Light-Driven Raman Coherence as a Nonthermal Route to Ultrafast Topology Switching in a Dirac Semimetal

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A grand challenge underlies the entire field of *topology-enabled* quantum logic and information science: how to establish topological control principles driven by quantum coherence and understand the time dependence of such periodic driving. Here we demonstrate a few-cycle THz-pulse-induced phase transition in a Dirac semimetal ZrTe₅ that is periodically driven by vibrational coherence due to excitation of the lowest Raman active mode. Above a critical THz-pump field threshold, there emerges a long-lived metastable phase, approximately 100 ps, with unique Raman phonon-assisted topological switching dynamics absent for optical pumping. The switching also manifests itself by distinct features: nonthermal spectral shape, relaxation slowing near the Lifshitz transition where the critical Dirac point occurs, and diminishing signals at the same temperature that the Berry-curvature-induced anomalous Hall effect magnetoresistance vanishes. These results, together with first-principles modeling, identify a mode-selective Raman coupling that drives the system from strong to weak topological insulators with a Dirac semimetal phase established at a critical atomic displacement controlled by the phonon coherent pumping. Harnessing of vibrational coherence can be extended to steer symmetry-breaking transitions, i.e., Dirac to Weyl ones, with implications for THz topological quantum gate and error correction applications.

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I. INTRODUCTION

Dynamic driving by periodic lattice vibrations represents a powerful approach to manipulate topological band structures, in stark contrast to equilibrium tuning methods, e.g., temperature, chemical substitution, and static strain, electric, or magnetic fields [1,2]. Ultrafast nonthermal manipulation of topology [3,4], particularly at preferred THz-cycle clock rates, is key for full implementation of the dynamical protocols needed to both match current information and sensing technologies and exceed their limits via topological functionalities [5–8]. Despite the recent intriguing studies of coherent phonon pumping in various quantum materials [9–12], topological semimetals dressed by optical phonon coherence have not been experimentally

explored, especially for Dirac materials. ZrTe₅ is a model Dirac system [13–15] for studying such a quantum topological switching by periodic driving from coherent phonons because of the minimal single nodal (Dirac) point and extreme sensitivity on the small structural changes across a broad range of phases, from strong topological insulators (STIs) to Dirac semimetal to weak topological insulators (WTIs). However, only thermal- or strain-induced transitions [16,17] in quasiequilibrium states have been reported.

We implement a dynamical topology-switching scheme using intense few-cycle THz pulses (red line) to excite a Raman active (A_{1g}) optical phonon mode, as illustrated in Fig. 1(a). The subpicosecond THz driving has broadband spectrum with central frequency of approximately 1.2 THz [gray shaded area, Fig. 1(b)]. The A_{1g} eigenmode [Fig. 1(c)] is a translational rigid-chain structure which mostly involves opposite displacements along the b axis of the dimer Te (Te_d), apical Te (Te_a), and Zr atoms from the neighboring Zr-Te units. This motion results in a modulation of the atomic positions along the b axis that determines the van der Waals coupling and, in turn, controls the band inversion at the Γ point between the p orbitals of Te_d and zigzag Te_z atoms. Most intriguingly, this mode

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FIG. 1. Intense THz-driven Raman phonon coherence in ZrTe_5 . (a) Schematic of the topological switching driven by the THz coherent excitation in Dirac semimetals. (b) Spectrum of the coherent phonon emission (red) obtained from the time-domain traces shown for maximum THz-field strengths $E_{\text{max}} = 736$ kV/cm. The pump THz spectrum is shown in gray. Dashed lines indicate the identification of the excited modes in the driven states with the measured Raman spectrum (blue). (c) Crystal structure of ZrTe_5 and the relative atomic displacements of the A_{1g} Raman mode leading to the modulation of interlayer spacing. The arrows indicate the direction and magnitude of the atomic displacements. (d) Time-domain traces of the measured coherent phonon emission for various THz-field strengths. The THz-driving pulse is shown in gray.

could effectively enable a topology change without symmetry breaking because it has extreme sensitivity to the band inversion while it still preserves the inversion symmetry of the lattice. Coherent excitation of the A_{1g} phonon is expected to create a periodically driven state via lattice vibrations that modulate the topological bands and switch from STI [top, Fig. 1(a)] to Dirac semimetal (middle) to WTI (bottom) phases. Dephasing of this topological coherent state via Dirac fermion-phonon interaction leads to the conversion of Raman phonon coherence into population, i.e., into finite atomic displacement associated with the establishment of a final state with highly nonthermal characteristics. However, the salient spectroscopy features for the ultrafast nonthermal phase transition in any Dirac materials have not been established until now.

