Letter

Unveiling the hybridization process in a weakly hybridized ferromagnet by ultrafast optical spectroscopy

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Utilizing ultrafast optical pump-probe spectroscopy, we present a comprehensive investigation on the photoexcited quasiparticle dynamics in single-crystal $CeAgSb_2$. We elucidate the emergence of collective c-f hybridization between conduction electrons and localized f moments below a critical temperature T^* (≈65 K), as indicated by the onset of an indirect gap of ∼9 meV via the fluence-dependent quasiparticle relaxation and phonon renormalization processes. Our experimental data also suggest that the c-f hybridization fluctuation should appear at a much higher-temperature value, i.e., $T^\dagger$ (≈180 K). Below ∼50 K, we further detect an additional quasiparticle relaxation channel with sub-ps timescale, which can be attributed to the crystalline electric field (CEF) excitation. These observations provide important insights into the Kondo lattice coherence and CEF effect in $CeAgSb_2$, shedding light on its unique properties and behaviors.

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Heavy fermion systems, also known as heavy fermion metals or Kondo lattice systems, constitute a unique class of materials within the realm of condensed matter physics [1,2]. These materials display intriguing electronic properties at low temperatures, marked by a pronounced increase in the effective mass of quasiparticles, strong electron-electron correlations, and unconventional superconductivity or magnetic phenomena [3]. The Kondo effect, occurring at individual atomic sites, fosters a hybridization between localized f electrons and itinerant conduction electrons, culminating in the formation of Kondo spin-singlet many-body states that effectively screen the localized magnetic moments. Concurrently, the localized f moments, facilitated by the conduction electrons through the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism, typically exhibit ordering, often in an antiferromagnetic order, at sufficiently low temperatures. The interplay of these interactions is delineated in the Doniach phase diagram, which dictates the system's ground state [4]. The emergence of heavy fermions is closely tied to the interaction between localized f moments and itinerant conduction electrons, with the Kondo effect playing a pivotal role in the hybridization between these two types of electrons [5]. Nevertheless, the evolution of Kondo lattice coherence with decreasing temperature, specifically the hierarchy of energy scales, continues to be a contentious topic. The coherence temperature T_{coh} and the single-ion Kondo temperature T_K, which can markedly diverge, are commonly utilized to delineate the initiation of heavy fermion characteristics [6]. Recent experimental findings suggest that the hybridization process or the development of Kondo lattice coherence in certain cerium-based materials is complex, with a distinct evolution at low and high temperatures [7–10].

In this Letter, we focus on an interesting heavy fermion compound [11,12], layered $CeAgSb_2$, which crystallizes in the tetragonal $ZrCuSi_2$-type structure, corresponding to the space group $P4/nmm$ [13–15]. $CeAgSb_2$ is categorized as a weakly hybridized ferromagnetic Kondo lattice [16], adding complexity to the interplay between Kondo physics and the RKKY interaction. For instance, the transition temperature, characterized by a maximum in zero-field resistivity or a sharp change in its slope, does not coincide with the Kondo lattice coherence temperature [14]. An analysis of susceptibility through multiplet states indicates a Kondo temperature T_K of approximately 65 K [17]. However, alternative methodologies, including inelastic neutron scattering and thermopower experiments, suggest a broader range for T_K, approximately 60–80 K, aligning with findings from muon spin relaxation studies [18,19]. An alternative perspective, based on the relationship between the Wilson ratio and the Kondo temperature for a doublet ground state, proposes a T_K value around 23 K [17]. This leads to speculation regarding the intricate relationship between ferromagnetic ordering and the Kondo effect, suggesting that T_K might lie within the range of 10–20 K [16]. In addition, no anomaly was found in the specific heat data of $CeAgSb_2$ above the ferromagnetic phase transition temperature [12,20]. Consequently, its associated Kondo energy scale remains elusive and is not yet fully comprehended.

