fast and slow axes of the crystal. This manifests itself as a non-circular ellipsoid after the quarter-waveplate (the orientation of which is not changed) and leads to an imbalance between the two orthogonal components recorded by the balanced photodetector, giving a non-zero signal $I_x - I_y \neq 0$. Effectively, the phase-modulation is thus transferred to a measurable intensity modulation.

Since the effect is linear, the two components can be described as:

$$I_x = \frac{I_0}{2} (1 + \sin(\Delta \phi)),$$

(4.11)

$$I_y = \frac{I_0}{2} (1 - \sin(\Delta \phi)).$$

(4.12)

For small angles, $\sin(\Delta \phi) = \Delta \phi$ holds. The physics explained above is visualised in fig. 4.3. By changing the arrival time of either pulse, one can hereby map the THz-field in time. To optimise the strength of the birefringence inscribed into the probe beam, the (linear) probe polarisation should be parallel or orthogonal to the THz-polarisation (for a circularly polarised THz-beam, the linear probe polarisation orientation is irrelevant).

In contrast to the generation with OR, the orientation of the EO-crystal should be set slightly different for optimal signal strength at $\theta = 90^\circ$, e.g. along the [110]-axis of an (110)-oriented zincblende crystal. For real media with dispersion and varying refractive index, the same implications on phase-matching and coherence length as with the generation of THz-radiation in OR apply. Additionally, the pulse duration of the pump pulse sets a limit to the temporal resolution, and the nonlinear susceptibility of a material is not equal across the entire bandwidth. Another spectral constraint arises from the thickness of the EO-crystal. For thicker crystals, experimental alignment and signal intensity increases, however, due to increased phonon and second order phonon absorption as well as prolonged phase-mismatching, the thick crystals show a narrower detection bandwidth than the thin crystals. Crystal thicknesses typically encountered are a few hundred $\mu$m, and suitable materials are ZnTe for detection at 800 nm, GaP in the 1 $\mu$m range, and GaAs at 1.35 $\mu$m.

Since the effect of the THz-field is small in most cases, EOS is usually connected to a lock-in amplifier, which typically records the intensity as a voltage linked to some sort of modulator (e.g. a chopping wheel). In some cases of extreme field strengths, the THz-beam must be attenuated to prevent an over-rotation of the probe-pulse. This is the case if the induced phase-delay is larger than $\frac{\pi}{2}$, resulting in a reduction of the recorded signal despite a stronger electric field. Interestingly, a method termed “spectral-domain interferometry THz-detection” does not encounter this issue.
4.3 Detection of THz-radiation

Figure 4.3. A sketch representing the EO sampling technique. The THz-beam (blue) induced birefringence in a crystal will change the polarization of the probe pulse (red). This modification leads to a non-circular polarization after the quarter waveplate (QWP), resulting in a difference in detection intensity on a balanced photodetector after the Wollaston prism (WP) has split the probe pulse into its two orthogonal polarization components.

The voltage-reading of the intensity difference at the maximum of the THz-pulse can also be used to estimate the real THz-field strength acting on the EO-crystal. For this, the probe intensity $I_0$-related voltage on the lock-in needs to be measured by modulating (chopping) the probe pulse and blocking one of the two polarizations incident on the balanced detector. Together with the difference signal $\Delta I = I_x - I_y$ and the relation \[ E_{\text{THz}} = \frac{c\Delta I}{I_0 L \omega n_{\text{op}}^3 r_{\text{max}}}, \] (4.13) this yields a result for the electric field strength $E_{\text{THz}}$ of the THz-beam with the constants $n_{\text{op}}$ (refractive index at the optical frequency), $r_{\text{max}}$ (electro-optic coefficient for the strongest tensor element), $L$ (EO-crystal thickness), $\omega$ (optical angular frequency) and $c$ the speed of light in vacuum.

In my opinion, this estimation method is problematic. For short optical probe pulses, the bandwidth can be substantial enough to cause issues with the assumption of a constant refractive index $n_{\text{op}}$. The same dispersion might also affect the electro-optic coefficient depending on the THz-bandwidth. Furthermore, it requires perfect angular alignment of the crystal with the THz- and probe beam (otherwise there is an underestimation). The perhaps most significant drawback is the fact that the THz- and optical beam’s spatial overlap in the crystal are assumed as perfect, meaning that the maximum lock-in voltage corresponds to the maximum THz-field strength. Unless the probe and THz-beam are perfectly collinear, this will not be the case. Hence, for an initial estimation of the field strength, this method is acceptable, but for accurate calculation, the following procedure is superior, given a strong enough THz-energy or THz-power for detection.
One can calibrate the on-axis field strength using

\[ E_{\text{cal}} = \Gamma \cdot E_{\text{raw}}, \quad (4.14) \]

with \( E_{\text{raw}} \) being the lock-in recorded EOS trace and \( \Gamma \) the scaling factor. To determine the scaling factor, the relation for the pulse energy \( U \) is given by the integral of the intensity:

\[ U = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{0}^{T} I(x, y, t) \, dt. \quad (4.15) \]

The spatial dimensions \( x \) and \( y \) are defined as the radius at \( 1/e^2 \) intensity of a Gaussian beam and thus the spatial expression becomes

\[ I(x, y) = \exp \left( -\frac{2x^2}{w_x^2} - \frac{2y^2}{w_y^2} \right), \quad (4.16) \]

where \( w_x \) and \( w_y \) are the Gaussian beam widths. Applying the spatial Gaussian integral

\[ \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp \left( -ax^2 - by^2 \right) \, dx \, dy = \frac{\pi}{\sqrt{a} \sqrt{b}} \quad (4.17) \]

and the relation between the intensity and the field

\[ I(t) = c\varepsilon_0 n \int_{0}^{T} E(t)^2 \, dt, \quad (4.18) \]

one can simplify to:

\[ U = \frac{\pi w_x w_y c\varepsilon_0 n}{2} \int_{0}^{T} E(t)^2 \, dt. \quad (4.19) \]

It is important to note that the often used relation \( I = \frac{c\varepsilon_0 n}{2} |E_0|^2 \) only holds for a time averaged sinusoidal electric field and thus cannot be used here. Together with the relation of power, repetition rate, and energy \( U = \frac{P}{f_{\text{rep}}} \), the scaling factor \( \Gamma \) is given by

\[ \Gamma = \frac{\sqrt{2P}}{f_{\text{rep}} \varepsilon_0 n \pi w_x w_y K \int_{0}^{T} E_{\text{raw}}(t)^2 \, dt}, \quad (4.20) \]

where \( c = 2.998 \cdot 10^8 \text{ m} \cdot \text{s}^{-1} \) is the speed of light in vacuum, \( \varepsilon_0 = 8.85 \text{ pF} \cdot \text{m}^{-1} \) is the vacuum permittivity, \( n \sim 1 \) the refractive index of air (or the surrounding medium), and \( K \) the setup-specific correction factor. To calculate the field strength with this method, one thus requires the corresponding THz-pulse energy (calculated from the recorded average THz-power \( P \) and repetition rate \( f_{\text{rep}} \)), the measured THz-focal spot size (with assumed Gaussian intensity distribution), and the integral of the measured EOS trace, which can be computed through \( \sum |E_{\text{raw}}|^2 \Delta t \) with the EOS time-step \( \Delta t \). The correction factor \( K \) depends on setup-related attenuation and the temporal scan-range of the EOS trace as will be explained. Through analysis of eq. (4.14) and eq. 4.20 it becomes clear that the calibration factor \( \Gamma \) is sensitive only to the shape of the EOS.
4.3 Detection of THz-radiation

trace, not its amplitude, while the measured beam spot, correction factor, and the filter-adjusted average THz-power are the sensitive input parameters. Distinct from other schemes, this makes the calibration of the THz-field strength using this approach insensitive to the alignment of the probe and THz-beams in the EOS detection, as the EOS trace amplitude is irrelevant. Additionally, it does not require any detection-crystal material parameters.

One must, however, pay careful attention to filter corrections, i.e., the input THz-power $P$ should not be compensated for any loss effects that affect the shape of the recorded EOS trace, such as humidity related loss. Objects that reduce transmission, but do not alter the pulse-shape (e.g., silicon wafers) must be corrected for. A drawback of this method is the assumption of the measured average THz-power being proportional to the recorded EOS trace. This can lead to overestimation of the field strength when taking time-limited EOS traces (up to several tens of picoseconds), as the power reading records an average including the contributions of all echoes and - when operating in standard atmosphere - water ringing in the system. If the setup is placed in dry air, where no water ringing is present, a moderate scan length can capture all relevant echoes in the EOS trace. But for traces recorded in humid atmosphere, one would require temporally long EOS-traces covering 100 ps or more as shown in fig. 4.4a). In practice, this is often non-viable due to the long duration such scans take while maintaining decent temporal resolution.

