

Ultrafast dynamics in 1T-TiS₂ single crystals: From quasi-flat-band-mediated carrier recombination to thermally triggered coherent acoustic phonons

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Carriers in flat bands (FBs) possess high effective masses and are predicted to exhibit unconventional transport properties. The study of the photocarrier dynamics in FBs is of fundamental importance for understanding many-body interactions and the emergence of exotic quantum phenomena. 1T-TiS₂, a layered transition-metal dichalcogenide featuring a quasiflat band near the Fermi level, provides an ideal platform for investigating nonequilibrium carrier dynamics in electronic flat-band systems. Here, we employed transient absorption microscopy to uncover a picosecond, quasi-flat-band-mediated bimolecular recombination of photoexcited electrons and holes confined within the flat-band states in the 80–295 K experimental window. In addition, the launch of coherent acoustic phonons in TiS₂ crystals at 45 GHz is demonstrated, from which sound velocity and Young's modulus are determined to be 4.39 km s⁻¹ and 62 GPa, agreeing well with the prediction from the first-principles calculation. The present study provides microscopic insight into ultrafast energy relaxation and the associated phonon generation in layered materials.

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I. INTRODUCTION

Layered transition-metal dichalcogenides (TMDs) have attracted considerable attention for their diverse electronic structures [1–5], strong light-matter interactions [6,7], and emergent many-body phenomena [8–10]. Their tunable band structures [11–16], van der Waals stacking [17–21], and sensitivity to dimensional confinement [22,23] make them an excellent platform for investigating nonequilibrium dynamics in low-dimensional systems. Among these materials, 1T-TiS₂ has long served as a prototype semimetal, characterized by a quasiflat band near the Fermi level that originates from Ti 3d–S 3p hybridization [24–26]. The existence of such a flat band leads to a high electronic density of states (DOS) and large effective mass, which thereby promotes carrier localization and enables flat-band-mediated carrier recombination and transport phenomena.

Previous studies have shown that TiS₂ exhibits both direct and indirect gaps, featuring a quasiflat band along the Γ –A direction and a smaller indirect valley near the L point [Fig. 3(d)] [27,28]. These quasi-flat-band electronic states dominate the low-energy optical response and give rise to a high density of localized electronic states near the Fermi energy. However, unlike conventional flat-band systems such as magic-angle twisted bilayer graphene [29–32] or kagome lattices [33–36]—which host strongly correlated phases arising from narrow bands—1T-TiS₂ belongs to a class of quasi-flat-band layered TMDs. In this family, including TiSe₂ and TiTe₂, flat bands emerge primarily from transition-metal-chalcogen p – d hybridization rather than moiré or geometric effects.

This intrinsic quasi-flat-band feature results in a heavy effective mass and suppressed band dispersion, which profoundly impact optical and transport behavior, including enhanced carrier localization and potential nonradiative recombination channels.

Several studies on TiSe₂ have revealed ultrafast photoinduced phase transitions and charge density wave melting, while recent investigations with time-resolved angle-resolved photoemission spectroscopy suggest that flat-band states may host nontrivial electron-phonon coupling pathways [37–39]. Similarly, 1T-TiTe₂, which shares a close structural and electronic affinity to TiS₂, has been demonstrated to host nearly dispersionless electronic bands near the Fermi level, resulting in a highly anisotropic electronic structure and strong

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electron-phonon interactions [40,41]. Despite these advances, the ultrafast carrier relaxation dynamics directly connected to quasiflat bands in TiS_2 itself remain largely unexplored. In addition, a challenge lies in the fact that the quasiflat bands in TiS_2 often overlap with other dispersive bands in energy, leading to competing relaxation channels and making it difficult to isolate flat-band-mediated recombination experimentally. Consequently, how photoexcited carriers relax within the flat-band manifold and couple to specific lattice modes on femtosecond timescales remains a critical open question.

Ultrafast pump-probe spectroscopy is a powerful technique for uncovering nonequilibrium carrier and phonon dynamics in layered materials, such as TaS_2 [42,43], MoS_2 [44–50], and MoSe_2 [51–55], which enables direct observation of photoexcited carrier relaxation across different probe wavelengths, providing valuable insight into electronic and lattice interactions in real time. In this work, we investigate the ultrafast carrier relaxation and lattice dynamics of high-quality 1T- TiS_2 single crystals by means of transient absorption microscopy (TAM). Fluence- and temperature-dependent measurements reveal a quasi-flat-band-mediated bimolecular recombination of photoexcited carriers within flat-band states in the 80–295 K experimental window. In addition, pump-launched coherent acoustic phonons (CAPs) at ~ 45 GHz were demonstrated, from which the sound velocity and Young's modulus are determined to be 4.39 km s^{-1} and 62 GPa, respectively, agreeing well with density functional theory (DFT) calculations. These results are consistent with carrier relaxation in 1T- TiS_2 being dominated by quasi-flat-band electronic recombination, while the ensuing lattice dynamics—including the 45 GHz coherent acoustic phonon and the slower thermal-diffusion-related decay observed at long delays—can be understood as consequences of the impulsive lattice heating produced during this ultrafast carrier relaxation. Thus, 1T- TiS_2 emerges as a benchmark system for exploring both flat-band-mediated ultrafast carrier and thermally triggered coherent phonon dynamics, offering insight into the interplay between correlated electronic states and lattice excitations in layered materials.

II. METHODS

Sample growth. High-quality 1T- TiS_2 single crystals were grown using the iodine-assisted chemical vapor transport method. High-purity titanium powder (99.99%) and sulfur powder (99.99%) were mixed in a stoichiometric molar ratio of 1:2 and sealed in evacuated quartz ampoules together with a small amount of iodine serving as the transport agent. The sealed tubes were placed in a two-zone furnace, where the source materials were heated to 700°C in the hot zone within 24 h and maintained at this temperature for 200 h. The TiS_2 single crystals were synthesized at the cold zone via vapor-phase transport, yielding platelike crystals with metallic luster suitable for optical measurements [56].

Pump probe. Two complementary ultrafast pump-probe configurations were used in this study. First, broadband femtosecond transient reflectivity measurements were performed

using a time-resolved TAM system. The system was driven by a high-repetition-rate femtosecond laser (Light Conversion) with a center wavelength of 1030 nm, a pulse duration of 190 fs, and a repetition rate of 100 kHz. The laser output was split into pump and probe beams using a beam splitter. The pump beam was directed into an optical parametric amplifier (TOPAS, Light Conversion) to generate 400 nm (3.10 eV) excitation pulses. The pump beam was modulated by a mechanical chopper operating at 200 Hz and guided to the sample by a series of reflective mirrors. The probe beam at the fundamental wavelength of 1030 nm passed through a motorized delay stage to control the temporal delay with respect to the pump pulse. The fundamental 1030 nm beam was focused into an yttrium aluminum garnet crystal to generate a supercontinuum white-light probe (550–900 nm). The probe beam was combined coaxially with the pump beam using a dichroic mirror. Both beams were focused onto the sample surface through a high-numerical-aperture microscope objective, enabling spatial overlap and micron-scale excitation. The reflected probe light was collected by the same objective and directed into a detection system consisting of an HRS-300 spectrometer and a Pixis-100BRX charge-coupled device detector (Princeton Instruments). The transient reflectivity change ($\Delta R/R_0$) was recorded as a function of probe wavelength and pump-probe delay, allowing broadband time-resolved mapping of the carrier dynamics with subpicosecond temporal resolution and micrometer spatial resolution. Temperature-dependent measurements were performed by mounting the sample in a cryostat, providing temperature control from 80 to 295 K. Second, two-color optical pump-optical probe microreflectivity measurements were carried out at room temperature using a separate femtosecond laser system. The laser source is a Ti:sapphire regenerative amplifier operating at a repetition rate of 1 kHz, delivering pulses with a duration of 120 fs and a center wavelength of 780 nm. The output beam was split into pump and probe paths using a beam splitter. The pump beam was frequency doubled in a β -barium borate crystal to generate 390 nm (3.18 eV) pulses. A short-pass filter was used to remove any residual 780 nm fundamental light. The 390 nm pump beam was modulated by a mechanical chopper operating at 500 Hz and directed to the sample through a series of mirrors. The probe beam at 780 nm passed through a motorized delay stage to control the temporal delay relative to the pump pulse. Both pump and probe beams were collinearly focused onto the same spot on the sample surface through a microscope objective. After reflection from the sample, the beams were collected by the same objective and directed through a Glan-Taylor prism for polarization selection. A bandpass filter placed in front of the detector was used to block the residual 390 nm pump light, ensuring that only the reflected 780 nm probe signal was detected. The transient reflectivity signal ($\Delta R/R_0$) was recorded using a silicon photodiode connected to a lock-in amplifier referenced to the chopper frequency, enabling phase-sensitive detection with high sensitivity. In both configurations, the pump and probe beams were arranged with mutually orthogonal polarization to suppress coherent artifacts in the transient reflectivity signal.