Given the uncertainties surrounding the temperature at which Kondo lattice coherence occurs, it is essential to identify alternative parameters that can elucidate the temperature-dependent evolution of the c-f hybridization in $CeAgSb_2$. Recently, ultrafast optical pump-probe spectroscopy has emerged as a valuable technique for examining the dynamics of f-electron hybridization in heavy fermion systems across a

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wide temperature range [8–10]. This method offers a unique investigative approach, enabling the determination of the coherence temperature—a pivotal temperature scale that marks the emergence of collective c-f hybridization [21]. Notably, previous studies employing this technique have successfully resolved discrepancies in understanding the underlying mechanisms in heavy fermion systems, as exemplified by the case of $CeCoIn_5$ [8].

In this study, we utilize ultrafast optical pump-probe spectroscopy to investigate the c-f hybridization in $CeAgSb_2$. Our measurements have uncovered a pronounced anomaly in the quasiparticle relaxation time, denoted as τ_1, near the critical temperature $T^\dagger$ (≈180 K). Below T^* (≈65 K), the fluence dependence of τ_1 becomes conspicuous. On the findings of prior research, our observations yield two significant inferences: (1) the emergence of a heavy electron state, which is concurrent with the formation of an indirect hybridization gap as the temperature descends below T^*; and (2) the manifestation of precursor hybridization fluctuations within the temperature interval spanning from $T^\dagger$ to T^*. Additionally, we have identified a quasiparticle relaxation process occurring on a subpicosecond timescale below approximately 50 K, which may be ascribed to the splitting of Ce 4f energy levels induced by the crystalline electric field (CEF) effects in the vicinity of the Fermi surface.

We conducted ultrafast time-resolved differential reflectivity measurements, denoted as $\Delta R(t)/R$, on single-crystal $CeAgSb_2$ over a temperature range from 5 K to room temperature. The measurements utilized a Yb femtosecond laser, which delivers pulses with a duration of approximately 50 fs at a central wavelength of 1030 nm. This corresponds to a photon energy of around 1.2 eV, a choice that circumvents the intricacies of quasiparticle dynamics that could arise from interband transitions at alternative excitation wavelengths or photon energies, such as 800 nm or approximately 1.55 eV (refer to the Supplemental Material [22]). The typical transient reflectivity $\Delta R(t)/R$ as a function of the probe delay time is depicted in Figs. 1(a) and 1(b). Further experimental details and data are available in the Supplemental Material [22] (see also Refs. [14,23–26] therein).

Following photoexcitation, an instantaneous increase is clearly observable at all examined temperatures, closely tracking the cross-correlation profile of the pump and probe pulses. Superimposed on the relaxation process is a subtle oscillatory response. Generally, the decay component of the $\Delta R(t)/R$ signal within the initial 10 ps can be ascribed to electron-electron (e-e) and electron-boson scattering processes that are characteristic of strongly correlated and metalliclike systems [27,28]. The bosons implicated in these processes may encompass phonons or other bosonic excitations [29,30].

We initially concentrate on the nonoscillatory aspect of the signal. As depicted in Figs. 1(c) and 1(d), the initial decay, occurring within approximately 6 ps, can be modeled using the subsequent function [31,32]

$$\Delta R(t)/R = \left(A_1 e^{-\tau_1/t} + A_2 e^{-\tau_2/t} + C\right) \otimes G(t), \quad (1)$$

where A_i and τ_i denote the amplitude and decay time, respectively. C is a time-independent constant. $G(t)$ is a Gaussian function standing for the pump-probe cross correlation. Note that the process characterized by τ_2 only appears below

