

Nonlinear terahertz phononics in the Dirac semimetal Cd_3As_2

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(Received 15 October 2024; revised 8 May 2025; accepted 11 July 2025; published 16 July 2025)

The coherent lattice dynamics in the three-dimensional (3D) Dirac semimetal Cd_3As_2 is investigated in the nonperturbative regime of light-matter interaction using coherent two-dimensional terahertz (THz) spectroscopy, as a function of time delay and field strength. The phase-resolved nonlinear signal reveals a coherent phonon mode at 1.48 THz, corresponding to the B_{2g} Raman-active mode, characterized by the periodic motion of two cadmium atoms in the substructure toward and away from two cadmium-vacant sites. The interaction of a strong THz electric field with the 3D Dirac semimetal Cd_3As_2 leads to transient charge carrier imbalance via tunneling ionization, which results in a displacive force that drives the lattice dynamics of a noninteracting Raman-active mode. The selective excitation of otherwise inaccessible phonon modes enables precise control over the material's properties at the atomic level, opening new pathways to manipulate its topological characteristics.

DOI: [10.1103/y2bx-tkkq](https://doi.org/10.1103/y2bx-tkkq)

The interaction of light pulses with solids offers a unique pathway to study and manipulate material properties, as these ultrafast interactions can trigger a variety of emergent phenomena at the atomic level. In particular, they can induce rapid nonequilibrium processes, leading to the excitation of phonons and other collective modes that are otherwise difficult to access. This ability to excite and control phonons through femtosecond light pulses opens up new opportunities for exploring ultrafast dynamics in materials with significant implications in optoelectronics and materials science [1–5].

In pump-probe experiments, optical phonons can be excited either directly through resonant electric dipole interactions [6,7] or indirectly via various mechanisms such as impulsive stimulated Raman scattering (ISRS) [5,8,9], displacive excitation of coherent phonons (DECP) [10–12], anharmonic lattice coupling [4,13], or two-photon sum-frequency generation [14,15]. In the ISRS mechanism, an ultrafast light pulse creates a coherent superposition of phonon states by impulsively modulating the electronic polarizability, leading to the generation of vibrational modes [16–19]. The DECP

mechanism involves a sudden shift in the equilibrium positions of atoms, driven by a strong electromagnetic field that transiently modifies the electron and hole distributions that couples to lattice distortions through the deformation potential [11,12,20]. Phonons can also be excited via anharmonic coupling, where nonlinear interactions transfer energy between different vibrational modes, resulting in the excitation of specific phonons [4,13]. Additionally, Raman-active phonons can be excited without invoking anharmonicity through the simultaneous absorption of two terahertz (THz) photons, i.e., THz sum-frequency generation [14,15].

Cd_3As_2 is a room-temperature stable three-dimensional (3D) Dirac semimetal, which crystalizes in a body-centered-tetragonal structure with the space group $I4_1/acd$ and a conventional cell consisting of 160 atoms which can be constructed as a $2 \times 2 \times 4$ supercell of ten-atom substructures [Fig. 1(a)] [21,22]. The distinguished physical properties of Cd_3As_2 are characterized by high carrier mobility, large Fermi velocity, and exceptional optical response [23,24], as well as its topological properties [25–29]. Despite the considerable attention that Cd_3As_2 has garnered in recent years, its lattice dynamics remains insufficiently explored. For instance, excitation resulting from the displacement of Cd atoms toward Cd vacancies, which induces a distortion of the As-Cd cube, could potentially trigger a structural transformation, offering deeper insights into the topological properties of this system [21], but has yet to be reported. This is largely due to the complexity of the lattice

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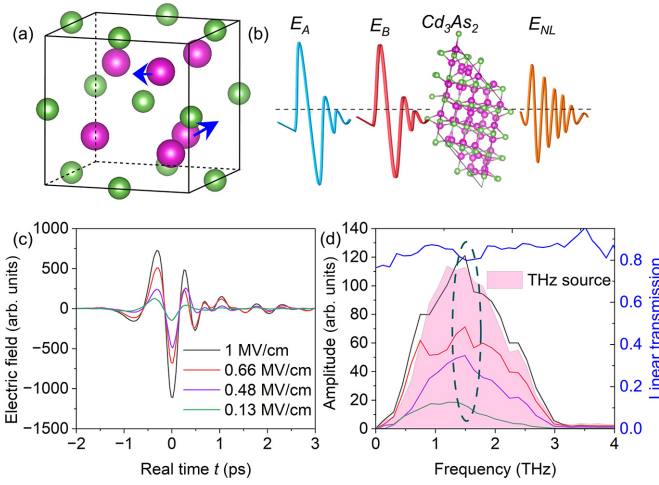


FIG. 1. (a) Ten-atom substructure of the Cd_3As_2 conventional cell. The blue arrows on the two cadmium atoms mark the atomic displacements driven by the B_{2g} mode. The displacements of other atoms are nonzero but relatively small, so we do not mark them in this schematic. (b) Schematic of the experimental configuration with the incident THz pulses A and B and the emitted nonlinear field. The signal is detected using a free-space electro-optic sampling. (c) Transmitted THz pulse E_A as a function of the real time t . (d) Corresponding frequency-domain spectra, obtained from the Fourier transformation of the transients in (c) compared with the spectra of the incident THz source and linear transmission spectrum (blue line, right abscissa). The source spectrum is slightly scaled to match the transmitted spectra. The transmission curve (blue curve, right abscissa) in the linear regime is essentially featureless, with no resolved IR modes.

structure, which presents significant challenges in studying its dynamics.

