Extremely low-energy collective modes in a quasi-one-dimensional topological system

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Received January 3, 2022; accepted January 10, 2022, published online March 31, 2022

We have investigated the quasiparticle dynamics and collective excitations in the quasi-one-dimensional material ZrTe$_5$ using ultrafast optical pump-probe spectroscopy. Our time-domain results reveal two coherent oscillations having extremely low energies of $\hbar \omega_1 \sim 0.33$ meV (0.08 THz) and $\hbar \omega_2 \sim 1.9$ meV (0.45 THz), which are softened as the temperature approaches two different critical temperatures ($\sim$54 K and $\sim$135 K). We attribute these two collective excitations to the amplitude mode of photoinduced dynamic charge density waves in ZrTe$_5$ with tremendously small nesting wave vectors. Furthermore, a peculiar quasiparticle decay process associated with the $\hbar \omega_2$ mode with a timescale of $\sim$1-2 ps is found below the transition temperature $T^*$ ($\sim$135 K). Our findings provide pivotal information for studying the fluctuating order parameters and their associated quasiparticle dynamics in various low-dimensional topological systems and other materials.

charge density waves, ultrafast quasiparticle dynamics, amplitude mode

PACS number(s): 71.45.Lx, 78.47.+p, 03.65.Vf

1 Introduction

In a many-body system, interaction between the quasiparticles, and/or interaction between the quasiparticles and other quantized modes may bring about various broken symmetry ground states [1], e.g., superconducting state, density wave state, and magnetically ordered state, accompanied simultaneously by abundant collective excitations. Examples of such excitations are phonons, density waves (charge or spin) and magnons obeying the Bose-Einstein statistics. Delicate balance among different interactions and phases may cause the fluctuating characteristics in some order parameters or collective excitations with a finite correlation time $\tau_F$ [1, 2].

ZrTe$_5$, a quasi-one-dimensional material with highly anisotropic crystal lattices and electronic structures [3-5], has recently attracted enormous attention. On the one hand, ZrTe$_5$ is associated with the topological phases of matter, i.e., the topological insulator (TI) [3, 6-9] or the Dirac semimetal

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which have been experimentally investigated by different equilibrium or quasi-equilibrium techniques, e.g., angle-resolved photoemission spectroscopy [6, 8], electronic transport [13, 14], infrared optical spectroscopy [10, 15-18], and scanning tunneling microscopy [6, 19]. On the other hand, many unusual electronic responses such as the chiral magnetic effect [20], anomalous Hall effect [21], and 3D quantum Hall effect [22, 23] have been reported. It is believed that those exotic phenomena are connected to the peculiar fermionic and bosonic excitations in ZrTe$_5$ with the anomalous phase transition around temperature $T^*$, where the resistivity presents a peak [24, 25]. However, any unique collective excitation and its associated dynamics in ZrTe$_5$ has never been observed or elucidated, although the related properties could be critical in understanding the emergent phenomena of this material previously observed via the quasi-equilibrium probing techniques.

By contrast, the ultrafast optical pump-probe technique is a powerful tool that not only can reveal the quasiparticle dynamics associated with the topological properties [26, 27] but also is capable of unveiling the collective phenomena in a broad correlation time- or length scale [28-30]. Here, using ultrafast optical pump-probe spectroscopy our measurements for the first time reveal two extremely low-energy collective amplitudes mode oscillations ($\sim$0.33 and 1.9 meV) at low temperatures in the high-quality ZrTe$_5$ single crystals. We unravel these two modes arising from the photoinduced dynamic charge density waves (CDWs) via the acoustic phonons. We also observe a novel quasiparticle dynamics associated with this type of bosonic mode emerging below $T^*$ with a ps-timescale.

2 Results and discussion

The experimental setup is schematically illustrated in Figure 1(a), and described in detail inside the Supplemental Material and refs. [15, 18, 27, 30-32]. The high quality ZrTe$_5$ sample was glued on a sapphire substrate. The temperature dependence of the resistivity is shown in Figure 1(b). The transition temperature is identified at around $T^*$ $\sim$135 K from the peak. Figure 1(c) shows the typical measured signals at low and high temperatures. Upon photoexcitation, all the $\Delta R(t)/R$ signals clearly exhibit an instantaneous rise, succeeded by lateral relaxation processes. Besides, immense damped ultrafast oscillations are superimposed on the non-oscillating decay background. These coherent oscillations can verify the high quality of our samples, as will be discussed in detail later.