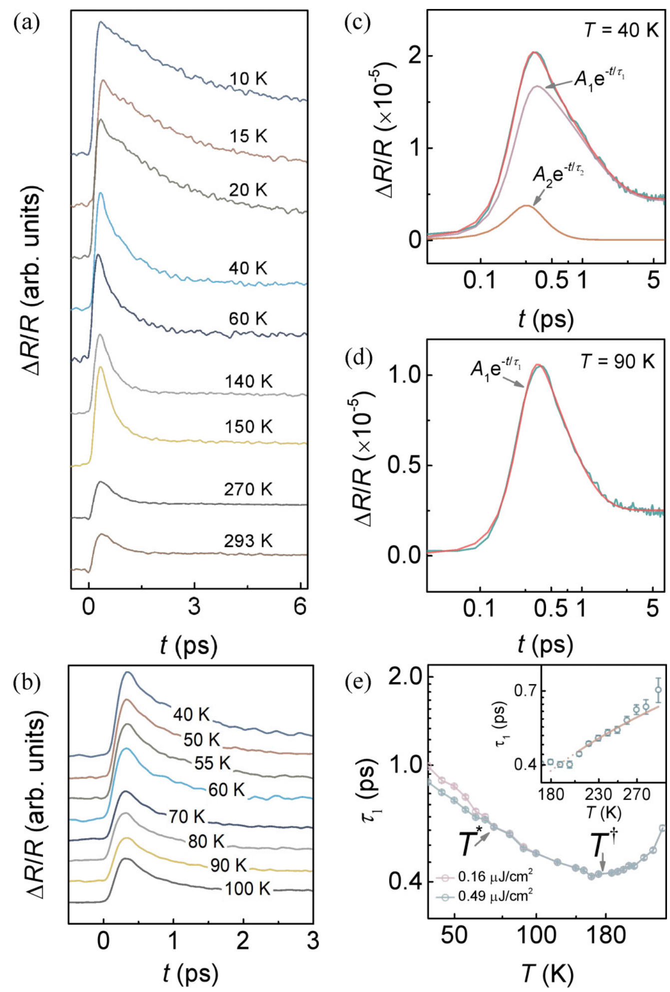

FIG. 1. (a) and (b) Time-resolved differential reflectivity $\Delta R(t)/R$ as a function of temperature in $CeAgSb_2$. (c) and (d) The experimental (green lines) and fitting results of $\Delta R(t)/R$ using Eq. (1) at 40 and 90 K, respectively. (e) Decay time τ_1 as a function of temperature at various pump fluences. Two critical temperatures, T^* and $T^\dagger$, can be defined. The temperature evolution of τ_1 from 180 to 293 K is shown in the inset, where the red line is a fitting due to the nonthermal process described in the main text.

~50 K, which means $A_2 = 0$ for $T > 50$ K, as demonstrated by Figs. 1(c) and 1(d). This process will be discussed later.

Figure 1(e) shows the temperature-dependent τ_1. Clearly, below a critical value of $T^* \approx 65$ K, $\tau_1(T)$ presents a fluence-dependent behavior within the experimental errors. Different from the previous works [8,9], the T^* of $CeAgSb_2$ here is much larger than the value of ~9.7 K, at which the ferromagnetic transition emerges, or the value of ~18 K where the resistivity reaches the maximum [14].

It has been demonstrated that the fluence-dependent behavior observed below T^* in heavy fermion systems can be effectively elucidated by the Rothwarf-Taylor (RT) model [8–10,33]. Specifically, the relaxation of excited quasiparticles, which possess an energy exceeding the energy gap ($\hbar\omega > 2\Delta$), is facilitated through the emission of high-frequency bosons. These bosons are capable of subsequently reexciting electron-hole pairs, as originally formulated by

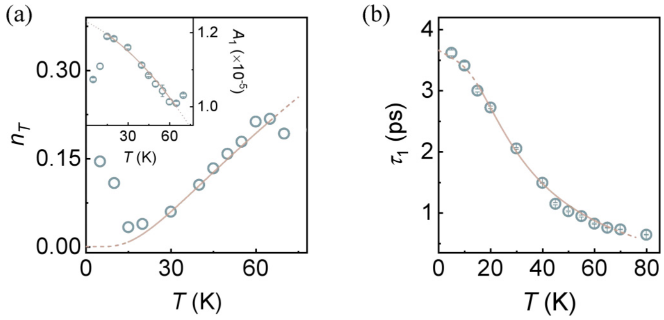

FIG. 2. (a) The density of thermally excited quasiparticles n_T as a function of temperature below T^*. The inset shows the temperature-dependent amplitude A_1. (b) Decay time τ_1 as a function of temperature below T^*. The solid lines are the fitting results using the RT model.

