

COLLAPSE OF KONDO LATTICE IN
 $\text{Ce}_{1-x}\text{La}_x\text{Pd}_3$ ($x = 0, 0.03$)

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The change of the electronic structure as well as the hybridization between the localized $4f$ state and the conduction band (cf hybridization) of $\text{Ce}_{1-x}\text{La}_x\text{Pd}_3$ ($x = 0, 0.03$) due to the La-substitution has been studied by the optical conductivity spectra in the infrared region. The width of the optical transition of $\text{Pd } 4d \rightarrow \text{Ce } 4f$ states that was mainly observed in the energy region shrinks by the La-substitution. This means that the cf hybridization is strongly suppressed by the absence of the periodicity of the Ce-ion.

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1. Introduction

CePd₃ that is known a typical Kondo lattice compound has been studied over a few decades. The main origin of the anomalous physical properties of Kondo lattice compounds is the hybridization between conduction bands and localized 4*f* states (*cf* hybridization). A heavy quasiparticle due to the *cf* hybridization in CePd₃ appears on the Fermi level below 100 K. In optical measurements, the Drude term with heavy effective mass and a long relaxation time due to the heavy quasiparticles appear in the far-infrared region [1]. The heavy-Fermion-like transport disappears in the case of the La-substitution of Ce or the Cu-substitution of Pd [2, 3]. In these cases, the electric resistivity that goes up with decreasing temperature looks like an impurity Kondo effect. Since the same La-substitution effect appears in Ce_{1-x}La_xCu₆, the behavior is thought to occur due to the absence of the periodicity of the Kondo lattice [4].

In this study, we investigate the change of the Ce 4*f* state due to the collapse of the Kondo lattice by the La-substitution using the optical conductivity ($\sigma(\omega)$) spectra of CePd₃ and Ce_{0.97}La_{0.03}Pd₃ in the infrared region. The main part of the Ce 4*f* state locates in the unoccupied states. The unoccupied states are expected to be observed by the inverse photoemission experiment [5]. However, the energy resolution is not sufficient to observe the tiny change of the 4*f* state. Then we measured the unoccupied Ce 4*f* states by the $\sigma(\omega)$ spectra through the optical transition of Pd 4*d* → Ce 4*f*.

The evidence of the *cf* hybridization is observed in the optical transition of the Pd 4*d* → Ce 4*f* in the mid-infrared region. In this paper, the temperature and the La-substitution dependences of the Ce 4*f* state as well as the *cf* hybridization state are mainly reported.

2. Experimental

The polycrystalline samples made by an arc melting method were polished up to mirror surfaces using diamond-rapping films. The optical measurement was done in the photon energy range of 5 meV–1.5 eV at several temperatures of 7 ~ 300 K by using a conventional infrared Fourier spectrometer. The spectra in the energy region of 1.2–30 eV at room temperature were measured at the beam line 7B of the synchrotron radiation facility, UVSOR, Institute for Molecular Science [6] and it was connected to their spectra below 1.5 eV for obtaining the optical conductivity ($\sigma(\omega)$) spectra via the Kramers–Kronig analysis. The Kramers–Kronig analysis is performed in the conventional method [7].

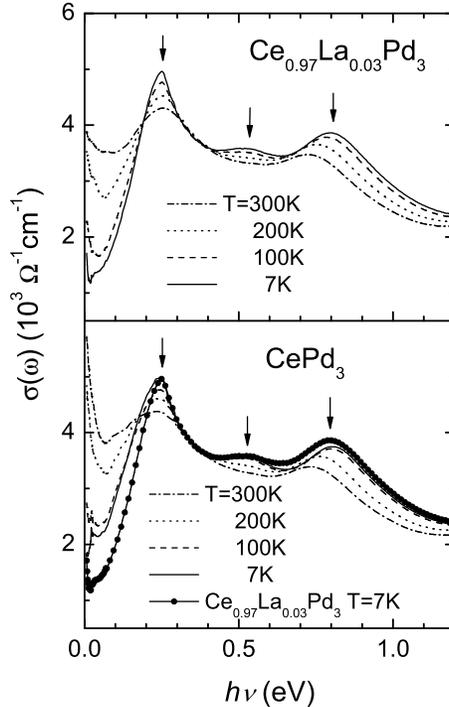


Fig. 1. Temperature dependence of the optical conductivity ($\sigma(\omega)$) spectra of $Ce_{1-x}La_xPd_3$ ($x = 0, 0.03$). In the lower figure for $CePd_3$, $\sigma(\omega)$ spectrum of $Ce_{0.97}La_{0.03}Pd_3$ at 7 K is also plotted as a reference.