In this study, we investigate strong THz field-driven excitation of lattice dynamics in the 3D Dirac semimetal Cd_3As_2 at room temperature. Using 2D THz spectroscopy, we achieve spectral and temporal separation of a coherent B_{2g} Raman phonon emission from other nonlinear signal contributions. Interaction with the strong THz pulses induces tunneling ionization near the Dirac point, in the vicinity of the Γ point, which transiently modifies the spatial charge distributions of carrier. This results in a transient crystal symmetry reduction and triggers a dispersive excitation of the B_{2g} phonon that cannot be linearly excited by the light fields.

High-quality (112)-oriented Cd_3As_2 thin films, 60 nm in thickness, were grown on a 70- μm mica substrate using a molecular-beam epitaxy system [30].

We achieve direct access to the transient nonlinear processes by performing a time-resolved two-dimensional coherent THz spectroscopy [31–33] [Fig. 1(b)]. Two phase-locked THz pulses are prepared by splitting an intense THz pulse generated via rectification of an 800-nm pulse in a LiNbO_3 crystal using tilted wave-front excitation. The stronger pulse A and the weaker pulse B are therefore synchronized with a tunable time delay τ . The THz electric field is measured as a function of real time t by electro-optic sampling. By varying the time delay τ and measuring the transmitted THz field E_{AB} of only one pulse A (or B) and E_{AB} of both pulses, we derive

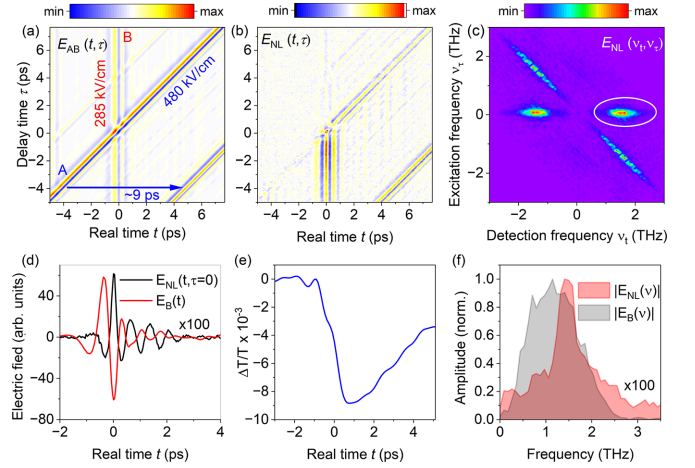


FIG. 2. (a) Contour plot of the THz electric fields of both pulses $E_{AB}(t, \tau)$ transmitted through the Cd_3As_2 sample. The vertical lines represent pulse B, which depends only on the real time t . The diagonal lines represent pulse A, which depends on both the real time t and the delay time τ . (b) Contour plot of the nonlinear emitted THz field $E_{NL}(t, \tau)$. (c) Two-dimensional Fourier amplitude of the emitted nonlinear signal $|E_{NL}(\nu_i, \nu_j)|$ of the nonlinear signal $E_{NL}(t, \tau)$. (d) Emitted THz nonlinear electric field $E_{NL}(t, \tau = 0)$ (red curve) plotted together with the transmitted probe THz field $E_B(t)$ for a zero delay between the pump and the probe (black curve); the pulse $E_B(t)$ is scaled to match $E_{NL}(t, \tau = 0)$. From the nonlinear signal, a coherent oscillation is observed up to about 6 ps. (e) THz differential transmission $\Delta T/T_0$. (f) Fourier transform of the emitted nonlinear signal amplitude $|E_{NL}(\nu)|$ together with the probe pulse $|E_B(\nu)|$. The spectrum of the emitted nonlinear signal exhibits a sharp peak at 1.48 THz.

the nonlinear THz response given by $E_{NL}(t, \tau) = E_{AB}(t, \tau) - E_A(t, \tau) - E_B(t)$. In the same setup, we can perform 1D THz transmission by blocking pulse E_B and using only pulse E_A .

Experimental results obtained with a single THz pulse $E_A(t)$ transmitted through the sample for various field strengths are plotted in real time t as presented in Fig. 1(c). The corresponding spectra, derived by fast Fourier transformation (FFT) of the transient signals, are shown in Fig. 1(d) along with the incident THz spectra (pink curve) from the LiNbO_3 source. With increasing THz electric field strength, one observes a distinct nonlinear behavior leading to pronounced changes of the spectral shape of the transmitted THz pulses, including the emergence of a new spectral feature at 1.5 THz. To explore whether any distinct spectral features exist around this frequency, we performed a linear transmission experiment using a commercially available THz spectrometer (Menlo Systems). The transmission spectrum in the linear response regime is essentially featureless, with no resolvable IR modes [Fig. 1(d), blue curve].

