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Unconventional charge density wave and photoinduced lattice symmetry change in the kagome metal CsV_3Sb_5 probed by time-resolved spectroscopy

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Recently, the kagome lattice metal AV_3Sb_5 (A = K, Rb, Cs) family has received wide attention due to its coexistence of superconductivity, charge density wave (CDW), and peculiar properties arising from a topological nontrivial electronic structure. With time-resolved pump-probe spectroscopy, we show that the excited quasiparticle relaxation dynamics can be explained by the formation of a temperature-dependent energy gap below the phase transition that is similar to a usual second-order CDW condensate. By contrast, the structural change shows a predominantly first-order phase transition. Furthermore, no CDW amplitude mode is identified in the ordered phase. The results suggest that the CDW order is very different from a conventional CDW condensate. We also find that a weak pump pulse can nonthermally melt the CDW order and drive the sample into its high-temperature phase, revealing a small difference in lattice potential between those phases.

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I. INTRODUCTION

Control over physical properties or electronic phases in quantum materials via external parameters is a central topic in condensed matter physics. Traditional ways of tuning the external parameters are to change the temperature, pressure, electric/magnetic field, or doping level of the material systems. In the last two decades, photoexcitation has emerged as a new method to probe and manipulate the properties of quantum materials, enabling ultrafast control over the material properties or quantum phases [1–16]. The ultrashort laser pulses can create a far from equilibrium distribution of the energy among the different degrees of freedom, triggering the formation of thermal or nonthermal metastable or even stable phases. Among different systems, correlated electronic systems with rich phase diagrams have been the most explored materials for practical manipulation, since the complex interactions or competition among different degrees of freedom make them very susceptible to external perturbation or stimuli [9].

Recently, a new family of kagome metals AV_3Sb_5 (A = Cs, Rb, K), composed of vanadium layers, antimony layers, and alkali ions sandwiched between the two layers, has attracted tremendous attention in the community [17–20]. These materials undergo a charge density wave (CDW) phase transition at $T_{\mathrm{CDW}} = 80$–100 K with a $2 \times 2 \times 2$ superlattice formation and a superconducting transition at $T_c = 0.9$–3.5 K. High-pressure measurements revealed the competition between the charge order and superconductivity by observing a double-dome superconductivity [21–25]. More intriguingly, a topological nontrivial electronic structure was found in the AV_3Sb_5 family (even in the CDW state) [17–21,26–32]. A giant anomalous Hall effect [33,34] was observed even in the absence of magnetic ordering [18,35]. Furthermore, transport and scanning tunneling microscopy (STM) measurements revealed that the sixfold rotation C_6 symmetry is further broken and reduced to C_2 symmetry at temperatures below 60 K [21,29,36–38]. A recent coherent phonon spectroscopy measurement [38] revealed the appearance of new phonon modes below 94 and 60 K, respectively, implying structural phase transitions. Thus, the AV_3Sb_5 series provides a new opportunity to study the competition between different phases and to understand the unconventional correlated physics emerging from itinerant kagome lattice electrons.

In this paper, we perform an ultrafast pump-probe reflectivity experiment on CsV_3Sb_5, aiming to investigate the CDW order across the phase transitions and possible photoexcitation control of different orders in the compound. Our measurements show that, while the quasiparticle relaxation dynamics upon weak pumping is governed by a continuous opening of an energy gap below the phase transition, the structural change is characterized by an abrupt change in the number of coherent phonon modes without showing clear softening at the CDW phase transition. Furthermore, no CDW amplitude mode can be identified in the ordered phase. Those results suggest that the CDW order is predominantly a first-order phase transition, and is very different from a traditional CDW condensate. More intriguingly, we show that even a small pumping fluence can nonthermally melt the C_2 ground state order and C_6 CDW order successively and drive the compound into a high-temperature phase. We shall discuss the implication of the results.

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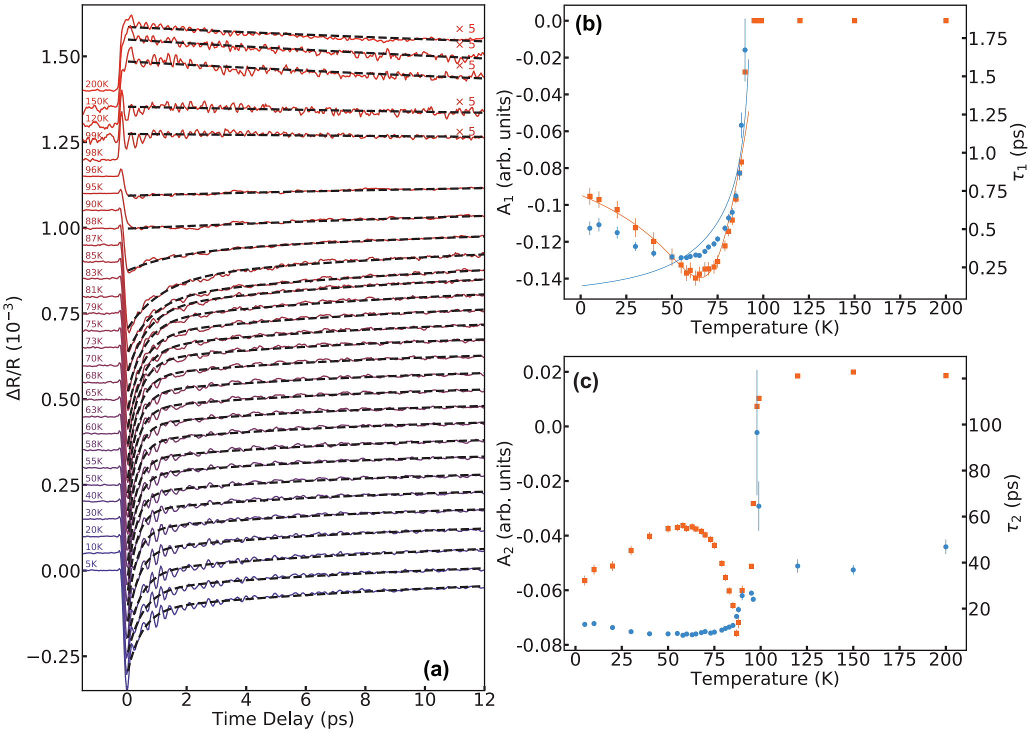