Generally speaking, for a typical THz-setup based on OR, the contributing time-delayed signals arise from the echoes within the generation crystal, filters used to remove the pump light (e.g., silicon wafers at 800 nm, or polytetrafluoroethylene (PTFE) at above 1 µm), and humidity related re-emission of THz-radiation (“water ringing”). For the generation crystals, thicknesses typically vary from 0.1-1 mm, with the refractive index roughly between 2-3.5 for various materials (organic and inorganic). A crude estimation based on these values leads to a spacing between subsequent echoes of 10 ps (thickness 0.5 mm, refractive index $n = 3$). Since the reflections are comparatively weak at $0^\circ$ incidence, i.e., below 6%, it suffices to include the first two echoes. In this case, a 20 ps scan-duration is sufficient to include these echoes in the EOS trace, which is usually within the scan-range of standard THz-TDS experiments. PTFE filters are usually very thin and scatter the IR-light while transmitting over 90% of the THz radiation. With a thickness of 100 µm and a THz-refractive index of $\sim 1.4$ [109], this leads to an echo-delay of only $\sim 1$ ps, which means these echoes are most definitely included in a THz-TDS scan. The same can not be said about Silicon wafers, which are often used to either filter out pump light at 800 nm, to attenuate a THz-beam, or to couple a probe beam collinearly into the THz-beam path. Here, the refractive indices for THz-frequencies lie above 3, leading to stronger Fresnel reflections and increased round-trip time. Typical production thickness is 525 µm, so the first echo appears at $\sim 11$ ps for perpendicular wafers and at $\sim 16$ ps for wafers angled at $45^\circ$.

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5One also requires large enough THz-power to be accurately measured with a power meter.
4.3 Detection of THz-radiation

One can calculate the transmission-contribution for $P$ and $S$ polarised waves of each echo to the overall power by using $T = 1 - R$ and the Fresnel equations (4.21), and (4.22)

$$R_P = \frac{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\theta_i) \right)^2 - n_2 \cos(\theta_i)}}{n_1 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\theta_i) \right)^2 + n_2 \cos(\theta_i)}}, \quad (4.21)$$

$$R_S = \frac{n_1 \cos(\theta_i) - n_2 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\theta_i) \right)^2}}{n_1 \cos(\theta_i) + n_2 \sqrt{1 - \left( \frac{n_1}{n_2} \sin(\theta_i) \right)^2}}, \quad (4.22)$$

with $n_i$ the specific media’s refractive index and $\theta_i$ the input-angle measured to the normal on the interface surface. In the case of unpolarised light, one can simply use the effective reflectivity $R_{\text{eff}} = 0.5 (R_S + R_P)$. For example, when calculating the contribution of the first three echoes of a Silicon wafer in air, one multiplies the number of internal reflections $R_{\text{SiAir}}$ for each THz-echo in the wafer at the silicon-air interfaces with the transmission coefficient at the silicon-air exit interface $T_{\text{SiAir}}$, adding them together as in eq. (4.23), being (for $P$-polarised light):

$$\left( R_{\text{SiAir}}^2 + R_{\text{SiAir}}^4 + R_{\text{SiAir}}^6 \right) T_{\text{SiAir}} = 6.89 \%. \quad (4.23)$$

Reflections at the initial air-silicon interface can be ignored, since the correction factor compensates for the truncation loss of the recorded EOS area only, and the reflected power does not participate here. Hence, by using equation (4.24), we receive $K = 1.074$ as the correction factor for a Silicon wafer at normal incidence in the THz-beam path (fourth and higher-order echoes are negligible). Equation (4.21) shows that the angle of attack and the (frequency dependent) refractive indices play an important role. Both lead the small differences observed between the theoretically calculated contribution and the obtained results one obtains through an empirical approach. Here, the THz-beam is affected by surface roughness, exact alignment, and minuscule absorption that all have an effect on the power transmission and thus the echo contribution.

Figure 4.4b) shows an EOS trace taken from a non-collinear THz-source (hence the absence of a generation-echo) propagated through dry air with a 525 $\mu$m thick high resistivity float zone silicon wafer at normal incidence in the beam path. The initial THz-waveform is followed by three declining echoes at specific time-delays of 11.9 ps, matching the calculated delay-time for the wafers. The power contribution of each waveform is shown in the inset as steps on an inverted logarithmic scale, each adding $\sim 7 \%$ of the power contained in the previous pulse. After the third echo the remaining signal strength is covered by noise.
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Figure 4.4. a) 100 ps scan of an ABCD recorded THz-trace (red) at 20 % RH. When scanning an EOS trace of 10 ps (dashed black), roughly 20 % of total power (blue) is not recorded in the EOS trace, which needs to be corrected for when calibrating the THz-field strength. b) EOS trace of a THz-pulse and its echoes from a 525 µm thick silicon wafer at normal incidence angle. The echoes are spaced 11.9 ps apart, corresponding to the round-trip time within the wafer, each containing ~7.5 % of the power of the previous pulse. After the third echo at ~40 ps, the signal is below the noise level. The inset shows the accumulated power on a log-scale.

Together with water ringing, which only phases out at about 80 ps for even moderate humidity levels as shown in fig. 4.4a), these echo contributions must be included in the correction factor $K > 1$ in front of the integral of the EOS trace in eq. (4.20), and can be calculated through:

$$K = \frac{C_1}{1 - C_1} + \frac{C_2}{1 - C_2} + \cdots + \frac{C_n}{1 - C_n} + 1,$$

(4.24)

where $C_n$ stands for the total power contribution after the temporal cut-off of the THz-trace for each attenuator. For example, with a 10 ps scan range and a 45°-angled silicon (Si) wafer in the beam path, the corresponding $C$-value for all echoes after a 10 ps scan range would be 0.075 (7.5 % of the total transmitted THz-power through the Si-wafer). The correction factor $K$ equals 1 if the sampled temporal window covers the full extent of the THz-pulse. In other words, this means that no THz-power resides outside the temporal window of the sampled signal. To conclude, all echoes lying outside of the recorded EOS time window are not included in the EOS trace, but contribute to the average power $P$ recorded by the power meter. Water ringing contributions

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6Si-echo data reproduced from figure 16 in [164] with permission from the author Peter Uhd Jepsen
are a little more complex to calculate and will be discussed in chapter 6. As a last measure, the noise in the EOS traces recorded before the onset of the THz-pulse should be excluded from the calibration as well.

4.3.2 Other detection methods

Just as with generation, there are several ways to detect THz-radiation that differ from EOS. The following section is held more brief due to the minor role of the other detection methods in this thesis. An alternative form of EOS is to use a different medium that allows for a modulation of the probe-light, specifically ambient air.

The method is termed “air-biased coherent detection” (ABCD) and exploits the third-order $\chi^{(3)}$-nonlinearity of air that is enhanced under an external bias-field, reported in 2008 [165]. If a strong enough electric field is applied to air, it can act as a second-harmonic generator through a four-wave-mixing process when a probe beam is spatiotemporally present. The strength of the SHG depends on the applied electric field. When focussing the THz-field into this biased-air, the electric field of the radiation will modulate the SHG properties, which leads to stronger or weaker SHG of the probe pulse. This can be used to achieve coherent detection [166] by filtering out the SHG signal after the generation and recording the intensity with a suitable detector (ideally an avalanche photodiode [167]). The hereby recorded signal is directly proportional to the changing THz-electric field and the (constant) bias-field. To filter out DC-components and increase sensitivity, the bias-field is typically synchronised to half the laser repetition rate in order to allow for lock-in detection [168]. The benefit of this detection method is the absence of any parasitic phonons that restrict the detection bandwidth, allowing for seemingly endless detection of THz-frequencies. On the downside, this setup requires modest field strengths in the kV·cm$^{-1}$-range and a high-voltage source, as well as the $\chi^{(3)}$ process being weaker than a $\chi^{(2)}$ one.

Perhaps the most frequently applied detection method for commercial THz-sources is through photo-conductive antennas. Here, the THz-generation method mentioned in chapter 4.2.2 is reversed in order to detect THz-radiation. An arrival-time controlled femtosecond pulse generates free carriers in the photoconductive switch in the gap between the antenna structure. Unlike generation, there is no external bias-field applied to accelerate the electrons. Instead, the incoming THz-field is focussed into the gap, the electric field of which allows for the flow of a photocurrent, ideally directed in such a way that it bridges the gap. The resulting instantaneous voltage across the antenna is thus linearly proportional to the THz-field, and the variable arrival time of the optical pulse inducing the free carriers allows for a temporal mapping of the THz [169]. Similar to the generation, the detection is limited in frequency by properties of the switch, e.g. the carrier life-time, and by material specific phonon absorption.

Electron field emission may be the newest approach to THz-detection. It offers great potential once its initial drawbacks regarding sensitivity and difficult calibration are addressed. Essentially, a THz-pulse focussed onto a metallic micro-antenna can lead to the emission of electrons. These can then be used for example to ionise surrounding gases, emitting light to be detected [170].
Another approach is to accelerate and multiply the free electrons in a photo-multiplier-tube [171], with the micro-antennas modifiable so that they can target specific frequency and energy ranges. THz-detection using electron field emission, however, is not yet widely known nor used and still requires some time and research before it can be considered a “go-to” method for THz-detection.
CHAPTER 5

Improving the efficiency of collinear OR

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5.1 Theory

As outlined in this thesis, standard EOS crystals used for THz-generation require high pulse energies to efficiently convert the optical pump into THz-radiation with field strengths beyond the kV·cm\(^{-1}\) regime. The pulse energy scales inversely with the repetition rate, so it is not feasible to use established THz-emitters as high repetition rate THz-sources. The approach in this thesis is to use a high-power laser combined with external pulse compression to improve the peak power, while applying the optical power to the highly efficient organic crystal (2-(4-hydroxy-3-methoxystyryl)-1-methylquinolinium2,4,6-trimethylbenzenesulfonate), abbreviated as HMQ-TMS [172]. This combination should allow for strong field strength THz-pulses at high repetition rates to be generated.

