

Light wave induced nanosecond-long persistent state in the Dirac semimetal Cd_3As_2

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Unraveling light induced ultrafast dynamics in three-dimensional Dirac materials is crucial for deepening our fundamental understanding and for advancing potential optoelectronic applications. While time-resolved pump-probe spectroscopy has predominantly revealed carrier dynamics governed by Dirac fermion cooling and population inversion, the full picture of relaxation pathways remains incomplete. In this Letter, we identify a relaxation channel in these materials. Using strong terahertz (THz) field excitation combined with a near-infrared (NIR) probe, we detect a long-lived carrier population persisting on the nanosecond timescale. This extended lifetime suggests that carriers become trapped in defect states, leading to sub-band-gap absorption in the NIR and pointing to the significant role of defect-assisted relaxation in the carrier dynamics of Dirac semimetals.

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Cd_3As_2 is an appealing three-dimensional (3D) Dirac semimetal, often regarded as the 3D analog of graphene. Unlike conventional semiconductors, it hosts massless Dirac fermions in three dimensions, making it a promising platform for exploring exotic quantum phenomena [1–7]. However, Cd_3As_2 inherently contains a significant number of defects, which can strongly influence its electronic and optical behavior. These defects such as vacancies, dislocations, and antisite defects arise from the material's metastable crystal structure and growth conditions. Acting as scattering centers, they impact carrier mobility and introduce localized states within the band structure, potentially modifying its quantum transport properties [8–13].

In conventional semiconductors, defect states commonly form within the band gap, between the conduction band (CB) and valence band (VB), where they act as carrier traps and significantly affect recombination dynamics [14–16]. In contrast, Dirac materials often exhibit enhanced robustness due to their linear band dispersion and symmetry-protected states, which can be harnessed to tailor electronic and magnetic properties. Notably, certain defects in these systems can induce localized magnetic moments or zero-energy modes, making them promising for applications in spintronics and quantum technologies [17,18]. Recent studies in Dirac semimetals, such as Cd_3As_2 and the closely related Na_3Bi , revealed that these defects shift the Fermi level (E_F) relative to the Dirac point

(E_D) by up to 20 meV [9,11,19]. Beyond their impact on carrier concentration, such defects can also introduce localized states near the Dirac nodes [12] or within the valence band [20], where they can act as carrier traps. Additionally, these localized electronic states may form deep levels that are significantly separated in energy from the band edge particularly under conditions such as band bending, surface oxidation, or interface induced potential fluctuations. In graphene, it has been demonstrated that defects can induce states that act as localized perturbations, breaking sublattice symmetry and resulting in midgap resonances and a partial opening of a band gap near the Dirac point [21,22]. In addition, these states can act as localized perturbations that mimic quantum dot behavior [19,23], exhibiting nanosecond-scale lifetimes that reflect strong carrier localization. Such long-lived, localized excitations open promising avenues for tuning near-infrared (NIR) and visible photonic responses [23].

Femtosecond pump-probe spectroscopy has been employed to investigate carrier dynamics in this system, with a primary focus on the Γ point in the Brillouin zone [24–27]. These studies have shed light on key aspects of Dirac fermion cooling dynamics. Two dominant relaxation pathways have been identified in 3D Dirac systems: (i) intraband relaxation via electron-phonon scattering, characterized by a relaxation rate that scales linearly with energy, and (ii) interband relaxation, indicated by the accumulation of Dirac fermions in the conduction band, an effect that has been proposed as a potential origin of population inversion [24,28,29]. Thus, probing relaxation dynamics at significantly higher photon energies could provide deeper insight into purely interband relaxation mechanisms, thereby enhancing our understanding of carrier behavior in 3D Dirac semimetals. However, only a limited number of experiments have been conducted at such higher photon energies, and these studies have not yet reached a clear

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consensus regarding the underlying relaxation mechanisms [30,31].

