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All-optical control of coherent phonons in the candidate type-II Weyl semimetal WTe_2

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ABSTRACT

Using ultrafast double-pulse spectroscopy, our experiment showcases coherent control over the 2.39 THz optical phonon mode within bulk WTe_2. Through precise manipulation of the delay between successive pump pulses, we successfully achieve modulation of the phonon amplitude at room temperature by a factor of two. This breakthrough showcases the potential for all-optical control and modulation of the unique properties in WTe_2 that are directly bound to its structural degrees of freedom.

Keywords: Coherent phonon, Weyl semimetal, ultrafast spectroscopy

1. INTRODUCTION

The large, non-saturating magnetoresistance of WTe_2[1] greatly increased the interest in this material. Since then, remarkable properties have been found, including ferroelectricity at room temperature[2] and possible type-II Weyl phase.[3] These characteristics have been shown to be linked with structural degrees of freedom, which can be directly influenced by ultrafast laser pulses. It has already been shown in WTe_2 that an ultrafast breaking of the lattice symmetry can be achieved.[4–6] Different time-resolved techniques, including transient reflection[6–8] and time- and angle-resolved photoelectron spectroscopy,[9] have been employed to investigate the generation of coherent phonons in WTe_2. Coherent phonons are vibrations of the crystal lattice exhibiting a well-defined phase relative to the optical activation field, making them fundamental to controlling the aforementioned properties.

The two most prominent low-frequency modes in WTe_2 correspond to A_1 zone-center optical phonons[10] at 0.24 and 2.39 THz. The first is attributed to out-of-phase shear displacements between adjacent layers in the material, while the latter involves complex femtometer in-plane and out-of-plane displacements of tellurium atoms against tungsten atoms.[10] It is accepted that the activation mechanism is the displacive excitation in which, for highly absorptive media, a long-lived carrier population in the conduction band leads to a coherent vibration of the nuclei around a new *displaced* equilibrium position.[11]

We target the higher frequency mode of these two vibrations. To do so, we employ an all-optical approach. By finely tuning a sequence of two excitation *pump* pulses we can selectively enhance (quench) the amplitude of coherent phonons in the material by constructive (destructive) interference of the phonon wavepackets. This approach has successfully been used to exert control in different systems, including silicon,[12] diamond,[13] materials with a well-defined charge-density-wave (CDW),[14–19] spin-density-wave (SDW)[20,21] and 2D thermoelectrics.[22] Here we report the first experimental demonstration of this approach to control coherent phonons in WTe_2.

2. EXPERIMENTAL METHODS

Samples

High-quality bulk crystals of WTe_2 are commercially available from HQ Graphene. Samples have a lateral dimension of ∼1 mm and are prepared by mechanical exfoliation using Kapton tape. The exfoliated flakes are subsequently encapsulated by the tape and a thin glass window (0.5 mm), thus allowing back-illumination

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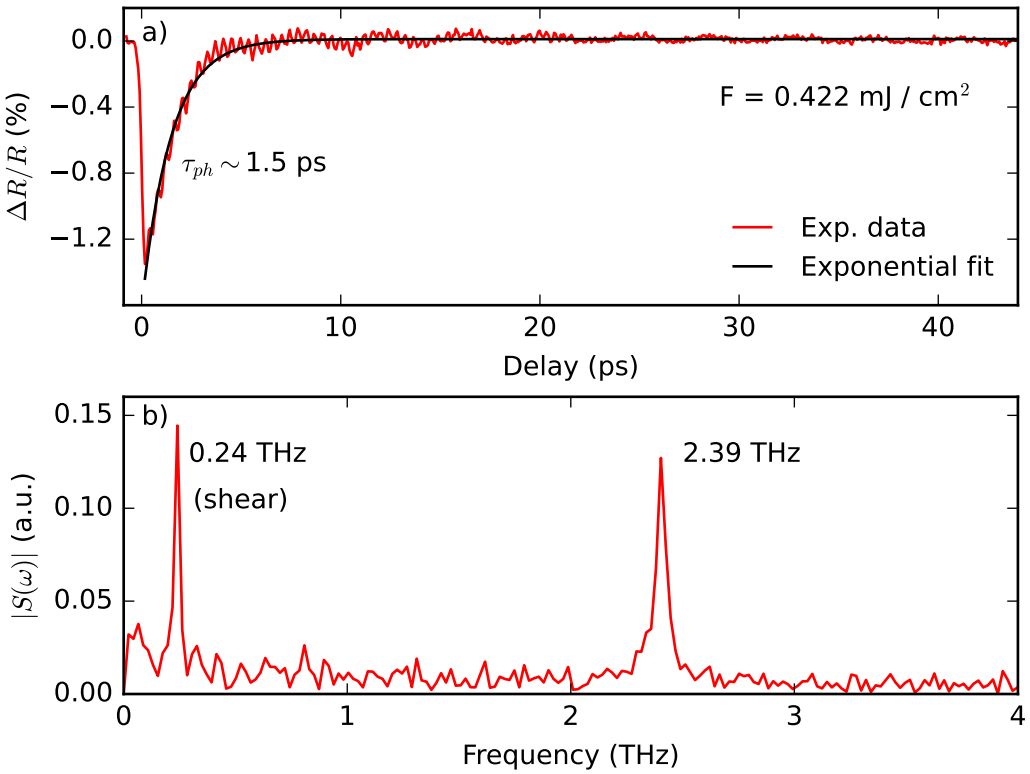

