Ultrafast dynamics in the Lifshitz-type 5*d* pyrochlore antiferromagnet $Cd_2Os_2O_7$

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We investigate the ultrafast dynamics of $Cd_2Os_2O_7$, a prototype material showing a Lifshitz-type transition as a function of temperature. In the paramagnetic metallic state, the photoreflectivity shows a subpicosecond relaxation, followed by a featureless small offset. In the antiferromagnetic state slightly below T_N, however, the photoreflectivity resurges over hundreds of picoseconds, which goes beyond the usual realm of the effective-temperature model. Our observations are consistent with the Lifshitz phase transition of $Cd_2Os_2O_7$ driven by the evolution of the local magnetic moment.

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Lifshitz showed that a Fermi surface topology of a metal can change during continuous deformation under pressure [1]. The Lifshitz transition due to various instabilities can be accompanied by a metal-insulator transition (MIT) as a function of pressure or temperature without doping [1–4]. In addition, various physical properties can exhibit anomalies across the Lifshitz transition [1,3–10]. In most of cases, however, Lifshitz transitions have been obscured by the presence of other bands at the Fermi level across the transition and/or extrinsic effects due to doping [5–10]. Therefore, various aspects of the Lifshitz transition remain largely unexplored.

The 5*d* pyrochlore $Cd_2Os_2O_7$ is a prototype all-in-all-out (AIAO) antiferromagnetic material exhibiting the Lifshitz transition accompanied by MIT. Because its resistivity increases abruptly below the antiferromagnetic transition temperature $T_N = 227$ K, the MIT is believed to be closely related to the magnetic order. Earlier studies on 5*d* pyrochlore $Cd_2Os_2O_7$ claimed a Slater-type MIT that occurs concurrently with unit cell doubling due to the magnetic order [11,12]. However, the strong spin-orbit coupling of the osmium 5*d* orbitals drives AIAO antiferromagnetic ordering [13–15]. The lack of unit cell doubling in the AIAO spin structure rules out the possibility of a Slater-type transition [16,17]. Recent studies have suggested a Lifshitz-type MIT driven by the magnetic ordering. Temperature-dependent evolution of the magnetic moment is suggested to push electron and hole bands away from the Fermi level [2]. Therefore, the system provides a good testbed to explore the temperature-dependent evolution of various properties across the Lifshitz transition.

The interactions between quantum degrees of freedom such as charge, spin, and lattice have been investigated extensively using ultrafast techniques [18–31]. Effective-temperature model provides the simplest picture of nonequilibrium dynamics. Figure 1 shows a schematic diagram of the relaxation process based on the effective temperatures of the electron (T_E), strongly coupled phonon (T_{SCP}), spin (T_S), and total lattice (T_L) subsystems. The ultrafast pump excitation generates hot electrons in the excited states, thus elevating T_E. Then, the hot electrons cool down by dissipating their excess energy to the other subsystems. The most effective cooling channel is the scattering by the strongly coupled phonons (SCPs), which dominates the subpicosecond (ps) dynamics [20–24]. This scattering process can be modified by the magnetic order, which introduces the spin subsystem into the ps region [24–27]. Investigating such ps dynamics is crucial to understanding the interplay between the electron, SCP, and spin subsystems. We note that T_L remains close to the equilibrium temperature during this ps relaxation. The further heat transfer from the electron-SCP-spin subsystems to the lattice, i.e., the entire phonon system, takes place via the phonon-phonon or spin-phonon scattering over hundreds of ps or longer. After those scatterings, all the subsystems finally result in a quasiequilibrium state at a slightly elevated temperature. The total system heating after the equilibration is much smaller than the initial heating of the electron subsystem, because the specific heat of the electron subsystem is only a small proportion of the total specific heat. Hence, the nonequilibrium electronic signal after cooling over hundreds of ps should become negligible in comparison to the ps region. However, we find that the photoreflectivity of $Cd_2Os_2O_7$ contrasts with this seemingly trivial expectation.