FIG. 1. Temperature-dependent dynamics for CsV$_3$Sb$_5$. (a) $\Delta R/R$ in the temperature range of 5–200 K. The lines in the high-temperature phase (from 98 to 200 K) are scaled by a fivefold factor. The offset is set as 0.05. The dashed lines are the double-exponential fitting curve: $\Delta R/R(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2}$. The data were cut at $t = 0$ ps where the absolute value of the transient reflectivity change is roughly at the maximum. (b), (c) A fast ($\tau_1 \sim 1$ ps) and a slow ($\tau_2 \sim 10$ ps) relaxation process, respectively. The amplitude A_1, A_2 (orange squares) and decay time τ_1, τ_2 (blue circles) of the reflectivity transients extracted from fits to the double exponential. We notice the amplitude of the fast relaxation process A_1 becomes negligible in the high-temperature phase, which means the fast relaxation process is absent in the high-temperature phase and the double-exponential function degrades into a single-exponential function. Error bars represent the standard deviation of the fit. The orange and blue lines are the fitting curves to the RT model.

II. EXPERIMENTS

Single crystals of CsV$_3$Sb$_5$ were grown from a Cs ingot (purity 99.9%), V powder (purity 99.9%), and Sb grains (purity 99.999%) using the self-flux method, similar to the growth of RbV$_3$Sb$_5$ [20]. We used an amplified Ti:sapphire laser system with an 800 nm wavelength and 35 fs pulse duration operating at a 1 kHz repetition frequency as the light source for the pump-probe measurement. The fluence of the probe beam is set below 3 μJ cm^{-2}, weaker than that of the pump beam. To reduce the noise caused by stray light, the pump and probe pulses were set to be cross polarized and an extra polarizer was mounted just before the detector. The signal-to-noise ratio (SNR) will be limited in systems with a lower repetition frequency. The spot size was determined to be 100 and 80 μm in diameter by a pinhole for the pump and probe spots, respectively. To improve the SNR, a balanced detection method was used. Before the probe light hit the sample, part of the light was separated by a splitter and detected by the same detector as the reference background signal. Each experimental data line is the average of three repeated measurements.

III. UNCONVENTIONAL CHARGE DENSITY WAVE STATE

Figure 1 presents the photoinduced reflectivity change of CsV$_3$Sb$_5$ as a function of time delay at different temperatures under a rather weak pumping excitation ~5 μJ cm^{-2}. Very prominently, $\Delta R/R$ changes sign, from positive to negative when temperature decreases across the CDW transition temperature T_{CDW}. At low temperature, the pump pulse induces an abrupt drop in reflectivity followed by a fast decay within a few picoseconds and a slower recovery process in the order of 10 ps, while there is only a slow decay dynamics at high temperature. We use a double-exponential function to fit the decay process, $\Delta R/R = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2}$, where $A_1 e^{-t/\tau_1}$ is the fast decay process and $A_2 e^{-t/\tau_2}$ presents the slower recovery process, respectively. The best fitting results are shown as dashed lines in Fig. 1(a). The fitting parameters of the double-exponential function are shown in Figs. 1(b) and 1(c). For the fast decay dynamics, the decay lifetime τ_1 diverges and the absolute value of amplitude A_1 drops precipitously to zero, as the temperature rises close to T_{CDW}. These behaviors signal the presence of a temperature-dependent energy gap that is closing near the critical temperature, in line with the second-order phase transition as is observed in many materials, for example, in 2H-TaSe$_2$ [39]. The relaxation dynamics of the photoexcited quasiparticles being significantly impeded by the gap opening was well described by the Rothwarf-Taylor (RT) model [40]. Similar to the situation of superconductivity from the condensate of electron-electron Cooper pairs, CDW is a condensate from electron-hole (e-h) pairs. The pump pulse