To directly access the nonlinear emitted field, we employed 2D THz spectroscopy. The obtained 2D data in the time domain are presented as contour plots in Figs. 2(a) and 2(b) for $E_{AB}(t, \tau)$ and $E_{NL}(t, \tau)$, respectively. Besides the main signal at real time $t = 0$ ps, a second spike along the diagonal observed at real time $t = 9$ ps [Fig. 2(a)] is due to the round-trip reflection of the initial terahertz pulse in the gold-coated diamond beam splitter, which is used to split and recombine

the generated THz pulse. By performing a 2D FFT along real time t and delay time τ , the nonlinear signal $E_{NL}(\nu_t, \nu_\tau)$ in the frequency domain as a function of detection frequency ν_t and excitation frequency ν_τ is determined and displayed as a contour plot in Fig. 2(c). The nonlinear response is characterized by pump-probe signals occurring at (ν_t, ν_τ) of (1.4, 0) THz and (1.4, -1.4) THz. The pump-probe signal is superimposed by a bright spot at frequencies $(\nu_t, \nu_\tau) = (1.48, 0)$ THz, highlighted by a white circle in Fig. 2(c). In Fig. 2(d) we compare the time traces of the emitted THz nonlinear electric field $E_{NL}(t, \tau = 0)$ to the transmitted THz field of the probe pulse (pulse B) for a pump pulse (pulse A) with a field strength of 480 kV/cm. The nonlinear field exhibits a π phase shift relative to the probe pulse, signifying an absorption increase due to the generation of carriers by the THz pulse [12]. In the time domain, the emitted nonlinear field shows a main subpicosecond transient, followed by long-lasting coherent oscillations persisting up to 6 ps. Figure 2(e) shows the time-integrated THz-pump/THz-probe signal as a function of the delay time τ , obtained from

$$\Delta T/T_0 = \frac{2 \int E_{Pr}(t, \tau) E_{NL}(t, \tau) dt}{\int |E_{Pr}(t, \tau)|^2}, \quad (1)$$

in which E_B is used as a probe pulse E_{Pr} . The THz differential signal displays a rapid transmission decrease on a timescale of 0.5 ps, followed by a relaxation over approximately 6 ps, characteristic of carrier relaxation dynamics reported in this system [34–37].

A Fourier transform of $E_{NL}(t, \tau = 0)$ is presented in Fig. 2(f). A narrowband emission at 1.48 THz is assigned to the excitation of the B_{2g} Raman-active phonon mode at the Γ point which is superimposed on the pump-probe signal. It is noteworthy that the emitted B_{2g} phonon mode present in the static Raman measurement [38] is absent in the linear THz experiment [Fig. 1(d)].

In order to elucidate the excitation mechanism, we carried out a study of the THz field-strength dependence. Figure 3(a) shows the nonlinear field signal $E_{NL}(t, \tau = 0)$ at various THz pump-field strengths at fixed delay time $\tau = 0$. With the pump field decreasing from a peak value of 0.92 MV/cm, the pronounced oscillation feature weakens rapidly but without changing its frequency. Such a rapid decrease reflects the THz field-induced nonlinearity. At 0.21 MV/cm a nonlinear signal is discernible just about ± 1 ps around $t = 0$ ps, while at a relatively low field of 96 kV/cm, coherent phonon oscillations are entirely absent. By performing a Fourier analysis, the amplitude of the coherent oscillation in the nonlinear-response channel is derived and plotted as a function of the THz field strength in Fig. 3(b). The nonlinear signal exhibits clearly a quadratic dependence on the THz pump field, as shown by the solid line in Fig. 3(b). For incident electric field strengths below 170 kV/cm, the nonlinear signal fields fall below the noise floor of our experiments. The symmetry of the Raman-active phononic excitation is studied by a polarization-dependent measurement of the nonlinear signal. In the experiment, the polarizations of pulses A and B were kept parallel to each other while the sample was rotated around its axis. To exclusively derive the polarization dependence of phonon emission while minimizing contributions