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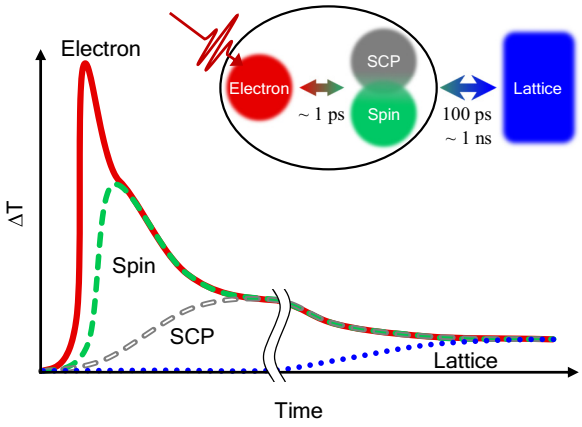

FIG. 1. Schematic diagrams of the relaxation dynamics in terms of the effective temperatures of the subsystems. Colored lines indicate the effective-temperature changes of the electron (solid line), spin (filled dashed line), strongly coupled phonon (SCP, open dashed line), and total lattice (dotted line) subsystems.

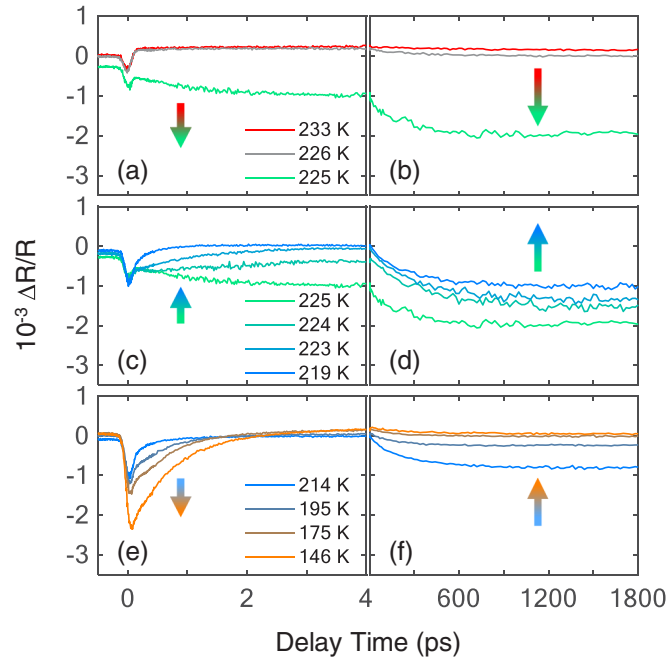

FIG. 2. Photoreflectivity $\Delta R/R$ in two temporal windows, spanning −0.5–4 ps (left panel), and 4–1800 ps (right panel) over temperatures from 146 to 233 K.

In this article we present the nonequilibrium dynamics of $Cd_2Os_2O_7$. In the antiferromagnetic state, the photoreflectivity initially shows the ps relaxation of the electrons. However, to our surprise, the photoreflectivity increases again during the successive electron cooling process. In particular, near T_N, the photoreflectivity after heating up the total system by 1 K in the nanosecond (ns) region is even larger than the initial value due to electron heating by more than 300 K. The huge resurgent behavior in the ns region suggests that an effective temperature of another subsystem has an exceptionally strong influence on the transient electronic structure. We discuss our observation in terms of the local magnetic moment in the process of the Lifshitz transition.

We measured the near-infrared photoreflectivity as a function of the pump and probe delay time (t_d) on $Cd_2Os_2O_7$ single crystals grown by the chemical transport method [32]. For the pump-probe measurements, we employed a commercial Ti:sapphire amplifier system, which generates pulses with a center energy of 1.55 eV and duration of 36 fs at a repetition rate of 250 kHz. For all measurements, the pump and probe fluences are set to 50 and 8.3 $\mu J/cm^2$, respectively. The spot sizes (full width half maximum) of the pump and probe beams are 84 and 44 μm, respectively. The pump and probe polarizations are perpendicular to each other to prevent the detection of scattered pump photons.