In this paper, we provide evidence for a distinct topology switching driven by THz Raman phonon coherence in a Dirac semimetal. Our results are corroborated by the calculations of A_{1g} mode-selective electronic band structures and topology changes. Harnessing the driven coherence of specifically tailored phonon modes with an intense near-single-cycle THz pulse may become a universal principle for steering topological phase transitions.

II. EXPERIMENTAL METHODS AND RESULTS

We use a bulk single-crystal ZrTe_5 sample that exhibits a 3D linear dispersion [18]. In equilibrium, our sample is close to charge neutral, and the Fermi level E_F is less than 5 meV and approximately 14 meV at 5 K and room

temperature, respectively. It has been reported that E_F is proportional to the Lifshitz transition temperature T_{Lifshitz} that depends on the Te content [19]. ZrTe_5 single crystals grown by Te self-flux, as we use, are close to the stoichiometric value and have lower E_F and T_{Lifshitz} , while those crystals grown by the chemical-vapor-transport method are sometimes Te deficient and have higher values. Intense near-single-cycle THz-pump pulses are generated through optical rectification employing the tilted pulse front scheme in LiNbO_3 using 800-nm 35-fs pulses from a 1-kHz Ti:sapphire regenerative amplifier system. A detailed description of the THz-pump, THz-probe, and THz-emission setups and data acquisition scheme can be found in Refs. [8,20–23].

Coherent phonon emissions after intense THz-pump excitation at 4.1 K for various field strengths $E_{\text{THz}} = 5, 22, 86, 184, 386, 552$, and 736 kV cm^{-1} are sampled in the time domain by a weak optical pulse, as shown in Fig. 1(d). A pronounced multicycle oscillation is clearly visible in the sample emission. The Fourier-transform spectra of these coherent beatings at 736 kV cm^{-1} display two dominant peaks centered at approximately 1.2 and 2.1 THz. The static Raman spectrum [blue line, Fig. 1(b)] from the same sample shown together, identifies their Raman symmetry (dash lines). Note that the strongest emission peak of approximately 1.2 THz in the driven states matches very well with the A_{1g} mode, unlike for the B_{2g} mode dominant in the static Raman spectra. Thus, the intense THz drive coherently excites the Raman A_{1g} mode resulting in a periodic modulation of the interlayer spacing. In addition, the three infrared (IR) active phonon modes with frequencies 0.63, 1.5, and 2.3 THz, as seen in linear THz transmission (see Fig. S2 in the Supplemental Material [24]) are negligibly small in the intense THz-driven state [Fig. 1(d)], i.e., the preferred coupling of the topological electronic bands to the A_{1g} mode. This Raman-mode-selective coupling attests the proposed excitation scheme in Fig. 1(c) due to the extreme sensitivity of the band inversion to the interlayer spacing, which is reproduced by density-functional simulations that we discuss later.

To characterize the observed periodically driven topological state dressed by Raman coherence, Fig. 2(a) plots the THz differential transmission $\Delta E/E_0$ (red circles) after excitation by the intense THz-pump pulse (gray shaded area) as a function of the pump-probe time delay Δt_{pp} at 4.1 K. Note that the pump-induced signals are normalized by the value of peak amplitude E_0 at the probe THz E field (gray shaded area, inset). The non-equilibrium signal $\Delta E/E_0$ measured at the gating pulse delay $t_{\text{gate}} = 0 \text{ ps}$ (inset) originates from multiple processes ranging from intra- and interband excitations assisted by coherent phonons, to phonon renormalization and recovery of the quasiequilibrium Dirac state, e.g., via scattering within and between the Dirac cones via electron-electron ($e-e$) and electron-phonon ($e\text{-ph}$) interactions. The