Refs. [33,34],

$$\frac{dn}{dt} = \beta N - Rn^2,$$
$$\frac{dN}{dt} = \frac{1}{2}[Rn^2 - \beta N] - (N - N_T)\tau_\gamma^{-1}, \tag{2}$$

where n is the total number of quasiparticles, R is the recombination rate of electron-hole pairs, N is the density of high-frequency bosons with an energy larger than 2Δ, β is the probability per unit time for generating the nonequilibrium quasiparticles by such bosons, τ_γ^{-1} is the escaping rate of the high-frequency bosons, and N_T is the thermal-equilibrium boson density. As long as R or τ_γ^{-1} is large enough [34], the quasiparticle relaxation dynamics will be dominated by the bimolecular recombination and display a clear fluence-dependent behavior. The gap formation in the density of states (DOS) can be studied by fitting $\tau(T)$ and $n_T(T)$ using the following equations [35–37],

$$\tau^{-1}(T) \propto \left[\frac{\delta}{\zeta n_T + 1} + 2n_T\right](\Delta + \alpha T \Delta^4),$$
$$n_T(T) = \frac{A(0)}{A(T)} - 1 \propto (T\Delta)^p e^{-\Delta/T}, \tag{3}$$

where α, ζ, and δ are fitting parameters, respectively. n_T is the density of quasiparticles thermally excited across the gap and p $(0 < p < 1)$ is a constant determined by the shape of gapped DOS [38]. For a typical DOS of the Bardeen-Cooper-Schrieffer (BCS) form, we may fix $p = 0.5$ and obtain a good fit as shown in Fig. 2. This yields a gap of $2\Delta \approx 9$ meV, indicating the formation of an indirect hybridization gap below T^* due to collective hybridization associated with the emergence of coherent heavy electron states near E_F. Due to the free-carrier absorption in the infrared spectroscopy and the requirement of large photon energies for achieving enough large electron scattering cross sections in angle-resolved photoemission spectroscopy (ARPES), such a small indirect gap is extremely difficult to be revealed by these two experimental techniques [8–10,39,40].

Above T^*, the fluence independence of τ_1 signals the disappearance of the indirect hybridization gap via the Kondo lattice coherence. The behavior of $\tau_1(T)$ can be further separated into two distinct regimes. For temperatures exceeding $T^\dagger$ ($\approx$180 K), τ_1 undergoes an increment with rising temperature. Such T dependence defies explanation through a conventional two-temperature model [28,41]. Instead, it points towards a nonthermal process, where the relaxation time due to electron-electron collisions ($\tau_{\text{e-e}}$) may exceed the electron-phonon relaxation time ($\tau_{\text{e-ph}}$) [28]. Alternatively, it suggests that the thermal distribution resulting from electron-electron scatterings is not instantaneously achieved, despite the scattered fermionic quasiparticles approaching near the Fermi energy (E_F). Such a process can be described by a nonequilibrium model [28], $1/\tau_{\text{e-ph}} = 3\hbar\lambda\langle\omega^2\rangle/(2\pi k_B T_l)$. Here, $\lambda\langle\omega^2\rangle$ denotes the electron-phonon coupling, and T_l signifies the lattice temperature. Indeed, if we assume $\tau_1 = \tau_{\text{e-ph}}$ in the high-temperature regime, specifically above $\sim$180 K, the model fits very well with the temperature-dependent τ_1, as illustrated in the inset of Fig. 1(e).