Figure 2 shows the temperature-dependent evolution of the photoreflectivity $\Delta R/R$ in two time windows: −0.5–4 ps (left panel) and 4–1800 ps (right panel). The ultrafast response above T_N is typical of a metal, which exhibits a sub-ps relaxation followed by a small offset [Figs. 2(a) and 2(b)]. As already discussed in Fig. 1, the sub-ps relaxation corresponds to the hot electron cooling process via scattering with the SCPs [20–24]. The finite offset lasting beyond the measurement time window is due to the heating of SCPs. Successive phonon-phonon scattering further cools down the electron and SCPs systems and also diffuses the excess heat out of the pump excited volume [22].

Figures 2(c) and 2(e) show that additional, richly varied ps dynamics appears below T_N, and is much slower than the sub-ps dynamics that dominates above T_N. In the magnetic state, the electron-spin scattering may provide an additional cooling path for hot electrons. Once the effective temperature of spin subsystem T_S has reached the same temperature as T_E, however, successive cooling of hot electrons should be accompanied by the cooling of the spin subsystem. This results in the slower relaxation dynamics of $Cd_2Os_2O_7$ below T_N. Furthermore, the magnetic order could restrict the phase space of the electron-phonon scattering. This also explains the slower relaxation dynamics in the magnetic state. A more quantitative understanding of the ps dynamics in the magnetic state requires us to consider the spin subsystem as well as the electron and SCP subsystems in terms of the effective temperatures [24,27].

Unexpected is the result shown in Figs. 2(d) and 2(f) that the photoreflectivity below T_N resurges over hundreds of ps. The photoreflectivity in the ns region is so unusually large that it becomes comparable to or even larger than the initial photoreflectivity. The large photoreflectivity in the ns region shows up abruptly below T_N. Hence the magnetic order should contribute to this behavior. In fact, the spin-lattice relaxation is frequently observed in the ns region. When the spin-charge coupling is weak, however, the influence of T_S on the electronic structure is marginal and the transient reflectivity does not show a significant change unless the heated magnetic state bears a resonance to the probe photon energy as in the case of $Eu_2Fe_2As_2$ [33]. The Os^{5+} d-electron spins in $Cd_2Os_2O_7$ are known to be strongly coupled with electronic structure [2,15,34,35]. Therefore, it is expected that the spin subsystem already plays a role in the ps region as discussed in the previous paragraph. In other words, T_S as well as T_E must monotonically decrease beyond 100 ps to reach the equilibrium state. Therefore, within the usual