FIG. 2. Light-driven topological phase transition above threshold THz-pump field and at low temperature where the Berry curvature dominates. (a) THz differential transmission ($\Delta E/E_0$, red circles) as a function of the pump-probe time delay (Δt_{pp}) for a peak pump E field of 736 kV/cm . The blue trace shows the simultaneously measured phonon emission. The THz-pump trace is shown in gray. The THz differential transmission starts to build up during the coherent phonon emission at much longer times than the pump THz pulse. Inset: Time-domain THz raw data. The transmitted probe E field through the sample E_{sample} (gray shaded area) and the pump-induced transmitted probe E -field change ΔE (red line) at $\Delta t_{\text{pp}} = 10 \text{ ps}$ are measured by scanning t_{gate} . We normalize the pump-induced signals by the value of peak amplitude E_0 at the probe THz E field. (b) THz differential transmission for the case of high-energy optical pump (1.55 eV) shows a much faster rise time. (c) Temperature dependence of the differential THz transmission in the metastable state ($\Delta t_{\text{pp}} = 10 \text{ ps}$, blue diamonds) indicates strong correlation with the Berry-curvature-induced anomalous Hall effect (black solid circles). (d) Pump E -field dependence of the THz differential transmission for the THz pump at ($\Delta t_{\text{pp}} = 10 \text{ ps}$), indicating a threshold pump field for the formation of the metastable state. Inset: The thermal topological change $\Delta E_{\text{thermal}}$ required for the thermally driven STI-DP-WTI transition is determined by the change of THz-field transmission between 4.1 and 160 K (T_{Berry}).

dynamics is further elaborated later. For clarity, shown together is the pump-induced coherent phonon emission (black line, overlaid) that is measured simultaneously as a function of Δt_{pp} . These results reveal the buildup of a metastable state, which occurs exclusively during the coherent Raman phonon oscillations in time domain. At the early times during the THz pulse marked by t_{pulse} [Fig. 2(a)], there is only small pump-induced $\Delta E/E_0$ signals. The delayed rise in ultrafast transient signals excludes the direct heating of electronic states near the Fermi surface as the dominant excitation channel, which will lead to quasi-instantaneous increase in the transient signals on the arrival of the pump pulse. In contrast, the transient-state evolution seen from the pronounced $\Delta E/E_0$ signals occurs only at later times after the pulse but before phonon dephasing marked as $t_{\text{dephasing}}$, i.e., during the period of the pronounced coherent phonon vibrations [blue line, Fig. 2(a)]. This emergent behavior after the incident THz excitation dominates the driven-state dynamics. The formation process is followed by a quasisteady temporal regime that marks the establishment of a final metastable state after the dephasing of the Raman phonon coherence by, e.g., the strong $e-e$ and e -ph coupling. In addition, the buildup behavior during coherent oscillations is absent for high-frequency pump pulses tuned at 1.55 eV [Fig. 2(b)]. Here we see only a sub-ps rise which now occurs mostly during the photoexcitation. This stark difference clearly indicates the nonthermal nature of the THz-driven state mediated by coherent Raman phonons.

Experimental evidence associating the observed phase evolution with the topological switching is presented in Figs. 2(c) and 2(d) as follows. The first evidence is to compare the temperature dependence of the pump-probe $\Delta E/E_0$ signals in the metastable states (at $\Delta t_{\text{pp}} = 10$ ps) with that of the anomalous Hall effect (AHE) magnetoresistance that directly probes the Berry curvature Ω_k generated by the Weyl nodes. As shown in Fig. 2(c), the nonlinear THz signal (blue diamond) quickly diminishes at the same temperature $T_{\text{Berry}} \sim 160$ K, where AHE sensitivity ρ_{AHE}^0 vanishes (black circles). In the AHE measurement, application of a magnetic field transforms a Dirac semimetal into a Weyl semimetal by breaking time-reversal symmetry. Consequently, Weyl nodes behave like magnetic monopoles that generate large Berry curvatures and act like an effective magnetic field. This field-induced Berry curvature gives rise to a nonzero $\rho_{\text{AHE}} = \Omega_k \times E$. Here we obtain the temperature dependence of ρ_{AHE}^0 , i.e., the saturation value of $\rho_{\text{AHE}}(B)$ (see Fig. S1 in the Supplemental Material [24]) by subtracting the ordinary Hall signals (linear background) at high magnetic field from the experimentally measured Hall resistivity [25]. It is clearly visible that ρ_{AHE}^0 in ZrTe₅ emerges below T_{Berry} when the dominant carriers are Dirac fermions with linear dispersion near the conical point with conserved chirality. The T_{Berry} correlates very

well with the critical transition temperature associated with the THz-driven metastable phase [blue diamond, Fig. 2(c)]. Therefore, the metastable phase has the same topological origin as the chiral transport and cannot be established by excitation of a normal Fermi surface dominant above T_{Berry} . Note also that the sign change of ρ_{AHE}^0 in the vicinity of $T_{\text{Lifshitz}} \sim 60$ K where the critical DP occurs separating the STI-WTI transitions [16] [see Fig. S1(b) in the Supplemental Material [24]] agrees with the rapid rise of the THz-pump-induced signals in the driven states marked by the black dashed line associated with the buildup of the new phase.

