Unraveling the Hybridization Process in CeAgSb$_2$ Kondo Lattice

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Abstract. We present a comprehensive investigation utilizing ultrafast optical pump-probe spectroscopy to elucidate the intricate quasiparticle dynamics in the CeAgSb$_2$ single crystal. Through our experimental endeavors, a nuanced two-phase evolution in the hybridization phenomena linking localized $f$ moments and conduction electrons has been unveiled. This intricate process is manifest in two distinct manifestations: firstly, the discernible manifestation of hybridization fluctuations spanning the temperature range of approximately 150 K (denoted as $T'$) down to approximately 65 K (referred to as $T$); secondly, the notable emergence of a collective hybridization phenomenon below the critical temperature, $T'$, as indicated by the onset of an indirect gap in the electronic structure. These discerning observations provide invaluable insights into the interplay of electronic configurations as a function of temperature in CeAgSb$_2$, shedding light on its unique properties and behaviors.

Keywords: ultrafast pump-probe spectroscopy; heavy electrons; collective hybridization.

1. Introduction

Heavy fermion systems, often referred to as heavy fermion metals or Kondo lattice systems, are a fascinating class of materials in condensed matter physics. These materials exhibit unusual electronic behavior at low temperatures, characterized by heavy effective masses of charge carriers, strong electron-electron correlations, and often unconventional superconductivity or magnetic properties [1, 2]. Central to the captivating attributes of heavy fermion systems is the phenomenon of delocalization of $f$ moments with temperature or external tuning parameters [3]. Specifically, at high temperatures, the localized $f$ electrons are weakly coupled with conducting electrons and behave as local moments. As the temperature is lowered, the conduction electrons begin to interact with the local magnetic moments, giving rise to the heavy fermion behavior. In a Kondo lattice, the Kondo temperature ($T_K$) represents the characteristic energy scale at which the interplay between the localized magnetic moments and the itinerant conducting electrons leads to a coherent state [4]. It constitutes a pivotal parameter essential for comprehending the intricate electronic characteristics inherent to heavy fermion systems. As a typical heavy fermion compound, CeAgSb$_2$ exhibits crystallization within the tetragonal ZrCuSi$_2$-type crystal structure, aligning with the space group $P4/nmm$ [5]. The determination of $T_K$ in CeAgSb$_2$ has attracted significant attention, resulting in a range of estimations. While susceptibility analysis [6] indicates a $T_K$ of approximately 65 K, alternative methods such as inelastic neutron scattering [7] and muon spin rotation experiments [8] propose a slightly broader range of $T_K$, around 60–80 K. Another perspective arises from an analysis of the relationship between the Wilson number and the Kondo temperature, which leads to a proposed value of $T_K$ around 23 K [6]. Moreover, considering the interplay between ferromagnetic ordering and the Kondo effect, there is speculation that $T_K$ could fall within the range of 10–20 K [9]. Overall, the Kondo energy scale associated with this compound remains elusive and not fully understood. It is thus important to study the evolution of $c$-$f$ hybridization with temperature in CeAgSb$_2$. Lately, ultrafast optical pump-probe spectroscopy has emerged as a viable approach for observing the coherent hybridization dynamics of $f$ electrons in heavy fermion systems across a broad temperature spectrum, presenting an alternative avenue for investigation [10].

In this work, through ultrafast optical pump-probe spectroscopy, the hybridization involving localized $f$ electrons and conducting electrons in CeAgSb$_2$ Kondo lattice is expounded upon through an exploration of quasiparticle dynamics. Specifically, we have noted that the relaxation time ($\tau_1$) of

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quasiparticles exhibits a distinct anomaly around the critical temperature \( T^* \approx 150 \) K. Furthermore, below \( T^* (\approx 65 \) K), the parameter \( \tau_1 \) demonstrates a conspicuous reliance on pump fluence, in contrast to its fluence-independent behavior observed above \( T^* \). These discoveries empower us to derive the following conclusions: (1) The emergence of a heavy electron state, concomitant with the establishment of an indirect hybridization gap as the temperature descends below \( T^* \), and (2) the presence of precursor hybridization fluctuations within the temperature range from \( T^* \) to \( T^\dagger \).

2. Results and discussions

Time-resolved measurements of differential reflectivity with ultrafast resolution, denoted as \( \Delta R(t)/R \), were conducted on a single crystal of CeAgSb$_2$, encompassing the temperature range from 5 K to ambient conditions. The experimental setup is shown in Figure 1. One-color pump-probe spectroscopic investigations were conducted in the reflection mode utilizing a femtosecond Ti:sapphire laser source. The laser pulse exhibits a width of approximately 55 fs (full width at half maximum) with a photon energy of approximately 1.55 eV (800 nm). The pump beam, maintained with \( p \) polarization, was focused onto the surface of the sample, yielding a focal spot diameter of around 100 \( \mu \)m, and it was aligned along the direction of normal incidence. In concert, the probe beam, upheld with \( s \) polarization, is incident upon the sample at an angle of approximately 15º relative to the sample's normal direction. The sample was affixed within a cryostatic environment. The temperature could be regulated within a range spanning from 5 to 300 K, with an accuracy of approximately 0.1 K.