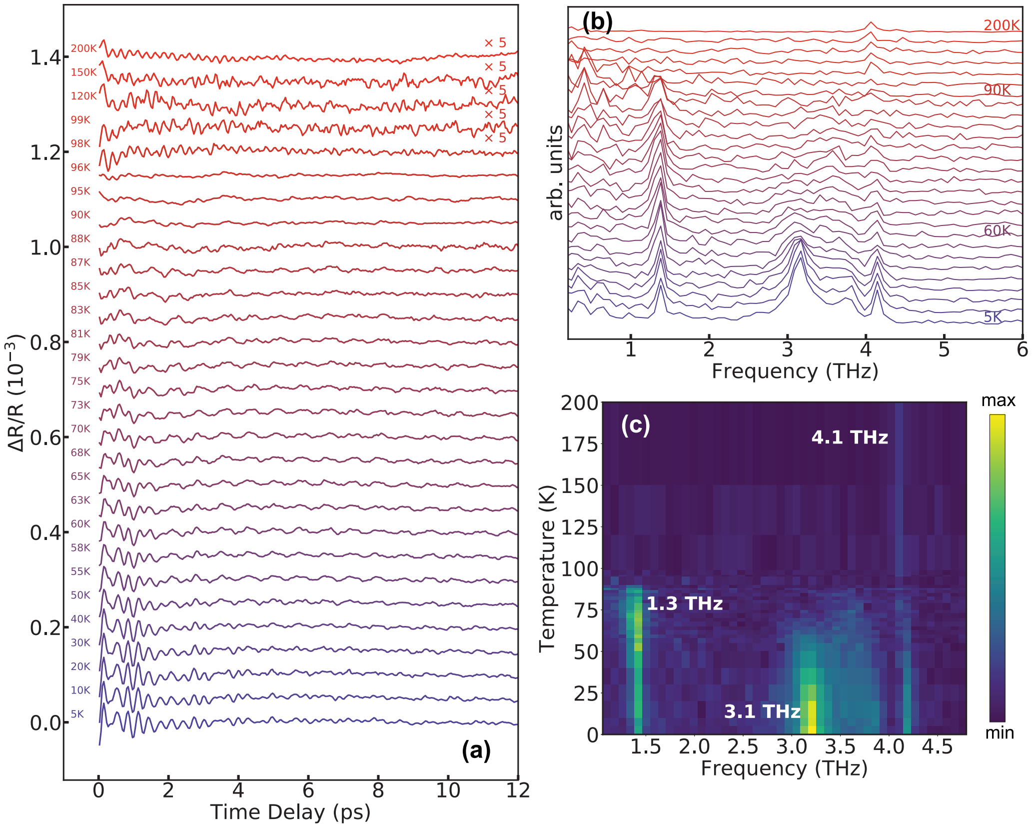

FIG. 2. Temperature-dependent coherent phonon spectroscopy for CsV$_3$Sb$_5$. (a) Coherent phonon oscillation in a time domain, where the decay background is subtracted. (b) The fast Fourier transformation of the data in (a). (c) Temperature dependence intensity map extracted from (b).

serves as an excitation to break *e-h* pairs across the energy gap of the CDW condensate, and the recombination of *e-h* pairs is accompanied by the emission of phonons with an energy higher than the energy gap $\Delta(T)$. τ_1 is the characteristic time for this recombination, since these excited phonons will in turn break additional *e-h* pairs, and decay time τ_1 is ultimately determined by the time required for these phonons to anharmonically decay into phonons with an energy less than Δ. The amplitude A represents the population of excited quasiparticles by a pump pulse. The change of relaxation time and the amplitude of the photoinduced reflectivity signal near the phase transition temperature in the RT model is given by following equations [40,41],

$$\begin{aligned}\tau(T) &\propto \frac{\ln\left[g+e^{-\Delta(T)/k_BT}\right]}{\Delta(T)^2},\\ A(T) &\propto \frac{\Phi/[\Delta(T)+k_BT/2]}{1+\Gamma\sqrt{2k_BT/\pi\,\Delta(T)}e^{-\Delta(T)/k_BT}},\end{aligned} \tag{1}$$

where a BCS-type energy band is assumed here as $\Delta(T) = \Delta_0\sqrt{1-T/T_{\mathrm{CDW}}}$ with $T_{\mathrm{CDW}} = 94$ K and the CDW gap at zero temperature $\Delta_0 = 40$ meV, which is taken from an angle-resolved photoemission spectroscopy (ARPES) measurement at the M point [42]. Qualitatively, the equations can reproduce the measurement results. The best fits yield the phenomenological fitting parameters $g = 0.18$ and $\Gamma = 17$. g and Γ are parameters of the photoexcited quasiparticle relaxation dynamics arising from electron-phonon coupling in the presence of gap formation, which is related to the density of states near the Fermi level and phonon frequencies.

