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Published in:
Applied Physics Letters

Link to article, DOI:
10.1063/1.4832825

Publication date:
2013

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):

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Citation: Applied Physics Letters 103, 221102 (2013); doi: 10.1063/1.4832825
View online: http://dx.doi.org/10.1063/1.4832825
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/103/22?ver=pdfcov
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Terahertz-induced Kerr effect in amorphous chalcogenide glasses

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(Received 18 September 2013; accepted 7 November 2013; published online 25 November 2013)

We have investigated the terahertz-induced third-order (Kerr) nonlinear optical properties of the amorphous chalcogenide glasses As2S3 and As2Se3. Chalcogenide glasses are known for their high optical Kerr nonlinearities which can be several hundred times greater than those of fused silica. We use high-intensity, single-cycle terahertz pulses with a maximum electrical field strength exceeding 400 kV/cm and frequency content from 0.2 to 3.0 THz. By optical Kerr-gate sampling, we measured the terahertz-induced nonlinear refractive indices at 800 nm to be \( n_2 = 1.746 \times 10^{-14} \text{ cm}^2/\text{W} \) for As2S3 and \( n_2 = 3.440 \times 10^{-14} \text{ cm}^2/\text{W} \) for As2Se3. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4832825]

Terahertz (THz) technology has the potential for application in a broad range of fields, e.g., biology, medicine,1 communication, and in the space-, defense-, and semiconductor-industry.2,3 While there has been tremendous progress in the development of THz sources, detectors, and passive linear components, many applications, such as ultrafast all-optical signal processing, require more versatile, nonlinear components. To construct these nonlinear components, materials with nonlinear interactions between the THz electric field and matter must be identified, developed, and quantified.

Chalcogenide glasses, first reported six decades ago,4 have attracted significant attention in recent years due to their unique properties such as high refractive index, mid-infrared transparency, and very large third-order (Kerr) nonlinearity.5 Their large nonlinearities in the optical, near-, and mid-infrared regions have made them promising candidates for fast nonlinear optical devices.5

The measurement of the THz-induced optical Kerr effect in solid-state materials has traditionally been hampered by a lack of high intensity THz sources. However, the recent introduction of tilted pulse front THz generation in lithium niobate (LiNbO3)7 now makes THz field strengths of the order of 1 MV/cm available.8,9 Such pulses were recently applied for the investigation of the THz-induced optical Kerr effect in liquids10 and in relaxor ferroelectrics.11

In this paper, we report the observation of the THz-induced optical Kerr effect in two bulk, amorphous chalcogenide glasses, As2S3 and As2Se3, and determine their third-order nonlinear coefficients. Both glasses have become very popular in optics due to their high nonlinear coefficients12–15 and flexibility towards micro- and nano-fabrication.16–18

Our experimental setup is shown schematically in Fig. 1(a). Briefly, an amplified femtosecond laser system (pulse energy 3.5 mJ, duration 100 fs, center wavelength 800 nm) drives a standard tilted pulse front scheme for generation of intense THz pulses in LiNbO3.8 The generated THz beam is tightly focused onto the sample by a combination of}

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frequency of the probe beam, and $L$ is the effective propagation length in the crystal. The probe beam intensity was kept at a low level to avoid any photodamage to the samples.

The recorded Kerr probe signals in As$_2$S$_3$ and As$_2$Se$_3$ at varying THz field strengths are shown in Fig. 2. The field strength was attenuated by a pair of wire grid polarizers placed immediately after the first paraboloidal mirror. To verify that the samples were not damaged during the characterization, each sample was measured at the highest field strength both at the beginning and at the end of the measurement series. The results were then compared to confirm that the samples had not been damaged during the characterization. As expected, the recorded signal increases with the THz pulse strength. At a field strength of 47 kV/cm, the THz-induced signal is barely noticeable but it grows significantly at higher field strengths. We used a 1 mm thick bulk silica glass sample as a reference. Even with the full field strength (400 kV/cm), no THz-induced birefringence was observed due to the very small third-order nonlinearity of silica compared to As$_2$S$_3$ and As$_2$Se$_3$.\textsuperscript{13}

For both the chalcogenide glasses, we observe that the temporal shape of the THz-induced Kerr signal closely follows the intensity profile of the THz pulse (dashed, red curves). This is indicative of a very fast nonlinear response for the two glasses, and is typical for nonresonant excitation. In contrast to some liquid samples, such as CH$_2$I$_2$,\textsuperscript{10} where the relaxation time can be rather long, there are no rotational dynamics of molecules in solid glasses. In the case of As$_2$S$_3$, the THz-induced signal instantly responds to the changes in the intensity profile of the THz pulse, as there is good phase matching between the THz wave and the optical probe in the glass. There is a slight indication of a delayed response from As$_2$Se$_3$. However, this might be due to the large velocity mismatch between the THz field and the optical probe, as will be discussed further below.