Here, we present evidence for a carrier relaxation pathway in the 3D Dirac semimetal Cd_3As_2 , revealed through pump-probe spectroscopy performed at various excitation and probe wavelengths. We observe a long-lived state persisting up to the nanosecond timescale under NIR probing, whereas this state is absent when using a THz probe. This long-lived response suggests suppressed carrier recombination, with carriers transiently trapped in newly formed localized states.

The sample consists of a $\text{Cd}_3\text{As}_2(112)$ thin film with a thickness of approximately 60 nm, grown using a PerkinElmer 425B molecular beam epitaxy (MBE) system. Freshly cleaved 2-in. mica (70 μm thick) was used as a substrate. A 10-nm-thick CdTe buffer layer was first deposited prior to the growth of $\text{Cd}_3\text{As}_2(112)$. The Cd_3As_2 bulk material was then evaporated onto the CdTe layer at a substrate temperature of 170 $^\circ\text{C}$. The Fermi energy and Fermi velocity were estimated to be $E_F = 118$ meV and $v_F = 7.8 \times 10^5$ m/s from Shubnikov–de Haas (SdH) oscillation measurements [32,33]. The shift of the Fermi level above the Dirac node is attributed to unintentional n -type doping introduced during sample preparation. These defects are mainly cadmium vacancies and arsenic interstitials.

We investigated the dynamics of photogenerated charge carriers in Cd_3As_2 by employing three distinct experimental methods: THz pump–optical probe, optical pump–THz probe, and THz pump–THz probe. Optical pulses (100 fs, 800 nm, 1 kHz) were generated by a Ti:sapphire amplifier. THz pulses were generated via optical rectification of the 800 nm pulse in LiNbO_3 using the tilted-wave technique to achieve phase matching. The resulting THz field was centered at 1 THz, with an electric field strength reaching 1 MV/cm [34–36].

The first method involved a THz pump–800 nm probe setup, where the THz field strength was varied to monitor the dynamics of the material under different field strengths and at varied delay times τ [Fig. 1(a)]. The second configuration can be realized by switching to an 800 nm pump–THz probe configuration within the same setup with a reduced THz pulse energy, enabling the examination of the system at different NIR pump energies [Fig. 1(a)]. Because of its low photon energy (4 meV), a THz pulse with low-field strength (i.e., a THz probe pulse) can sensitively detect the Drude-like free-carrier response near the band edges. However, when the field strength is sufficiently high, the THz field can drive carriers into a nonperturbative regime, inducing intraband acceleration across the Brillouin zone, as well as interband excitation [37,38]. In contrast, NIR light with a photon energy of 1.5 eV couples to high-energy states far from the band edges and therefore probes a different subset of carriers. When used as a pump, NIR excitation can access a much broader range of electronic states in energy space compared to THz excitation [Fig. 1(a), right panel].

The third experimental method, pump-probe THz spectroscopy, was conducted using pairs of THz pulses: a primary pulse A and a weaker pulse B. A delay stage was used to adjust the time delay τ of pulse A relative to pulse B [Fig. 1(b)]. Two optical chopper wheels, operating at 250 and 500 Hz, respectively, enabled independent measurements of the transmitted electric-field transients— E_{AB} for both pulses, E_A for pulse A