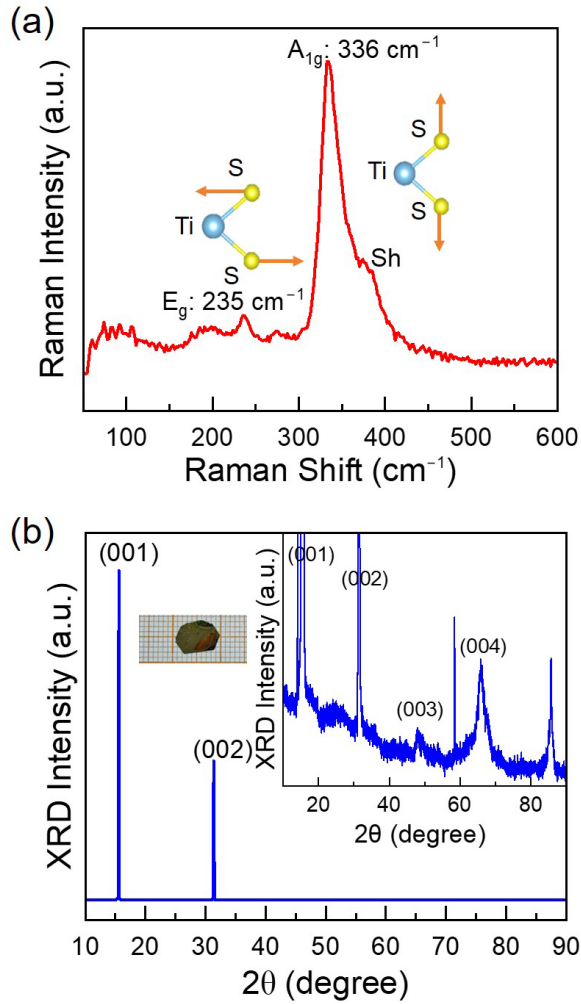


FIG. 1. (a) Raman shift of 1T-TiS₂, showing the characteristic E_g mode at 235 cm⁻¹ and A_{1g} mode at 336 cm⁻¹, together with a weak shoulder near 379 cm⁻¹. The insets illustrate the corresponding in-plane (E_g) and out-of-plane (A_{1g}) modes. (b) XRD pattern of TiS₂ crystal, exhibiting sharp (00l) diffraction up to (004). The inset shows an optical photograph of the as-grown TiS₂ crystal with a metallic luster.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the room-temperature Raman spectrum of the 1T-TiS₂ single crystal. Two prominent Raman-active modes are observed at approximately 235 and 336 cm⁻¹, corresponding to the in-plane E_g mode and the out-of-plane A_{1g} mode, respectively, consistent with the characteristic vibrational fingerprints of the 1T phase. A weak shoulder feature near 379 cm⁻¹ is also detected, which has been reported previously in layered TiS₂ and is commonly attributed to higher-order or defect-related phonon scattering [57,58]. The crystalline quality and phase purity were further examined by x-ray diffraction (XRD), as shown in Fig. 1(b). Only sharp (00l) diffraction peaks up to the (004) reflection are observed, indicating a highly oriented layered structure with the *c* axis perpendicular to the crystal surface. The absence of impurity-related peaks confirms the high phase purity of the as-grown crystals [59]. An optical photograph of a representative TiS₂

crystal is shown in the inset of Fig. 1(b), displaying a uniform surface and metallic luster.

We begin by investigating the ultrafast carrier dynamics of 1T-TiS₂ using broadband transient reflectivity measurements on freshly cleaved single-crystal surfaces. Figure 2(a) presents the wavelength-resolved transient reflectivity spectra ($\Delta R/R_0$) recorded within the first 10 ps following 400 nm photoexcitation at a pump fluence of 0.737 mJ/cm². A pronounced wavelength dependence is observed over the probe range from 550 to 900 nm, indicating that the transient optical response sensitively depends on the probed electronic states.

The pronounced spectral dispersion observed here qualitatively reflects the differing sensitivity of each probe wavelength to the various stages of the underlying relaxation cascade, including ultrafast electron-electron and electron-phonon thermalization, flat-band-mediated bimolecular electron-hole recombination, CAPs generation, and slower lattice thermal diffusion. Microscopically, this differing sensitivity arises because each probe wavelength samples electronic transitions of a specific energy: Any pair of valence and conduction states whose energy difference matches the probe photon energy can in principle contribute to the signal, regardless of where these states lie in the Brillouin zone, with the relative weights set by the joint density of states and the optical matrix elements. Accordingly, near 630 nm the transient reflectivity is dominated by a photoinduced absorption that is most sensitive to the subsequent bimolecular electron-hole recombination stage, whereas near 710 nm a positive photobleaching contribution that tracks the subpicosecond electron-phonon energy transfer becomes the dominant feature; the 780 nm trace lies in between. To further resolve these dynamics, transient traces were extracted at three representative probe wavelengths of 630, 710, and 780 nm, as shown in Fig. 2(b). At 630 and 780 nm, the transient signals exhibit clear biexponential decay behavior, consisting of a fast relaxation component on a picosecond timescale followed by a slower decay extending to longer delays. To accurately extract the ultrafast component, the analysis in Fig. 2 focuses on the first 10 ps after photoexcitation. We also present the full decay dynamics over a 2000 ps time window (see the Appendix), where all probe wavelengths consistently exhibit a long-lived relaxation component attributed to lattice thermal diffusion.

The transient reflectivity signals were quantitatively fitted using a biexponential response function convolved with the instrument response [60]:

$$\frac{\Delta R}{R_0} = \sum_{i=1,2} A_i \exp\left(\frac{-t}{\tau_i}\right) \times \text{erfc}\left(\frac{\omega}{\tau_i} - \frac{t}{2\omega}\right), \quad (1)$$

where A_i and τ_i denote the amplitudes and lifetimes of the fast (τ_1) and slow (τ_2) relaxation components, respectively. ω denotes the pulse width and $\text{erfc}(x) = 1 - \text{erf}(x)$ is a complementary error function. The solid lines in Figs. 2(b), 2(d), and 2(f) represent the fitting results, demonstrating excellent agreement with the experimental data for both pump fluence- and temperature-dependent measurements.