Organic crystals like HMQ-TMS offer an advantage in customising the structure of the electron donors and acceptors arranged in a non-centrosymmetric array. This can be exploited to create structures with very large polarisability (called hyperpolarisability). It is implemented by arranging the molecules “in line”, i.e. in such a way that their p-orbitals are maximised through conjugation and electrons have a group of atoms they belong to, rather than a single atom, increasing their mobility and through this the material’s polarisability. What makes HMQ-TMS stand out from other organic crystals is the almost zero angle between the polar axis (i.e. the “in-line” axis of the p-orbitals) and the axis along which the molecules are ordered, meaning an almost ideal configuration [172]. Other organic crystals tend to have an angle between 20° and 30° [117]. Together with the ultrafast response time of the carriers and its general resistivity to environmental effects (e.g. humidity) compared to other organic crystals, HMQ-TMS is therefore an ideally engineered THz-generation crystal [173].
As explained in chapter 4.2.1, the THz-conversion in OR relies on the large displacement of electrons achieved by an incident electro-optical field in order to create a strong induced polarisation. The molecular structure and packing of HMQ-TMS allows for such strong displacement, while also having a fairly low mismatch between the optical \( \sim 1 \mu m \) pump wavelength and the THz-frequencies refractive index of \( \Delta n < 0.3 \) \( [174-176] \). This allows for decent phase-matching and coherence lengths shown in fig. 5.1. A very crude estimate using a general refractive index mismatch of 0.25 turns the denominator in eq. (4.9) to 0.5, meaning that the coherence length for each generated THz-frequency is double its wavelength and decreases for larger THz-frequencies.

![Figure 5.1. Colourmap showing the coherence length of HMQ-TMS as a function of pump wavelength and generated THz-frequency. Despite larger coherence lengths for longer wavelengths, HMQ-TMS is also well suited to be pumped at \( \sim 1 \mu m \). (shaded area)](image)

Figure 5.1 indicates that the optimal pump-wavelength is above 1300 nm, accessible by optical parametric amplifiers, which currently cannot readily provide the required high repetition rates and powers. The pump-wavelength accessible through the system used in this thesis is highlighted with a shaded area above 1000 nm and still offers decent coherence lengths above 0.2 mm for generation of THz-radiation up to almost 6 THz. The electro-optic coefficient of HMQ-TMS lies at a high \( r_{333} = 50 \text{ pm} \cdot \text{V}^{-1} \) \( [117, 172] \) (measured at 633 nm, though expected not to change too much at longer wavelengths), with the main contribution to the molecular nonlinearity coming from the HMQ-cations. Due to the almost identical alignment of the polar axis and the crystal packing orientation in HMQ-TMS (angular mismatch less than 1\( ^\circ \)), the hyperpolarisability tensor elements \( \beta_{ijk} \) derived in the supplementary information of \( [172] \) show a clear trend towards a single optimal value for \( \beta_{333}^{\text{eff}} = 185 \cdot 10^{-30} \text{ esu} \). This means that for HMQ-TMS all effective first hyperpolarisability tensor elements other than the \( \beta_{333}^{\text{eff}} \) are negligibly small and can be considered as zero. The pump polarisation should therefore be aligned parallel to the 333
axis of the crystal. This aligns the crystal to the pump polarisation for maximum THz-generation efficiency, while other cut-angles or rotation alignment will drastically reduce the THz-generation efficiency. In general, the typical organic crystals used for THz-generation all outperform their inorganic counterparts in terms of the electro-optic coefficient $r_{ijk}$, which generally lies in the range from 50 to 90 pm·V$^{-1}$ [117]. The HMQ-TMS samples used in this thesis were provided by Prof. O-Pil Kwon from Ajou University in South Korea, who has several more co-publications regarding this organic crystal. Many interesting applications, such as tuneable THz-generation, have been presented, and the reader is directed to these publications for more information on the growth process, chemical information and molecular structure, and further applications of HMQ-TMS [95, 172, 174, 175, 177–181].

The expected THz-output when pumping the crystal at a given wavelength can be modelled analytically. The majority of the simulation work was conducted by Dr. Mojca Jazbinsek at ZHAW School of Engineering in Winterthur, Switzerland. The approach is based on that proposed in [182] and is used to evaluate the generated THz-spectra from 0.25 mm thick HMQ-TMS as a function of the transform-limited (TL) pump pulse duration at an equal pump energy (and pump fluence). For this, a non-depleted plane-wave pump approximation is adopted, the frequency dependent Fresnel transmission at each crystal-air interface area is accounted for, and realistic material parameters are used, including the dispersion of the refractive index and absorption in the THz- and optical range for HMQ-TMS [174, 175]. The results are shown in fig. 5.2 for different pump pulse durations at otherwise equal parameters, i.e. pulse energy and pump spot size. The resulting estimates are not restricted by any spectral limitations due to THz-wave propagation or detection, i.e. they model the THz-output directly after the generation crystal.

![Graph](image)

**Figure 5.2.** Generated THz-amplitude over frequency in HMQ-TMS for various pump pulse durations at 1035 nm and equal pulse energy. The THz-radiation drastically improves in amplitude and bandwidth for shorter pump pulses. All curves are normalised to the 20 fs peak.
5.1 Theory

The model shows that the output spectral bandwidth scales favourably with a decreasing input pulse duration, showing higher frequency spectral components and an overall higher spectral amplitude compared to the output when pumped with longer pulses. For a clear comparison, all curves are normalised to the peak of the TL 20 fs trace. There is a remarkable enhancement to the generated spectra for pulse durations below 50 fs compared to 88 fs and 95 fs. The pulse durations were chosen in accordance with the pump durations used on GaP at high repetition rates reported by other groups [183, 184]. Furthermore, the reduced pulse duration allows for operation at a lower average thermal load while maintaining suitably high peak powers, allowing for efficient operation in a non-critical thermal regime of the THz-generation crystal. The difference between the generated THz-radiation from the uncompressed output of the laser at 250 fs and from the compressed pulse duration of 20 fs is most striking. Especially the high-efficiency area of HMQ-TMS between 3-5 THz reaches an outstanding improvement. The higher frequencies also benefit strongly from the shorter pump pulse duration, while the low frequency components below 2 THz improve only slightly. This proves that external pulse compression is necessary not only to achieve higher peak power, but also an extended bandwidth of the pump pulse that allows for the generation of higher frequency components through OR. The spectral range of the modelling is limited to 12 THz due to the lack of data on HMQ-TMS beyond this frequency, but our own TDS-data suggests that there is little frequency content beyond 15 THz either way, see fig. 5.3a).

To verify and to gain confidence in the parameters used for HMQ-TMS, THz-TDS was conducted on the HMQ-TMS crystal. The THz-system used was an in-house broadband THz-source based on laser driven gas ionisation that provides a frequency range covering up to 18 THz at -20 dB using 40 fs pump pulses at 800 nm centre wavelength at a 1 kHz repetition rate. The optical power of 500 mW for generation was focussed to a spot size of 22 µm in diameter at FWHM. To detect such a large THz-bandwidth without phonon restrictions, the ABCD-method was used. For this, the high-voltage was set to 5 kV over an electrode gap distance of 5 mm, leading to a biased electric field of about 10 kV cm$^{-1}$, probed with a 80 mW 40 fs pulse. The results of the THz-TDS using the HMQ-TMS generation crystal as the sample oriented along its polar-axis (e.g. as it would be to generate maximum THz-radiation) are shown in fig. 5.3.

The spectral traces of the reference and sample measurements (calculated through FT of the recorded time-traces) are depicted in fig. 5.3a), and the refractive index and absorption coefficient extracted for a crystal of 0.26 mm thickness are shown in fig. 5.3b). Compared to fig. 5.2, the recorded spectra of the sample trace shows great overlap with the expected THz-generation resulting from HMQ-TMS. This makes sense, since the crystal should be transparent at the frequencies one can generate from it, while it is opaque for other frequencies. The results resemble the shape of the reported values used for the simulations in [174, 175], but the exact values differ, attributed to the inaccuracy of the sample-thickness. They are therefore to be viewed with caution, i.e. the refractive index values reported in [175] are 0.2 lower on average. A more recent study from 2020 [176] also indicates slightly lower values for the refractive index, meaning that the crystal thickness might be underestimated.
5.1 Theory

Figure 5.3. THz-TDS results for a 0.26 mm thick sample of HMQ-TMS. a) The spectra of the sample (red) and reference (blue) trace. b) The refractive index (blue) and the absorption coefficient (red) for HMQ-TMS extracted from the TDS measurement. Sample thickness uncertainty explains the mismatch in value to other literature reports [174–176]. Measurements were conducted along the polar-axis of the crystal.

To verify the preliminary modelling experimentally, HMQ-TMS was used for OR to generate THz-radiation, while the EOS detection was applied with a GaP crystal. The obtained results will be discussed in the following chapter.
5.2 HMQ-TMS: Experimental results

5.2.1 THz-setup

Since the initial HMQ-TMS measurements were run with the first compression method producing 30 fs pulses, and the subsequent experiments were conducted at the improved compressed pulse duration of 22 fs with higher average power, this section introduces two setups: The 22 fs pulse duration setup, which is referred to as the main setup, and the 30 fs pulse duration setup which was used for the first HMQ-TMS experiments.

Figure 5.4. Overview of the THz-setup. The few-cycle pulses are split into a pump and probe beam. The pump beam passes a variable neutral density filter (NDF) and is focussed into the generation crystal through a 250 mm lens and then filtered out with a PTFE filter. The collimated THz-radiation can be attenuated (Filter). The THz- and the variably time-delayed probe beam are combined through a Si-wafer. Both are then focussed into the detection crystal followed by the standard setup for EOS with a balanced photodetector (BPD). The detection crystal (GaP) can be exchanged with the THz-power meter (PM) (when blocking the probe beam). A chopper in the pump beam is connected to the lock-in amplifier.