The second evidence is a distinct nonlinear pump fluence dependence of the THz differential transmission $\Delta E/E_0$. It exhibits both photoexcitation threshold behavior and larger magnitude than the change required for the thermally driven topological phase transition denoted as $\Delta E_{\text{thermal}}$. Specifically, by increasing the temperature from 4.2 to 160 K (T_{Berry}), as we show in the inset of Fig. 2(d), the $\Delta E_{\text{thermal}}$ can be directly determined as approximately 0.09 corresponding to the change of THz-field transmission during the STI-DP-WTI transition in equilibrium. Here we compare the $\Delta E_{\text{thermal}}$ with the THz-pump field dependence of the differential transmission $\Delta E/E_0$ signals at a fixed time $\Delta t_{\text{pp}} = 10$ ps [Fig. 2(d)]. We emphasize two key points. First, it is clearly visible that the pump-induced $\Delta E/E_0$ is negligibly small at THz-field strengths less than $E_{\text{th}} \sim 75$ kV/cm. This threshold behavior of the formation dynamics [Fig. 2(a)] is not limited by our noise floor, which is a hallmark of the nonequilibrium phase transition to a THz-driven metastable state. Second, at slightly higher field above the E_{th} , the pump-induced differential transmission surpasses the $\Delta E_{\text{thermal}}$ value for the thermally induced STI-DP-WTI topological switching in equilibrium (black dashed line). Thus, we conclude that the sufficiently large lattice displacement above the E_{th} drives the system across the topological phase boundary to new band structures determined by phonon vibration amplitudes.

Next, we further identify some distinguishing spectral and temporal features associated with the THz-driven metastable topological phase that are different from the thermal states. First, Fig. 3(a) reveals a distinct nonthermal spectral shape in the nonequilibrium response function. At equilibrium, the frequency-dependent conductivity $\sigma_1/\sigma_{\text{dc}}$ [inset, Fig. 3(a)] reveals two strong IR-active phonons along the a axis (probe direction) with resonant peaks at $\omega_{\text{IR}}^{1,2} \sim 1.5$ and approximately 2.5 THz in the 4.2-K trace (red line). These IR phonon modes can contribute to the Raman A_{1g} phonon generation via the ionic Raman mechanism that involves the IR modes as a mediator and IR Raman coupling due to anharmonicity [26]. At elevated temperatures, these modes progressively shift to higher frequencies up to 200 K (magenta line). In the

FIG. 3. The distinct spectral and temporal features of the highly nonthermal, metastable topological states. (a) Pump-induced change in conductivity ($\Delta\sigma_1/\sigma_{dc}$, red circles) at $\Delta t_{pp} = 10$ ps and $T = 4.2$ K compared with the thermal conductivity change (black circles) between 120 and 4.2 K, i.e., STI and WTI states. Inset shows the shift in the IR phonon frequencies in the static conductivity along the a axis as a function of the temperature. (b) Differential THz transmission profiles ($\Delta E/E_0$) for various temperatures show that the relaxation of the metastable state is the longest near $T_{\text{Lifshitz}} \sim 60$ K. Inset: THz response $\Delta\sigma_1/\sigma_{dc}$ as a function of the frequency for time delays $\Delta t_{pp} = 2.5, 10, 30$, and 50 ps at $E_{\text{THz}} = 736$ kV/cm at 4.2 K.

THz-driven phase, the pump-induced conductivity change $\Delta\sigma_1/\sigma_{dc}$ at $\Delta t_{pp} = 10$ ps shows spectral oscillations [red circles, Fig. 3(a)] with pronounced absorptive features (red arrows). In contrast, the normal-state thermalization leads to dominantly inductive spectral shape [black line, Fig. 3(a)], which can be obtained by subtracting σ_1 traces at higher temperature and 4.2 K, i.e., between the STI ground and WTI thermal states. This result highlights the difference between the driven phase evolution and temperature- or laser-heating-induced phase thermalization process. Second, Fig. 3(b) plots the relaxation dynamics that measures the lifetime of the THz-driven state from 4.2 to 120 K. The temporal profile is consistent with the slow buildup peaked at approximately 10 ps, and approximately 100-ps decay of transient Dirac fermion population and phonon renormalization, as seen in the σ_1/σ_{dc} spectra shown for various Δt_{pp} delays [inset, Fig. 3(b)]. The