Notably, at the probe wavelength of 710 nm, an additional ultrafast component with a characteristic lifetime of approximately 0.18 ps appears at early delay times, producing a

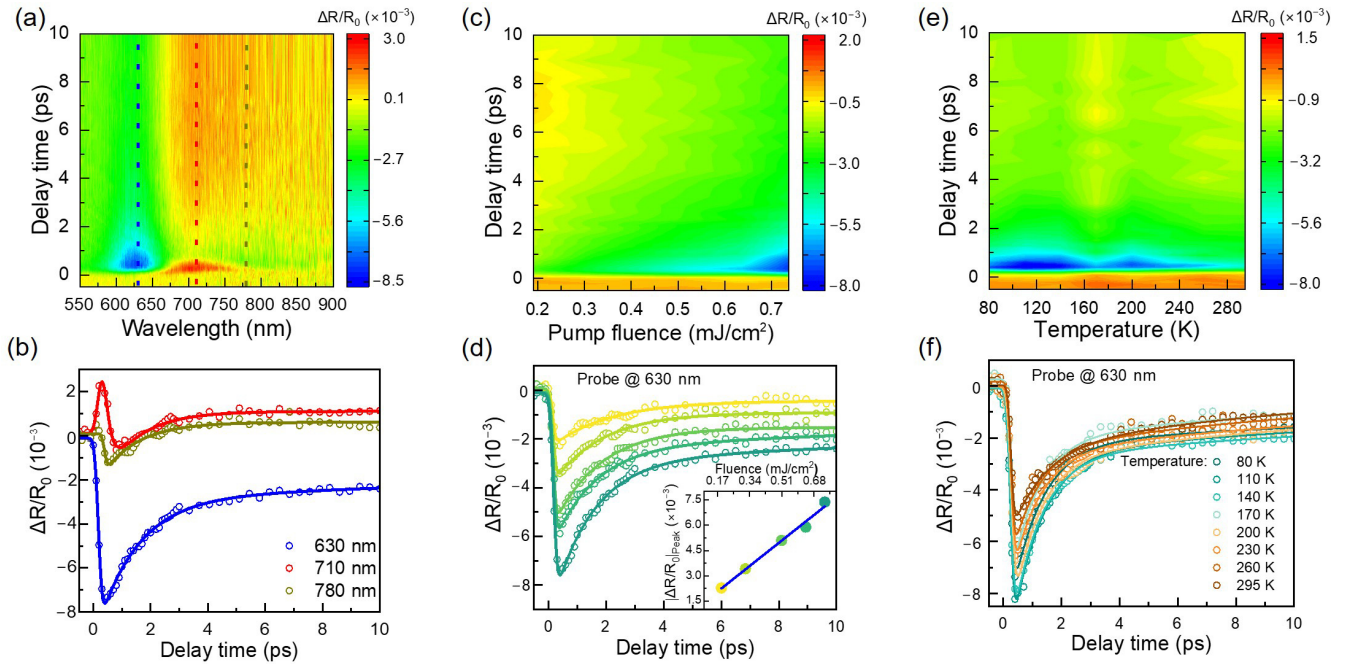


FIG. 2. (a) Broadband transient reflectivity spectra ($\Delta R/R_0$) of 1T-TiS₂ measured under 400 nm excitation with a pump fluence of 0.737 mJ/cm², collected within 10 ps over the probe wavelength range of 550–900 nm. The blue, red, and brown dashed lines mark the representative probe wavelengths of 630, 710, and 780 nm, respectively. (b) Time-resolved transient traces at 630, 710, and 780 nm extracted from panel (a), together with biexponential fits to the data. (c) Pump-fluence-dependent transient maps at a probe wavelength of 630 nm. (d) Transient reflectivity traces obtained at five different pump fluences; the inset shows the corresponding absolute peak amplitudes with respect to pump fluence. (e) Temperature-dependent transient maps recorded at a fixed pump fluence of 0.509 mJ/cm². (f) Transient reflectivity traces at eight representative temperatures together with biexponential fitting curves. In all panels, open circles denote the experimental $\Delta R/R_0$ data and solid lines are biexponential fits.

transient positive feature that is most clearly resolved at this wavelength [red curve in Fig. 2(b)]. We note that this 0.18 ps component is not unique to the 710 nm probe wavelength: In the broadband transient absorption data of Fig. 2(a), the fast positive feature is resolved continuously across the entire 650–780 nm window, and the 710 nm trace was selected in Fig. 2(b) because, near this probe wavelength, the positive photobleaching contribution associated with the ultrafast carrier thermalization becomes relatively pronounced, while the slower negative photoinduced-absorption background arising from carrier recombination is comparatively weak. The combined spectral contrast therefore makes this fast positive component most clearly visible at 710 nm. The DFT calculated band structure of Fig. 3(d) shows that multiple pairs of valence and conduction states with the appropriate energy difference are available across the Brillouin zone throughout the 1.6–1.9 eV probe range, which is consistent with the picture introduced above, in which the 710 nm probe is most pronounced in the subpicosecond electron-phonon energy-transfer stage of the relaxation cascade rather than reflecting a single, isolated momentum-space transition.

We assign the 0.18 ps component to an effective electron-phonon coupling time. Because the transient signal across the 650–780 nm probe window reflects a weighted average of electron-electron scattering and electron-phonon coupling, this effective time is shorter than the electron-phonon coupling times commonly reported for comparable semiconductors. The assignment is further supported on phys-

ical grounds by a self-consistency argument: The subsequent CAP and the long-lived lattice heating both require that a sizeable fraction of the absorbed photon energy be deposited into the lattice on a subpicosecond timescale, whereas the bimolecular electron-hole recombination at τ_1 only releases an energy on the order of the small (~ 0.2 eV) direct gap per recombination event, which is not enough to account for the observed lattice excitation; an ultrafast electron-phonon channel is therefore needed to transfer the hot-carrier excess energy to the lattice, and the 0.18 ps component is the natural candidate. A more detailed microscopic decomposition of this subpicosecond thermalization process is beyond the scope of this work. In the following, we focus on the subsequent quasi-flat-band-mediated bimolecular recombination and the subsequent CAP generation.

Figures 2(c) and 2(d) display the pump-fluence-dependent transient reflectivity measured at a representative probe wavelength of 630 nm. The peak amplitude of $\Delta R/R_0$ increases linearly with pump fluence, reflecting the increasing density of photoexcited carriers. Importantly, the temporal profiles at different excitation densities exhibit nearly identical decay shapes, indicating that the underlying relaxation mechanism remains the same across the investigated fluence range. The near-linear dependence of the peak amplitude on pump fluence [inset of Fig. 2(d)] confirms that the measurements are performed within the linear excitation regime.