The THz-generation setup through OR in the organic crystal HMQ-TMS is shown in fig. 5.4. The 22 fs compressed output of the external compression stage is sent through a beamsplitter to separate the probe and pump pulses and both beams are transmitted through variable neutral-density filters to allow for individual attenuation. The pump beam with a maximum power of 3 W (2.5 W for the 30 fs setup) is modulated by a chopper set at 673 Hz (500 Hz for the 30 fs setup), which acts as the reference signal to the lock-in amplifier. It is then focussed into a HMQ-TMS crystal of a selected thickness (0.25 mm or 0.45 mm), or into a GaP crystal (0.3 mm, 1.0 mm). The focussing lens has a focal length of 250 mm for the main setup (150 mm for the HMQ-TMS vs. GaP comparison at 30 fs), resulting in a minimum focal spot size of 0.17 mm at 1/e²-diameter (0.07 mm in the 30 fs setup). The rotation stage mounted crystal is placed on a lateral translation stage with a 25 mm travel range so that it can be moved in or out of the focus to vary the fluence, rather than changing the pump-power. The spot size of the 30 fs setup as a function of stage position is shown in fig. 5.5a) alongside the calculated fluence for pulses with 0.2 μJ pulse energy in fig. 5.5b).
Figure 5.5. a) The measured spot size diameter of the pump beam at 0.2 µJ as a function of the stage-position. The minimum is reached at 1500 µm of the 2500 µm travel range. b) The corresponding pump fluence, with the red shaded area indicating the damage threshold range for thin HMQ-TMS crystals.

Since the damage threshold lies at values as shown by red shaded area in fig. 5.5b), it becomes clear why the 250 mm focal length lens was chosen for the main setup with the 22 fs pump pulse duration, as this leads to a maximum fluence of 2.6 mJ·cm² at the focal waist when operating at 0.25 µJ pulse energy. This way, it is possible to vary the pump fluence within the uncertainty of the damage threshold, but not beyond as in the 30 fs setup (which is essentially “lost” operation area). A less tight focal spot size also allows for a slightly lower divergence of the generated THz-beam. The individual pulse energy is not reduced by the chopper, so for its calculation, double the value of the average power measured behind the chopper is taken and divided by the repetition rate of 10 MHz. After the pump pulse is transmitted through the crystal and has generated the THz-frequencies, the pump-light is removed by using two PTFE layers of 75 µm thickness each, attenuating through reflection and strong scattering of the IR light, while transmitting ∼89 % of the THz-power. The remaining THz-light is collimated with a two-inch focal length off-axis paraboloidal mirror, reflected off a flat gold-coated mirror and transmitted through a 525 µm thick Si-wafer angled at 45°.

This is the rendezvous point with the probe beam, allowing for a THz-power transmission of 57.8 % for the initial pulse, and a collective 62.6 % power transmission when including echoes from the Si-wafer. The probe beam is guided through a delay-stage and towards the silicon wafer by silver-coated mirrors to minimise dispersive effects to the pulse duration so that the high
temporal resolution of the ultrashort pulses is maintained. Both beams are aligned to propagate collinearly after the overlap through the Si-wafer and then focussed into the same spot by using a three inch focal-length mirror. At the position of the focal point, the detection crystal, a 300 µm thick <110> oriented GaP crystal attached to an index-matched 1 mm thick <100> GaP crystal, which mitigates etalon effects, is placed on a lateral translation stage to optimise alignment with the focal point. The detection crystal is also set in a rotation stage to optimise the detection scheme as outlined in chapter 4.3.1. A 50 mm focal length lens is used to collimate the probe beam which is then guided towards the quarter waveplate (Thorlabs model WPQ05M-1030), the Wollaston prism (Thorlabs model WP10) and the balanced photo-receiver (Newport 2107-FS-M), acting as the lock-in input signal, as described in chapter 4.3.1. The photo-receiver has adjustable low- and high-pass filters which were set to 100 Hz and 10 kHz, with the gain set to 1. This way, the probe beam of 10^10 pulses per second is modulated at 673 Hz by the THz-induced birefringence. Each modulation period therefore contains ~7,429 pulses, compared to the one pulse encountered in a 1 kHz system chopped at 500 Hz. The chopping frequency was chosen so that the signal of the photo-receiver on an oscilloscope when modulating the probe beam at 673 Hz would show as a sharp signal-response with very short rise-time in an almost square-pulse shaped pattern.

For the THz-power measurements, another two layers of PTFE are placed behind the first paraboloidal mirror to remove any collimated stray light from the pump and the THz-beam was modulated at 15 Hz to be detected with a calibrated pyro-electric detector (Gentec-EO). To remove the influence of any residual IR incident on the final THz-power value, the generation crystal was moved far out of the focal-spot to prevent any meaningful THz-generation, and the remaining reading recorded by the THz-power meter at this setting was used as the background-signal to deduct from the final power-measurements. Despite using several PTFE and Si-filters this was still necessary, as the measured THz-power values would be in the low µW-regime, and pumping at over 2 W optical power, the residual IR in the µW-regime is still prevalent even after a -60 dB attenuation. Any additional filters placed in the beam path were accounted for by back-calculation of their transmission values. The power transmission values of Si-wafers and PTFE were recorded with an in-house broadband THz-system (reaching 30 THz [185]) through THz-TDS. Values were extracted for the relevant spectral range of 0.5-5 THz (for HMQ-TMS) and 0.5-3.5 THz (for GaP)^2, rather than applying standard literature values for each material, as batch-to-batch thickness, production quality, and individual assembly of the PTFE-filters will affect the absolute transmission value. Furthermore, the use of stacked silicon wafers was avoided, since the transmission function is sensitive to the angular alignment of consecutive wafers and is not given by the simple approximation of 0.5^n [164].

With collinear setups, alignment is straightforward, as one can remove the IR-filter after the generation crystal to trace the (attenuated) IR-pump beam which will overlap with the THz-beam. This allows for visual alignment (using detector cards), knowing that the pump and THz-beam should follow the same trajectory. The important parameters for the THz-TDS therefore lie with the

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1One modulation consists of an opening and shutting of the chopper wheel, hence the number of pulses is \( f_{\text{rep}} / f_{\text{chop}} / 2 \)

2These are the frequency windows corresponding to the -20 dB bandwidth of their respectively generated THz-bandwidths when used as a generation crystal
lock-in detector (Stanford Research Systems SR830), which has to be set correctly to resolve the THz-time trace accurately, whilst ideally keeping measurement times as short as possible. Whenever the time-delay stage moves, vibrations to the mirrors add noise to the recorded signal, further necessitating a "wait-time"\(^3\) after each step that is longer than the integration constant of the lock-in amplifier. Depending on the Slope/Oct value, the ideal ratio of wait time to integration constant varies between 5 and 10. The specific settings depend on the signal quality and strength, which lead to integration time constants used in this thesis being between 300 ms and 1 s, a high-reserve setting and a medium-high sensitivity, as the initial system noise fed into the lock-in was high compared to the actual signal containing the information.

![Comparison of the expected THz-output in HMQ-TMS from a transform-limited pump pulse (blue) at 25 fs and the real compressed 25 fs pulse (red). The cleaner pulse shows a better performance in amplitude. This is due to the increased amount of peak power in the TL pulse, whereas the real pulse contains part of its energy in side-lobes.](image)

**Figure 5.6.** Comparison of the expected THz-output in HMQ-TMS from a transform-limited pump pulse (blue) at 25 fs and the real compressed 25 fs pulse (red). The cleaner pulse shows a better performance in amplitude. This is due to the increased amount of peak power in the TL pulse, whereas the real pulse contains part of its energy in side-lobes.

Lastly, the pulse durations presented here, 30 fs and 22 fs, are the pulse durations measured through IAC. The ND-filters add a slight amount of dispersion and the focusing lens also distorts the pulse temporally [39]. This partly explains the observed differences between experimental result and theoretical prediction together with the difference between a perfect, transform-limited pulse and the real experimental trace with its temporal structuring. This is shown in fig. 5.6, which depicts the expected THz-amplitude over frequency for an optimal 25 fs and the experimental pulse of 25 fs measured through FROG\(^4\). This highlights the effect of the quality of the pump pulse, as OR requires all pulse-internal frequencies to interact with each other within the crystal. A non-transform-limited pulse will have frequency components outside of the main pulse (see fig. 3.9).

\(^3\) Additionally to the time required from the processing of the lock-in amplifier itself

\(^4\) 25 fs is chosen over 22 fs, since this is the duration recorded by the FROG
5.2 HMQ-TMS: Experimental results

Reducing the bandwidth used for OR when the main pulse with the large intensity reaches the crystal. Especially high THz-frequency conversion efficiency experiences a downgrade when a non-perfect temporal pulse is used for THz-generation through OR. A detailed study on the effects of TOD in the pump pulse for OR in PPLN comes to similar conclusions [186].

For the calculation of the THz-field strength it was necessary to record the THz-spot size in the focus where the GaP detection crystal stood. The exact position of the focus was found by using an uncooled micro-bolometer (NEC THz Imager IR/V-T0831) with a pixel size of 25.5 µm and moving it along the transmission path of the THz-beam. The position of maximum intensity on the camera coincides with the optimal position of the GaP detection crystal.