After subtracting the dynamics background (the fitting curves from two exponential decay terms), we obtained the coherent phonon oscillations, as shown in Fig. 2(a). The fast Fourier transform result is presented in Fig. 2(b). A two-dimensional intensity map as a function of temperature and frequency is displayed in Fig. 2(c). The coherent phonon spectroscopy is similar to a previous study on CsV$_3$Sb$_5$ [38]. Above $T_{\mathrm{CDW}} = 94$ K, there is only one peak at 4.1 THz, which is in good agreement with the density functional theory (DFT) calculations for the A_{1g} mode of the phonon spectra [28,38]. Below T_{CDW}, a new mode at the 1.3 THz peak abruptly appears, yielding evidence for CDW structural modulation. Below 60 K, a weak and broad feature emerges near 3.1 THz, which evolves into a sharp and strong peak below 30 K. This mode was suggested to be linked to the uniaxial order observed by STM experiments which breaks the C_6 rotational symmetry [38]. It deserves to be mentioned that, for a conventional CDW condensate, the CDW amplitude mode usually appears as the strongest oscillation in the pump-probe measurement. As the temperature increases to the transition temperature T_{CDW}, the mode frequency softens and behaves as an order parameter [43]. However, no such CDW amplitude mode could be identified from the above spectra, suggesting that the CDW order in CsV$_3$Sb$_5$ is very different from a conventional CDW condensate. The abrupt change of the coherent mode near 1.3 THz further indicates that the structural transition is predominantly a first-order phase transition. On the other hand, the symmetry change from C_6 to C_2 is a crossover behavior. A short range of C_2 nematic order, corresponding to the broad feature, is seen roughly near 60 K, which gradually evolves into a static C_2 order below 30 K.

The above analysis reveals a rather peculiar situation: While the excited electron relaxation dynamics upon weak pumping can be interpreted by a continuous opening of an energy gap below the phase transition, which is similar to a usual second-order CDW condensate, the structural change is primarily a first-order phase transition, which is characterized by an abrupt change in the number of coherent phonon modes without showing clear softening at the CDW phase transition. Additionally, no CDW amplitude mode is present in the condensate state. Those results suggest that the CDW order is rather unconventional. It implies an unusual and weak coupling between the electron and lattice degree of freedoms. We notice that nuclear magnetic resonance (NMR) measurements on CsV_3Sb_5 indicated a similar situation [44]. A first-order structural phase transition associated with orbital ordering is seen as the sudden splitting of the orbital shift in the ^{51}V NMR spectrum at T_{CDW}. By contrast, a typical second-order transition behavior is seen in the quadrupole splitting which appears gradually below T_{CDW}. The NMR measurement also suggests that the CDW order is a secondary electronic order.

The physical origin for the peculiar CDW properties is not clear. Nevertheless, a hint is obtained from the fact that both the lattice and electronic structures are intrinsically unstable and the instabilities from the two degrees of freedom have the same wave vector. According to the first-principles DFT calculations, the ideal CsV_3Sb_5 kagome lattice has imaginary phonon frequencies near the M and L points in the phonon dispersions, that is, energetically unstable; an inverse star-of-David distortion with a π-phase shift between neighboring layers is energetically favored [27,38]. This instability also explains the rotational symmetry breaking from C_6 to C_2 and is indeed confirmed by the structural studies [27,28,37,38]. On the other hand, the DFT calculations [27] and ARPES measurement [42] also revealed an electronic structure instability at the M point, i.e., a saddle point near E_F or van Hove singularity. As the two instabilities have the same wave vector, even for an extremely weak electron-phonon coupling, an electronic instability would trigger the lattice instability to occur at the same temperature, leading to a first-order structure phase transition. However, the electronic response seems to be less affected and keeps showing a continuous opening of an energy gap below the CDW phase transition. Apparently, further studies are needed to solve the puzzle.

IV. PHOTOINDUCED NONTHERMAL STRUCTURE PHASE TRANSITION

In addition to varying temperature, we also performed a measurement of the photoinduced reflectivity change at different pump fluences in the CDW phase. The measurement results at 20 K are shown in Fig. 3. We found that a surprisingly small pump fluence can melt the C_2 nematic and then the C_6 CDW order. As shown in Fig. 3(a), at 5 μJ cm^{-2} fluence, the fast decay process shows a lifetime of hundreds of femtoseconds. With the pump fluence increasing, the absolute value of negative $\Delta R/R$ continues growing, and reaches a maximum value at around $F^{\text{melt}} \sim 55$ μJ cm^{-2}. Then it turns back and goes into the positive direction, passes through zero, and gradually grows larger. This behavior is very similar to the warming process of the temperature-dependent measurement. From the double-exponential decay approach, we can extract the relaxation time of the fast decay dynamics, as shown in Fig. 3(b). As the fluence increases to F^{melt}, the relaxation time sharply increases. Similar to the above analysis for temperature variation, we assume that a CDW energy gap is present up to the melting fluence as $\Delta = \Delta_0(1 - F/F^{\text{melt}})$, and the decay time is inversely proportional to the energy gap near the critical melting fluence $\tau \propto 1/\Delta$. Indeed, the fast decay time can be reasonably reproduced by the relation as shown in Fig. 3(b). The fluence-dependent relaxation dynamics suggests that a pump flence as small as 55 μJ cm^{-2} can melt the CDW order in CsV_3Sb_5.