As shown in Figure 2(a), an immediate and pronounced transient response is discerned in the acquired data from 5 K to 293 K, subsequent to the photoexcitation. The relaxation dynamics governing the evolution of \( \Delta R(t)/R \) over the initial ten picoseconds (ps) is commonly dominated by the scattering process of e-e (electron-electron) and electron-boson in strongly correlated electron systems [11]. These bosons could encompass phonons or other types of bosonic type excitations [12]. As shown in Figure 2(b), \( \Delta R(t)/R \) can be fitted using the following function:

\[
\Delta R(t)/R = A_1 e^{-\tau_1/t} + A_2 e^{-\tau_2/t} + C
\]  

(1)
where $A_1$ (or $A_2$) and $\tau_1$ (or $\tau_2$) represent the amplitude and decay time, respectively. $C$ is a $t$-independent offset. In Figure 2(c), we present the derived $\tau_1$ as a temperature-dependent function at different pump fluences. Within the confines of our experimental errors, a fluence-dependent $\tau_1$ is observed below a characteristic temperature of 65 K, denoted as $T^*$. The fluence-related behavior of the decay time $\tau_1$ below $T^*$ can be explained through the Rothwarf-Taylor (RT) model [13, 14].

Within the framework of this theoretical model, the kinetics of quasiparticles ($n$) and bosons ($N$) are dominated by a narrow energy gap opened in the density of states (DOS). The gap size can be investigated by fitting the $\tau(T)$ and $n(T)$ using the subsequent equation,

$$\tau^{-1}(T) \propto \left( \frac{\delta}{\Delta_{nT+1}} + 2n_T \right) \left( \Delta + \alpha T \Delta^4 \right)$$

and

$$n_T(T) = \frac{A(0)}{A(T)} - 1 \propto (\Delta T)^p e^{-\Delta T^c}$$

where $\alpha$, $\zeta$, and $\delta$ are fitting parameters. $n_T$ denotes the density of quasiparticles that are excited across the narrow energy gap, and $p$ ($0 < p < 1$) represents a constant that is determined by the shape of DOS [15]. A satisfactory fitting result is attainable, as illustrated in Figure 3, yielding a gap magnitude of $2\Delta \approx 9$ meV. This result indicates the establishment of collective hybridization between $f$-electrons and conducting electron.

Fig. 2 (a) Time-resolved differential reflectivity, denoted as $\Delta R(t)/R$, of CeAgSb$_2$ single crystal as a function of temperature. (b) $\Delta R(t)/R$ at 5 K and the fitting results using Eq. (1). (c) The evolution of decay time $\tau_1$ as a function of temperature at various pump fluences. Two critical temperatures, $T^*$ and $T^\dagger$ are defined.

Above $T^\dagger$, the absence of fluence dependence suggests the closure of the indirect hybridization gap. As for $T > T^\dagger$ (~ 150 K), the parameter $\tau_1$ initially attains saturation, thereafter demonstrating a modest increment with ascending temperature. Between $T^*$ and $T^\dagger$, $\tau_1$ increases with lowering the temperature. Similar temperature-dependent relaxation process has been observed in the CeCoIn$_5$ heavy fermion compound [10], where precursor hybridization fluctuations are claimed to dominate the reduction in the $T$-dependent decay rate $\gamma (= 1/\tau)$ below $T^\dagger$ [16]. Specifically, the scattering
process of electron-electron (e-e) and electron-phonon are both suppressed when the fluctuating f moments are involved in the hybridization process between the itinerant conducting electrons and localized f electrons. The hybridization fluctuations process can lead to the renormalization of the conduction bands, even within the high-temperature domain (e.g., > 70 K). The aforementioned findings showcase a two-phase hybridization process within the CeAgSb₂ single crystal, that is, the initiation of hybridization fluctuations below \( T^\dagger \) and the coherent hybridization accompanied by an indirect gap in DOS below \( T^* \). The schematic representation of this two-stage hybridization process is presented in Figure 4.

![Fig. 3](image_url) (a) The evolution of density of stimulated quasiparticles, \( n_T \), with temperature below \( T^* \). The inset illustrates the dependence of amplitude \( A_1 \) on temperature. (b) Dependence of decay time \( \tau_1 \) on temperature below \( T^* \). The red curves represent the fitted outcomes utilizing the Rothwarf-Taylor (RT) model.

![Fig. 4](image_url) Illustration of two stage hybridization process in CeAgSb₂ single crystal. The critical temperature \( T^\dagger \) represents the emergence of hybridization between localized f electrons and conducting electrons, however, a coherent hybridization state has not formed within the entire lattice. The critical temperature \( T^* \) marks the further development of such hybridization into a coherent heavy electron state.

3. Summary

In summary, our findings elucidate the intricate hybridization process in the Kondo lattice compound CeAgSb₂ single crystal across a broad spectrum of temperatures. The increase of relaxation time \( \tau_1 \) below \( T^\dagger \sim 150 \) K implies the presence of hybridization fluctuations. Below the
critical temperature of ~65 K, we confirm the formation of an indirect energy gap with the size of approximately 9 meV, which protects the coherent heavy fermion state.

References