5.2.2 HMQ-TMS vs. GaP

The first set of experiments were conducted with a 30 fs pump pulse on a HMQ-TMS crystal of 0.25 mm thickness and compared to the THz-output of 1.0 mm GaP at the same pump parameters. As GaP is the typical "work-horse" for THz-generation with collinear OR for emission wavelengths at about 1 µm, any new crystal aiming to replace it must show superior properties. These can be a combination of conversion efficiency, THz-generation bandwidth, damage threshold, size-scalability, or manufacturing costs. To show that HMQ-TMS is such an alternative that offers great advantages, a collinear OR setup was built and the generated THz-radiation was detected using a standard EOS-setup with a GaP detection crystal. The pump-beam was a 30 fs, 0.25 µJ train of pulses at 10 MHz repetition rate, focussed through a 150 mm optical lens into the 0.25 mm thick HMQ-TMS or 1 mm thick GaP crystal.

Experiments conducted at 2 W pump-power in dry-air filled environment on 0.3 mm thick GaP crystals showed no significant bandwidth change compared to the 1 mm thick GaP crystal, apart from slightly more frequency content above 4 THz, though this was below -20 dB. The THz-field amplitude was just 35 % of that achieved with the 1 mm GaP crystal, as shown in fig. 5.7. For the ease of alignment and detection, and due to the relative similarity of the bandwidth, the thicker 1 mm GaP was chosen to be compared to HMQ-TMS.

Due to the tight focal spot size, pulse energies above 0.2 µJ were not applicable as it instigated immediate damage occurrence on HMQ-TMS, which is why the pump-beam was attenuated to below 2 W of average power. The generation crystal was moved slightly out of focus to a spot size of 0.1 mm at 1/e^2 diameter to avoid the critical fluence. When replacing the generation crystal to GaP, the stage-position was not changed. This insured that GaP and HMQ-TMS both experienced the same fluence, with a minor uncertainty arising from the exact lateral position of the crystals in their rotation mounts and the difference in thickness.
Figure 5.7. a) THz-electric field strengths of a 0.3 mm (red) and 1 mm thick (blue) GaP crystal normalised to the thick crystal’s peak. b) THz-spectrum of the two crystals. The thin crystal (red) offers a broader bandwidth (at very low dB), whereas the overall spectral power for the thick crystal (blue) is significantly larger.

Figure 5.8 shows the recorded EOS traces when pumping either crystal with 0.068 µJ pulses (0.680 W of average power before the chopper wheel) and their respective frequency-domain conversion, both normalised to the maximum value of the HMQ-TMS signal. The THz-spectral power obtained from the HMQ-TMS crystal shows an improvement over GaP of two orders of magnitude, consistent with previous reports [174]. The spectral bandwidth is improved up to 6 THz at -30 dB for HMQ-TMS, almost twice the bandwidth achieved with GaP. As demonstrated earlier, a thinner GaP crystal can produce a larger bandwidth than what is shown in fig. 5.8. GaP has been reported to provide large bandwidths of up to 7 THz with a 0.15 mm thick crystal at 1030 nm [184], although with a notable shift of the spectrum to higher frequencies overall. Additionally, the extended spectrum for GaP suffers from a low spectral power density. The HMQ-TMS spectrum, in contrast, shows a smooth, almost gap-less, flat-top shape from 2-5 THz (in a dry-air atmosphere). This bandwidth falls in line with previous work on 0.22 mm thick HMQ-TMS crystals pumped with 65 fs pulses at 1000 nm centre wavelength [175].

The direct comparison between HMQ-TMS and GaP here was held at 680 mW pump power to securely avoid damage to the crystal, and each measurement was taken several times to compute an average trace with minimal noise. This process required very long illumination times exceeding two hours, giving further incentive to use a low pump fluence. For the direct comparison of the conversion efficiency, the setup could not be purged by dry-air due to space
constraints resulting from the THz-power meter. Since this added a \( \sim 40\% \) power loss to the measurements\(^5\), the GaP signal was too close to the noise floor at a pump power of 680 mW. To accurately measure the THz-power, the pump power was increased therefore to 2 W for both crystals. Precaution was taken by moving the stage position further away from the focus. The recorded value of the THz-power meter placed at the position of the detection crystal was corrected for any attenuation and filters present in the setup as well as for the humidity-loss.

The 1 mm thick GaP crystal provided an average power of 8.8 µW when pumped at 2 W (0.2 µJ), corresponding to a conversion efficiency of \( 4.0 \cdot 10^{-6} \) which is comparable to efficiencies reported recently by other groups working on high repetition rate OR with GaP \([118, 183, 187]\), though cryogenic cooling might lead to higher values \([188]\). The 0.25 mm thin HMQ-TMS crystal provided 380 µW of average THz-power at equal pump settings to that of GaP, leading to a conversion efficiency of \( 1.86 \cdot 10^{-4} \). This is \( \sim 50 \) times larger than that achieved with GaP under equal conditions. The results indicate an electro-optic coefficient at the pump wavelength of 1035 nm for HMQ-TMS larger than 50 pm\( \cdot \)V\(^{-1}\) (based on that from GaP at 1 pm\( \cdot \)V\(^{-1}\)). The actual value might be even larger, as the GaP crystal here is four times thicker than the HMQ-TMS crystal. This leads to more THz-power and thus a better conversion efficiency, as showcased in fig. 5.7.

\(^5\) Loss was determined from EOS traces in dry and humid air, and verified computationally.
Previous work on HMQ-TMS at 1030 nm has reached conversion efficiencies of 26·10^{-4} at 500 Hz in a 0.63 mm thick HMQ-TMS crystal [178], and 3.5·10^{-5} at 800 nm [177] (repetition not specified, but assumed at 1 kHz). Further studies at 1100-1500 nm have shown that the crystal provides a high conversion efficiency at many pump wavelengths [179]. This thesis shows that MHz operation with HMQ-TMS is just as feasible in terms of generation efficiency and achieved bandwidth as with low-repetition rate sources.

The previously reported damage threshold for HMQ-TMS at low repetition rates due to multi-photon absorption lies at a fluence of >20 mJ·cm^{-2} at 100 Hz [175]. This is in sharp contrast to the significantly lower damage threshold encountered in this thesis and can be traced back to the change in repetition rate leading to a thermal damage mechanism. While performing standard EOS experiments at different pump powers, a recurring damage pattern was observed, which was a burnt hole in the generation crystal at the focal point of the pump. The hole was surrounded by shapes resembling molten plastic, as seen in fig. 5.9 in the black area beneath the burnt-through hole.

![Figure 5.9](image_url) Close-up view of a fragment of HMQ-TMS destroyed by thermal damage. The black melted area around the initial pump-spot is far larger than the pump-beam spot size of \(\sim 100 \mu\text{m}\) in diameter, which is in contrast non-thermal laser-induced damage.

It seems obvious that due to the change in repetition rate, the deposited heat accrued through absorption of the IR light can not be dissipated fast enough. This leads to a positive net flux of heat incident on the crystal after a given threshold power is reached, causing it to heat up to its melting point at 274 °C [172] and to destroy the crystal in the process. This threshold power is termed the damage threshold in this thesis and varies for different sets of crystals between 1.8-3.6 mJ·cm^{-2}. This low threshold is the main disadvantage compared to GaP with a
MHZ-damage threshold of 4.3 mJ·cm$^{-2}$ [189]. In terms of peak irradiance, the damage threshold corresponds to 52-107 GW·cm$^2$. A positive observation was the long-term stability of HMQ-TMS over several hours of operation. Typically, organic crystals are supposed to be stored in dry-air environments to prevent the incorporation of water into the crystal structure. This seemed unnecessary for HMQ-TMS, as samples that were facing ambient environment over a prolonged period of time did not show a degraded performance. It must be noted that this damage threshold does not hold when the pump beam is not chopped, as the vital "cool down" periods between subsequent irradiation times is removed. The constant illumination leads to a faster thermal build up as this heat-removing aspect is reduced. A detailed study of the damage was not conducted due to the limited supply of the HMQ-TMS crystals.

The field strength for the traces recorded at their best THz-power performance at the point of tightest THz-focus was calculated using the spot size and the method from eq. (4.20). An exemplary THz-spatial intensity distribution is depicted in fig. 5.10. The discrepancy between the x- and y-axis radius results from focusing characteristics of the paraboloidal mirror combined with a not perfectly collimated and straight incidence THz-beam. Despite this, the Gaussian approximation to the THz-intensity distribution is verified through the fits in fig. 5.10. The strongest THz-field strengths achieved with the 30 fs pump pulses were calculated to be over 7 kV·cm$^{-1}$ at an average THz-power of 0.38 mW (corrected for all losses, including humidity) and a measured average Gaussian radius of 360 µm.

![Figure 5.10](image.jpg)

**Figure 5.10.** THz-camer recorded spot size (exemplary) of the focussed THz-beam at the point of EOS detection. The $1/e^2$-intensity radius is 157 µm on the y-axis and 207 µm at the x-axis. The discrepancy comes from misalignment on the focussing paraboloidal mirror. Measured values (blue circles) are well represented with a Gaussian fit (blue line).
5.2.3 Crystal thickness impact

After improvement of the pulse-compression to 22 fs, similar experiments to the HMQ-TMS vs. GaP procedures were carried out with HMQ-TMS crystals of thickness 0.25 mm and 0.45 mm in order to see the impact of the crystal thickness on THz-generation. As expected from the theory presented in chapter 5.1, the THz-output bandwidth and generation efficiency improved when using the shorter and higher power pump pulse compression setup. Due to the fact that up to 3 W of average power was available and the damage threshold for HMQ-TMS is known from the previous experiment, the chosen focusing lens was 250 mm in focal length, allowing for a less tight focal spot with a maximum fluence just above the damage threshold. This way, the generation crystal could be placed within the Rayleigh range of the focused beam with a Gaussian diameter of 0.17 mm and the power was controlled using a neutral density filter. Experiments were conducted in ambient atmosphere with a relative humidity between 20-30 %, leading to the strong absorption dips in the spectrum seen in fig. 5.11, with the pump power set to 2.5 W (0.25 µJ).