transient phase decays with a single exponential profile over approximately 120 ps; see, e.g., the 60-K trace (black line). Such a relaxation is nearly 2 orders of magnitude longer than that reported for the case of optical excitation using higher-energy photons [27,28]. This observation is due in part to the minimal electronic or phonon heating and negligible excitation of other topological trivial bands from the low-frequency THz pumping. Intriguingly, the relaxation time exhibits a nonmonotonic temperature dependence. It becomes longer initially with temperature increase from 4.2 K (gray line), reaches a maximum at approximately 60 K (black line), and finally decreases with a temperature increase up to 120 K (blue line), as shown in Fig. 3(b). Critical to note is that the longest lifetime appears at approximately T_{Lifshitz} at which the Fermi surface crosses the Dirac point [16]. The prolonged decay times and their correlation with the Lifshitz transition are exceptional in ZrTe_5 , which reflects the unique dynamics of Dirac bands, instead of other topologically trivial ones. This correlation further underscores the topological origin of the THz-driven phase transition as we discuss below.

Coherent and nonthermal THz photoexcitations alter the thermodynamic equilibrium of electronic states and lead to a series of dynamic processes arising from the generation and decay of transient Dirac fermion and phonon population in ZrTe_5 . During and immediately following the single-cycle THz excitation, strongly driven coherent phonons modulate the Dirac bands and decrease the gap size and Fermi energy. These periodic gap closing by coherent phonons assist the interband excitation between the Dirac cones and the intraband Fermi sea pumping. The excitation is followed by buildup of a quasiequilibrium Dirac state after the dephasing. Then, for longer times this quasiequilibrium Dirac state relaxes via scattering within and between the Dirac cones via intraband and interband transitions. These processes determine the decay dynamics and slowing behavior observed in Fig. 3(b). We emphasize two key points. First, the linear Dirac spectrum gives rise to slow imbalance relaxation. As it is rigorously formulated in graphene, the number of transient electrons and holes in the upper and lower Dirac bands are individually conserved up to second order in the Coulomb interaction [29]. Second, near the Lifshitz transition when the Fermi energy approaches the conical point, there will be further slowing in the decay due to kinematic phase-space constraints imposed on the relaxation process: less scattering due to reduced density of states between the Dirac cones (interband) and within them (intraband) via phonons. Note that our observed slow decay is consistent with similar “tens of” for clarity of ps lifetimes reported in carrier-density-tuned topological insulators [30] and should be distinguished from the dynamic slowing mechanism in symmetry-breaking phase transitions [31].

III. ELECTRONIC STRUCTURE CALCULATIONS AND DISCUSSIONS

To put the observed Raman-mode-selective topological phase transition on stronger footing, we carry out phonon and electronic band structure calculations based on density-functional theory (DFT) that provide theoretical support for the topological switching by the coherent A_{1g} phonon driving. Among the calculated zone-center optical phonon modes with the relaxed unit cell (see Supplemental Material [24]), the lowest Raman A_{1g} mode of 1.12 THz and B_{2g} mode of 2.18 THz compare well with the experimental values [32] of 1.18 and 2.16 THz, respectively. To study the effect of this A_{1g} mode on the electronic structure and band topology of ZrTe_5 , we change the atomic positions according to the A_{1g} mode [see Fig. 1(c)] with a lattice distortion factor (λ) and calculate the new band structure. For $\lambda = 1.0$, the corresponding atomic displacements are 0.033 Å for Zr, Te_d , and Te_a , and 0.017 Å for Te_z . The band structure of ZrTe_5 along $Z - \Gamma - Y$ for different displacements λ is shown in Fig. 4. First, without any atomic displacements from equilibrium ($\lambda = 0.0$) [Fig. 4(b)], ZrTe_5 has a narrow band gap of 0.04 eV. Both the valence- and conduction-band edges appear along $\Gamma - Y$ and are off the Γ point toward the Y point. The projection (green shadow) on the Te_d p orbitals clearly shows the band inversion between the valence and conduction bands, which agrees with the gapped massless Dirac state observed in experiments [13]. As one of the indicators, the 2D topological index on the $k_z = 0$ plane is 1 [see Fig. S3(a) in the Supplemental Material [24]] shown by the odd number of crossings for the Wannier charge centers (WCCs) moving along k_y . The overall topological invariant index is (1;110) for the initial state. For $\lambda = -3.0$ [Fig. 4(a)], the band gap increases to 0.07 eV, but the band inversion along the $Z - \Gamma - Y$ directions remains. The system is still a gapped massless Dirac state [33]. Most interestingly, in contrast to the above, by moving in the other direction with more positive λ , the band gap decreases. The valence and conduction bands touch at the Γ point when $\lambda = 2.15$ [Fig. 4(c)]. Then, the band gap reopens as λ increases further. For $\lambda = 3.0$ [Fig. 4(d)], the reopened band gap has no band inversion along $Z - \Gamma - Y$, as seen from the orbital projection of the Te_d p orbitals (inset, green line). The corresponding 2D topological index on the $k_z = 0$ plane is now 0, shown by the even number of crossings for the WCCs moving along k_y in Fig. S3(b) of the Supplemental Material [24]. The overall topological index becomes (0;110) for a WTI. Thus, with the coherently excited Raman A_{1g} mode within one THz-pulse cycle above threshold E_{th} , ZrTe_5 can be driven into coherent topological oscillations between the STI and WTI states, with an interesting critical Dirac point in between. The dephasing of the phonon vibration from