Temperature-dependent measurements further demonstrate the robustness of the relaxation dynamics. Figure 2(e) shows

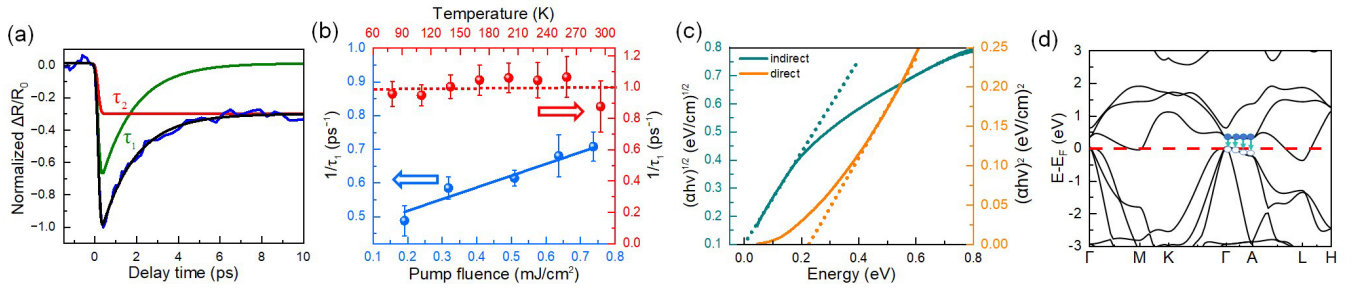


FIG. 3. (a) Decomposition of the transient reflectivity signal at a probe wavelength of 630 nm into individual exponential components obtained from biexponential fitting, showing the respective contributions of the fast (τ_1) and slow (τ_2) relaxation channels. (b) Fluence and temperature dependence of the inverse fast relaxation time ($1/\tau_1$), showing linear fluence scaling and negligible temperature variation. (c) Tauc plots derived from the absorption spectrum: direct-allowed form $(\alpha h\nu)^2$ vs $h\nu$ giving a direct optical gap of about 0.22 eV, and indirect-allowed form $(\alpha h\nu)^{1/2}$ vs $h\nu$ whose low-energy tangent extrapolates essentially to zero energy gap, consistent with the semimetallic band overlap predicted by DFT. (d) DFT calculated electronic band structure of 1T-TiS₂. The arrows in the figure indicate the flat-band-mediated recombination between electron and hole in the flat band.

the transient reflectivity maps recorded at a fixed pump fluence of 0.509 mJ/cm² over a wide temperature range. Although the signal amplitude gradually varies with temperature, the overall relaxation dynamics remain unchanged. As shown in Fig. 2(f), all transient traces can be well described by the same biexponential model from 80 to 295 K, yielding a fast and a slow relaxation component.

The transient reflectivity signals were decomposed into individual exponential components, as shown in Fig. 3(a), where the relative contributions of the fast and slow relaxation channels are clearly resolved, enabling an unambiguous identification of the fast decay component associated with τ_1 . Figure 3(b) presents the inverse fast time $1/\tau_1$ with respect to pump fluence (blue line) and temperature (red line). It is evident that the magnitude of $1/\tau_1$ shows a linear dependence on pump fluence, while exhibiting almost no dependence on temperature. Such behavior is a hallmark of bimolecular recombination driven by carrier-carrier interactions, where the recombination rate scales with the product of electron and hole densities. Specifically, following photoexcitation, the recombination rate of electrons and holes can be expressed as $dn/dt = -Bnp$, where n and p are, respectively, the photoexcited electron and hole densities, and B is the bimolecular recombination coefficient. Under above-bandgap excitation and charge neutrality, $n \approx p$, yielding $dn/dt = -Bn^2$. By matching the initial decay rate of the analytic bimolecular solution $n(t) = n_0/(1 + Bn_0t)$ to that of an effective single-exponential model $n(t) \approx n_0 \exp(-t/\tau_1)$ at $t = 0$, one obtains $1/\tau_1 = Bn_0 \propto$ pump fluence, which directly accounts for the observed linear scaling of $1/\tau_1$ with pump fluence. The full step-by-step derivation, together with the corresponding Shockley-Read-Hall (SRH) and three-particle Auger rate-equation forms used to rule out those channels are provided in the Appendix. Additional two-color pump-probe measurements using 390 nm excitation and 780 nm probing (see the Appendix) further confirm the linear scaling of $1/\tau_1$ with pump fluence.

The temperature independence of $1/\tau_1$ between 80 and 295 K, together with its linear-in-fluence scaling, provides a qualitative constraint on the dominant recombination channel within the experimental window. A defect-dominated monomolecular (SRH) channel would give a fluence-

independent $1/\tau_1$, and a three-particle phonon-assisted Auger channel would produce a superlinear rise of $1/\tau_1$ at the highest fluences; neither behavior is observed in the broadband TAM data of Fig. 3(b) or in the lock-in-detected two-color pump-probe data (see the Appendix). Moreover, if a phonon-assisted indirect channel between the Γ -A quasi-flat-band manifold and the L -valley conduction-band minimum dominated the decay, the population of the mediating phonon would increase between 80 and 295 K and produce a measurable temperature dependence of $1/\tau_1$, which is not seen. We further note that the weak Raman shoulder near 379 cm⁻¹ has recently been attributed to a combination of intrinsic anharmonic two-phonon scattering and a defect-induced A_{1g} -mode splitting by excess interlayer Ti atoms [57,58]. The clearly linear scaling of $1/\tau_1$ with pump fluence is characteristic of bimolecular recombination rather than of a fluence-independent monomolecular SRH process. It therefore rules out a defect-dominated decay as the dominant mechanism in the carrier-density range studied here, although a small residual SRH contribution from a low density of Ti-interstitial defects cannot be entirely excluded. Taken together, the observed dynamics are most naturally interpreted as flat-band-mediated bimolecular recombination within the Γ -A manifold, within the 80–295 K experimental window.

To correlate the observed dynamics with the electronic structure, complementary optical absorption and DFT analyses were performed. Figure 3(c) shows the Tauc plot of both the direct allowed $[(\alpha h\nu)^2$ versus $h\nu$] and the indirect allowed $[(\alpha h\nu)^{1/2}$ versus $h\nu$] derived from absorption spectrum of 1T-TiS₂. Tangent extrapolation of the direct-allowed plot yields a direct optical gap of $E_g^{\text{dir}} \approx 0.22$ eV, in close agreement with the small band separation between the quasiflat valence and conduction bands along Γ -A in the DFT band structure of Fig. 3(d). Tangent extrapolation of the low-energy linear region of the indirect-allowed plot extrapolates essentially to zero bandgap, consistent with the semimetallic band overlap at the L point predicted by DFT, where the conduction-band minimum lies at or just below the Γ -A valence-band maximum. The flat-band-mediated bimolecular recombination claim in the present work is associated specifically with this small direct transition near Γ -A. DFT calculations [Fig. 3(d)] reveal a quasiflat conduction band along the Γ -A direction, coexisting with a lower-energy

indirect valley near the L point, consistent with previous reports [61]. The optimized 1T-TiS₂ crystal structure, together with side and top views, is presented in Fig. 8(a), while the projected electronic band structures highlighting the respective contributions from Ti and S atoms are displayed in Figs. 8(b) and 8(c) (see the Appendix). These calculations confirm the layered 1T-phase symmetry and demonstrate that the quasiflat band near the Fermi level is predominantly derived from Ti 3d states, providing a microscopic basis for the enhanced carrier localization and flat-band-mediated recombination dynamics discussed above.

Although 1T-TiS₂ is a semimetal with the conduction-band minimum near the L point, the presence of quasiflat bands near Γ enables a small-direct-gap electron-hole recombination channel near Γ -A. Photoexcitation generates a high carrier density near Γ -A, where the large density of states and heavy effective mass associated with the flat band strongly enhance carrier localization and carrier-carrier interactions. These features suppress momentum relaxation and support flat-band-mediated bimolecular recombination within the 80–295 K temperature range, consistent with the experimentally observed fast decay component $\tau_1 < 2$ ps.

Building on the carrier-recombination analysis above, we next examined the coherent lattice response of 1T-TiS₂. Here, we used the two-color pump-probe microreflectivity configuration with lock-in detection; this phase-sensitive detection scheme provides a substantially higher signal-to-noise ratio than the white-light TAM setup on the weak $\Delta R/R_0 \approx 10^{-4}$ oscillatory signal, allowing the long-lived coherent acoustic component and its damping to be resolved cleanly without averaging over many supercontinuum spectra.