Figure 1. **Transient reflectivity of WTe_2.** a) Experimental data (red) and exponential fit (black) measured at a fluence of 0.422 mJ/cm^2. b) Fast Fourier Transform (FFT) of a), clearly depicting the two A_1 phonon modes at 0.24 and 2.39 THz.

through the glass surface. Reference measurements showed that the glass window has no transient response, thus ensuring that the response comes exclusively from WTe_2.

Transient reflectivity

The source laser is a Ti:Sapphire regenerative amplifier (Solstice Ace^{TM} by SpectraPhysics) that produces pulses of $\sim$ 100 fs with a center wavelength of 800 nm at a repetition rate of 1 kHz. The output of the laser was split into three components: two high-intensity pulses for the *pumps* and a heavily attenuated *probe*. The latter is left stationary for an enhanced signal-to-noise (SNR) ratio, while the pumps are delayed with respect to the probe by suitably changing the path length using mechanical delay stages. Both pumps are frequency-doubled in $\beta-$barium-borate (BBO) to generate high-intensity pulses centered at 400 nm.

The transient response from WTe_2 was recorded using a homodyne detection scheme with a reference frequency of 500 Hz provided by an optical chopper. The signal was obtained from a balanced photodiode and filtered by chaining a boxcar integrator, and a lock-in amplifier locked to the reference frequency. For the double-pulse measurements, the second pump was left unchopped, thus making it invisible to the lock-in amplifier but leaving its effects on the sample itself.

3. COHERENT PHONON DYNAMICS IN WTE_2

The transient reflection of WTe_2 under 400 nm excitation is shown in Figure 1a. Due to the pump-probe conditions, a single exponential fit (black) with a time constant of $\tau_{ph} \sim 1.5$ ps is sufficient to describe the thermalization of high-energy photoexcited electrons with the lattice. The fluence is kept at $F = 0.422$ mJ/cm^2 to ensure that we remain below anharmonic excitation of various phonon modes.[6] Figure 1b shows the Fast Fourier Transform (FFT) of the data set after subtraction of the exponential fit. We can clearly distinguish the two lowest frequency A_1 phonon modes at 0.24 and 2.39 THz. The first corresponds to interlayer shear displacements, while the higher-frequency mode is described by intralayer out-of-phase displacements of tellurium