FIG. 4. Electronic band structure calculations for the topological phase switching via excitation of the A_{1g} phonon mode in ZrTe_5 . The band structure along $Z - \Gamma - Y$ for different magnitudes of the lattice distortion factor λ corresponding to the A_{1g} mode [see Fig. 1(c)]: (a) -3.00 , (b) 0.00 , (c) 2.15 , and (d) 3.00 . For $\lambda = 1.0$, the corresponding atomic displacements for the A_{1g} mode are 0.033 Å for Zr, Te_d , and Te_a , and 0.017 Å for Te_z . The green shadow indicates the projection of Te_d p orbitals denoting band inversion. This procedure identifies the states with $\lambda = -3.00, 0.00$ (a),(b) as STI having a topological index of (1;110), whereas the critical Dirac point occurs for $\lambda = 2.15$ (c). Absence of band inversion for $\lambda = 3.00$ (d) indicates a WTI with the topological index (0;110). (e) Energy gap at the Γ point as calculated from the band structure as a function of the atomic displacement magnitudes λ .

multiple Dirac fermion-phonon scattering and/or disorder effects results in nonthermal phonon populations, which lead to a renormalized spectral shape different from thermal ones shown in Fig. 3(a). This critical bulk DP point at $\lambda = 2.15$ is marked by the vertical arrow in the λ dependence of the band gap [Fig. 4(e)], which can exist at the Γ point in ZrTe_5 as the phase boundary between the gapped Dirac state and WTI. Such a transition can be driven by the A_{1g} Raman phonon mode consistent with the measured THz-driven states. We note that a recent study reported a direct measurement of the atomic displacement of a low-frequency shear phonon mode vs THz-field strength in a Weyl semimetal [9]. Comparing the two excitation conditions, we can estimate an atomic displacement of approximately 0.07 Å for the 750-kV/cm field and is on the same order of magnitude

needed for driving the topological phase transition by the A_{1g} mode at the critical Dirac point ($\lambda = 2.15$) in the simulation.

It is also important to note that unlike the low-energy THz excitation, the huge mismatch between the optical pump vs A_{1g} mode energy may lead to many spurious effects and mask the intrinsic Dirac dynamics, e.g., the excitation of topological trivial bands, incoherent electron and phonon heating, and more rapid loss of coherence that inhibits the formation of the metastable state. In addition, we also theoretically examine the other modes such as B_{2g} excited in the driven state [Fig. 1(b)]. Unlike the A_{1g} mode involving the lattice distortion along the stacking b axis [Fig. 1(c)], the B_{2g} mode at 2.16 THz involves atomic displacements of Te along the a axis, a torsion of the Te dimer, and thereby costs higher energy, i.e., not as effective as the A_{1g} mode to change the band inversion between the p orbitals of the Te_d and Te_z atoms.

IV. CONCLUSION

In summary, we identify a previously inaccessible topological tuning scheme via mode-selective Raman coherence. We implement it by demonstrating THz-driven topological phase transition during coherent lattice oscillations in a Dirac semimetal. This topological quantum switching dynamics warrant further investigation to resolve them through direct electronic structure probes within the phonon oscillation cycle. This dynamic lattice-driven scheme may also be extended to diverse complex materials and driven phenomena, such as symmetry-breaking transitions to induce Weyl states [34], magnetic materials [35,36], photovoltaic semiconductors [22,37], and superconductors [38,39].

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