As shown in Fig. 4(a), periodic oscillations are superimposed on the long-time transient reflectivity signal measured at 780 nm under 390 nm excitation, persisting up to ~ 40 ps. The time-domain signal is well fitted by an exponentially decaying background combined with a damped oscillatory term [62]:

$$\frac{\Delta R}{R_0} = A_1 \exp\left(\frac{-t}{\tau_1}\right) + A_p \exp\left(\frac{-t}{\tau_p}\right) \cos(2\pi \nu_p t + \varphi) + C, \quad (2)$$

where the first term represents fast carrier recombination and the second term describes the coherent phonon oscillation with frequency ν_p , which decays with a damping time τ_p .

The extracted oscillation frequency is approximately 45.0 GHz, obtained directly from time-domain fitting of the transient reflectivity traces, corresponding to a coherent longitudinal acoustic (LA) phonon mode propagating along the out-of-plane direction.

This assignment is strongly supported when alternative origins are considered in detail. The observed oscillation is incompatible with a confined standing-wave acoustic mode, since previous studies on multilayer transition-metal dichalcogenides have shown that such standing-wave coherent acoustic modes arise only in finite-thickness few-layer structures where the phonon frequency is quantized by the sample thickness [63]. In contrast, the present specimens are bulk single crystals whose dimensions are far beyond the few-layer

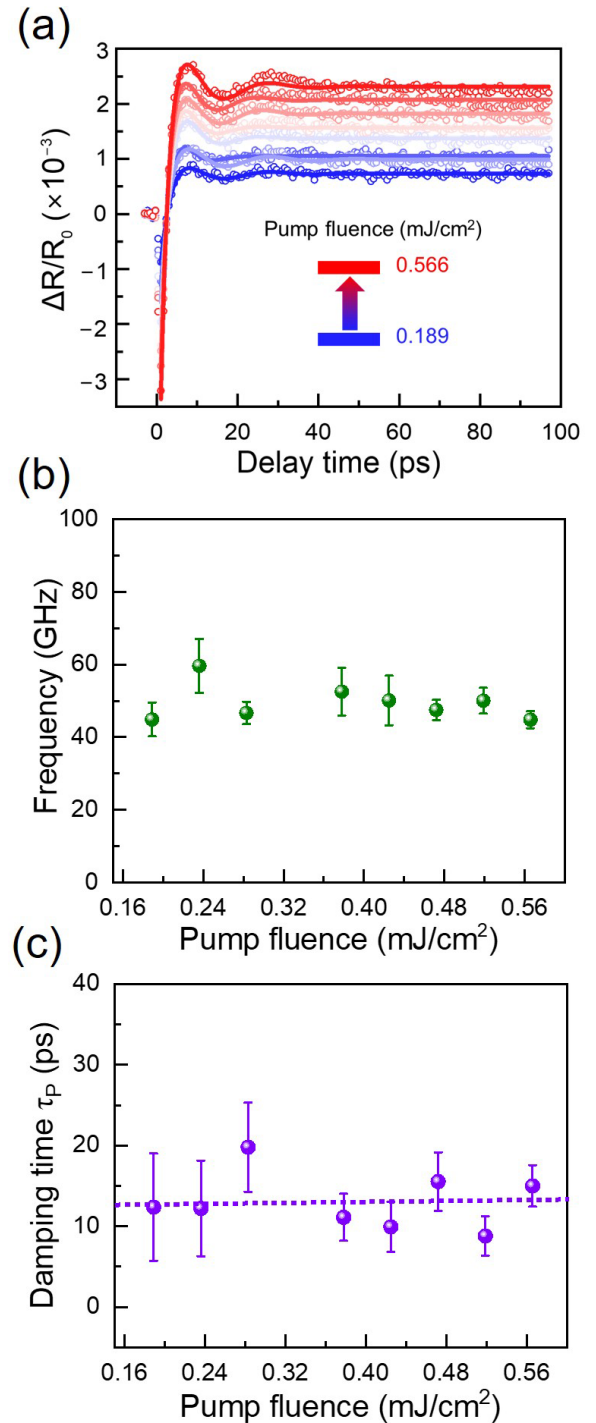


FIG. 4. (a) Transient reflectivity ($\Delta R/R_0$) of 1T-TiS₂ measured at 780 nm under 390 nm excitation with various fluences, exhibiting periodic oscillations superimposed on an exponential background. The curves are color coded by pump fluence, from blue (lowest, 0.189 mJ/cm^2) to red (highest, 0.566 mJ/cm^2). Open circles are the experimental data and solid lines are single-exponential-plus-damped-oscillation fits. (b) Fitted oscillation frequency and (c) fitted damping time τ_p of the 45 GHz coherent acoustic oscillation with respect to pump fluence. In panels (b) and (c), symbols denote the fitted parameters, and the error bars represent their standard errors, with the dashed line in panel (c) indicating the mean value.

regime required for acoustic confinement. The oscillation is therefore more naturally understood as a propagating bulk LA phonon detected through time-domain Brillouin scattering (TDBS), rather than as a thickness-confined standing-wave mode. An interlayer breathing optical-phonon origin can likewise be excluded. Layer-dependent coherent phonon studies in layered chalcogenides such as PdSe₂ have demonstrated that interlayer breathing optical modes typically appear in the low-wave-number Raman range of several to tens of cm⁻¹, corresponding to frequencies in the subterahertz to terahertz regime [64]. By comparison, the present 45 GHz oscillation corresponds to only ~ 1.5 cm⁻¹, more than one order of magnitude lower than the characteristic frequency range of coherent interlayer breathing modes, thereby ruling out an optical-phonon origin. Further support for the LA-TDBS interpretation comes from the fluence dependence of the oscillatory response. As shown in Fig. 4(b), the oscillation frequency remains unchanged over the explored excitation range, indicating that the mode frequency is governed by the intrinsic LA phonon dispersion rather than by carrier-density-induced nonlinear frequency renormalization or strong photoinduced lattice softening. Consistently, the fitted damping time $\tau_p \approx 12$ ps also remains fluence independent within experimental uncertainty [Fig. 4(c)]. The absence of measurable fluence dependence in either the oscillation frequency or the damping behavior demonstrates that the coherent phonon evolves entirely within the linear excitation regime. Under these conditions, the decay of the oscillatory amplitude is most naturally attributed to intrinsic phonon dephasing processes, predominantly anharmonic phonon-phonon scattering of the LA mode itself, rather than to carrier-density-dependent damping channels. Taken together, these observations consistently identify the 45 GHz oscillation as a propagating coherent LA phonon detected via TDBS.

For a LA phonon propagating along the *c* axis, the sound velocity *V* is related to the oscillation frequency *f*, probe wavelength λ , and refractive index *n* via the relation $V = \lambda f / (2n)$. Using $f = 45$ GHz, $n = 4.0$ [65], we obtain an out-of-plane sound velocity of $V = 4.39 \times 10^3$ m s⁻¹. The corresponding out-of-plane Young's modulus Y_Z is then estimated from $Y_Z = \rho V^2$, where $\rho = 3.22 \times 10^3$ kg m⁻³ is the mass density of TiS₂. Substituting the above parameters yields $Y_Z = 62$ GPa, in excellent agreement with DFT calculations ($Y_Z^{\text{DFT}} = 70.4$ GPa). This quantitative consistency validates the assignment of the oscillatory component to coherent LA phonons and demonstrates the reliability of extracting elastic properties from ultrafast optical measurements. Complementary first-principles calculations of the elastic constants, including the anisotropic Young's moduli, were performed to support this analysis (see the Appendix).