We first focus on the non-oscillatory signals. A close inspection of the rising curve (see the inset to Figure 1(c)) reveals that it actually comprises two parts in the time-domain: (1) one exactly overlaps with the pump-probe cross correlation profile; (2) the other one shows relatively slow rising before the $\Delta R/R$ signal reaches the maximum value. We identify that the relative slow-rising can be well described by a sub-ps exponential decay process having amplitude with a different sign from that of the subsequent relaxation dynamics [33, 34] (See Supplemental Material for further details). However, the succeeding relaxation process does not exactly follow a single exponential decay at all investigated temperatures. Rather, an extra decay process can be unambiguously observed below a critical temperature, which is nearly the same as the transition temperature $T^*$ $\sim$135 K. According to these observations, we can fit our data using the following formula [27, 35]:

$$\Delta R(t)/R = (A_1 e^{-t/T_1} + A_2 e^{-t/T_2} + A_3 e^{-t/T_3} + C) \times G(t),$$  \hspace{1cm} (1)$$

where $A_j$ and $T_j$ ($j = 1, 2, 3$) are the amplitudes and decay times, respectively. $A_1 > 0$ and $A_2, A_3 \leq 0$. Here, $T_1$ and $T_2$ represent the sub-ps and extra decay processes, respectively, $T_2$ is the subsequent decay after $T_1$ or $T_3$. $C$ is a constant, and $G(t)$ is an Gaussian function standing for the pump-probe cross correlation. As seen in Figure 1(d) and (e), the fitted curves are in excellent agreement with the experimental data at low and high temperatures. Specifically, we find that $T_1$ is less than $\sim$0.3 ps, $T_2$ is $\sim$5-18 ps, and $T_3$ is $\sim$1-2 ps. The decay process characterized by $T_1$ emerges below $\sim$135 K ($T^*$), or $A_j = 0$ for $T \geq T^*$. The extracted $T_j$ ($j = 1, 2, 3$) as a function of temperature are shown in Figure 2.

In general, the relaxation processes of photoexcited quasiparticles in conventional nonmagnetic materials disclosed by $\Delta R(t)/R$ can include the e-e thermalization, the e-phonon scattering, the e-h recombination, and the thermal diffusion processes [36-38]. The e-e thermalization usually has a timescale less than $\sim$100 fs. If this process is faster than the excitation pulse and out of the time resolution limit, it is often concealed in the initial rising signal, and then follows the pump-probe cross correlation profile. This is well consistent with our experiments. On the other hand, the thermal diffusion process and the e-h recombination across a direct gap with radiative decay have timescales larger than $\sim$100 ps, and clearly do not contribute to the three processes characterized by $T_j$ ($j = 1, 2, 3$) shown in Figures 1 and 2.

Since $T_1$ and $T_2$ decay processes exist at all investigated temperatures, and experience a continuous change across $T^*$, they should represent a generic dynamical responses of ZrTe$_5$, independent of any physics underlying the transition around $T^*$. In fact, the sub-ps process characterized by $T_1$ can be attributed to the photoexcited hot-carrier cooling via e-phonon scattering (see Figure 2(b)), which in turn leads to the optical phonon emission. Its $T$-dependence is quite similar with
those of e-ph thermalization times observed in metals, semiconductors and topological materials [33, 36, 39-42]. Such $T$-dependent behavior can be described by the two-temperature model (TTM) [39, 43]. Figure 2(a) demonstrates that the fitted results using TTM agree very well with the experimental data. The fluence-dependent $\tau_1$ further confirms the effectiveness of TTM (see Supplemental Material). Employing $\tau_{\text{e-ph}}^{-1} = \hbar \lambda(\omega^2) / (\pi k_B T_d)$, we can further obtain the dimensionless e-ph coupling constant $\lambda$ to be $\sim 0.17$ via $\tau_1(T)$. During the calculations, we obtained $\lambda(\omega^2) = 1.8 \times 10^{25}$ Hz$^2$ = 78.2 meV$^2$, where the second moment of the phonon spectrum $\langle \omega^2 \rangle$ is $\sim 6.2 \times 10^4$ K$^2$ extracted via the Debye model.

The $\tau_2$ decay process, with a timescale of $\sim 3-18$ ps, illustrates a totally different $T$-dependent behavior as that of the $\tau_1$ process (see Figure 2(c)). After the e-ph thermalization, the nonequilibrium electrons (holes) can still accumulate in the conduction (valence) bands. We might thus consider the phonon-assisted e-h recombination [44, 45], where the electron and hole recombine with the assistance of e-ph scattering between the electron and hole pockets, as illustrated in Figure 2(d). In fact, the conduction and valence band extrema for ZrTe$_5$ are in close proximity within the Brillouin zone [3, 46], in favor of such interband e-h scattering. Therefore, we assign the $\tau_2$ decay to this type of recombination process. This inference is not only consistent with the fluence ($F$) dependence of $\tau_2^{-1}$ (inset of Figure 2(c)) [30, 47], but also is strongly justified by the excellent agreement between the experimental $\tau_2$ and the theoretical fitted results based on such recombination, as shown in Figure 2(c).