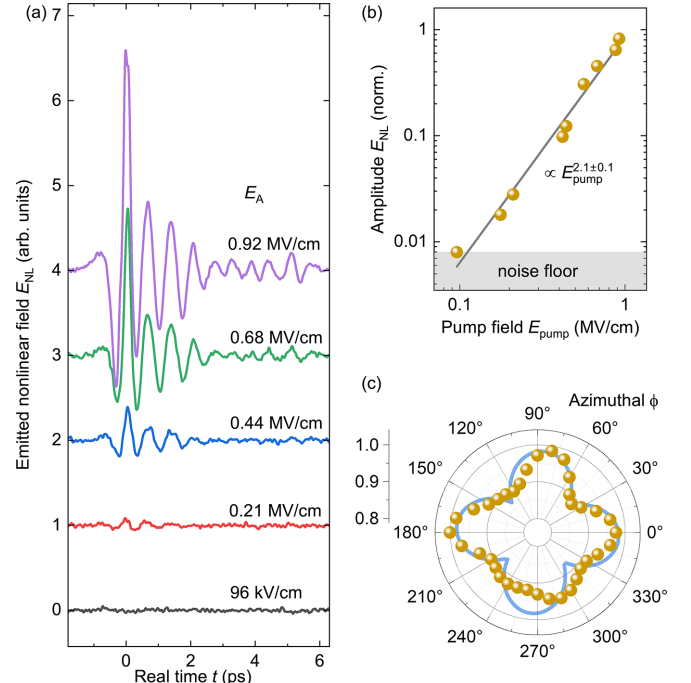


FIG. 3. (a) Field dependence of the coherent nonlinear B_{2g} signal. With decreasing pump-field strength, the oscillation behavior becomes less pronounced, which below $E_{pump} \sim 0.1$ MV/cm is not resolvable within the experimental uncertainties. (b) Amplitude of the emitted coherent nonlinear field versus the pump-field strength exhibiting a quadratic power-law dependence of $E_{nonlinear} \propto E_{pump}^{2.1 \pm 0.1}$. (c) Angular dependence of the time-integrated nonlinear signal $\int |E_{NL}(t, \tau = 0)| dt$, which can be approximately described by a fourfold rotation symmetry as shown by the solid line.

from the pump-probe signal, which primarily occurs at $t = 0$ ps, the phonon emission is analyzed at $t = 3$ ps for different azimuthal angles Φ . The polarization dependence obtained is shown in Fig. 3(c). A pronounced angular dependence with fourfold symmetry is observed, with the pump-probe signal being modulated by up to 15%. This fourfold symmetry is a hallmark of the highly nonperturbative character of the excitation, since excitation limited to the third perturbative order $\chi^{(3)}$ would lead to a purely isotropic signal [12,39,40].

The investigation of THz field-driven Raman phonons has been a central focus in the study of various materials. However, the lattice dynamics of Cd_3As_2 has primarily been explored through interband excitation using femtosecond near-infrared pulses at low temperatures ranging from 6 to 200 K [41]. In these studies, coherent phonon beating, attributed to A_{1g} optical phonon modes arising from counter helices formed by cadmium vacancies, was observed. These modes become inactive as the temperature exceeds 200 K [41]. Notably, the absence of B_{2g} modes under resonant excitation, coupled with their presence under THz excitation even at room temperature, excludes the possibility of impulsive Raman excitation of the B_{2g} mode in the context of THz excitation. For an impulsive Raman excitation, the femtosecond light field induces oscillatory vibrations around the equilibrium atomic positions without any net displacement of the lattice. In contrast, the displacive mechanism involves

nonperturbative THz field excitation, which alters the spatial charge distribution of carriers, inducing a transient reduction in crystal symmetry and enabling the displacive excitation of Raman modes. This phenomenon was recently reported in the semimetal bismuth [11,12,20]. Additionally, in topological insulators, lattice dynamics is triggered by anharmonic coupling [13]. However, our linear transmission experiments [see Fig. 1(d), blue curve] alongside previously reported reflectivity data [42], confirm the absence of any infrared optical phonon modes within our THz range. Consequently, phonon excitation via anharmonic coupling can be confidently ruled out. The THz sum-frequency mechanism is also unlikely, as the observed Raman phonon frequency falls within the spectrum of our THz pulse. Furthermore, such a mechanism has only been reported in systems that are insulating in the terahertz regime [14,15,43].

The observed anisotropy in the excitation of the B_{2g} mode upon changing the azimuthal angle Φ [Fig. 3(c)] indicates that the system has moved into the nonperturbative regime of light-matter interaction. Thus, the most probable excitation mechanism is displacive excitation, in which lattice distortions are strongly coupled to the distribution of electrons and holes via the deformation potential. As a result, the equilibrium positions of certain optical phonons are displaced when carriers are excited into the conduction band. Moreover, Cd_3As_2 , with its conelike electronic structure, is similar to both graphene and the semimetal bismuth, meaning that its interaction with a strong THz pulse leads to nonperturbative carrier generation near the Γ point through mechanisms such as field-induced tunneling, Rabi flopping, and impact ionization [44]. When an external THz pulse with vector potential $A(t)$ is applied, the conduction-band population initially undergoes intraband motion driven by $A(t)$, up to the field strength E_F/ev_F . Once the field exceeds E_F/ev_F , electrons in the valence band, outside the range E_F/ev_F in k space, experience interband transitions due to the large transition dipole near the Γ point and the available states in the conduction band. As a result, the electrons undergo both intra- and interband motion. In the absence of scattering events, the electron dynamics are fully reversible, with interband excitation and deexcitation occurring symmetrically under the THz pulse. However, in the presence of electron-electron or electron-phonon coupling, the coherence of the electron motion is disrupted within a 100 fs, leading to the generation of additional populations in the conduction band after the THz pulse excitation [12]. The observed nonlinear emitted field in Fig. 2(d), along with the transmission changes as a function of delay time Fig. 2(e), provides strong evidence of carrier generation induced by the terahertz field in this system.