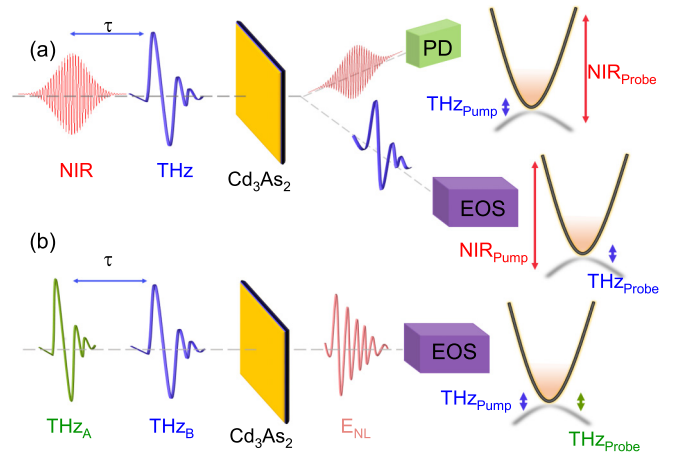


FIG. 1. Investigation of ultrafast carrier dynamics in the Dirac Semimetal Cd_3As_2 . (a) Schematic illustration of the ultrafast THz pump/NIR probe experiment used to measure carrier dynamics following strong-field THz excitation, as a function of the delay between the THz pump and the NIR probe. The transmitted NIR pulse is detected using a single photodiode (PD). The same setup also allows NIR pump/THz probe measurements, in which the transmitted THz pulse is detected using free-space electro-optic sampling (EOS). The right panel shows a representative band diagram near the Γ point, indicating the photon energies of both pump and probe pulses. (b) Schematic of the experimental configuration with the incident THz pulses A and B and the emitted nonlinear field. The signal is detected using EOS. The right panel presents a schematic band diagram highlighting the energy range and interaction of the THz pulses with the band structure.

alone, and E_B for pulse B alone—so we derive the nonlinear THz response given by $E_{NL}(t, \tau) = E_{AB}(t, \tau) - E_A(t, \tau) - E_B(t)$ [39–42].

Figure 2(a) presents the transient transmission of the NIR pulse as a function of the delay between the THz pump and the NIR probe, measured for various applied THz field strengths. The transmitted signal is normalized to the NIR transmission in the absence of the THz field. Notably, even at the lowest THz field strength, the transmitted NIR signal displays a long-lived plateau. This steplike behavior becomes more and more pronounced with increasing the THz pump field strength. The emergent state persists beyond the nanosecond timescale without any observable relaxation. [Fig. 2(a) Inset]. To gain deeper insight into the underlying carrier dynamics, we evaluated the rise and decay times of the transient signal, as summarized in Table I. The rise time was extracted from the temporal position of the maximum, while the relaxation time was obtained by fitting the decay with a single-exponential function. With increasing THz pump field strength, the rise time increases from 2.9 to 4.6 ps, and the relaxation time from

TABLE I. Extracted rise and relaxation times.

THz electric field (MV/cm)	0.17	0.31	0.56	0.72	0.96
Rise time (ps)	2.91	3.18	3.16	3.70	4.60
Decay time (ps)	3.00	4.26	4.57	4.98	5.87

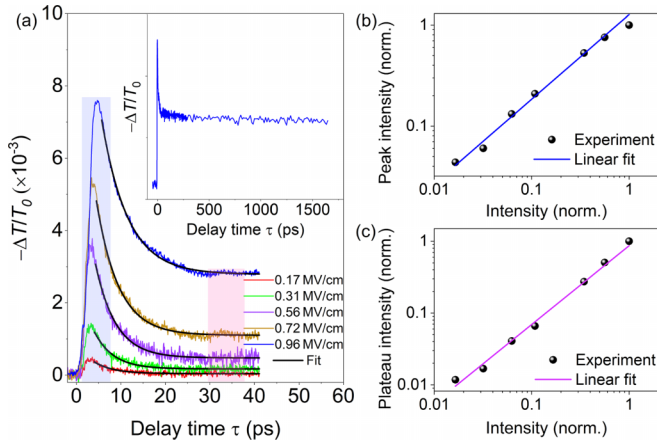


FIG. 2. Strong THz field induced long-lived state in Cd_3As_2 . (a) THz pump field induced amplitude changes of the transmitted NIR probe pulse as a function of pump-probe delay for different THz pump field strengths. The signal is normalized to the NIR amplitude T_0 corresponding to the transmitted NIR probe without THz excitation. A long-lived state is observable even at the lowest THz field strength, while the steplike signal becomes more pronounced as the THz pump field increases. The relaxation time was obtained by fitting the decay with a single-exponential function (solid black curve). Inset: Changes in NIR transmission at long pump-probe delays, extending up to the nanosecond range. Evolution of the peak area (b) and plateau integrated intensity (c) as a function of the pump intensity, where intensity refers to the square of the electric field amplitude. The integrated plateau intensity and peak area, extracted from (a) as indicated by the blue and pink rectangles.