At longer delay times, transient reflectivity signals measured up to 3000 ps reveal a slow relaxation component with a characteristic lifetime $\tau_2 \approx 125$ ps [Fig. 5(a)]. The extracted τ_2 values show no dependence on pump fluence [Fig. 5(b)], indicating that this relaxation channel is governed by lattice thermal diffusion rather than carrier-density-dependent recombination. To verify this assignment quantitatively, we estimate the characteristic thermal diffusion time using the measured optical penetration depth and reported thermal pa-

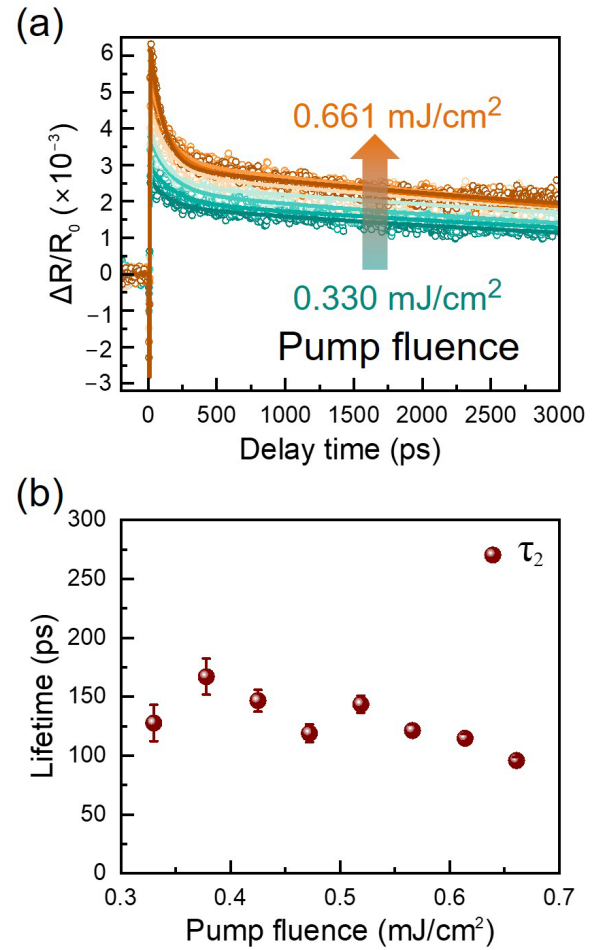


FIG. 5. (a) Long-delay (0–3000 ps) transient reflectivity ($\Delta R/R_0$) traces of 1T-TiS₂ under 390 nm excitation, showing the slow relaxation component associated with lattice thermal diffusion ($\tau_2 \approx 125$ ps). The traces are color coded by pump fluence, from teal (lowest, 0.330 mJ cm⁻²) to orange (highest, 0.661 mJ cm⁻²); the arrow indicates increasing pump fluence. Open circles are the experimental data and solid lines are biexponential fits. (b) Pump-fluence dependence of the slow lifetime τ_2 . Symbols denote the fitted τ_2 and the error bars represent their standard errors.

rameters of 1T-TiS₂. At the pump wavelength of $\lambda = 390$ nm, the extinction coefficient is $\kappa = 1.9$ [66], yielding an optical penetration depth $L = \lambda / (4\pi\kappa) \approx 16.3$ nm. The photocarriers and the associated lattice heating are confined within a near-surface layer of approximately 16.3 nm. Thus, the thermal diffusivity α is given by $\alpha = K / (\rho C_p)$, where the out-of-plane thermal conductivity $K \approx 4.2$ W m⁻¹ K⁻¹, and specific heat $C_p \approx 6.0 \times 10^2$ J kg⁻¹ K⁻¹ (derived from $C_p \approx 67.34$ J mol⁻¹ K⁻¹ with $M = 112$ g mol⁻¹) [67,68]. Substituting these values yields $\alpha \approx 2.17 \times 10^{-6}$ m² s⁻¹.

For one-dimensional heat flow across the optically excited layer, the characteristic diffusion time can be approximated as $t = L^2 / \alpha$, which gives $t \approx 123$ ps. This value agrees remarkably well with the experimentally extracted $\tau_2 \approx 125$ ps, confirming that the long-lived decay component originates from lattice thermal diffusion rather than slow electronic processes or defect trapping. Importantly, the observation of this

well-defined thermal diffusion timescale provides a crucial link to the incomplete recovery of the transient reflectivity signal even at a delay time of 3000 ps. Once the heat has diffused across the initially excited layer on a ~ 125 ps timescale, the subsequent relaxation is governed by much slower heat dissipation from the photoexcited region into the surrounding bulk and external environment. The persistent offset at long delays therefore directly reflects residual lattice heating and gradual thermal coupling between the optically excited volume and its surroundings. Such multistage thermal relaxation behavior is characteristic of layered materials with low cross-plane thermal conductivity, where interlayer heat transport and interfacial thermal exchange constitute the rate-limiting processes.

IV. CONCLUSIONS

In summary, we have systematically investigated the ultrafast carrier and lattice dynamics in layered semimetallic 1T-TiS₂ using broadband and two-color pump-probe reflectivity spectroscopy. The measurements reveal that photoexcited carriers rapidly relax into quasiflat conduction-band states along the Γ -A direction, where the high density of states and carrier localization are consistent with a quasi-flat-band-mediated bimolecular electron-hole recombination with a characteristic timescale of ~ 1.5 ps within the 80–295 K experimental window, evidenced by the linear fluence dependence and negligible temperature dependence of the recombination rate. Concurrently, ultrafast electronic relaxation induces coherent longitudinal acoustic phonons, from which an out-of-plane sound velocity of 4.39×10^3 m s⁻¹ and a Young's modulus of ~ 62 GPa are extracted, in good agreement with DFT calculations. At longer delay times, a slow relaxation component with a lifetime of ~ 125 ps is quantitatively identified as thermal diffusion within the optically excited surface layer, which in turn leads to a much slower and incomplete recovery of the transient reflectivity signal up to 3000 ps, reflecting residual lattice heating and gradual thermal coupling between the photoexcited region and the surrounding environment. Together, these results outline a complete ultrafast energy-relaxation pathway in 1T-TiS₂, in which the bimolecular electron-hole recombination is consistent with a quasi-flat-band-mediated process within the explored 80–295 K window, while the longitudinal acoustic phonons and the multistage lattice heat dissipation can be accounted for as consequences of the impulsive lattice heating that accompanies this ultrafast carrier relaxation.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are available from the corresponding authors upon reasonable request.

APPENDIX

This appendix contains information about supplementary experimental data and analysis including long-time transient reflectivity, two-color pump-probe study, carrier rate-equation analysis, and DFT calculation of band structure and Young's modulus.

1. Long-time transient reflectivity measurements

To fully capture the relaxation dynamics of photoexcited carriers in 1T-TiS₂, transient reflectivity ($\Delta R/R_0$) traces were recorded over an extended time window of up to 2000 ps at room temperature (295 K) using probe wavelengths of 630, 710, and 780 nm under 400 nm excitation with a pump fluence of 0.737 mJ cm⁻². Figures 6(a)–6(c) show the full-time window traces for the three wavelengths. In all cases, a pronounced long-lived decay is observed, corresponding to the slow thermal relaxation of the lattice following electron-phonon interactions. To accurately extract the ultrafast component associated with electron-hole recombination (τ_1), fitting was restricted to the first 10 ps of the transient signal. The full 2000 ps traces here confirm the existence of the slow thermal process and justify the short-time analysis for characterizing fast electronic relaxation.