Within the temperature interval spanning from T^* to $T^\dagger$, the decay time τ_1 exhibits an increase as the temperature declines. Similar temperature-dependent behavior has been documented in various other heavy fermion systems, such as $CeCoIn_5$, $CeRh_6Ge_4$, and single-crystal cerium [8–10]. This observation implies that an indirect hybridization gap may not be present. Concurrently, precursor hybridization fluctuations, which manifest on a brief correlation timescale or length scale, are thought to be responsible for the increase in decay time τ_1 (or the reduction in decay rate $1/\gamma$) observed below $T^\dagger$ [42]. Specifically, both electron-electron (e-e) and electron-boson scattering processes are expected to diminish as the conduction electrons become engaged in hybridization with the fluctuating f moments. Consequently, our findings indicate that the f moments remain coupled to the conduction electrons even within the high-temperature regime.

In contrast to previous studies, our work uncovers a different subpicosecond decay process, characterized by τ_2, which emerges below a critical temperature T_{CEF} ($\approx$50 K) in the quasiparticle dynamics of $CeAgSb_2$ (refer to the Supplemental Material [22]). Notably, T_{CEF} closely approximates the energy scale associated with the first excited crystalline field splitting of the Ce $4f$ level. The CEF effect induces a differentiation of the originally sixfold-degenerate $4f$ energy levels into three Kramers doublets, with excitation energies Δ_{CEF1} and Δ_{CEF2} corresponding to transitions from the ground state to the first and second excited states, respectively. This CEF-induced splitting has been demonstrated to be pivotal in establishing the magnetic ground state of $CeAgSb_2$ [12,17]. An analysis predicated on magnetic entropy suggests that $\Delta_{\text{CEF1}} \sim 48$ K [12]. The existence of such an excited CEF splitting level could potentially trigger an orbital crossover [20], thereby introducing an additional relaxation pathway for nonequilibrium quasiparticles. At elevated temperatures, the pertinent states become populated by thermally excited electrons, which results in the cessation of the τ_2 process above 50 K.

In terms of the oscillatory component, a satisfactory signal-to-noise ratio was achieved when the photon energy was adjusted to approximately 1.55 eV, which is the setting used for the subsequent analysis. Figure 3(a) displays the oscillatory components with the nonoscillatory background subtracted. Two distinct modes in the terahertz (THz) frequency range are evident at all examined temperatures,

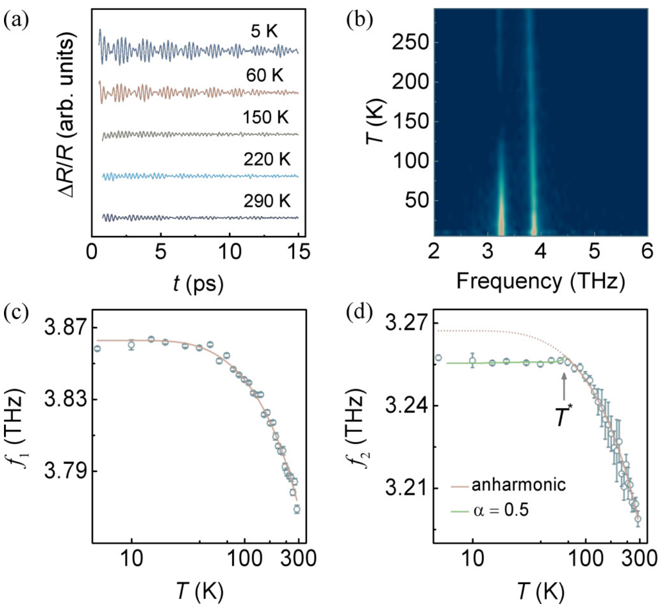

FIG. 3. (a) Extracted oscillations at some typical temperatures. (b) The fast Fourier transform spectra of oscillations in the frequency domain. (c), (d) The derived $f_j = \omega_j/2\pi$ $(j = 1, 2)$ as a function of temperature using Eq. (4). The red lines are the fitting curves via the anharmonic phonon decay model, and the green line only considers the contribution of Kondo singlets, as described in the main text.

specifically at around 3.8 and 3.2 THz. These modes are typically linked to the coherent optical phonon modes arising from the photoinduced Raman process or displacive excitation [8–10]. The oscillations can be quantitatively characterized using the following formula,

$$(\Delta R/R)_{\text{osc}} = \sum_{j=1,2} A_j e^{-\Gamma_j t} \sin(\Omega_j t + \phi_j), \tag{4}$$

where A_j, Γ_j, Ω_j, and ϕ_j $(j = 1, 2)$ are the amplitude, damping rate, frequency, and phase, respectively. Ω_j and ϕ_j are related to an underdamped harmonic oscillator. $\Omega_j = \sqrt{\omega_j^2 - \Gamma_j^2}$, where ω_j $(= 2\pi f_j)$ is the natural frequency.