The peculiar $\tau_2$ process emerges below $T^*$, implying that it is associated with the physics beyond the transition. Therefore, one may argue whether the $\tau_2$ process is associated with the Fermi level shifting from the valence band to the conduction band as the temperature decreases across $T^*$ [48]. However, previous ultrafast optical studies on graphene [49] demonstrated that when the Fermi level was continuously tuned across the Dirac point, no emergence or disappearance of any quasiparticle decay channel was observed except for the change of the e-ph relaxation time. Therefore, this effect should be irrelevant here.

Since $T$-dependent behavior of $\tau_2$ well resembles that of $\tau_1(T)$, we suspect that this $\tau_2$ process should microscopically resemble the e-ph scattering but perhaps involves unknown bosonic excitations emerging below $T^*$. Numerically, we still can use $\tau_{\text{e-ph}}^{-1} = 3/5 \lambda(\omega^2) / (\pi k_B T_d)$ to fit $\tau_2$ based on TTM. However, the fitted parameter $\lambda(\omega^2)$ ($\sim 5.47$ meV$^2$) only gives an upper limit because the TTM now evolves into a three-temperature model [50]. At this stage, although the type of boson associated with $\tau_2$ is unknown (to be identified later), we can tell its energy is not very high and probably close to scale of the acoustic phonons based on its timescale (1-2 ps) [26].

Now let us focus on the “rich” oscillations observed in $\Delta R(t)/R$. Figure 3(a) shows the typical time domain signals at temperatures below and above $T^*$. The temperature evolution...
Figure 2  (Color online) (a) $\tau_1$ as a function of temperature. Red line is a fit via TTM. (b) Schematic of the hot carrier excited by hv photon cooling via e-phon scattering. (c) $\tau_2$ as a function of temperature. Red line is a fit based on the phonon-assisted e-h recombination process. (d) Schematic of the phonon-assisted e-h recombination. (e) $\tau_3$ as a function of temperature. Red line is a fit via TTM. (f) Schematic of the hot carrier excited by hv photon cooling involved with unknown bosonic excitations emerging below $T^*$. Insets in (a), (c) and (e) show $1/\tau_1$, $1/\tau_2$ and $1/\tau_3$ as a function of the pump fluence ($F$) at 5 K, respectively.

Figure 3  (Color online) (a) Extracted oscillations for three typical temperatures. (b) The temperature-dependent Fourier transform spectra for the extracted oscillations. (c) The two low-energy modes, $\omega_1$ and $\omega_2$, as a function of temperature in the frequency domain. The red dashed curves are guides to the eyes. (d) At 5 K, wavelength dependence of the two low-energy modes, $\omega_1$ and $\omega_2$, indicated by the red dashed lines.

The oscillatory signals can be revealed by the Fourier transform data in the frequency domain (see Figure 3(b)), where five modes with distinct frequencies are clearly observed. In general, the oscillations with THz frequency in pump-probe spectroscopy arise from the coherent optical phonon modes near the $\Gamma$ point due to the coherent Raman scattering or displace excitation [51]. The exact modes can be revealed by a direct comparison with the Raman spectroscopy data [52, 53] (see also the Supplemental Material). We thus obtain that the three modes with center frequencies near $\sim 1.2$, $\sim 3.7$, and $\sim 4.6$ THz are the $A_k$ phonon modes. These modes are characteristics of a high quality ZrTe$_5$ single crystal, first-time observed simultaneously in the time-resolved measurements. Temperature-dependence of all these modes can be well described by the anharmonic phonon model. We note that the laser pulse chirping could affect the amplitudes of coherent optical phonons [54, 55]. However, since the fs pulses arriving at the samples are nearly transform limited via our pulse compression system, this effect can be safely neglected. Moreover, even if there exists slight chirp effect, its influence on characterizing the sample quality can be ignored as the corresponding phonon frequencies remain unchanged.