In the electronically excited state, the equilibrium position of the optical phonon coordinate is shifted spatially compared to its position in the crystal's ground state. This shift is analogous to the origin displacement of the vibrational potential observed in molecules, while in a crystal the displacement of an optical phonon mode alters the crystal's electronic band structure. We conduct *ab initio* calculations of the electronic band structure using density functional theory and examine the changes induced by B_{2g} Raman-active lattice distortions in the Cd_3As_2 band structure. The equilibrium electronic band structure (blue solid lines) of Cd_3As_2 is shown

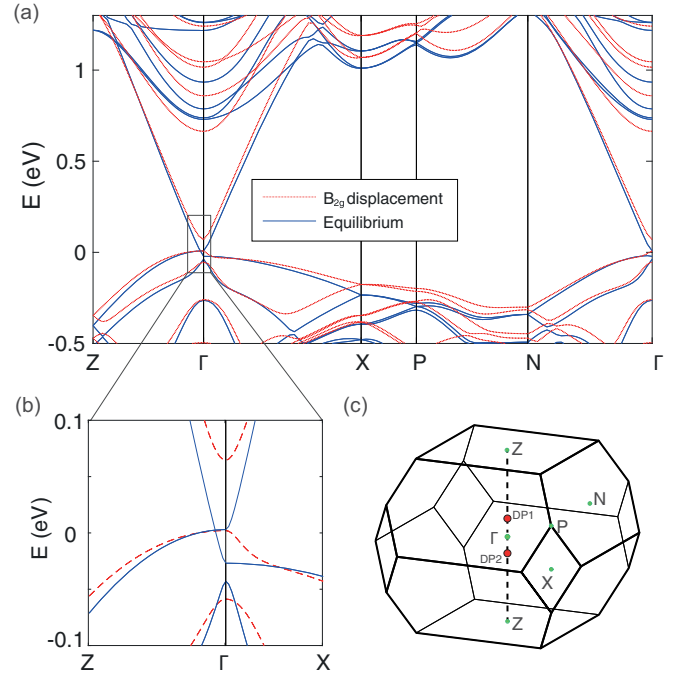


FIG. 4. (a) Blue lines show the electronic band structure of unexcited Cd_3As_2 . Red dashed lines show the deformed electronic band structure for a 0.63-Å displacement of the two Cd atoms describing the B_{2g} optical phonon at the Γ point. (b) Close-up near the Γ point. The B_{2g} phonon displacement modifies the band structure and leads to 0.06-eV modulation at the Γ point. (c) Corresponding first Brillouin zone of the Cd_3As_2 , with the positions of the pair of the Dirac points DP1 and DP2 marked by red dots.

in Fig. 4(a), where a pair of Dirac points is located on the Z- Γ k path [21,22]. These Dirac points are protected by the C_4 symmetry of the lattice. The lattice distortion associated with the excitation of the B_{2g} phonon mode, quantified by the displacement of the two predominantly involved cadmium atoms as illustrated in Fig. 1(a), leads to a significant modification of the electronic band structure, as depicted by the red dashed lines in Fig. 4(a). Besides the substantial changes along the Γ -X-P path, the B_{2g} excitation induces a significant modification at the Γ point, where the Dirac points merge into a single gapped Dirac cone with a finite band gap of approximately 60 meV for a 0.63-Å displacement of the two cadmium atoms [Fig. 4(b)]. Such displacement results in a small perturbation of the bond lengths, approximately 0.07 Å for both Cd-Cd and Cd-As bonds, corresponding to about 2%. To establish a correlation between atomic displacement and the applied THz field, we reference a previous study that reported a direct measurement of atomic displacement for a low-frequency shear phonon mode as a function of THz field strength in a Weyl semimetal [45]. By comparing the two excitation conditions, we estimate an atomic displacement of approximately 0.07 Å for a field strength of 750 kV/cm. Given the quadratic dependence of the phonon amplitude on the incident THz field, we can further establish the relationship between the atomic displacement and THz field strength. Our results suggest that Cd-As bond distortions lead to transient alterations in the system's topological properties. These

findings pave the way for future investigations into the impact of THz phononics on the topological properties of this system, using time- and angle-resolved photoemission spectroscopy to directly map the electronic band structure and its dynamic response to phonon amplitudes.

In summary, we have explored the THz excitation of the Raman-active B_{2g} phonon in Cd_3As_2 in the nonperturbative regime of light-matter interaction. Our findings reveal that this phonon is excited through the DECP mechanism, where the strong interaction with the THz field induces both intra- and interband transitions via tunneling ionization, generating displacive forces that initiate phonon oscillations. Our findings highlight the intricate phonon dynamics present in topological materials, which may open a route to control their topological phases. In particular, such band-gap modulation may be a key to inducing an ultrafast topological phase

transition, which was predicted for this system and in the closely related Na_3Bi under the application of a static electric field [46,47].

We would like to thank Michael Woerner, Klaus Reimann, and Thomas Elsaesser for valuable discussions. We acknowledge partial support from Mercator Research Center Ruhr via Project No. Ko-2021-0027 and from the European Research Council under the Horizon 2020 research and innovation program, Grant Agreement No. 950560 (DynaQuanta). F.X. was supported by the National Natural Science Foundation of China (Grants No. 52225207, No. 11934005, and No. 52350001), the Shanghai Pilot Program for Basic Research of Fudan University 21TQ1400100 (Grant No. 21TQ006), and the Shanghai Municipal Science and Technology Major Project (Grant No. 2019SHZDZX01).