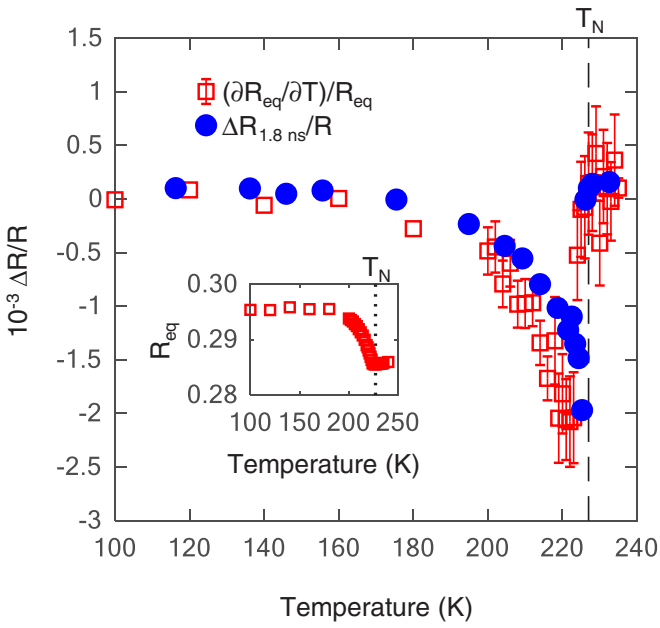

FIG. 3. Temperature-dependent evolution of $\Delta R/R$ (circles) measured at $t_d = 1.8$ ns and $(\partial R_{\rm eq}/\partial T)/R_{\rm eq}$ (squares) obtained from the equilibrium reflectivity $R_{\rm eq}$. Inset: The $R_{\rm eq}$ at 1.55 eV measured by spectroscopic ellipsometry at 20 K intervals from 100 to 200 K, and at 1 K intervals from 200 to 235 K.

effective-temperature model, we cannot explain the huge resurgence in photoreflectivity in the ns region in terms of T_S.

The simple thermodynamic simulation of the change in equilibrium reflectivity confirms that the resurgence in the reflectivity change can be attributed to the heating of the whole system. We assume that the injected pump energy is initially absorbed entirely by the electron subsystem. When the base temperature prior to pumping is close to T_N, we find that the pump excitation should heat up the electron subsystem by more than 300 K. After the heat has dissipated to all of the other subsystems, however, the temperature of the entire system is estimated to increase by only 1 K from the base temperature (see the Supplemental Material for further details [36]). In Fig. 3 we compare the photoreflectivity at $t_d = 1.8$ ns ($\Delta R_{1.8\rm ns}/R$) with the thermomodulation of the reflectivity $(\partial R_{\rm eq}/\partial T)/R_{\rm eq}$. Note that the thermomodulation corresponds to the change in reflectivity due to temperature increment by 1 K in the equilibrium state. The good agreement between $\Delta R_{1.8\rm ns}/R$ and $(\partial R_{\rm eq}/\partial T)/R_{\rm eq}$ suggests that the system indeed reaches a quasiequilibrium state at $t_d = 1.8$ ns, where the temperature of all subsystems is slightly elevated by 1 K, as shown in Fig. 1.

As $\Delta R_{1.8\rm ns}/R$ can be explained in terms of the equilibrium reflectivity ($R_{\rm eq}$), the large value of $\Delta R_{1.8\rm ns}/R$ slightly below T_N may seem trivial following from the temperature-dependent evolution in the equilibrium state. However, it is peculiar that small heating in the whole system induces a huge transient reflectivity comparable to or even larger than ΔR due to the electron heating. We note that the reflectivity is directly determined by the electronic structure of the material. As far as we know, all the reports based on the effective-temperature model have assumed that the ultrafast photoreflectivity depends only on T_E [20–24]. Within the effective-temperature model, it has never been expected that the huge resurgence in photoreflectivity may occur due to the tiny heating after completion of the ps relaxation to $\Delta R/R \sim 0$.

There are other examples of a large transient electronic response in the ns region. In superconductors, the photoinduced reflectivity change may increase over hundreds of ps under weak pump excitation [18,19]. This slow response is due to the small superconducting pairing energy and can be well understood in terms of the quasiparticle dynamics. Some manganite systems also exhibit the relaxation dynamics over hundreds of ps with a large transient optical response, comparable to that observed in the ps region [37–39]. It has been suggested that manganite systems enter a photoexcited metastable state, which does not correspond to any equilibrium state at elevated temperatures [39]. These are different from the case of $Cd_2Os_2O_7$. As far as we know, there is no other example where the ns response comparable to the ps response in photoreflectivity is induced by the tiny heating in the quasiequilibrium state.