Unexpectedly, as shown in Figure 3(b)-(d), we also clearly observed two low-energy modes with frequencies of $\omega_1/2\pi \sim 0.08$ and $\omega_2/2\pi \sim 0.45$ THz, respectively. These two modes cannot be attributed to the optical phonons since their frequencies are well under the low frequency limit of the Raman-active optical phonons, i.e., $\sim$1 THz, obtained by our theoretical calculations. In specific, the $\omega_1$ mode, emerging below $\sim$60 K, exhibits a clear softening down close to zero as the temperature increases. The $\omega_2$ mode is non-observable above $\sim T^*$. One might suspect whether they originate from the low-energy acoustic phonons. However, (1) the acoustic phonon will not suddenly disappear above some critical temperature, (2) the estimated phonon frequency and the corresponding probe-wavelength dependent shift significantly deviate from our observations, and (3) the coherent acoustic phonons will not experience an abrupt $\pi$-phase change as the probing wavelength varies slightly (see Figure 3(d) and Supplemental Material (sects. F-I)). Therefore, according to the $T$-dependent and wavelength-dependent behaviors of these two modes, we can exclude such a possibility [56-58]. On the other hand, the long-range CDW order stabilized using the strong magnetic field has been proposed to explain the 3D quantum Hall effect in ZrTe$_5$ [22,23]. Therefore, one can question if the static CDWs give rise to the two collective modes observed here. However, up to now no conventional thermodynamic measurements report the existence of CDWs without magnetic field, the instability of static CDWs can hardly contribute to the low-energy modes.
We notice that there exists subtle correlation between the anisotropic crystal lattices and electronic structures in ZrTe$_5$ [3-5]. Photoexcitation in our optical pump-probe experiments can not only introduce non-equilibrium carriers within the electronic bands but also lead to strong coherent motion of the lattices, as already demonstrated above. These facts enable us to suspect that the photoinduced CDWs [59] may emerge in this material even within the current pump-fluence regime. The photoinduced dynamic CDWs may give rise to two collective excitations: the amplitude mode (amplitude) and the phase mode (phason). Because the phason has similar dispersion relation as that of acoustic phonon, it is expected to bear the same wavelength dependence [58] and thus cannot contribute to these two low-energy modes. We thus finally arrive at the most reasonable candidate for these two modes, i.e., the amplitudes of photoinduced dynamic CDWs.

Particularly, the $T$ dependence of these two modes is quite similar with the temperature evolution of the fluctuating CDWs [1, 28, 58], as can be seen in Figure 4(a) and (b). $\omega_1$ and $\omega_2$ as a function of $T$ can well be fitted by a mean-field-like $T$ dependence [1,28]: $\omega_\lambda = \omega_\lambda(0)(1 - T/T_c)^{\beta}$, where $T_c$ is defined as a transition temperature. Satisfactory fits to the data in Figure 4(a) and (b) give: $\beta_{\omega_1} = 0.25$ and $\beta_{\omega_2} = 0.1$ corresponding to $T_c^{\omega_1} \approx 54$ K and $T_c^{\omega_2} \approx 135$ K, respectively. $\beta_{\omega_1}$ having a value of $\approx 1/4$ indicates that $\omega_1$ mode has a pure one-dimensional origin [1]. In fact, such character is consistent with our theoretical calculations. Specifically, the obtained phonon linewidth in ZrTe$_5$ has a quasi-one-dimensional character, i.e., the corresponding values of three acoustic modes along the $\Gamma-Z$ direction (inter-plane) are in general larger than those along other directions. Since the e-ph interactions dominate the order parameter [23], it is not surprising that a fluctuating CDW can emerge along the inter-plane direction ($b$-axis). By contrast, the $\omega_2$ mode cannot belong to a simple one-dimensional case. The intrinsic $\omega_2$ mode might have higher dimensional characteristics, and needs to be unveiled by further investigations. Interestingly, in contrast to the $\omega_2$ mode a $\tau$-phase change was observed for the $\omega_1$ mode as the probe wavelength tunes from 800 to 820 nm in Figure 3(d), which implies that the wavelength dependence of the dielectric tensor components $\epsilon_j$ ($j = a, b, c$) can provide dimensionality information of different CDW modes (see Supplemental Material).

The $T$-dependent damping rate $\Gamma_1$ ($\Gamma_2$) for the $\omega_1$ ($\omega_2$) mode can be extracted by fitting the oscillatory signals via a damped sine function, as shown in Figure 4(c) and (d). In order to compare with the theoretical calculations [60, 61], we used a power law ($\propto T^n$) to fit the data, and obtain that $\Gamma_1$ and $\Gamma_2$ scale well as $T$ and $T^n$, respectively. Such $T$-scalings clearly deviate from the theoretical intrinsic damping [29, 60, 61], i.e., $T^2$ away from $T_C$ and $T^3$ close to $T_C$, respectively. Indeed, decay of the amplitude includes both the intrinsic damping and correlation time of the fluctuating CDW. Therefore, $\Gamma_1^{-1}$ and $\Gamma_2^{-1}$ here provide a lower bound of the correlation time ($\tau_{\Gamma_1}$ and $\tau_{\Gamma_2}$). We note that the exact value of the damping rate is material-dependent, and could be quite different between various systems [29, 58]. In addition, the phase mode of dynamic CDWs can be revealed by damping of the signals via the transient grating technique [29], which, however, is only effective for the energies of CDW modes far away from the acoustic and optical phonons. Since energies of the dynamic CDWs observed here are too close to these two type of phonons, it will be extremely difficult to extract the information of phonons in ZrTe$_5$.