The analysis of the peak values of the THz-induced signal in glasses reveals a standard third-order nonlinear behavior, shown in Fig. 3. For the small values of the phase retardation observed here, the recorded values are accurately fitted by a quadratic fit $D/V = V_0/C^2$, where $V_0$ is a constant and $E$ is the electrical field strength. This demonstrates that the observed THz-induced signals in the chalcogenide samples are due to the Kerr effect ($\Delta n = 2KE^2$), where $\Delta n$ is the change in the refractive index, $k$ is the wavevector, and $K$ is the Kerr constant. The nonlinear refractive index can be written as $n_2 = \Delta n/I$ where $I$ is the intensity of the THz pulse averaged over either the propagation length in the glass or the physical length of the sample, whichever is shorter.

**FIG. 1.** (a) A schematic illustration of the LiNbO$_3$ THz-TDS system used for THz Kerr measurements. (b) THz pulse in time- (black curve) and frequency-domain (red curve). 84% of the pulse energy is concentrated between 0.2 and 1.0 THz, and is highlighted by dashed area. (BS) beam splitter, (OPM) optical parabolic mirror, (PD) photodiode, and (PBS) polarizing beam splitter.

**FIG. 2.** The THz-induced birefringence in (a) As$_2$S$_3$ (0.716 mm) and (b) As$_2$Se$_3$ (1.059 mm) bulk samples as a function of the peak THz field strength. The intensity of the THz pulse normalized to the maximum recorded signal is indicated by the dashed, red curve. The black curve is for a 1 mm thick silica glass sample measured with 400 kV/cm field strength for comparison.
As we have previously reported, chalcogenide glass has the universal characteristics of an amorphous system at lower terahertz frequencies with a monotonic increase of the absorption coefficient. However, crystalline phases may exist within the amorphous matrix. To verify that our samples are truly amorphous, we performed the Kerr measurements with the full electrical THz field strength at different orientation angles of the As$_2$S$_3$ sample (Fig. 4). No significant change in the Kerr signal is observed when the sample was rotated from $0^\circ$ to $90^\circ$.

To determine the magnitude of the Kerr effect, we measured both the refractive index and absorption coefficient of the two chalcogenide glasses in the terahertz range with a Picometrix T-Ray 4000 THz-TDS system, as shown in Fig. 5. The refractive indices for both glasses in the frequency range 0.2–1.0 THz are nearly constant (2.80 for As$_2$S$_3$ and 2.86 for As$_2$Se$_3$). The slightly higher As$_2$Se$_3$ refractive index can be explained by the higher density of As$_2$Se$_3$ (4.75 g/cm$^3$) compared to As$_2$S$_3$ (3.42 g/cm$^3$). The group indices for the 800-nm optical probe beam are 2.76 for As$_2$S$_3$ and 2.86 for As$_2$Se$_3$. The larger THz Kerr effect for As$_2$Se$_3$ is consistent with the significantly larger third-order nonlinear optical coefficients. For further analysis, we calculated the weighted average of the absorption coefficient over the pulse spectrum in the range 0.2–1.0 THz. This range contains 84% of the total intensity of the THz pulse (see Fig. 1(b)). Averaged absorption coefficients $\alpha = 6.2$ cm$^{-1}$ and 10.8 cm$^{-1}$ are found for As$_2$S$_3$ and As$_2$Se$_3$ with a characteristic penetration depth ($L_{abs}$ = $1/\alpha$) of 1.61 mm and 0.93 mm, respectively. We use these values to calculate the local THz field inside the bulk sample taking absorption and Fresnel transmission coefficients into account.