- [1] S. Hunsche, K. Wienecke, T. Dekorsy, and H. Kurz, Impulsive softening of coherent phonons in tellurium, *Phys. Rev. Lett.* **75**, 1815 (1995).
- [2] U. Ritzmann, P. M. Oppeneer, and P. Maldonado, Theory of out-of-equilibrium electron and phonon dynamics in metals after femtosecond laser excitation, *Phys. Rev. B* **102**, 214305 (2020).
- [3] H. Handa, Y. Okamura, R. Yoshimi, A. Tsukazaki, K. S. Takahashi, Y. Tokura, and Y. Takahashi, Terahertz field driven giant nonlinear phonon response in ferroelectric semiconductor In-doped $(\text{Sn,Pb})\text{Te}$, *Phys. Rev. B* **109**, L081102 (2024).
- [4] M. Först, C. Manzoni, S. Kaiser, Y. Tomioka, Y. Tokura, R. Merlin, and A. Cavalleri, Nonlinear phononics as an ultrafast route to lattice control, *Nat. Phys.* **7**, 854 (2011).
- [5] M. Kozina, M. Fechner, P. Marsik, T. van Driel, J. M. Glowina, C. Bernhard, M. Radovic, D. Zhu, S. Bonetti, U. Staub, and M. C. Hoffmann, Terahertz-driven phonon upconversion in SrTiO_3 , *Nat. Phys.* **15**, 387 (2019).
- [6] Z. Fu and M. Yamaguchi, Coherent excitation of optical phonons in GaAs by broadband terahertz pulses, *Sci. Rep.* **6**, 38264 (2016).
- [7] T. Huber, M. Ranke, A. Ferrer, L. Huber, and S. L. Johnson, Coherent phonon spectroscopy of non-fully symmetric modes using resonant terahertz excitation, *Appl. Phys. Lett.* **107**, 091107 (2015).
- [8] T. F. Nova, A. Cartella, A. Cantaluppi, M. Först, D. Bossini, R. V. Mikhaylovskiy, A. V. Kimel, R. Merlin, and A. Cavalleri, An effective magnetic field from optically driven phonons, *Nat. Phys.* **13**, 132 (2017).
- [9] G. A. Garrett, T. F. Albrecht, J. F. Whitaker, and R. Merlin, Coherent THz phonons driven by light pulses and the Sb problem: What is the mechanism? *Phys. Rev. Lett.* **77**, 3661 (1996).
- [10] H. J. Zeiger, J. Vidal, T. K. Cheng, E. P. Ippen, G. Dresselhaus, and M. S. Dresselhaus, Theory for displacive excitation of coherent phonons, *Phys. Rev. B* **45**, 768 (1992).
- [11] A. Koç, I. Gonzalez-Vallejo, M. Runge, A. Ghalgaoui, K. Reimann, L. Kremeyer, F. Thiemann, M. Horn-von Hoegen, K. Sokolowski-Tinten, M. Woerner, and T. Elsaesser, Quantum pathways of carrier and coherent phonon excitation in bismuth, *Phys. Rev. B* **107**, L180303 (2023).
- [12] M. Runge, A. Ghalgaoui, I. Gonzalez-Vallejo, F. Thiemann, M. Horn-von Hoegen, K. Reimann, M. Woerner, and T. Elsaesser, Ultrafast carrier dynamics and symmetry reduction in bismuth by nonperturbative optical excitation in the terahertz range, *Phys. Rev. B* **107**, 245140 (2023).
- [13] T. G. H. Blank, K. A. Grishunin, K. A. Zvezdin, N. T. Hai, J. C. Wu, S.-H. Su, J.-C. A. Huang, A. K. Zvezdin, and A. V. Kimel, Two-dimensional terahertz spectroscopy of nonlinear phononics in the topological insulator MnBi_2Te_4 , *Phys. Rev. Lett.* **131**, 026902 (2023).
- [14] F. Giorgianni, M. Udina, T. Cea, E. Paris, M. Caputo, M. Radovic, L. Boie, J. Sakai, C. W. Schneider, and S. L. Johnson, Terahertz displacive excitation of a coherent Raman-active phonon in V_2O_3 , *Commun. Phys.* **5**, 103 (2022).
- [15] S. Maehrlein, A. Paarmann, M. Wolf, and T. Kampfrath, Terahertz sum-frequency excitation of a Raman-active phonon, *Phys. Rev. Lett.* **119**, 127402 (2017).
- [16] A. M. Kalashnikova, A. V. Kimel, R. V. Pisarev, V. N. Gridnev, P. A. Usachev, A. Kirilyuk, and T. Rasing, Impulsive excitation of coherent magnons and phonons by subpicosecond laser pulses in the weak ferromagnet FeBO_3 , *Phys. Rev. B* **78**, 104301 (2008).
- [17] A. Bartels, T. Dekorsy, and H. Kurz, Impulsive excitation of phonon-pair combination states by second-order Raman scattering, *Phys. Rev. Lett.* **84**, 2981 (2000).
- [18] A. Ghalgaoui, K. Reimann, M. Woerner, T. Elsaesser, C. Flytzanis, and K. Biermann, Resonant second-order nonlinear terahertz response of gallium arsenide, *Phys. Rev. Lett.* **121**, 266602 (2018).
- [19] A. Ghalgaoui, K. Reimann, M. Woerner, T. Elsaesser, C. Flytzanis, and K. Biermann, Frequency upshift of the transverse optical phonon resonance in GaAs by femtosecond electron-hole excitation, *Phys. Rev. Lett.* **125**, 027401 (2020).
- [20] B. Cheng, P. L. Kramer, M. Trigo, M. Liu, D. A. Reis, Z.-X. Shen, J. A. Sobota, and M. C. Hoffmann, Terahertz-induced tunnel ionization drives coherent Raman-active phonon in bismuth, *arXiv:2408.05770*.
- [21] M. N. Ali, Q. Gibson, S. Jeon, B. B. Zhou, A. Yazdani, and R. J. Cava, The crystal and electronic structures of Cd_3As_2 , the three-dimensional electronic analogue of graphene, *Inorg. Chem.* **53**, 4062 (2014).
- [22] S. Yue, H. T. Chorsi, M. Goyal, T. Schumann, R. Yang, T. Xu, B. Deng, S. Stemmer, J. A. Schuller, and B. Liao, Soft phonons