Figure 5.11 shows the calibrated THz-field time trace for both crystal thicknesses and their corresponding spectra, which span almost 6 THz at -20 dBm·THz⁻¹ for either crystal. The achieved peak field in ambient air from the thin crystal is ~6.3-6.4 kV·cm⁻¹, 20 % stronger than that achieved with the thick crystal (~5.3-5.4 kV·cm⁻¹). This may initially seem counter-intuitive, as the field strength for GaP increased with the thicker generation crystal as shown in the previous chapter. However, a closer look at figures 5.1 and 5.2 explains this. The main part of the THz-energy when pumping with an ultrashort pump pulses is centred between 3-5 THz, where the coherence length is generally below 0.4 mm. The thicker crystal benefits from the good phase-matching and long coherence lengths at low frequencies, which manifests itself in the stronger low-frequency content seen in fig. 5.11b). However, most of the frequency content is found at higher frequencies, where the thicker crystal is at a disadvantage, leading to an overall spectral power content of only 80 % of that produced by the 0.25 mm crystal. Furthermore, the conversion efficiency relates quadratically to the THz-frequency, meaning that higher frequency components have a stronger effect on the overall generation efficiency [112]. Considering that water vapour absorption significantly affects this spectral region, the observed difference will only increase when placing the setup into dry-air conditions.

The peak field of the thin crystal reaches 6.4 kV·cm⁻¹ for a THz-average power of 0.904 mW (not corrected for water losses) at 10 MHz repetition rate. A sizeable portion of the initial THz-pulse energy is contained in the oscillations in the wake of the main pulse caused by the humidity. Comparing this to the dry-air trace in fig 5.8 which reaches peak fields of 7 kV·cm⁻¹ at ~1/3 of the THz-power of the 6.4 kV·cm⁻¹ trace, shows the strong influence of water vapour on broadband THz-pulses and will be discussed in more detail in chapter 6. Therefore, by operating at low humidity or by focussing the THz-beam more tightly, peak on-axis field strengths beyond 20 kV·cm⁻¹ can readily be achieved. Such moderately strong few-cycle i.e. femtosecond THz-pulses at MHz repetition rates could offer an improvement for various applications, e.g. broadband THz-STM measurements [190] allowing for high spatial and temporal resolution.
Figure 5.11. a) The recorded and calibrated THz-field generated in a 0.25 mm (blue) and a 0.45 mm (red) thick HMQ-TMS crystal. The peak field strength for the thinner crystal is \( \sim 20\% \) larger. b) The corresponding spectral power density. The thick crystal (red) has a higher yield at low frequencies, but overall, it loses out on power due to the increased high frequency components accessed through the thinner crystal (blue).

For the efficiency calculation, the full THz-power of \( \sim 1.38\) mW (corrected for humidity-loss) for the thin crystal is used. This leads to an optical-to-THz conversion efficiency of \( 5.5 \times 10^{-4} \) for the thin crystal, while the thick crystal conversion efficiency is slightly lower at \( 3.3 \times 10^{-4} \). Compared to the efficiency achieved with the 30 fs setup, this is a threefold increase gained from using a shorter pump pulse duration. Additional conversion efficiency increase can therefore be expected when using shorter and temporally less structured pulses. Lastly, the observed damage threshold from the HMQ-TMS vs. GaP experiments was verified, though for the thick crystal the value is shifted to a slightly lower fluence. This is attributed to the higher absorption of IR-pump light due to the extended interaction length. As the surface area remains the same, the increased absorption leads to a faster thermal build-up. For this reason, a fluence limit below 1.8 mJ cm\(^{-2}\) is suggested for safe operation with HMQ-TMS.

Pumping HMQ-TMS in a collinear OR setup with a high repetition rate source and ultrashort pulses is hereby proven to be a suitable and easily implemented method to obtain mW-level broadband THz-radiation with field strengths of tens of kV cm\(^{-1}\). The main drawbacks of the organic crystal HMQ-TMS are its comparatively low damage threshold and the very brittle nature of the crystal, leading to difficult handling when mounting or combining it to other substrates.
Despite this, the average THz-power of 1.38 mW achieved here is, in theory, scaleable through several factors:

- The crystal can be grown to large sizes. Combined with a high-power laser source and a large beam-area that offers a fluence of $\sim 1.5 \text{ kV cm}^{-2}$, this would allow for generating several milliwatts of THz-power. Tightly focusing such a high-power THz-beam might lead to field strengths approaching hundreds of kV cm$^{-1}$, though thermal power handling remains a strong issue.

- Cryogenic cooling of the HMQ-TMS sample, or simply mounting the sample to a transparent substrate to act as a heat sink - such as sapphire - would greatly improve the power-handling capability as heat removal contributions are increased. This would allow for stronger optical pumping, leading to higher output powers and conversion efficiencies.

- Even shorter pulses with broader internal bandwidth should lead to an increase of the conversion efficiency and higher frequency components. The solid-core concept provided in this thesis is ultimately limited as demonstrated in chapter 3.5, but hollow-core fibre compression could lead to higher-power and shorter pulse duration pump beams to be used for the OR process.

This concludes the chapter on providing analysis of the work on generating a high-power THz-source at MHz repetition rates using the highly efficient organic crystal HMQ-TMS. The following chapter will give a brief overview of the absorption of water-vapour through analytical modelling, succeeded by the conclusion to this thesis.
In chapter 4, the impact of air humidity on the THz-pulses and power was briefly mentioned, as the high-power, high repetition rate THz-source introduced in this thesis is also in part aimed at applications with significant power loss. Water vapour is the main atmospheric component affecting THz-radiation in the approximate frequency range of 0.1-17 THz. The next strongest contributor to attenuation, carbon dioxide, exerts its first impactful absorption at $\sim$20 THz, which is rarely reached with typical THz-sources and therefore the following discussion will focus on water only. With the growing impact of THz-science in real-life applications, the water related effects need to be addressed more thoroughly as it is often not possible to operate in purged atmosphere.

The first THz-TDS paper was in fact carried out on water vapour [19], and with increased interest in THz-radiation from telecommunication applications or remote sensing and detection, one would expect many detailed studies addressing this topic. However, qualitative and detailed studies are not in abundance. They mostly cover the low THz-frequency range below 3 THz (relevant for telecommunication applications) and rarely exceed 40 % RH values [191–198], though some offer long distance measurements of over 100 m. There is scarce experimental information at higher frequencies exceeding 3 THz, with only very few dedicated studies analysing the impact of varying humidity levels explicitly, e.g. up to 14 THz [199], though these are limited to below 40 % RH. Despite this lack of published experimental data, several spectral databases offer information on the absorption line-widths for these less extensively studied frequencies. The (probably) most widely known source being the high-resolution transmission molecular absorption (HITRAN) database 1. As for the data in the HITRAN database, the Harvard–Smithsonian Center for Astrophysics - which maintains the database - states on its website 2 that “The parameters in HITRAN are sometimes direct observations, but often calculated”.

The available data one can download for different isotopes from HITRAN gives the user a set of absorption line-strengths and line-widths rather than more practical absorption coefficients. Converting these into absorption coefficients for a given concentration of the selected molecule (i.e. relative humidity in the case of water) requires moderate mathematical analysis and an assumption of the absorption to be Lorentzian in shape. This assumption is valid for weak absorption lines, as the symmetry is mostly conserved, and holds for the THz-absorption of water. The data from HITRAN was converted to absorption coefficient values with the help of a python code written by Peter Uhd Jepsen. The code also allows for propagation simulation of a chosen input pulse

2https://www.cfa.harvard.edu/hitran/ - accessed 10.11.2020
6 THz-absorption in humid air

(Experimental or simulated), which was used to compute and study the effects of RH and propagation distance. It establishes the connection between the HITRAN line strength and width, the Lorentzian line profile, and the Lorentzian oscillator model of the permittivity based on the work presented in [200]. In principle, it offers a path to reach the dielectric function required to propagate a simulated pulse based on the absorption data obtained from HITRAN. A few assumptions allow for an easier approach. These include setting the refractive index of water to unity, allowing for first order approximation of the refractive index, as well as only viewing frequencies close to resonance \( f = f_0 \). The simplifications result in an expression for the extinction coefficient \( \kappa \) derived from the Lorentzian oscillator model, which is linked to the absorption coefficient through \( \alpha = 4\pi f_0 \kappa \). Through this connection, the two line profiles (HITRAN and Lorentzian oscillator model) can be equated, leading to a direct connection between the HITRAN data and the corresponding parameters in the permittivity model.

This chapter will focus in more detail on THz-absorption and power loss through water vapour, as well as the behaviour for different relative humidity (RH) values and for changing propagation distances. The results will show that water vapour absorption cannot simply be approximated by using a given overall absorption coefficient for a certain bandwidth in combination with Beer-Lambert’s law.