Our measurement shows that only the highest phonon survives when the fluence is higher than 55 μJ cm^{-2}. However, the earlier study by Ratcliff *et al.* [38] clearly showed the presence of other phonon modes at twice that fluence, ~0.1 mJ/cm^2. The CDW order did not melt in their study. We notice that the pulse durations are different. In the present study, the pulse duration is 35 fs, while theirs is 100 fs. As the peak electric field is related to the pulse duration by $E_p = \sqrt{2Z_0F/\tau}$, the peak electric field in the present study is stronger. Another possibility is that the pump pulse wavelengths in the two studies are different. The pulse wavelength of Ref. [38] is 1515 nm, different from 800 nm. One expects that the penetration depth and reflectivity are different in terms of the infrared spectroscopy data, which could lead to the difference in threshold.

The coherent phonon spectroscopy provides more direct evidence for ultrafast optical melting of those orders in CsV_3Sb_5. We performed a Fourier transformation of the pump-probe measurement after subtracting the relaxation background and obtained the coherent phonon spectra for different fluences, as displayed in Fig. 3(c). The intensity plot as a function of both frequency and pump fluence is shown in Fig. 3(d). At small fluence, for example, at 5 μJ cm^{-2}, three resonance peaks centered at 1.3, 3.1, and 4.1 THz can be observed. As the pump fluence rises to 23 μJ cm^{-2}, the peak at 3.1 THz that is associated with C_2 symmetry [38] disappears. As the fluence increases beyond 55 μJ cm^{-2}, the phonon peak at 1.3 THz, which is related to CDW order [38], is not visible. The critical pump fluence is in agreement with the analysis from the relaxation dynamics. The 4.1 THz resonance is present in all fluences. The strength of this mode is further enhanced up to the highest used fluence 300 μJ cm^{-2}. This behavior is expected normally for the coherent phonon generation in a pump-probe experiment.

We emphasize that the melting of the low-temperature orders cannot be attributed to a trivial thermal effect owning to the very small pump fluence. The temperature rise ΔT can be phenomenologically estimated by the energy conservation law $S\delta_0\rho/M \int_{T_0}^{T_0+\Delta T} C_p(T)dT = (1-R)FS$, where S is the excitation area, the mass density $\rho = 5.2$ g/cm^3, the molar mass $M = 894.5$ g/mol, the penetration depth $\delta_0 \approx 64$ nm, and the reflectivity $R = 0.42$ at 800 nm for CsV_3Sb_5 from

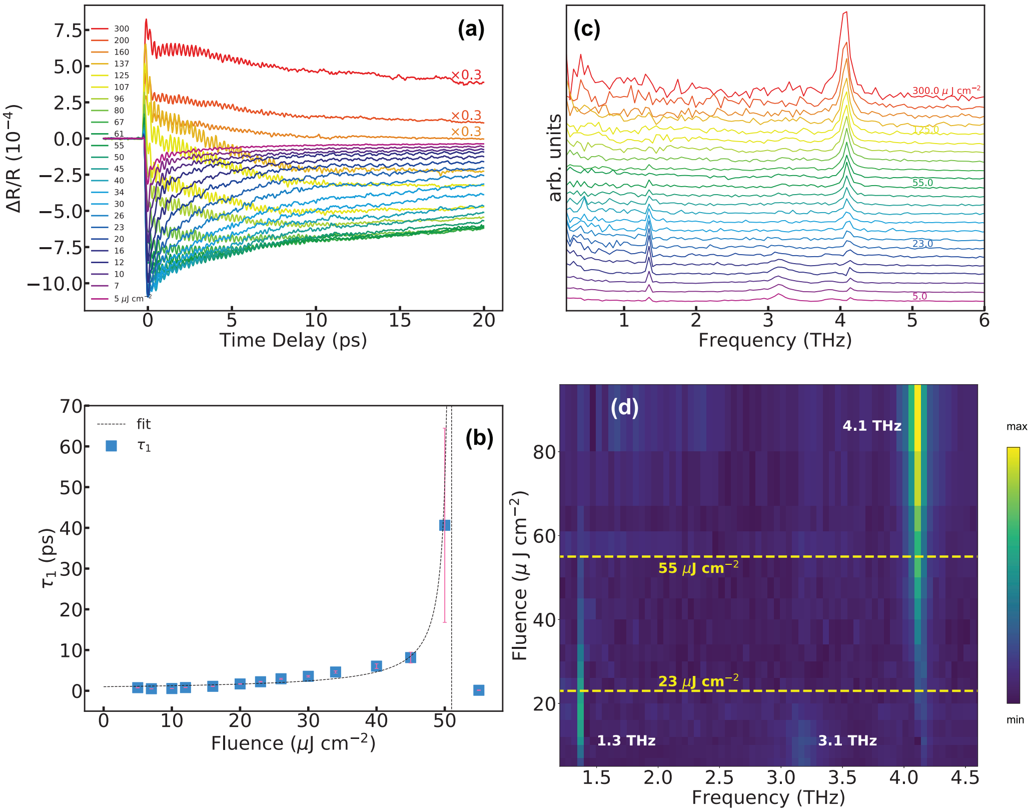

FIG. 3. Fluence-dependent pump-probe reflectivity signal of CsV_3Sb_5. (a) Photoinduced reflectivity $\Delta R/R$ as a function of time delay vs pump fluence in the range from 5 to 300 μJ cm^{-2} of CsV_3Sb_5 at 20 K. The data of the three highest fluence are scaled by a factor of 0.3. (b) Fluence dependence of decay time τ of the reflectivity transients extracted from fits to the double-exponential decay function. (c) The fast Fourier transformation of the oscillation parts by subtracting the decay background of $\Delta R/R$ in (a). Three resonance peaks at 1.3, 3.1, and 4.1 THz can be seen at the lowest fluence 5 μJ cm^{-2}. (d) Two-dimensional intensity map of these three resonance peaks.