3. Results and discussion

Obtained $\sigma(\omega)$ spectra of $CePd_3$ and $Ce_{0.97}La_{0.03}Pd_3$ are shown in Fig. 1. The characteristic of the spectra is three peaks at 0.26, 0.52 and 0.78 eV commonly observed in both materials. Three peaks that become narrow with decreasing temperature have the same temperature dependence in both materials. Because the peak intensity at 0.26 and 0.78 eV is strong, the peaks originate from allowed transitions, in the other hand, the peak at 0.52 eV seems to be due to a forbidden optical transition. Therefore the peaks are considered to originate from the transition of Pd $4d \rightarrow Ce 4f$. Usually, the Ce $4f$ state splits in two peaks of $J = 5/2$ and $7/2$ with the splitting energy of 0.26 eV due to the spin-orbit interaction. The energy difference of the peaks at 0.26 and 0.78 eV is twice of the spin-orbit separation and the peak at 0.52 eV locates at the center of these peaks. Therefore, in the case that it is assumed that the Pd $4d$ density of states has a double peak structure (Pd $4d_{5/2}$ state at -0.52 eV and Pd $4d_{3/2}$ state at -0.26 eV from the Ce $4f_{5/2}$ state), the transitions from these peaks to the spin-orbit separated Ce $4f$

states makes three peaks with the energy separation of 0.26 eV. The allowed transitions of Pd $4d_{3/2} \rightarrow$ Ce $4f_{5/2}$ and Pd $4d_{5/2} \rightarrow$ Ce $4f_{7/2}$ make two peaks at 0.78 and 0.26 eV, respectively. The transitions of Pd $4d_{3/2} \rightarrow$ Ce $4f_{7/2}$ and Pd $4d_{5/2} \rightarrow$ Ce $4f_{3/2}$ that should appear at the same energy of 0.52 eV are forbidden but a small peak may appear by the hybridization effect.

Since the Pd $4d$ state does not change with the La-substitution, the peak width mainly reflects the Ce $4f$ band width as well as the cf hybridization intensity. The detail temperature dependence of the peak parameters will be given elsewhere [8]. Here, only the peak width at 0.25 eV is discussed.

The peak width in $\text{Ce}_{0.97}\text{La}_{0.03}\text{Pd}_3$ at 7 K is about 80 % in comparison with that in CePd_3 in spite that the width at 300 K is similar to each other. This means that the cf hybridization intensity is reduced by the La-substitution. As a reason, we may consider that the periodicity of the Ce-ion as well as the cf hybridization is broken, *i.e.*, the Kondo lattice is collapsed by the La-substitution. The spectral change indicates the change of the electronic structure due to the collapse of the Kondo lattice.

According to the analysis of mid-infrared peaks of Kondo insulators by Okamura *et al.*, similar peaks are observed in CeNiSn (Kondo Temperature, $T_K \sim 40$ K), CeRhSb ($T_K \sim 60$ K) but there is no significant peak in CeRhAs ($T_K \sim 1300$ K) [9, 10]. The absence of the peak in CeRhAs is considered to originate from the so strong cf hybridization because of its so high T_K . Therefore the peak width is scaled by the Kondo temperature as well as the cf hybridization intensity. The interpretation is consistent with ours. Therefore, in CePd_3 , the narrowing of the peak width by the La-substitution indicates the decreasing of the cf hybridization intensity as well as the change of the electronic structure due to the collapse of the Kondo lattice.

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