The acoustic phonon may soften significantly at $q = 2k_f$ upon the phase transition of CDW [1]. In the $q = 0$ limit, $\omega_A = \lambda^{1/2} \omega_{2k_f}$, where $\omega_{2k_f}$ is the frequency of the softening phonon above the phase transition connected to the frequency of amplitude ($\omega_A$) via the e-ph coupling constant $\lambda$. Therefore, using a linear dispersion approximation with sound velocity of $v_s \sim 10^3$ m/s [62], $k_f$ can be estimated to have values of 0.053 Å$^{-1}$ and 0.34 Å$^{-1}$ for $\omega_1$ and $\omega_2$ modes, respectively. Although we do not have accurate knowledge of anisotropic $v_s$ associated with $\omega_1$ and $\omega_2$ modes, based on these two estimated values we can conclude that their corresponding modulation wave vectors should be very small. Such uncommonly small $k_f$ values indicate that our observed photoinduced density waves have tremendously long lattice modulation periods in the real space. These results are directly due to the extremely low energies of $\omega_1$ and $\omega_2$ modes. In-
terestingly, these derived values are consistent with the work demonstrating the 3D quantum Hall effect [22].

Since \( \omega_1 \) and \( \omega_2 \) modes now can be attributed to the fluctuating CDWs that obeys the Bose-Einstein statistics, they can naturally be assigned to the bosons contributing to the new scattering process characterized by \( \tau_s \) emerging below \( T^* \). However, the \( \omega_2 \) mode is the mostly possible candidate due to three reasons: (1) appearance temperature of this mode coincides with that of the \( \tau_s \) process; (2) no clear anomalies at \( T_C^{\omega_2} \) are found in the \( T \)-dependent \( \tau_s \) and \( \Delta_s \); (3) energy of the \( \omega_1 \) mode is nearly one order of magnitude smaller than that of the \( \omega_2 \) mode so that the timescale of energy exchange between the nonequilibrium quasiparticles and \( \omega_1 \) mode can mix up with the long thermal diffusion process, where a large number of low-energy acoustic phonons over-whelm the CDW modes.

3 Conclusion

In summary, using the ultrafast optical spectroscopy we present a detailed investigation on the quasi-one-dimensional system \( \text{ZrTe}_5 \). Our results reveal that below \( T^* \), there emerges a new quasiparticle decay process with a timescale of \(-1\text{-}2\) ps, in addition to the two relaxation dynamics persisting at all temperatures with timescales of \(-0.15\text{-}0.3\) and \(-3\text{-}18\) ps, respectively. We address that this new decay process is most likely due to appearance of some novel collective excitations in \( \text{ZrTe}_5 \), while the other two can be attributed to the e-ph scattering and the phonon-assisted e-h recombination processes. Surprisingly, our experimental data for the first time unambiguously reveal two coherent oscillations \( (\omega_1 \) and \( \omega_2 \) with extremely small energies occurring below two distinct temperatures \(-54 \) and \(-135 \) K, respectively. We argue that they arise from the amplitude mode of photoinduced dynamic CDWs triggered by acoustic phonons, and can well explain the newly observed quasiparticle decay below \( T^* \). Whether these findings are connected to the other emergent phenomena in this system deserves further explorations.

This work was supported by the National Natural Science Foundation of China (Grant Nos. 11974070, 11724006, 11922408, and 11921004), the Frontier Science Project of Dongguan (Grant No. 2019ZX2101004), the National Key R&D Program of China (Grant Nos. 2016YFA0300500, and 2016YFA0300500), the Strategic Priority Research Program of Chinese Academy of Sciences (Grant No. XDB19000000), the K. C. Wong Education Foundation (Grant No. GJTD-2018-01), the Beijing Natural Science Foundation (Grant No. Z180008), the Beijing Municipal Science and Technology Commission (Grant No. Z191100007219313), and the CAS Interdisciplinary Innovation Team. We acknowledge the valuable discussion from Hrvoje Petek and Jure Demar.

Supporting Information

The supporting information is available online at phys.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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