3.0 to 5.8 ps, indicating fast localization into the emergent quantum state, where carriers become trapped on the nanosecond timescale.

To further investigate this phenomenon, we conducted additional experiments at low temperatures ($T = 10$ K, data not shown), and observed no noticeable difference compared to room temperature; both the rise time and the plateau intensity remained unchanged, where the long-lived state was consistently observed. The persistence of the long-lived state

strongly indicates carrier localization into a transiently formed quantum state.

In Fig. 2(b) we plot the peak area as indicated by the blue rectangle in Fig. 2(a) transmitted versus the normalized intensity which is proportional to the electric field square. We applied the same analysis for extracting the integrated plateau intensity versus the pump fluence intensity [Fig. 2(c)] as indicated by the pink rectangle in Fig. 2(a). In our analysis, we used the peak area rather than the signal maximum, as it provides a more robust and reliable measure against experimental uncertainty. Interestingly, both the integrated plateau and the rapid signal rise at zero delay exhibit a linear dependence on the pump fluence, indicating that carrier recombination, saturation effects, or heat accumulation do not dominate the dynamics within the measured fluence range. Thus, the persistence of the plateau on the nanosecond timescale suggests that the system reaches a quasiequilibrium state following the initial excitation, characteristic of trapped carrier populations.

The long-lived state is not observed when using a NIR pump in combination with a THz probe. Figure 3(a) shows the transmitted THz pulse as a function of the pump-probe delay following interband excitation at 800 nm. The observed relaxation behavior reflects the interband relaxation dynamics and is consistent with previously reported results. The initial population of photoexcited carriers at high-energy continuum states is scattered back to the Γ point by intraband cooling within a few hundreds femtoseconds, a process that can be probed by the THz pulse. However, this intraband cooling occurs much faster than the oscillation period of the THz probe pulse (1 ps). Consequently, the observed carrier dynamics on the picosecond timescale primarily reflect interband relaxation processes near the Γ point, consistent with previous reports [28,29]. Notably, the corresponding relaxation time increases only slightly, despite a 30-fold increase in pump fluence from 0.01 to 0.33 mJ/cm^2 . As directly resolved by the Time- and angle-resolved photoemission spectroscopy (Tr-ARPES) experiment, this relatively slow interband relaxation is attributed to a population-inverted state formed at the Dirac cones [24,28,29].