$Cd_2Os_2O_7$ is a unique system, in that its photoreflectivity in the ns region is not explained only with T_E. In the nonequilibrium state, the heating of any subsystem may affect the photoinduced signal, such that

$$\Delta I = A_E \Delta T_E + A_{\rm SCP} \Delta T_{\rm SCP} + A_S \Delta T_S + A_L \Delta T_L, \quad (1)$$

where ΔT_E, $\Delta T_{\rm SCP}$, ΔT_S, and ΔT_L are the effective-temperature changes of electron, SCP, spin, and lattice subsystems. The coefficients A_E, $A_{\rm SCP}$, A_S, and A_L are evaluated based on what is measured as the photoinduced signal. Although these coefficients may depend on the equilibrium temperature, they can be regarded as constants under small temperature variation. Reflectivity is directly determined by the electronic structure and ΔT_E is much larger than the other effective-temperature changes right after pumping. The reflectivity change, therefore, has been considered to be dominated by the electron subsystem, such that $\Delta R/R \sim A_E \Delta T_E$. However, the resurgence in the photoreflectivity of $Cd_2Os_2O_7$ cannot be explained only by ΔT_E, because the second law of thermodynamics states that T_E must continue to decrease after ps relaxation. To explain the resurgent behavior over hundreds of ps, another effective temperature other than T_E should be considered. Note that $\Delta T_{\rm SCP}$ and ΔT_S already play roles in the ps region as discussed with Figs. 2(c) and 2(e). Therefore, the remaining term is the total lattice system ΔT_L within Eq. (1). We note that the photoreflectivity could be governed by other unknown subsystems not included in Eq. (1) instead of ΔT_L. However, the dynamics shown in Figs. 2(d) and 2(f) is well described by a single exponential component and the effective temperatures of all subsystems are quasiequilibrated in the ns region. Therefore, we may use ΔT_L to stand for the subsystem playing the important role. Considering $\Delta T_E^{\rm max} \sim 300$ K, $\Delta T_L^{\rm max} \sim 1$ K (see the Supplemental Material [36]), and the value of $\Delta R_{1.8\rm ns}/R$ comparable to the initial electronic response below T_N, we find that the additional effective temperature coined by ΔT_L has an extremely strong influence, such that $\Delta R/R \sim A_E \Delta T_E + A_L \Delta T_L$ with $A_L/A_E \sim 300$ near T_N.

Why should ΔT_L exert such a strong influence over the electronic structure of $Cd_2Os_2O_7$? What should be modified

by ΔT_L? Although the lattice shows a slight thermal expansion, our density functional theory calculations find that the effect of lattice expansion on the band structure is negligible (see the Supplemental Material for details of this calculation [36]). Instead, it is expected that the local magnetic moment of osmium m_{Os}, especially the longitudinal fluctuations, which is not included in ΔT_S, may play a certain role for the modification of electronic structure in the ns region [34,40]. Note that the band structure of $Cd_2Os_2O_7$ strongly depends on the local magnetic moment of osmium m_{Os}. The local spin density approximation calculations by Shinaoka *et al.* revealed that $Cd_2Os_2O_7$ undergoes a Lifshitz-type MIT as the electron correlation strength U varies [2]. This corresponds to that m_{Os} decreases as temperature increases and induces the MIT as well as the AIAO magnetic transition in $Cd_2Os_2O_7$ [2,34,41]. We conjecture that the longitudinal fluctuations of m_{Os} may explain the observed slow dynamics coined by ΔT_L. Unfortunately, it is still unveiled that how the heating of the total lattice modifies m_{Os}, which in turn strongly influence the electronic structure in the ns region. The temperature-dependent subtle evolution of the effective correlation strength could be more important than a direct interaction between the lattice and m_{Os}. Further studies are required to understand the coupling between the lattice temperature and m_{Os} and its role on the Lifshitz transition in $Cd_2Os_2O_7$.