our own reflectance measurement by a Fourier transform infrared spectrometer. With the initial temperature $T_0 = 20$ K and the temperature-dependent thermal capacity $C_p(T)$ being extracted from previous data [18], the values of ΔT are calculated to be approximately 10 and 19 K at fluences of 23 and 55 μJ cm^{-2}, respectively. From this estimation, the possibility of a thermal effect can be unambiguously ruled out.

The nonthermal melting at such a small pump fluence is another key finding in this work. Although photoinduced lattice symmetry changes or phase transitions have been observed in many different systems, the threshold fluences for completely changing the lattice potentials are usually much higher than the present measurement. For example, $TiSe_2$, a well-studied CDW system, undergoes a second-order phase transition on cooling to a CDW phase at 200 K. Photoexcitation at 80 K can melt the CDW order [45,46]. However, the required pump threshold fluence is ~0.21 mJ cm^{-2} for the 800 nm pump-probe measurement with a similar pulse duration <40 fs. If we consider the effect of penetration depth and convert to the energy per unit cell via $E_{\text{unit cell}} = F(1-R)/\delta \times M/\rho N_A$, the energy would be 8.8 meV per unit cell for CsV_3Sb_5, which is roughly half of the value ~16.5 meV for $TiSe_2$ [45]. Our measurement suggests that the lattice potential difference between those different phases is relatively small. The weak pump pulses can excite a sufficient number of electrons and their perturbation to the lattice potential is large enough to modify the symmetry. As a result, it drives the structural phase transition nonthermally. A recent x-ray scattering measurement [28] on CsV_3Sb_5 revealed that, while the CDW is long-range ordered, the integrated CDW superlattice peak intensity that is proportional to the CDW order parameter is extremely small. Comparing with the fundamental Bragg peaks, the CDW peak intensity is 3–5 orders of magnitude weaker, demonstrating small lattice distortions. We expect our result to be correlated with this observation. It may shed light on the energy difference between different lattice phases identified from the x-ray measurement.

V. SUMMARY

To summarize, laser pulses serve as a tool not only to probe the dynamic behavior but also to drive phase transitions in the kagome metal compound CsV_3Sb_5. Our measurement reveals a peculiar CDW phase transition, i.e., the quasiparticle relaxation dynamics can be explained by the formation of an energy gap below the phase transition being similar to a usual second-order CDW condensate, so the structure change is predominantly a first-order phase transition. Furthermore, no CDW amplitude mode can be identified in the ordered phase. We also show that even a small pumping fluence can nonthermally melt the C_2 ground state order and then C_6 CDW order successively and drive the compound into its high-temperature phase, suggesting that the lattice potential difference between those different phases is relatively small.

ACKNOWLEDGMENTS

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[1] A. Cavalleri, C. Tóth, C. W. Siders, J. A. Squier, F. Ráksi, P. Forget, and J. C. Kieffer, Phys. Rev. Lett. **87**, 237401 (2001).

[2] S. Tomimoto, S. Miyasaka, T. Ogasawara, H. Okamoto, and Y. Tokura, Phys. Rev. B **68**, 035106 (2003).

[3] H. Okamoto, Y. Ishige, S. Tanaka, H. Kishida, S. Iwai, and Y. Tokura, Phys. Rev. B **70**, 165202 (2004).

[4] N. Takubo, Y. Ogimoto, M. Nakamura, H. Tamaru, M. Izumi, and K. Miyano, Phys. Rev. Lett. **95**, 017404 (2005).

[5] M. Matsubara, Y. Okimoto, T. Ogasawara, Y. Tomioka, H. Okamoto, and Y. Tokura, Phys. Rev. Lett. **99**, 207401 (2007).

[6] A. Tomeljak, H. Schäfer, D. Städter, M. Beyer, K. Biljakovic, and J. Demsar, Phys. Rev. Lett. **102**, 066404 (2009).

[7] S. Wall, D. Wegkamp, L. Foglia, K. Appavoo, J. Nag, R. F. Haglund, J. StäCurrency Signhler, and M. Wolf, Nat. Commun. **3**, 721 (2012).

[8] L. Stojchevska, I. Vaskivskyi, T. Mertelj, P. Kusar, D. Svetin, S. Brazovskii, and D. Mihailovic, Science **344**, 177 (2014).

[9] C. Giannetti, M. Capone, D. Fausti, M. Fabrizio, F. Parmigiani, and D. Mihailovic, Adv. Phys. **65**, 58 (2016).