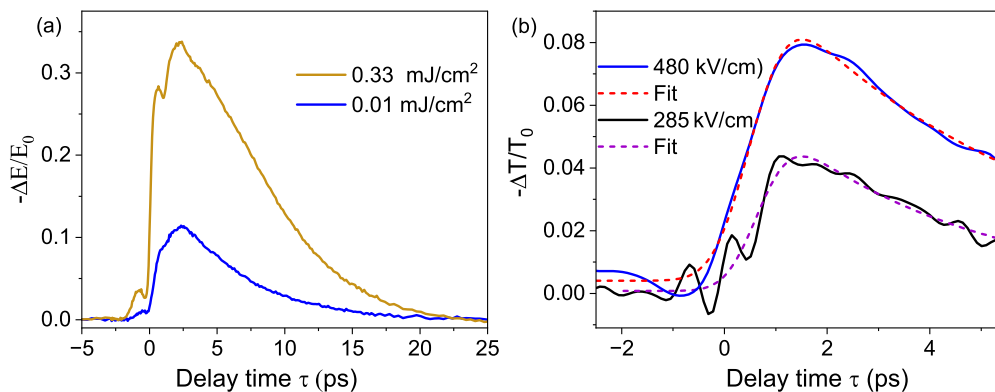


FIG. 3. Carrier relaxation dynamics in Cd_3As_2 near the Γ point for different excitation wavelengths. (a) Pump induced amplitude changes of the transmitted electric field of the terahertz probe pulse after optical pump excitation at 1.55 eV as a function of pump-probe delay for two different pump fluences. The signal is normalized with the THz probe amplitude E_0 , corresponding to the transmitted THz pulse without NIR pump excitation. (b) Evolution of the THz pump/THz probe signal as a function of pump-probe delay for two different electric field strengths. The dashed curve indicates the fitted relaxation wavelength. In contrast to NIR probing, measurements near the Γ point reveals the absence of long-lived states, independent of the excitation wavelength.

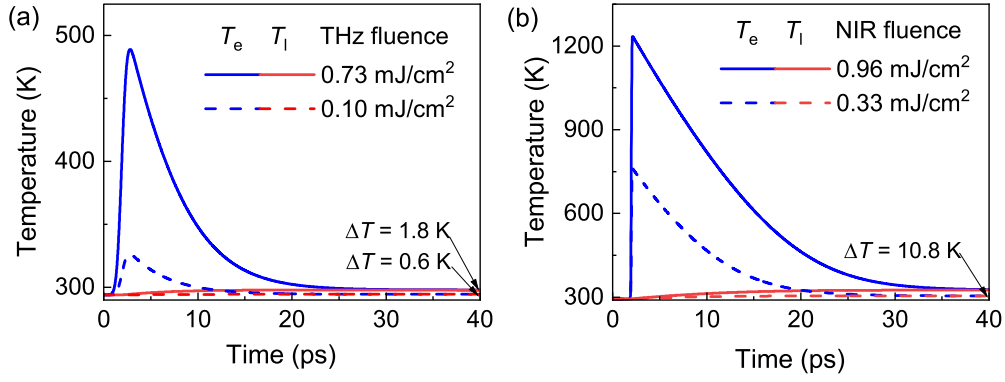


FIG. 4. Two-temperature model. (a) Time evolution of the electronic (T_e , blue dashed curve) and lattice (T_l , red curve) temperatures in a Cd_3As_2 sample under THz excitation at two different fluences. The THz oscillation period is 1 ps, and the shape and duration of the THz pulse were extracted using a THz camera. For the highest fluence, the electronic temperature T_e rises to approximately 500 K before relaxing to the lattice temperature over a timescale of 10–20 ps. The lattice temperature T_l reaches a maximum of about 1.8 K for the highest fluence, indicating negligible heating effects from the THz excitation. (b) Evolution of the electronic T_e and lattice T_l temperatures under two different NIR fluences used in the experiment. The NIR pulse duration is 100 fs. For the highest fluence, the electronic temperature T_e reaches approximately 1200 K.

To gain deeper insight into the carrier dynamics, we performed a THz pump–THz probe experiment. Figure 3(b) presents the time-integrated THz pump/THz probe signal as a function of the delay time τ , for various applied THz pump field strengths obtained from

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

in which E_B is used as a probe pulse. The resulting THz differential signal exhibits a rapid increase in transmission within approximately 0.5 ps, followed by a relaxation process lasting about 4–5 ps. When the THz pump field strength is nearly doubled, the initial fast rise becomes more pronounced, indicating increased absorption due to enhanced carrier generation, while the relaxation time remains in the same range (5 ps). A similar relaxation behavior is observed in the 800 nm–THz probe configuration, consistent with interband relaxation dynamics in this system [24,27–29,42].

It is noteworthy that a similar long-lived state was previously reported using a NIR pump–NIR probe setup in reflection geometry [30,31]. This feature becomes less pronounced when employing a probe with significantly lower photon energy. While the exact origin of this behavior remains inconclusive, it has been attributed to either multiband transitions or the involvement of low-frequency acoustic phonons [30].