Figures 3(c) and 3(d) show the temperature dependence of f_j. $f_1(T)$ aligns well with the anharmonic phonon decay model [8,24,25], and the same for T-dependent f_2 at $T > T^*$ as well (see Supplemental Material for fitting details [22]). Below T^*, exotic behavior is found for the f_2 mode, which exhibits a notable softening compared with the anharmonic model. Clearly, the anomaly around T^* cannot be attributed to the phonon-magnon coupling effect, since the long-range ferromagnetic ordering in $CeAgSb_2$ only emerges below the Curie temperature of ∼9.7 K [12].

To quantitatively apprehend the behavior of f_2 below T^*, we determined the values of δf_2, which denote the discrepancy between the predicted values from the anharmonic model and the experimentally observed f_2. This deviation is posited to stem from the Kondo lattice coherence—or the collective hybridization [8,26], $\delta f_j \propto \langle b_i \rangle^\alpha$, where the density of Kondo singlets $\langle b_i \rangle$ is proportional to $[1 - n_T(T)/n_T(T^*)]$. Utilizing this formula, an optimal fit with a fitting parameter $\alpha = 0.5$ was achieved. The observed frequency softening thus offers additional substantiation that coherent heavy electron states manifest below T^*.

Our findings in $CeAgSb_2$ distinctly demonstrate that collective c-f hybridization, or Kondo lattice coherence, should emerge below $T^* \approx 65$ K, accompanied by the formation of an indirect hybridization gap of approximately 9 meV, although precursor hybridization fluctuations might manifest at significantly higher temperatures, potentially up to ∼180 K, these results closely parallel those observed in other Ce-based heavy fermion systems [8–10], underscoring the universality of these hybridization phenomena across various materials. Consequently, we anticipate that band bending near the Fermi level E_F in the electronic structure of $CeAgSb_2$ should be evident at considerably high temperatures, specifically above 100 K. This expectation could be substantiated by future ARPES experiments.

Notably, the zero-field resistivity $\rho(T)$ exhibits a peak at approximately 18 K, a temperature significantly lower than T^*. This observation contrasts with that in other heavy fermion systems, such as $CeCoIn_5$ and $CeRh_6Ge_4$, where these temperatures are aligned. Such a discrepancy is likely a consequence of the complex interplay between the Kondo interaction and the RKKY interaction. In this context, the RKKY interaction may exert a more pronounced influence, contributing to the weakly hybridized nature of the system, as 18 K is much closer to the ferromagnetic ordering temperature (approximately 9.7 K) than T^* (≈65 K) [12,17]. Our findings indicate that the phonon frequency f_2 is highly sensitive to collective hybridization. Consequently, if hybridization were significantly dampened by ferromagnetic ordering, an additional phonon anomaly would be expected around the Curie temperature. However, no such anomalous behavior was detected. This absence of an anomaly suggests that the ferromagnetic order is not tightly coupled to the lattice degrees of freedom. Therefore, the abrupt change in the resistivity slope $d\rho/dT$, observed at the Curie temperature, is likely primarily indicative of a reduction in spin-disorder scattering due to the emergence of ferromagnetism.

Our research offers insights into the c-f hybridization process and the Kondo lattice coherence in $CeAgSb_2$. The identification of a subpicosecond decay process underscores the pivotal role of the CEF in shaping the band structure of this material. These discoveries not only deepen our understanding of $CeAgSb_2$, a compound whose physical properties warrant continued investigation, but also contribute to the broader knowledge of hybridization phenomena and electronic behavior across heavy fermion materials.

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