2. Two-color pump-probe results

To further verify the fluence dependence of ultrafast carrier relaxation, additional two-color pump-probe measurements were performed using 390 nm excitation and 780 nm probing. Figure 7(a) displays the transient reflectivity traces under several pump fluences, where each trace is fitted with a bi-exponential function to extract the fast decay component τ_1 . The inset confirms that the peak amplitude of $\Delta R/R_0$ scales linearly with pump fluence, indicating the system operates within the linear excitation regime and excluding saturation effects. Figure 7(b) shows the inverse fast decay time ($1/\tau_1$) versus pump fluence. The observed linear relationship in Fig. 7(b), together with the linear scaling of $1/\tau_1$ with pump fluence in the broadband transient absorption data of Fig. 3(b), confirms that the fast relaxation is dominated by a two-particle bimolecular electron-hole recombination process within the explored fluence range. In addition, the absence of any superlinear acceleration of $1/\tau_1$ at the highest fluences excludes a dominant three-particle phonon-assisted Auger channel, which would otherwise produce $1/\tau \propto Cn^2$ scaling. These results provide robust evidence that, within the 80–295 K experimental window, flat-band-mediated bimolecular recombination governs the ultrafast carrier dynamics in 1T-TiS₂.

3. Rate-equation analysis of carrier relaxation

The fast inverse time $1/\tau_1$ is extracted from a single-exponential fit $n(t) = n_0 \exp(-t/\tau_1)$ to the early-delay portion of the photoexcited carrier-density transient. Here, we test which of the three recombination processes—monomolecular (SRH), bimolecular (electron-hole), and three-particle (Auger)—is consistent with the measured dependence of $1/\tau_1$ on the absorbed pump fluence F . For each

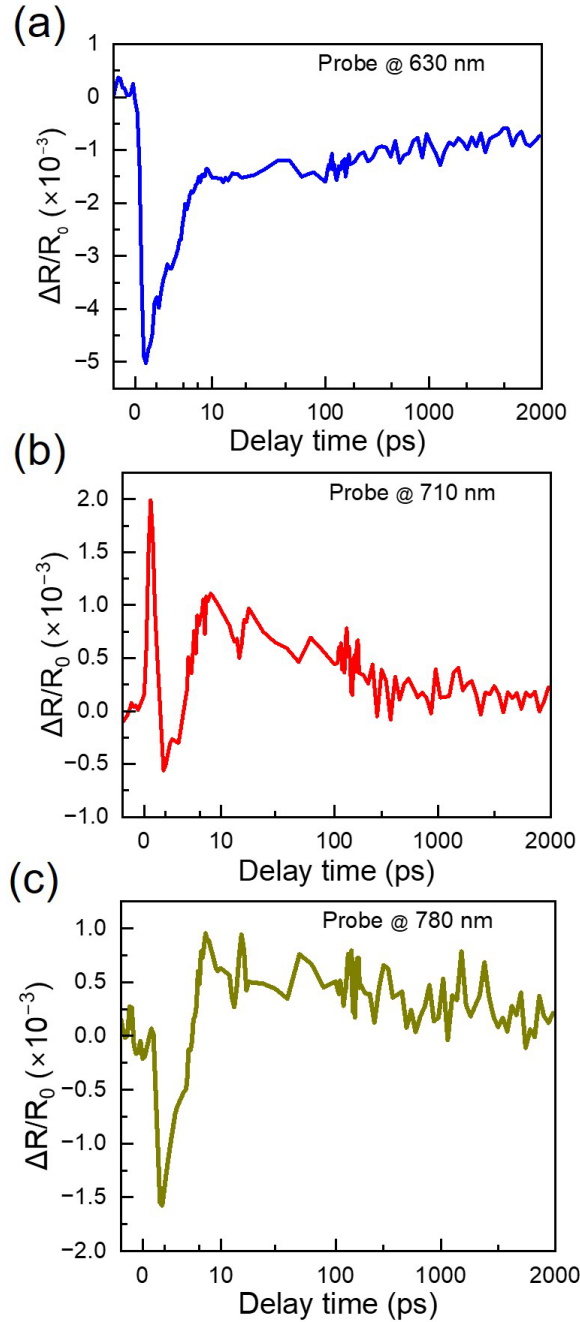


FIG. 6. Transient reflectivity change ($\Delta R/R_0$) traces of 1T-TiS₂ measured at probe wavelengths of 630 [panel (a)], 710 [panel (b)], and 780 nm [panel (c)] under 400 nm excitation with a pump fluence of 0.737 mJ cm^{-2} at room temperature (295 K). The traces capture the full relaxation dynamics over a 2000 ps time window, revealing a long-lived, slow decay component associated with lattice thermal diffusion.

process, we solve the rate equation, identify the effective early-time exponential to which a single-exponential fit corresponds, and read off the predicted scaling of $1/\tau$ with F .

The most general single-band rate equation for the photoexcited carrier density $n(t)$, including all three recombination channels together with a photoexcitation

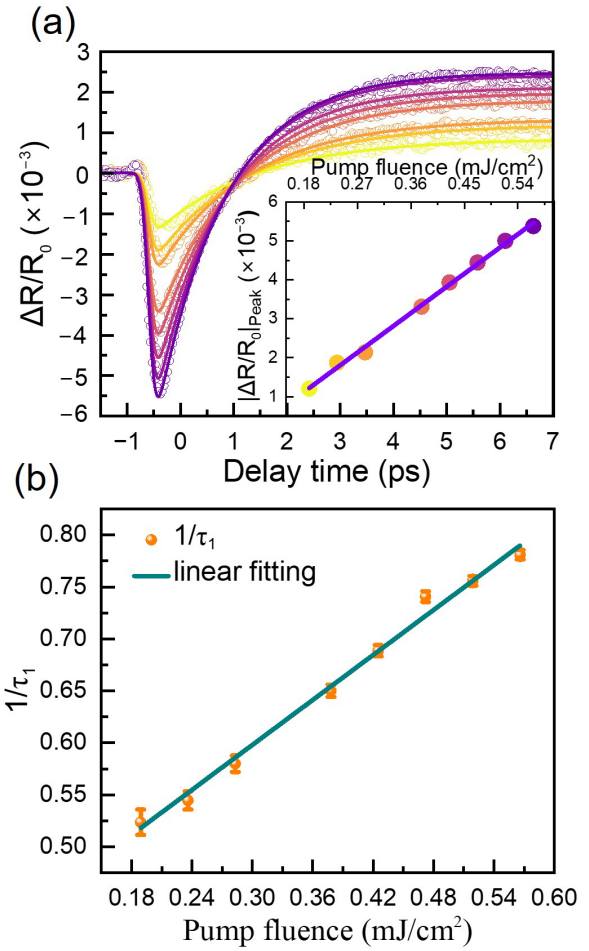


FIG. 7. (a) Transient reflectivity ($\Delta R/R_0$) traces of 1T-TiS₂ measured at a probe wavelength of 780 nm under various pump fluences with 390 nm excitation, fitted using a biexponential function to extract the fast decay time τ_1 . The inset shows the linear relationship between the peak signal and pump fluence. (b) Inverse fast decay time ($1/\tau_1$) in units of ps^{-1} plotted vs pump fluence, demonstrating a linear scaling characteristic of bimolecular electron-hole recombination.

source term $G(t)$, reads

$$dn/dt = G(t) - An - Bn^2 - Cn^3, \quad (\text{A1})$$

where $G(t)$ is the optical generation rate and A , B , C are the monomolecular, bimolecular, and Auger coefficients, respectively. The pump pulse is much shorter than the decay time of interest, so it deposits all carriers essentially instantaneously at $t = 0$, fixing the initial density $n(0) = n_0 \propto F$. For $t > 0$, the source term vanishes, $G(t) = 0$, and Eq. (A1) reduces to $dn/dt = -An - Bn^2 - Cn^3$. We solve this equation under the approximation of monomolecular, bimolecular, and three-particle processes as follows.