The absence of a long-lived state when employing both NIR and THz pumps in combination with a THz probe, along with the observed linear dependence of the plateau intensity on THz pump fluence and the consistent behavior observed even at low temperature, rules out the possibility that the long-lived state arises from pump induced heating. Furthermore, other potential contributions such as low thermal lattice conductivity or the presence of low-frequency acoustic phonon soft modes can be safely excluded. If these effects were responsible, a similar long-lived state would also be expected when using a THz probe in combination with either a THz or NIR pump. Our conclusion is further supported by

simulations based on the two-temperature model. The results from the two-temperature model are presented in Fig. 4. We computed the electron (T_e) and phonon (T_l) temperatures using the methods and material parameters described in Ref. [27]. We observe an increase in the phonon temperature of 1.8 K for the highest THz fluence used in the experiment, compared to 0.6 K for a pump field strength of 170 kV/cm [Fig. 4(a)]. In both cases, the temperature change is too small to explain the distinct change of THz or NIR absorption observed in the experiments. In contrast, NIR pumping leads to strong temperature increases, e.g., of 10 K for a pump fluence of 0.33 mJ/cm² [Fig. 4(b)]. Nonetheless, the experimental data do not show any long-lived contributions neither for NIR nor for THz pumping combined with a THz probe. Therefore, it can safely be excluded that the long-lived state is induced by heating.

The nonlinear currents within the valence and conduction bands are governed by the time-dependent vector potential $A(t)$, which drives electrons via intraband motion across a distance in reciprocal space given by $\Delta k = \frac{2\pi e}{h} A(t)$, according to the semiclassical equation of motion,

$$\frac{h}{2\pi} \frac{dk(t)}{dt} = -eE(t). \quad (2)$$

where $E(t)$ is the applied THz field [43,44].

The interaction of Cd_3As_2 with the strong THz field is highly dependent on the field strength. At low THz field amplitudes, carrier dynamics are primarily dominated by intraband motion, in which electrons are accelerated within the same energy band. However, as the field strength increases, the system enters a nonlinear regime, leading to nonperturbative carrier generation near the Γ point [45]. In this high-field regime, electron-hole pairs are generated through both intraband acceleration and interband transitions, enabling complex ultrafast dynamics. The generation process is irreversible, predominantly due to electron-electron and electron-phonon scattering.

For our experimental THz field amplitudes of around 0.6 MV/cm, and frequency of 1 THz, we can obtain results