![Figure 6.1](image)

**Figure 6.1.** a) The HITRAN-data line strengths for water absorption over THz-frequencies. b) Absorption coefficient (blue) obtained from the line strengths and power absorption (red) for a 45 cm beam path at 25 % RH.
To begin with, fig. 6.1a) shows the water absorption line-strengths using HITRAN data for THz-frequencies up to 20 THz for isotopes $\text{H}_2^{16}\text{O}$ and $\text{H}_2^{18}\text{O}$, which together make up 99.87317% of all their isotopes, according to HITRAN. The line-strength is not equivalent to the absorption, which is illustrated in fig. 6.1b) where the corresponding absorption over 20 THz is shown at a RH of 25% and a propagation distance of 45 cm (right axis) as well as the general absorption coefficient at 25% RH (left axis), are shown. Clearly, large parts of a broadband THz-pulse’s spectrum will be completely absorbed and lost. The conversion from the HITRAN line-strength data to absorption coefficient is given by eq (6.1) and is computed using the aforementioned code:

$$\alpha(\tilde{\nu}) = \rho S_{ij} f(\tilde{\nu}), \quad (6.1)$$

where $f(\tilde{\nu})$ is the normalised spectral line profile in units of [1/cm$^{-1}$], $\tilde{\nu}$ the wavenumber in units of [cm$^{-1}$], and $\rho$ the molecular density in [molecules/cm$^3$]. As stated by HITRAN, a Lorentz-shaped line profile for $f(\tilde{\nu})$ is a valid approximation to real line shapes, and therefore eq. (6.2) can be used to define $f(\tilde{\nu})$:

$$f(\tilde{\nu}) = \frac{\tilde{\gamma}}{\pi \tilde{\gamma}^2 + (\tilde{\nu} - \tilde{\nu}_{ij})^2}. \quad (6.2)$$

Here, $\tilde{\gamma}$ is the HITRAN data for the half-width at half maximum of the absorption line profile in units of [cm$^{-1}$], whereas $\tilde{\nu}_{ij}$ is the central wavenumber in [cm$^{-1}$]. By setting $\tilde{\nu} = \tilde{\nu}_{ij}$, the peak absorption coefficient of a HITRAN line becomes $\alpha = \rho S_{ij} / (\pi \tilde{\gamma})$, which easily allows for converting the HITRAN data to absorption coefficient for a given density of water molecules, i.e. the relative humidity.

To verify the accuracy of the (mostly calculated) HITRAN data as well as the aforementioned code, the recorded “dry” THz-traces are computed to propagate at a given RH and distance, and are then compared to equivalent-settings traces obtained experimentally. To ease the computational load and time, a threshold value is set which cuts off absorption lines that lie below 0.01% of the strongest line-strength and the CO$_2$ concentration is set to zero. This is justified, as any significant CO$_2$ concentration is prevented by purging the setup with nitrogen and then adding the humidity for a certain RH value. The humidity is generated through a commercial room-humidifier running on distilled water and guided into the setup box using tubes fed with nitrogen. Simulation temperature is set to room-temperature of the laboratory at 23°C and the atmospheric pressure is assumed to be at 1 bar. The experimental systems used were a commercial photoconductive antenna system from Toptica Photonics (5 THz bandwidth at -80 dB), and a two-colour laser system generating THz-radiation through air-plasma (up to 20 THz bandwidth at -20 dB) using ABCD to record the THz-field.

The extremely low noise level of the Toptica system results from averaging several thousands of pulses over one minute. This high accuracy level and averaging was not possible in the laser-plasma system due to a single scan duration of half an hour that was necessary to achieve the desired temporal and spectral resolution. Since these scans required such a long time, no averaging over several scans was possible and therefore the noise level remained high. Adding to this was the issue of maintaining a stable RH in the box over such a long period, which is why an average of the logged RH was taken as the reference humidity. Problematically, a decrease or
increase over the time of the measurement would affect the amplitude of the recorded water ringing. Lastly, system instabilities of the source laser and thus also of the generated THz-radiation over the course of the entire measurement process, further added to the increased inaccuracy of the data. Nonetheless, the position of the absorption peaks recorded with the two-colour system matched very well with the computed ones, strengthening the confidence in the HITRAN data.

To show the accuracy of the code, an exemplary recording exhibiting good overlap between a recorded trace at 25 % RH and the computed trace is depicted in fig. 6.2a) (traces at other RH demonstrate similarly good overlap). The input for the computed trace was a reference trace recorded under dry circumstances with the Toptica system. The absorption peaks in fig. 6.2b) overlap perfectly, and the absorption strength is highly accurate even at -60 dB. This proves that the code provides a tool that accurately propagates a THz-field through a simulated atmosphere and distance.

![Figure 6.2.](image)

**Figure 6.2.** a) Temporal and b) spectral traces of an experimentally recorded THz-trace at 25 % RH (blue) and the computed result from propagating the recorded dry reference trace (dashed red). The two curves show almost perfect overlap, highlighting the accuracy of the code used to simulate propagation in humid atmosphere.

After verifying the accuracy of the code experimentally, further simulations are conducted to observe the behaviour of water-absorption for varying distance and RH. Since water absorption
has a strong impact on THz-pulses, it is of interest to quantify the behaviour of the absorption coefficient with increasing RH for a given bandwidth over varying distances. According to eq. (6.1), the absorption coefficient relies linearly on the density of molecules, which equates to the RH. Therefore, the typical assumption [199] is that increasing RH relates linearly to the absorption coefficient, which is tested here by propagating a simulated THz-pulse with a centre frequency of 5 THz for a fixed distance of 100 cm at increasing RH levels of 5%. A linear increase of the overall absorption coefficients should transpose to an exponential decay for the pulse’s total transmitted spectral power of the form \( T(RH) = \exp(-\alpha(RH)100) \), with \( \alpha(RH) \) the absorption coefficient for a given RH. Indeed, for a single frequency at a chosen absorption line, the absorption coefficient increases linearly, as can be seen in fig. 6.3a). However, as fig. 6.3b) shows, the overall transmission over the entire bandwidth for increasing RH is not represented by a simple exponential decay. This is contrary to the common assumption regarding water vapour absorption, which is based on the limited RH-range of studies in the established literature. The misrepresentation of the absorption though a simple exponential decay increases with broader bandwidth THz-pulses. In fact, the decay is accurately represented by a stretched exponential function of the form \( \exp(-\alpha^\zeta) \).

![Absorption coefficient α at 4.5161 THz over relative humidity.](image)

**Figure 6.3.** a) Absorption coefficient \( \alpha \) at 4.5161 THz over relative humidity. Due to the linear dependence of \( \alpha \) on the molecular density, the graph is strictly linear. b) Transmission of a computed broadband THz-pulse in % over increasing RH. The y-axis is on a log scale, so that an exponential decay is resembled by a straight line. However, comparing the measured data (black stars) with a simple exponential fit (blue line) shows that it is not exponential. Instead, a stretched exponential (red) is necessary.

The interpretation of this behaviour relates in part to the fact that the line-widths are not delta-
functions. Instead, they have a finite line-width that is not square-shaped and overlaps with neighbouring absorption lines, especially at the base of the Lorentzian line-profile. A stronger impact results from the varying absorption strength for the different frequency absorption lines. Assuming a cw THz-source at a single frequency, the resulting decay of power will in fact be purely exponential. But with a broadband THz-pulse, the different frequencies will experience varying strengths of attenuation that could be described with a sum of several exponential decay functions for each absorption line of the form $e^{-\alpha_1 x} + e^{-\alpha_2 x} + e^{-\alpha_3 x} + \ldots$. However, adding each absorption line is strenuous, and the decay is thus represented more concisely by the stretched exponential function introduced above. The physical meaning of the stretched exponential parameter $\zeta$ is hard to define, as it varies depending on the amount of data points taken from the same curve. For example, a fit to data plots up to 50 % RH will have a different $\zeta$-value than a fit to the same data, but using all points up to 100 % RH. The extrapolation of the shorter fit matches very well with the long fit, but the parameters can be quite different. Therefore, only trends can be extracted, e.g. a change in RH will lead to an increase or decrease of the parameter.

The same behaviour is also observable for a fixed relative humidity and varying transmission distance, as shown in fig. 6.4. Here, the RH is set to 35 % and the distance is increased in 5 cm steps from 0 to 100 cm. The overall spectral power transmission is plotted over the distance for a THz-pulse with a centre frequency of 3 THz. Typically, broader THz-pulses will experience lower transmission, as more absorption lines are affecting the pulse, while increased RH also leads to increased transmission loss.

Figure 6.4. Transmission over beam path for a computed broadband THz-pulse at 35 % RH. As with fig. 6.3, a stretched exponential (red) is necessary to adequately fit the data (black stars), rather than a standard exponential (blue).
A comprehensive experimental study using the air-plasma system was conducted, but additional real-world effects resulted in a far lower transmission of power than was expected from theoretical modelling of the dry reference pulse. As shown in fig. 6.5, the line representing the experimental overall transmission lies well below the calculated values, with the mismatch increasing for growing values of RH, i.e. a higher concentration of water in the air. Since the computed result predicts higher transmission, the logical conclusion is that the experimental signal strength recorded was weaker than it should have been for a purely water absorption related loss. The missing signal strength can result from a reduced detection efficiency of the ABCD method (placed within the humidity-box). Humidity affects the conductivity of air and its break-down threshold, both possibly leading to an effect on the $\chi^{(3)}$-value of air, which governs detection. The humidifier used in the experiment generated a visible stream of fog, which may have affected the broadband pulse through scattering processes. Fog droplets have a wide range of size distribution depending on several parameters. They can range from sub-micrometre diameters [201] to sizes of 50 µm [202] and beyond [203], though no larger than 100 µm. Hence, the scattering impact for THz-frequencies above 6 THz (50 µm) cannot be dismissed before the size-distribution of the fog droplets produced by the humidifier is analysed. Scattering effects reduce the amount of THz-radiation that reaches the detection point, which translates to a power-loss in the analysis, though it is in fact not related to the absorption coefficient of the water molecules. Lastly, especially at high humidity, condensation of water droplets or thin films on optics in the beam path such as mirrors or lenses (e.g. in the Toptica system) can have a severe impact due to the stronger absorption coefficients of liquid water compared to vapour [204]. Despite this, the general shape of the stretched exponential is preserved for the experimental trace.