[10] M. Y. Zhang, Z. X. Wang, Y. N. Li, L. Y. Shi, D. Wu, T. Lin, S. J. Zhang, Y. Q. Liu, Q. M. Liu, J. Wang, T. Dong, and N. L. Wang, Phys. Rev. X **9**, 021036 (2019).

[11] E. J. Sie, C. M. Nyby, C. D. Pemmaraju, S. J. Park, X. Shen, J. Yang, M. C. Hoffmann, B. K. Ofori-Okai, R. Li, A. H. Reid, S. Weathersby, E. Mannebach, N. Finney, D. Rhodes, D. Chenet, A. Antony, L. Balicas, J. Hone, T. P. Devereaux, T. F. Heinz *et al.*, Nature (London) **565**, 61 (2019).

[12] T. F. Nova, A. S. Disa, M. Fechner, and A. Cavalleri, Science **364**, 1075 (2019).

[13] X. Li, T. Qiu, J. Zhang, E. Baldini, J. Lu, A. M. Rappe, and K. A. Nelson, Science **364**, 1079 (2019).

[14] A. S. McLeod, J. Zhang, M. Q. Gu, F. Jin, G. Zhang, K. W. Post, X. G. Zhao, A. J. Millis, W. B. Wu, J. M. Rondinelli, R. D. Averitt, and D. N. Basov, Nat. Mater. **19**, 397 (2020).

[15] A. Kogar, A. Zong, P. E. Dolgirev, X. Shen, J. Straquadine, Y.-Q. Bie, X. Wang, T. Rohwer, I.-C. Tung, Y. Yang, R. Li, J. Yang, S. Weathersby, S. Park, M. E. Kozina, E. J. Sie, H. Wen, P. Jarillo-Herrero, I. R. Fisher, X. Wang, and N. Gedik, Nat. Phys. **16**, 159 (2020).

[16] Q. M. Liu, D. Wu, Z. A. Li, L. Y. Shi, Z. X. Wang, S. J. Zhang, T. Lin, T. C. Hu, H. F. Tian, J. Q. Li, T. Dong, and N. L. Wang, Nat. Commun. **12**, 2050 (2021).

[17] B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M. McQueen, and E. S. Toberer, Phys. Rev. Materials **3**, 094407 (2019).

[18] B. R. Ortiz, S. M. L. Teicher, Y. Hu, J. L. Zuo, P. M. Sarte, E. C. Schueller, A. M. M. Abeykoon, M. J. Krogstad, S. Rosenkranz, R. Osborn, R. Seshadri, L. Balents, J. He, and S. D. Wilson, Phys. Rev. Lett. **125**, 247002 (2020).

[19] B. R. Ortiz, P. M. Sarte, E. M. Kenney, M. J. Graf, S. M. L. Teicher, R. Seshadri, and S. D. Wilson, Phys. Rev. Mater. **5**, 034801 (2021).

[20] Q. Yin, Z. Tu, C. Gong, Y. Fu, S. Yan, and H. Lei, Chin. Phys. Lett. **38**, 037403 (2021).

[21] H. Zhao, H. Li, B. R. Ortiz, S. M. L. Teicher, T. Park, M. Ye, Z. Wang, L. Balents, S. D. Wilson, and I. Zeljkovic, Nature (2021), doi: 10.1038/s41586-021-03946-w.

[22] K. Y. Chen, N. N. Wang, Q. W. Yin, Y. H. Gu, K. Jiang, Z. J. Tu, C. S. Gong, Y. Uwatoko, J. P. Sun, H. C. Lei, J. P. Hu, and J.-G. Cheng, Phys. Rev. Lett. **126**, 247001 (2021).

[23] F. Du, S. Luo, B. R. Ortiz, Y. Chen, W. Duan, D. Zhang, X. Lu, S. D. Wilson, Y. Song, and H. Yuan, Phys. Rev. B **103**, L220504 (2021).

[24] X. Chen, X. Zhan, X. Wang, J. Deng, X.-B. Liu, X. Chen, J.-G. Guo, and X. Chen, Chin. Phys. Lett. **38**, 057402 (2021).

[25] Z. Zhang, Z. Chen, Y. Zhou, Y. Yuan, S. Wang, J. Wang, H. Yang, C. An, L. Zhang, X. Zhu, Y. Zhou, X. Chen, J. Zhou, and Z. Yang, Phys. Rev. B **103**, 224513 (2021).

[26] Y. Wang, S. Yang, P. K. Sivakumar, B. R. Ortiz, S. M. L. Teicher, H. Wu, A. K. Srivastava, C. Garg, D. Liu, S. S. P. Parkin, E. S. Toberer, T. McQueen, S. D. Wilson, and M. N. Ali, arXiv:2012.05898.

[27] H. Tan, Y. Liu, Z. Wang, and B. Yan, Phys. Rev. Lett. **127**, 046401 (2021).