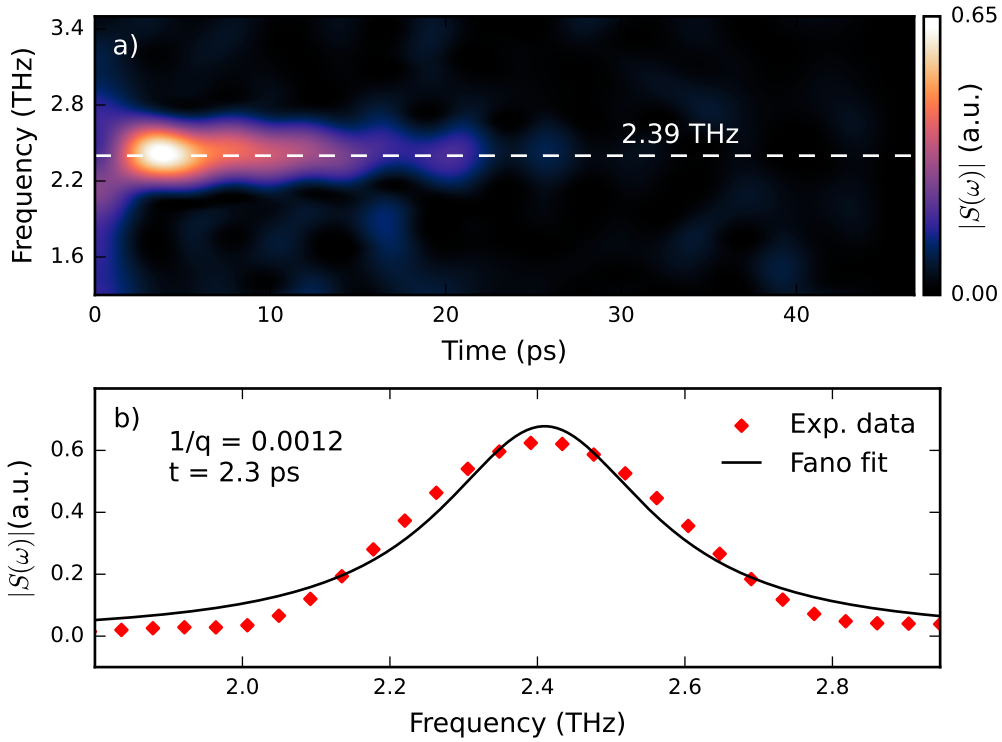

Figure 2. **Sliding-window Fourier Transform (SWFT) analysis.** a) SWFT spectrum for the 2.39 THz phonon mode. b) Vertical cut at 2.3 ps (red diamonds) of a) and its corresponding Fano fit (solid black line).

atoms against tungsten atoms.[6,10] To the author's best knowledge, it is the first experimental demonstration of activation of these phonon modes under the *current* pump-probe conditions.

To gain further insight into the dynamics of the high-frequency mode, we performed a sliding-window Fourier transform (SWFT) analysis to the time trace in Figure 1a. The resulting SWFT spectrum is shown in Figure 2a, showcasing the time evolution of the 2.39 THz phonon mode. After ~ 30 ps, the oscillations fully decay.

We can further fit the line shape of the spectral magnitude $|S(\omega)|$ at 2.39 THz by a Fano function of the form:

$$|S(\omega)| = \frac{(q + (\omega - \omega_0)/\gamma)^2}{1 + ((\omega - \omega_0)/\gamma)^2}$$

where q, ω_0 and γ are the Fano parameter, the resonance frequency, and the linewidth, respectively. A Fano resonance occurs from the coupling of two oscillator amplitudes,[23] leading to the asymmetry of the spectral line shape. In our case, fitting the line shape will shed light on the electron-phonon coupling in the material, with a factor of $1/q = 0$ corresponding to a perfect Lorentzian emission profile: No electron-phonon interaction. Figure 2b shows a Fano fit to the SWFT at a time of 2.3 ps. With a corresponding value for the inverse of the Fano factor of $1/q = 0.0012$, the phonon mode at 2.39 THz follows a quasi-Lorentzian profile, indicating a potentially weaker electron-phonon coupling compared to other tungsten-based semimetals, such as WP_2.[24]

By carefully adding a second pump pulse with a similar fluence to the already excited crystal, we can control the coherent phonon mode at 2.39 THz. Figure 3a and b show the 2D ultrafast dynamics of WTe_2 in the time and frequency domain, respectively. The effects of the arrival of the modulation pulse can clearly be seen in Figure 3a as the bright diagonal that corresponds to Delay $= -t_{mod}$. Here, a negative τ_{mod} corresponds to a trailing modulation (or control) pulse, arriving a few picoseconds after the arrival of the first pump pulse. For