Finally, we briefly discuss the phase transition behaviors of the ps relaxation dynamics in Figs. 2(c) and 2(e). We obtained the temperature dependence of the relaxation time (τ_2) and the amplitude (A_2) of the ps component that shows up below T_N in detail by fitting our data to a biexponential decay model in the 10 ps region:

$$\Delta R/R(t_d) = A_1 e^{-t_d/\tau_1} + A_2 e^{-t_d/\tau_2} + C, \qquad (2)$$

where τ_1 and A_1 are the relaxation time and amplitude of the sub-ps component, respectively, and C is a constant offset. While the sub-ps component of τ_1 ~0.1 ps does not vary significantly, the ps component exhibits a strong temperature dependence, as shown in Fig. 4 (see the Supplemental Material for details of the fitting process [36]). τ_2 exhibits critical slowing down behavior as it approaches T_N due to the AIAO phase transition [26,27]. It suggests that the excitation and recovery of the magnetic order with a given local magnetic moment m_{Os} take place in this ps time window. The saturation behavior of the ps component under high fluence at low temperature well below T_N also supports that the ps relaxation reflects a dynamics of an order parameter, while the ns one shows a linear response to the fluence (see the Supplemental Material for further details [36]).

Figure 4 shows another anomaly in the τ_2 and the A_2 at approximately T^* ~216 K. It has been claimed that this system exhibits a Lifshitz MIT at T^* that is separate from the AIAO magnetic phase transition at T_N [2,15,42]. As mentioned above, the Lifshitz transition is driven by the correlation effect as a function of m_{Os}. In other words, the energy gap in the insulating state should open between the electron

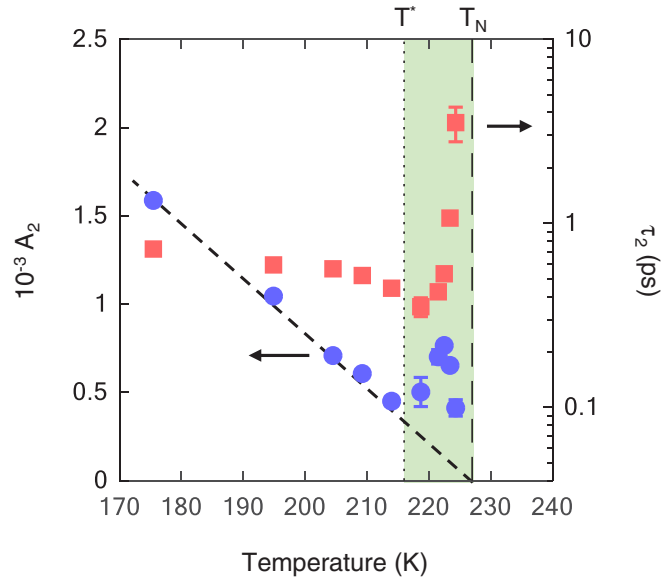

FIG. 4. Temperature dependence of the relaxation time τ_2 (squares) and amplitude A_2 (circles) of the ps components below T_N, based on biexponential fitting to the photoreflectivity $\Delta R/R$.

correlation-induced bands. In contrast to the slow electronic relaxation by recombination process in semiconductors and band gap insulators over more than 100 ps [43,44], the relaxation of correlated insulators has been reported to take place on the ps timescale, which is comparable to the relaxation of their metallic state [45–48]. Therefore, the gradual evolution of the carrier relaxation dynamics across T^* is consistent with the correlation-induced nature of the Lifshitz transition in $Cd_2Os_2O_7$.

In summary, we performed pump-probe reflectivity measurements on $5d$ pyrochlore $Cd_2Os_2O_7$, a prototype material exhibiting the Lifshitz transition. The nonequilibrium dynamics reveals an exceptionally large resurgence in the photoreflectivity, over hundreds of ps. It shows that a slight heating of the total system has a huge influence on the electronic structure much stronger than the heating of electrons. These findings are consistent with the Lifshitz-type transition controlled by the local magnetic moment of osmium m_{Os} that depends sensitively on temperature. The additional anomaly in the ps dynamics at T^* ~216 K supports that the Lifshitz-type MIT occurs below T_N. Our study of the prototypical response of the correlation-induced Lifshitz transition will provide a guide to understanding the ultrafast dynamics of correlated systems that exhibit emergent phenomena.

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