- and ultralow lattice thermal conductivity in the Dirac semimetal Cd_3As_2 , *Phys. Rev. Res.* **1**, 033101 (2019).
- [23] S. Kovalev, R. M. A. Dantas, S. Germanskiy, J.-C. Deinert, B. Green, I. Ilyakov, N. Awari, M. Chen, M. Bawatna, J. Ling, F. Xiu, P. H. M. van Loosdrecht, P. Surówka, T. Oka, and Z. Wang, Non-perturbative terahertz high-harmonic generation in the three-dimensional Dirac semimetal Cd_3As_2 , *Nat. Commun.* **11**, 2451 (2020).
- [24] B. Cheng, N. Kanda, T. N. Ikeda, T. Matsuda, P. Xia, T. Schumann, S. Stemmer, J. Itatani, N. P. Armitage, and R. Matsunaga, Efficient terahertz harmonic generation with coherent acceleration of electrons in the Dirac semimetal Cd_3As_2 , *Phys. Rev. Lett.* **124**, 117402 (2020).
- [25] Z. K. Liu, J. Jiang, B. Zhou, Z. J. Wang, Y. Zhang, H. M. Weng, D. Prabhakaran, S.-K. Mo, H. Peng, P. Dudin, T. Kim, M. Hoesch, Z. Fang, X. Dai, Z. X. Shen, D. L. Feng, Z. Hussain, and Y. L. Chen, A stable three-dimensional topological Dirac semimetal Cd_3As_2 , *Nat. Mater.* **13**, 677 (2014).
- [26] M. Neupane, S.-Y. Xu, R. Sankar, N. Alidoust, G. Bian, C. Liu, I. Belopolski, T.-R. Chang, H.-T. Jeng, H. Lin, A. Bansil, F. Chou, and M. Z. Hasan, Observation of a three-dimensional topological Dirac semimetal phase in high-mobility Cd_3As_2 , *Nat. Commun.* **5**, 3786 (2014).
- [27] S. Borisenko, Q. Gibson, D. Evtushinsky, V. Zabolotnyy, B. Büchner, and R. J. Cava, Experimental realization of a three-dimensional Dirac semimetal, *Phys. Rev. Lett.* **113**, 027603 (2014).
- [28] T. Liang, Q. Gibson, M. N. Ali, M. Liu, R. J. Cava, and N. P. Ong, Ultrahigh mobility and giant magnetoresistance in the Dirac semimetal Cd_3As_2 , *Nat. Mater.* **14**, 280 (2015).
- [29] C. Zhang, Y. Zhang, X. Yuan, S. Lu, J. Zhang, A. Narayan, Y. Liu, H. Zhang, Z. Ni, R. Liu, E. S. Choi, A. Suslov, S. Sanvito, L. Pi, H.-Z. Lu, A. C. Potter, and F. Xiu, Quantum Hall effect based on Weyl orbits in Cd_3As_2 , *Nature (London)* **565**, 331 (2019).
- [30] Y. Liu, C. Zhang, X. Yuan, T. Lei, C. Wang, D. Di Sante, A. Narayan, L. He, S. Picozzi, S. Sanvito, R. Che, and F. Xiu, Gate-tunable quantum oscillations in ambipolar Cd_3As_2 thin films, *NPG Asia Mater.* **7**, e221 (2015).
- [31] A. Ghalgaoui, L.-M. Koll, B. Schütte, B. P. Fingerhut, K. Reimann, M. Woerner, and T. Elsaesser, Field-induced tunneling ionization and terahertz-driven electron dynamics in liquid water, *J. Phys. Chem. Lett.* **11**, 7717 (2020).
- [32] J. Lu, X. Li, H. Y. Hwang, B. K. Ofori-Okai, T. Kurihara, T. Suemoto, and K. A. Nelson, Coherent two-dimensional terahertz magnetic resonance spectroscopy of collective spin waves, *Phys. Rev. Lett.* **118**, 207204 (2017).
- [33] M. Woerner, A. Ghalgaoui, K. Reimann, and T. Elsaesser, Two-color two-dimensional terahertz spectroscopy: A new approach for exploring even-order nonlinearities in the nonperturbative regime, *J. Chem. Phys.* **154**, 154203 (2021).