[28] H. X. Li, T. T. Zhang, T. Yilmaz, Y. Y. Pai, C. E. Marvinney, A. Said, Q. W. Yin, C. S. Gong, Z. J. Tu, E. Vescovo, R. G. Moore, S. Murakami, H. C. Lei, H. N. Lee, B. J. Lawrie, and H. Miao, Phys. Rev. X **11**, 031050 (2021).

[29] H. Chen, H. Yang, B. Hu, Z. Zhao, J. Yuan, Y. Xing, G. Qian, Z. Huang, G. Li, Y. Ye, Q. Yin, C. Gong, Z. Tu, H. Lei, S. Ma, H. Zhang, S. Ni, H. Tan, C. Shen, X. Dong *et al.*, Nature (2021), doi: 10.1038/s41586-021-03983-5.

[30] S. Ni, S. Ma, Y. Zhang, J. Yuan, H. Yang, Z. Lu, N. Wang, J. Sun, Z. Zhao, D. Li, S. Liu, H. Zhang, H. Chen, K. Jin, J. Cheng, L. Yu, F. Zhou, X. Dong, J. Hu, H.-J. Gao, and Z. Zhao, Chin. Phys. Lett. **38**, 057403 (2021).

[31] Y.-X. Jiang, J.-X. Yin, M. M. Denner, N. Shumiya, B. R. Ortiz, G. Xu, Z. Guguchia, J. He, M. S. Hossain, X. Liu, J. Ruff, L. Kautzsch, S. S. Zhang, G. Chang, I. Belopolski, Q. Zhang, T. A. Cochran, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, Z. Wang, R. Thomale, T. Neupert, S. D. Wilson, and M. Z. Hasan, Nat. Mater. **20**, 1353 (2021).

[32] Y. Hu, S. M. L. Teicher, B. R. Ortiz, Y. Luo, S. Peng, L. Huai, J. Z. Ma, N. C. Plumb, S. D. Wilson, J. F. He, and M. Shi, arXiv:2104.12725.

[33] S. Y. Yang, Y. Wang, B. R. Ortiz, D. Liu, J. Gayles, E. Derunova, R. Gonzalez-Hernandez, L. Šmejkal, Y. Chen, S. S. Parkin, S. D. Wilson, E. S. Toberer, T. McQueen, and M. N. Ali, Sci. Adv. **6**, eabb6003 (2020).

[34] F. H. Yu, T. Wu, Z. Y. Wang, B. Lei, W. Z. Zhuo, J. J. Ying, and X. H. Chen, Phys. Rev. B **104**, L041103 (2021).

[35] E. M. Kenney, B. R. Ortiz, C. Wang, S. D. Wilson, and M. J. Graf, J. Phys.: Condens. Matter **33**, 235801 (2021).

[36] Y. Xiang, Q. Li, Y. Li, W. Xie, H. Yang, Z. Wang, Y. Yao, and H.-H. Wen, arXiv:2104.06909.

[37] H. Li, H. Zhao, B. R. Ortiz, T. Park, M. Ye, L. Balents, Z. Wang, S. D. Wilson, and I. Zeljkovic, arXiv:2104.08209.

[38] N. Ratcliff, L. Hallett, B. R. Ortiz, S. D. Wilson, and J. W. Harter, arXiv:2104.10138.

[39] J. Demsar, L. Forró, H. Berger, and D. Mihailovic, Phys. Rev. B **66**, 041101(R) (2002).

[40] V. V. Kabanov, J. Demsar, B. Podobnik, and D. Mihailovic, Phys. Rev. B **59**, 1497 (1999).

[41] T. Lin, L. Y. Shi, Z. X. Wang, S. J. Zhang, Q. M. Liu, T. C. Hu, T. Dong, D. Wu, and N. L. Wang, Phys. Rev. B **101**, 205112 (2020).

[42] Z. Wang, S. Ma, Y. Zhang, H. Yang, Z. Zhao, Y. Ou, Y. Zhu, S. Ni, Z. Lu, H. Chen, K. Jiang, L. Yu, Y. Zhang, X. Dong, J. Hu, H.-J. Gao, and Z. Zhao, arXiv:2104.05556.

[43] R. Y. Chen, S. J. Zhang, M. Y. Zhang, T. Dong, and N. L. Wang, Phys. Rev. Lett. **118**, 107402 (2017).

[44] D. W. Song, L. X. Zheng, F. H. Yu, J. Li, L. P. Nie, M. Shan, D. Zhao, S. J. Li, B. L. Kang, Z. M. Wu, Y. B. Zhou, K. L. Sun, K. Liu, X. G. Luo, Z. Y. Wang, J. J. Ying, X. G. Wan, T. Wu, and X. H. Chen, arXiv:2104.09173.

[45] E. Möhr-Vorobeva, S. L. Johnson, P. Beaud, U. Staub, R. De Souza, C. Milne, G. Ingold, J. Demsar, H. Schaefer, and A. Titov, Phys. Rev. Lett. **107**, 036403 (2011).

[46] M. Porer, U. Leierseder, J.-M. Ménard, H. Dachraoui, L. Mouchliadis, I. E. Perakis, U. Heinzmann, J. Demsar, K. Rossnagel, and R. Huber, Nat. Mater. **13**, 857 (2014).