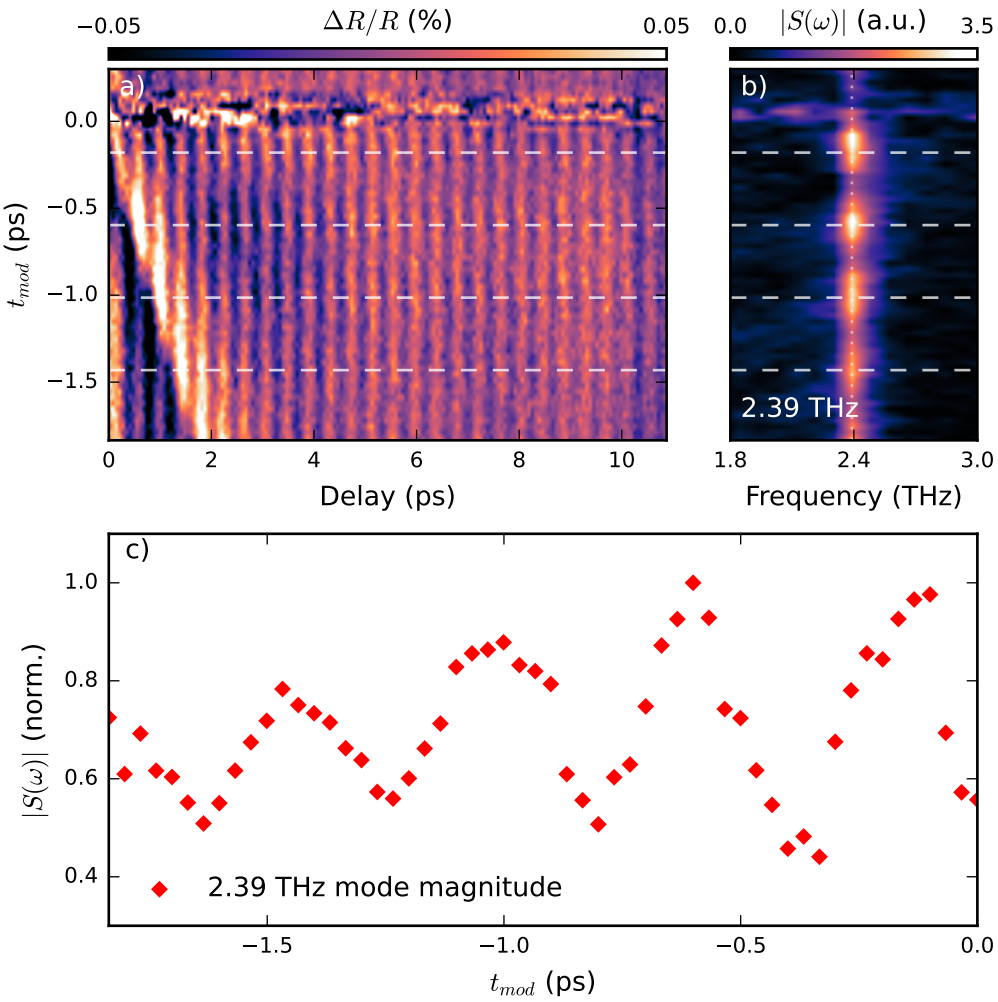

Figure 3. **Coherent phonon control on WTe_2 via two-pulse excitation.** a) 2D ultrafast dynamics of WTe_2 in the time domain. The horizontal axis represents the delay between the pump and the probe. t_{mod}, on the vertical axis, represents the modulation time between pumps. A negative t_{mod} corresponds to a trailing modulation pulse. b) 2D ultrafast dynamics in the frequency domain. The vertical axis is shared with a) as depicted by the white dashed lines. c) Periodic modulation of the 2.39 THz phonon mode magnitude as a function of t_{mod}.

$t_{mod} \sim 0$ we observe a highly anharmonic response of the transient reflection that we attribute to the interference of the carrier frequency of both pump pulses. In contrast, when the pumps are separated by $|t_{mod}| > 200$ fs the material response is dominated by the pulse envelope.

In Figure 3a and b the quenching and enhancement of the dominant phonon mode are evident, showcasing on-demand control. The maxima of the spectral magnitude as a function of time are highlighted by horizontal dashed white lines, spaced exactly by one phonon period of 417 fs. By taking a vertical cut at 2.39 THz in Figure 3b we can trace the time-evolution of the magnitude of the phonon mode as a function of the modulation time. This is depicted in Figure 3c) where we can clearly see the periodic modulation, normalized to its maximum value. Here, we achieve modulation of the fast phonon mode exceeding a factor of two. Notably, for a smaller separation of the pump pulses, we obtain a stronger modulation of the phonon mode. This, however, is indicative of the detection method we are using, as the second (modulation) pump is unchopped, we are exclusively looking at the contribution of the chopped pump. The response, in this case, is proportional to the product of both pump signals, rather than their sum, thus increasing if the magnitude of the responses is larger as a function of

time.

In conclusion, our results successfully showcase the modulation of the phonon mode at 2.39 THz using an all-optical approach. With this, we open the possibility of the potential use of light pulses for on-demand control of the remarkable properties of WTe_2.

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