![Figure 6.5](image.png)  
**Figure 6.5.** Transmission of an air-plasma broadband THz-pulse for increasing RH in 67.5 cm of air. The experimental data (blue) is well approximated with a stretched exponential (green). The lower transmission values compared to the same pulse’s simulated transmission (red) are a result of additional system loss. Data above 80 % RH was not recorded due to breakdown of the ABCD-bias field.
Through THz-TDS one can technically extract the absorption coefficient and refractive index of dry air mixed with water vapour, though the figure shows that the results obtained experimentally will also be affected by additional loss contributions. These distort the absorption coefficient results when assuming pure water absorption, especially with broadband THz-pulses at higher RH or longer propagation distances, so special care must be taken when performing and analysing such measurements. An additional error in this specific case occurred from system instabilities of the pump laser and the required long scan times of up to thirty minutes. It should also be noted, that the 100 % transmission value in fig. 6.5 is set at 9 % RH, which was the recorded level of the humidity at the “dry” reference trace for the particular setup. This air plasma setup in a self-constructed purge box had the THz-generation placed at constant humidity, while the THz-detection was within the varying humidity environment, potentially affected the detection efficiency. Overall, the air-plasma study on the effects of humidity was superior to the Toptica system only in bandwidth, as the long scan-times and lack of averaging greatly reduced the data quality. This highlights a potential application for a high-power broadband THz-source operating at several MHz, such as the one introduced in this thesis.

This chapter has shown that simple exponential decay resulting from water absorption in air for THz-frequencies is only valid in specific circumstances, e.g. very narrow bandwidth, low RH or short propagation distances. When considering the overall contribution of water on broadband THz-absorption, a stretched exponential function describes the power decay more accurately, as the many absorption lines acting upon the pulse are of varying strength. What initially seems like a simple experiment to determine absorption coefficients of water through THz-TDS, is in fact not conducted as straight forwardly as one might hope for. This is due to additional power loss contributions, such as scattering, that reduce the recorded THz-power and thus lead to an overestimation of the absorption coefficients extracted through THz-TDS. Practically, this might not affect the applications that rely on water absorption values, such as telecommunications, as overall loss - be it through absorption or scattering - is loss either way and thus can be included in the effective absorption coefficient. From a scientific standpoint though, these contributions need to be viewed separately by calculating the absorption loss through numerical methods and then comparing it with the experimental values to extract additional loss.
In summary, this thesis has outlined a feasible method to combine two exciting areas of nonlinear optics to allow for efficient and high-power THz-radiation generation at MHz repetition rates. The aim of part I was to build and demonstrate the use of a stable external compression setup driven by the powerful output of a (commercial) femtosecond fibre-laser which was introduced and characterised in chapter 3.1. External pulse compression, the maths and physical principles were explained in chapters 2.1-2.4. The classical understanding of this method was put to test by using a LMA-PCF rather than a standard SMF at peak powers above 1 MW. At such high peak powers, several nonlinear effects take place within the fibre, exerting their influence onto the pulse. An undesired effect is SRS, which distorts the linear chirp of the broadened pulse, leading to complications with the subsequent compression when using a pair of glass prisms. To find a balance between spectral broadening and undesired phase accumulation, the SRS-threshold and the optimal fibre length for classical compression were compared to each other in detail in chapter 3.3. The results offer a starting point for the chosen peak power and fibre length for the experiment, and the results of the broadening and the compression were showcased in chapter 3.4. A new approach was introduced by using a polarisation maintaining LMA-PCF, preventing polarisation cross-talk and allowing for a cleanly polarised output, which is desirable for the THz-generation process.

With this method, it was possible to achieve a tenfold decrease in pulse duration to few-cycle pulses of 22 fs FWHM at a centre wavelength of 1035 µm, coinciding with a tenfold increase of the spectral bandwidth to ∼100 nm. The corresponding peak power was increased by a factor of six to 8.28 MW, considering system loss acquired after the fibre-exit and energy lost in the temporal side-lobes of the compressed pulse. Disregarding the side-lobes, the peak power reached 13.8 MW, almost a tenfold increase. The drawbacks of this solid-core setup, such as the limited power-threshold of 2.1 MW discussed in chapter 3.5 and the fibre-input coupling efficiency of 50 %, are tolerable for the desired outcome.

Overall, the simple compression setup has a small footprint of less than 1 m², requires few optical components, and remains stable over long periods of time, i.e. months, before needing readjustment. The economic aspect is also favourable, as all components used required only marginal investments. Hence, for many research labs using high-power, high repetition rate femtosecond lasers, the compression method outlined in this thesis can significantly boost the peak power of their systems. The resulting access to linearly polarised few-cycle pulses can be operated at various repetition rates, as the limitations are peak power dependent.
Part II of this thesis dealt with the applications of the compressed pulses to efficiently generate THz-pulses. Chapter 4 introduced the reader to a few applications and a more elaborate discussion on the various generation and detection methods used in THz-physics, with a strong focus on the method of OR and EOS. In line with the low-complexity employed for the compression, collinear OR was the chosen generation method, as it is the least complex option for THz-generation using free-space laser beams. The novelty in this thesis was to apply the compressed, broadband few-cycle pulses to the organic crystal HMQ-TMS for THz-generation at the MHz repetition rates provided by the fibre-laser. The importance of a broadband, ultrafast pump pulse was highlighted in chapter 5.1, further strengthening the motivation for part I. The following chapter 5.2 introduced the results obtained with HMQ-TMS and compared them to those achieved with GaP. The effect of crystal thickness was also studied, with thin HMQ-TMS crystals offering a better performance. As a result, a 10 MHz source of broadband femtosecond THz-pulses covering more than 6 THz in bandwidth with an average power of over 1 mW was demonstrated. The optical to THz-conversion efficiency reached $5.5 \times 10^{-4}$ - two orders of magnitude higher than that achieved with similar efforts using inorganic crystals. The high average power manifests itself in field strengths of tens of kV·cm$^{-1}$, making this type of THz-source a useful tool for many promising THz-applications.

A consistent issue with THz-radiation in most applied fields results from absorption in water molecules in air (i.e. the humidity). This issue was addressed in the final chapter 6 of the thesis, by experimentally confirming and then applying a computer code that simulates propagation of THz-pulses through humid atmosphere. This resulted in demonstrating that the power absorption of broadband THz pulses in air is better represented through a stretched exponential decay rather than a simple exponential one, such as Beer-Lambert’s law suggests.

During the external research stay at Kyoto University towards the end of this thesis, several months were spent trying to detect Landau-level transitions in cryogenically cooled graphene using THz-TDS. Much to the dismay to everybody involved, the experiments did not bear fruit due to the low signal-to-noise ratio present in the applied setups, and were thus not included in this thesis. In retrospect, the high repetition rate, high-power, broadband THz-source developed in this thesis would have been ideal to obtain the targeted results in Japan. The mW-power would have allowed for stronger THz-detection, mitigating the loss-heavy reflection geometry that was necessary for the experiment. The high repetition rate connected with a high-speed lock-in amplifier would allow for very low-noise signal recordings at fast scanning speeds - a strong contrast to the twenty minute scans that were necessary when using the 1 kHz THz-source present in the laboratory. This issue of low repetition rate sources also became clear in the humidity analysis, where it could be directly compared to a high repetition rate source that produced far cleaner and more accurate data.

Measuring Landau-level transitions in graphene or conducting a detailed study on water vapour are just two examples where the work in this thesis could have provided a benefit. Such a tool therefore can advance THz-science in other fields of physics. Until recently, high THz-field strengths have been restricted to low repetition rate sources due to the requirement of high driving pulse energies. Consequently, this imposed long data acquisition times to various applications.
This limitation was overcome through accessing a modern high-power femtosecond laser, optimising the bandwidth and peak power with external compression, and by using an optimised, highly efficient THz-generation crystal. Furthermore, the combination of related, but different research fields (femtosecond lasers and THz-TDS) demonstrated in this thesis may encourage more research groups to employ applicants from non-directly related fields and backgrounds. Cross-disciplinary knowledge is often an advantage that outweighs the initial delay caused by adapting to a new scientific field. I hope that my efforts over the last 40 months will accelerate the efforts of the THz-community towards real-life applications that will benefit society. A reduction of resource-waste could be achieved by optimising industrial processes through in-situ THz-imaging and monitoring. And in fundamental science, a better understanding of chemical processes, e.g. involving catalysts through ultra-fast THz-TDS could greatly improve process efficiencies, for example in batteries.
Appendices
### Abbreviations

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<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>ABCD</td>
<td>Air-Biased Coherent Detection</td>
</tr>
<tr>
<td>AC</td>
<td>Alternating Current</td>
</tr>
<tr>
<td>AR</td>
<td>Anti-Reflective</td>
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<td>(2-(4-hydroxy-3-methoxystryl)-1-methylquinolinium2,4,6-trimethylbenzenesulfonate)</td>
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