a. Monomolecular recombination

In the monomolecular limit, each photoexcited carrier is captured independently by a deep trap, and Eq. (A1) reduces to the linear form $dn/dt = -A n(t)$, where A is a material-only coefficient, set by intrinsic properties of the sample and

independent of the photoexcited carrier density. Direct integration gives the exact solution $n(t) = n_0 \exp(-At)$, which is a single-exponential function with $\tau_{\text{SRH}} = 1/A$. Since A does not depend on n_0 , and hence not on F , $1/\tau_{\text{SRH}}$ is independent of the pump fluence (F).

b. Bimolecular recombination

In the bimolecular limit, two distinct photoexcited carriers—one electron and one hole—must encounter each other to recombine, so the rate is proportional to the product of the electron and hole densities. Under above-bandgap excitation and overall charge neutrality, $n_e(t) = n_h(t) \equiv n(t)$, and Eq. (A1) reduces to $dn/dt = -Bn^2(t)$. Separating variables and integrating from $t = 0$ [with $n(0) = n_0$] yields the solution $n(t) = n_0/(1 + Bn_0t)$. By considering the approximation of $Bn_0t \ll 1$, $n(t) = n_0/(1 + Bn_0t) \approx n_0 \exp(-Bn_0t)$, since both expressions share the same initial value and the same initial slope at $t = 0$. A single-exponential fit to the early-time portion of the transient therefore returns a $1/e$ time τ_1 satisfying $1/\tau_1 = Bn_0 \propto F$, in which the $1/\tau_1$ shows a linear dependence on pump fluence.

c. Three-particle (Auger) recombination

In the Auger limit, three carriers must meet simultaneously: An electron-hole pair recombines and transfers its energy nonradiatively to a third (electron or hole) carrier. Two microscopic channels exist—electron-electron-hole (eeh, rate $\propto n_e^2 n_h$) and hole-hole-electron (hhe, rate $\propto n_h^2 n_e$)—but under charge neutrality both yield a rate $\propto n^3$, and Eq. (A1) reduces to $dn/dt = -Cn^3(t)$, with $C \equiv C_{\text{eeh}} + C_{\text{hhe}}$. Separating variables and integrating from $t = 0$ [with $n(0) = n_0$] yields the exact analytic solution, $n(t) = n_0/\sqrt{1 + 2Cn_0^2 t}$. In the early-delay window, $2Cn_0^2 t \ll 1$, so that a Taylor expansion of $n(t)$ gives $n(t) \approx n_0(1 - Cn_0^2 t + \dots) \approx n_0 \exp(-Cn_0^2 t)$, since both expressions share the same initial value and the same initial slope at $t = 0$. A single-exponential fit to the early-time portion of the transient therefore returns a $1/e$ time τ_{Auger} satisfying $1/\tau_{\text{Auger}} = Cn_0^2 \propto F^2$, in which $1/\tau_{\text{Auger}}$ shows quadratic dependence on pump fluence.

4. DFT calculations of the band structure and Young's modulus of 1T-TiS₂

First-principles calculations based on DFT were performed using the Vienna Ab initio Simulation Package. The exchange-correlation energy was treated within the generalized gradient approximation using the Perdew-Burke-Ernzerhof functional [69]. The interaction between valence electrons and ionic cores was described using the projector augmented wave method [70,71]. To accurately capture the interlayer van der Waals (vdW) interactions, the DFT-D3 correction scheme was included [72].

The kinetic energy cutoff for the plane-wave basis was set to 500 eV, and the total energy convergence criterion was 10^{-5} eV. Structural optimization was performed using the conjugate gradient method until the residual forces on each atom were below $0.01 \text{ eV } \text{\AA}^{-1}$. After full relaxation with vdW correction, the optimized lattice constants of 1T-TiS₂ were determined to be $a = 3.34 \text{ \AA}$ (in-plane) and $c = 5.52 \text{ \AA}$ (out of plane), in excellent agreement with previous reports [73,74].

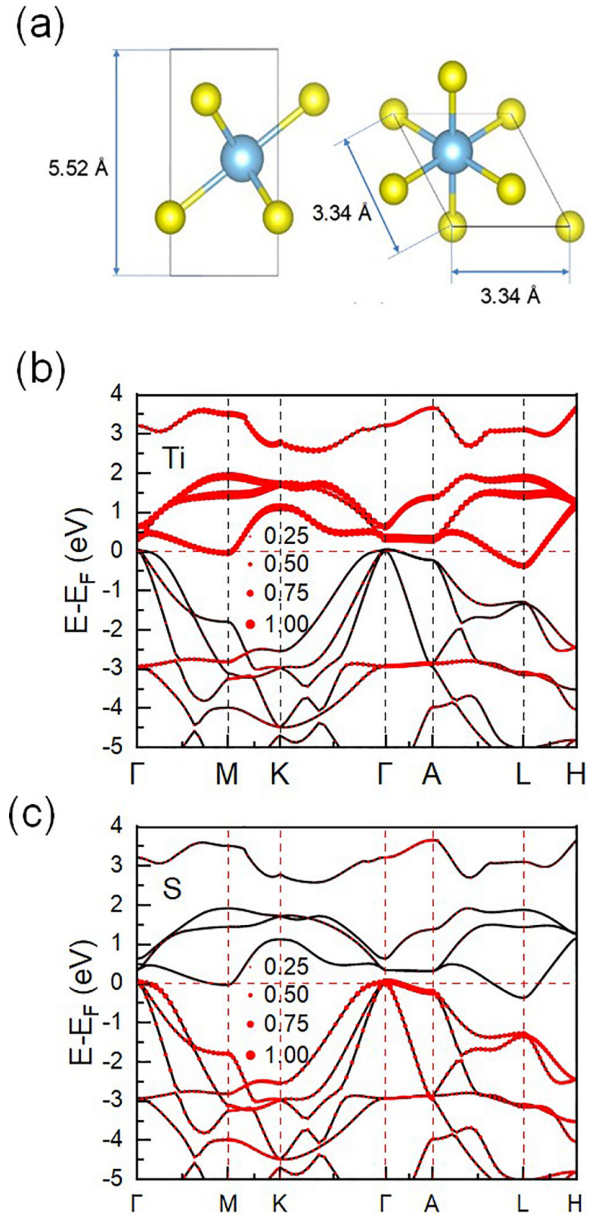


FIG. 8. (a) Side and top views of the optimized 1T-TiS₂ crystal structure. (b), (c) Projected electronic band structures showing the contributions from Ti and S atoms, respectively.

Figures 8(b) and 8(c) present the projected band structures of 1T-TiS₂, highlighting the orbital contributions from Ti and S atoms, respectively. The calculated results confirm the semimetallic nature of 1T-TiS₂, with a quasiflat dispersion near the Fermi level along the Γ –A direction. This flat band results in a high DOS and a large carrier effective mass, both of which enhance carrier localization and support the flat-band-mediated bimolecular recombination ($\tau_1 \approx 1.5 \text{ ps}$) observed experimentally within the 80–295 K experimental window.

The calculated elastic properties (Table I) reveal pronounced anisotropy in 1T-TiS₂. The in-plane and out-of-plane Young's moduli were determined to be 124.1 and 70.4 GPa, respectively, in close agreement with experimental estimates derived from coherent acoustic phonon oscillations

TABLE I. Calculated mechanical properties of 1T-TiS₂ with DFT-D3.

Parameter	Y(100)	Y(001)	B	G
This work (DFT-D3)	124.1 GPa	70.4 GPa	48.2 GPa	34.4 GPa

(~62 GPa). The bulk modulus (B) and shear modulus (G) were obtained as 48.2 and 34.4 GPa, respectively. These results demonstrate the layered mechanical nature of 1T-TiS₂, characterized by strong intralayer bonding and weak interlayer coupling.

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