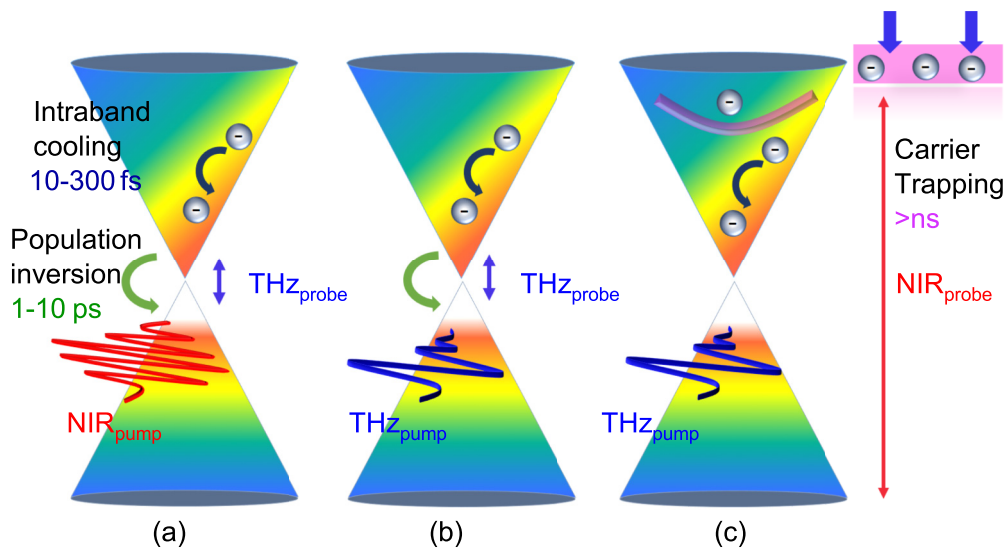


FIG. 5. Carrier relaxation pathways in Cd_3As_2 . (a) Under strong NIR excitation, the carrier dynamics are dominated by Dirac fermion cooling, which occurs on a timescale of a few hundred femtoseconds, followed by population inversion lasting between 1 and 10 ps, depending on the NIR fluence. (b) A strong THz field can induce both intraband and interband carrier motions, leading to similar relaxation pathways as in NIR excitation—namely, Dirac fermion cooling and population inversion within a few picoseconds. Dirac fermion cooling occurs on a much shorter timescale than the oscillation period of the THz probe (1 ps). (c) NIR light with a photon energy of 1.5 eV couples to high-energy electronic states far from the band edges and thus probes a different subset of carriers, which are often trapped in defect states.

in an excursion amplitude of approximately $\Delta k = 8 \text{ nm}^{-1}$ displacing electrons significantly from their initial k -space positions. Such a large electron excursion allows electrons to traverse nearly the entire in-plane Brillouin zone, reaching high-symmetry points such as P or N. The photoexcited carriers then follow two distinct relaxation pathways: (i) A portion of the carriers undergoes intraband relaxation, primarily mediated by electron-phonon and interband scattering processes. This relaxation is evidenced by THz pump–THz probe and NIR pump–THz probe measurements, which reveal carrier relaxation occurring within approximately 4–5 ps [Figs. 5(a) and 5(b)]. (ii) In addition to phonon-mediated relaxation, a fraction of the photoexcited carriers may become trapped in defect states, which can serve as long-lived traps that influence carrier mobility and alter recombination dynamics. This effect is evidenced when using a NIR probe instead of a THz probe. These are primarily localized defect states within the valence band that act as selective hole traps, leaving conduction-band electrons largely unaffected [20]. This energetic and spatial separation suppresses electron-hole recombination, enabling Dirac electrons in the conduction band to maintain their high mobility and extended lifetimes [20]. Alternatively, weakly localized defect states may form near the Dirac node [12], where the low density of states and symmetry protection further inhibit fast recombination processes. Such states may also reside at higher energies within the conduction-band continuum, or just below the conduction-band minimum. This interpretation is supported by two key observations: first, the large electron excursions induced under intense THz fields, during which localized electronic states may form deep levels that are significantly separated in energy from the band edge; and second, the presence of a long-lived signal exclusively in the NIR probe, absent in the THz probe, which indicates

that these defect states are located at relatively high energies and are therefore inaccessible to the low-photon-energy THz probe [Fig. 5(c)]. Remarkably, similar long-lived absorption signals were observed in experiments using NIR excitation and NIR probing [30,31], indicating that the formation of trapped carrier populations is not exclusive to THz-driven dynamics. This consistent behavior across both excitation schemes, despite their fundamentally different mechanisms, strongly supports the conclusion that the observed long-lived states originate from carrier trapping in defect induced localized states, rather than being determined by the specifics of the excitation method.

In summary, our results reveal a relaxation pathway for photoexcited carriers in Cd_3As_2 , involving long-lived trapping in defect induced localized states. This finding highlights the critical influence of native defects on carrier dynamics, particularly through selective trapping that decouples electrons and holes. These insights underscore the urgent need for further experimental studies using advanced spectroscopic techniques, such as X-ray absorption spectroscopy, to investigate defect states that remain inaccessible to scanning tunneling microscopy (STM) or optical methods. In parallel, theoretical investigations are essential to fully elucidate the role of these defects in shaping the electronic band structure and transport properties of Dirac semimetals. Beyond their impact on relaxation dynamics, defect states may also offer functional advantages, such as enhancing NIR absorption, which is particularly beneficial for broadband photodetection [46]. Moreover, the potential of defect engineering to activate and control photocatalytic properties [47–50] opens exciting opportunities for expanding the application landscape of Cd_3As_2 and related materials.

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Data Availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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