We determined the THz-induced nonlinear refractive indices $n_2 = 1.746 \times 10^{-14}$ cm$^2$/W and $n_2 = 3.440 \times 10^{-14}$ cm$^2$/W at 800 nm for As$_2$S$_3$ and As$_2$Se$_3$, respectively, in Table I. The larger THz Kerr effect for As$_2$Se$_3$ is consistent with the significantly larger third-order nonlinear optical coefficient in As$_2$Se$_3$ as compared to As$_2$S$_3$. The measured 3.65 for As$_2$S$_3$ and As$_2$Se$_3$, respectively. The resulting velocity mismatch between the optical probe beam and the THz pulse may result in smearing of the recorded Kerr signals, and consequently a slight underestimation of the extracted nonlinear coefficient. This effect will be most profound for As$_2$Se$_3$. The absorption coefficient monotonously increases from nearly zero to approximately 20–22 cm$^{-1}$ for both samples between 0.2 THz and 1.0 THz. This behavior is universal for a wide range of glasses, and confirms the amorphous nature of chalcogenide glasses at low THz frequencies.

![FIG. 4. The THz-induced Kerr signal as a function of As$_2$S$_3$ rotation angle at a THz electric field strength of 400 kV/cm. The rotation spans from $0^\circ$ to $90^\circ$ in steps of $10^\circ$.](image)

![FIG. 5. The refractive index (solid curves) and absorption coefficient (dashed curves) for As$_2$S$_3$ (red) and As$_2$Se$_3$ (black) samples as measured by THz-TDS.](image)

![FIG. 3. The peak values of the THz-induced Kerr signals for As$_2$S$_3$ (squares) and As$_2$Se$_3$ (triangles). The red, dashed curves are quadratic fits to the measured data points.](image)

![TABLE I. THz-induced optical parameters of As$_2$S$_3$ and As$_2$Se$_3$ glasses.](table)

<table>
<thead>
<tr>
<th></th>
<th>As$_2$S$_3$</th>
<th>As$_2$Se$_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta n [10^{-6}]$</td>
<td>5.34</td>
<td>6.30</td>
</tr>
<tr>
<td>$n_2 [10^{-14} \text{cm}^2/\text{W}]$</td>
<td>$1.75 \pm 0.09$</td>
<td>$3.44 \pm 0.17$</td>
</tr>
<tr>
<td>$n_{2,\text{meas}} [10^{-14} \text{cm}^2/\text{W}]$</td>
<td>2.5</td>
<td>11.4</td>
</tr>
<tr>
<td>$\chi^{(3)} [10^{-20} \text{m}^2/\text{V}^2]$</td>
<td>$4.84 \pm 0.24$</td>
<td>$9.96 \pm 0.50$</td>
</tr>
<tr>
<td>$I_{peak} [10^8 \text{W/cm}^2]$</td>
<td>$3.06 \pm 0.15$</td>
<td>$1.83 \pm 0.09$</td>
</tr>
</tbody>
</table>

$^a\Delta n$ is the modulation of the refractive index.

$^b$n$_2$ is the nonlinear refractive index.

$^c$n$_{2,\text{meas}}$ is the nonlinear refractive index measured at near infrared.$^{12,14}$

$^d}\chi^{(3)}$ is the nonlinear susceptibility ($\chi^{(3)} = \frac{i}{4} \times 4\pi \chi^{(2)} n_2$, where $n_0$ is linear refractive index).

$^e}I_{peak}$ is the peak intensity averaged through samples.
nonlinear refractive index for $\text{As}_2\text{S}_3$ is comparable with the values that were reported for the near infrared, $n_{2,nir} = 2.5 \times 10^{-14}$ cm$^2$/W,\textsuperscript{12} while that of $\text{As}_2\text{Se}_3$ is approximately 3 times lower, $n_{2,nir} = 1.4 \times 10^{-14}$ cm$^2$/W.\textsuperscript{14}

In summary, we have reported the observation of the THz-induced optical Kerr effect in solid materials, using intense THz pulses of up to 400 kV/cm. Both chalcogenide samples, $\text{As}_2\text{S}_3$ and $\text{As}_2\text{Se}_3$, reveal large nonlinear refractive indices. Furthermore, due to the nonresonant nature of the third-order nonlinearity in the amorphous glasses, a near-instantaneous (faster than the time resolution of the experiment) response of the Kerr signal was observed.

Financial support from the Danish Research Council for Technology and Production Sciences through Project No. 09-070397-FTP (THz-COW) and Project No. 11-106748-FTP (HI-TERA) is acknowledged.

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