- [34] C. Zhu, P. Pilch, R. M. A. Dantas, A. Reinold, Y. Yang, F. Xiu, A. Bedoya-Pinto, S. S. P. Parkin, R. Moessner, and Z. Wang, Population inversion and ultrafast terahertz nonlinearity of transient Dirac fermions in Cd_3As_2 , *Phys. Rev. B* **109**, L201111 (2024).
- [35] C. Zhu, X. Yuan, F. Xiu, C. Zhang, Y. Xu, R. Zhang, Y. Shi, and F. Wang, Broadband hot-carrier dynamics in three-dimensional Dirac semimetal Cd_3As_2 , *Appl. Phys. Lett.* **111**, 091101 (2017).
- [36] C. Bao, Q. Li, S. Xu, S. Zhou, X.-Y. Zeng, H. Zhong, Q. Gao, L. Luo, D. Sun, T.-L. Xia, and S. Zhou, Population inversion and Dirac fermion cooling in 3D Dirac semimetal Cd_3As_2 , *Nano Lett.* **22**, 1138 (2022).
- [37] W. Zhang, Y. Yang, P. Suo, K. Sun, J. Peng, X. Lin, F. Xiu, and G. Ma, Semiconductorlike photocarrier dynamics in Dirac-semimetal Cd_3As_2 films probed with transient terahertz spectroscopy, *Phys. Rev. B* **106**, 155137 (2022).
- [38] A. Sharafiev, V. Gnezdilov, R. Sankar, F. C. Chou, and P. Lemmens, Optical phonon dynamics and electronic fluctuations in the Dirac semimetal Cd_3As_2 , *Phys. Rev. B* **95**, 235148 (2017).
- [39] M. I. Aroyo, J. M. Perez-Mato, C. Capillas, E. Kroumova, S. Ivantchev, G. Madariaga, A. Kirov, and H. Wondratschek, Bilbao crystallographic server: I. Databases and crystallographic computing programs, *Z. Kristall. Crystall. Mater.* **221**, 15 (2006).
- [40] M. I. Aroyo, A. Kirov, C. Capillas, J. M. Perez-Mato, and H. Wondratschek, Bilbao crystallographic server. II. Representations of crystallographic point groups and space groups, *Acta Cryst. A* **62**, 115 (2006).
- [41] F. Sun, Q. Wu, Y. L. Wu, H. Zhao, C. J. Yi, Y. C. Tian, H. W. Liu, Y. G. Shi, H. Ding, X. Dai, P. Richard, and J. Zhao, Coherent helix vacancy phonon and its ultrafast dynamics waning in topological Dirac semimetal Cd_3As_2 , *Phys. Rev. B* **95**, 235108 (2017).
- [42] D. Houde, S. Jandl, M. Banville, and M. Aubin, The infrared spectrum of Cd_3As_2 , *Solid State Commun.* **57**, 247 (1986).
- [43] C. L. Johnson, B. E. Knighton, and J. A. Johnson, Distinguishing nonlinear terahertz excitation pathways with two-dimensional spectroscopy, *Phys. Rev. Lett.* **122**, 073901 (2019).
- [44] W. Kuehn, P. Gaal, K. Reimann, M. Woerner, T. Elsaesser, and R. Hey, Terahertz-induced interband tunneling of electrons in GaAs, *Phys. Rev. B* **82**, 075204 (2010).
- [45] E. J. Sie, C. M. Nyby, C. D. Pemmaraju, S. J. Park, X. Shen, J. Yang, M. C. Hoffmann, B. K. Ofori-Okai, R. Li, A. H. Reid, S. Weathersby, E. Mannebach, N. Finney, D. Rhodes, D. Chenet, A. Antony, L. Balicas, J. Hone, T. P. Devereaux *et al.*, An ultrafast symmetry switch in a Weyl semimetal, *Nature (London)* **565**, 61 (2019).
- [46] H. Pan, M. Wu, Y. Liu, and S. A. Yang, Electric control of topological phase transitions in Dirac semimetal thin films, *Sci. Rep.* **5**, 14639 (2015).
- [47] J. L. Collins, A. Tadich, W. Wu, L. C. Gomes, J. N. B. Rodrigues, C. Liu, J. Hellerstedt, H. Ryu, S. Tang, S.-K. Mo, S. Adam, S. A. Yang, M. S. Fuhrer, and M. T. Edmonds, Electric-field-tuned topological phase transition in ultrathin Na_3Bi , *Nature (London)* **564**, 390 (2018).