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RAPID COMMUNICATION

Optical study on intermediate-valence compounds $Yb_{1-x}Lu_xAl_3$*

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We report an optical spectroscopy study on intermediate valence system $Yb_{1-x}Lu_xAl_3$ with $x = 0$, 0.25, 0.5, 0.75, and 1. The Kondo temperature in the system is known to increase with increasing Lu concentration. Therefore, it is expected that the energy scale of the hybridization gap should increase with increasing Lu concentration based on the periodic Anderson model. On the contrary, we find that the spectral structure associated with the hybridization effect shifts monotonically to lower energy. Furthermore, the Lu substitution results in a substantial increase of the free carrier spectral weight and less pronounced plasma frequency reduction upon lowering temperature. We attribute the effect to the disruption of the Kondo lattice periodicity by the random substitution of Yb by Lu. The work highlights the importance of the lattice periodicity of the rare earth element for understanding the Kondo lattice phenomena.

Keywords: heavy fermions, intermediate valence system, optical conductivity, hybridization effect

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1. Introduction

Strongly correlated, rare-earth element based intermetallic compounds, including heavy fermion (HV) and intermediate valence (IV) systems, have attracted considerable interest due to the wide range of novel behaviors. In those systems, the Kondo interaction plays a very important role. At high temperature, the f-electrons of the rare-earth elements are localized and form local moments, which give rise to a Curie–Weiss like susceptibility. The conduction electrons are strongly scattered by the local moments. With decreasing temperature, the conduction electrons tend to screen the f-electron local moment collectively and to form a Kondo singlet state. Its effect is to build a narrow resonance feature with a width of Kondo temperature (T_K) in the density of states near the Fermi level. Due to the periodical arrangement of the moments in the lattice, the Kondo resonance at each f-electron site transforms into a narrow electron band. This narrow band hybridizes with the conduction electron band, leading to the formation of a hybridization energy gap in the vicinity of the Fermi level in $\boldsymbol{k}$-space. Such physics is well captured by the periodic Anderson model (PAM), which gives the hybridization energy gap of $\Delta_{\rm dir} \sim \sqrt{T_K W}$ in the mean-field approximation, where W is the band width of the conduction electrons.[1–5] In addition to the Kondo interaction, there exists the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction between the local moments being mediated by the conduction electrons, which tends to order the local moments in those intermetallic compounds. The ground states are determined by the competition between the RKKY-type magnetic interaction and the Kondo screening interaction.[1–3]

The strengths of Kondo and RKKY interactions vary in different intermetallic compounds. The IV compounds generally have stronger hybridization strength between the conduction electrons and rare earth 4f electrons than the HF compounds due to the reduced lattice constants and have moderate enhancement of effective masses being about 10–50 m_e at low temperature. The rare-earth ions in those compounds have a non-integer valence. Typical IV compounds include $YbXCu_4$ (X = Ag, Cd, In, Mg, Tl, Zn), $CePd_3$, $EuRh_2$, TmSe, YbB_{12}, $YbAl_2$, and $YbAl_3$.[6–12] They usually reside away from a quantum critical point locating at a delicate balance between RKKY and Kondo interactions, but can still have different ground states such as Kondo insulator (e.g., YbB_{12}[10]) or moderate heavy mass Fermi liquid state (e.g., $YbAgCu_4$,[6] $CePd_3$,[7] and $YbAl_3$[12]).

Among IV systems, $YbAl_3$ is one of the most studied compounds. It has a relatively high Kondo temperature T_K in Yb-based IV compounds ($\sim$ 600 K) and a fully coherent state is formed below the temperature $T_{\rm coh}$ ($\sim$ 40 K).[13–15] It was revealed that, when Yb is partially substituted by Lu (i.e., in $Yb_{1-x}Lu_xAl_3$), the temperature of the broad peak in the magnetic susceptibility, $T_{\rm max}$, dramatically shifts to higher temperatures,[15,16] so that the Kondo temperature T_K, which is estimated as $T_K \sim$ (4–6) $T_{\rm max}$, further increases with increas-

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ing x.[14–18] Therefore, it is expected that the energy scale of the hybridization gap should increase with increasing Lu concentration based on the periodic Anderson model. The Lu-substituted $YbAl_3$ system provides another opportunity to study the evolution of the hybridization strength versus T_K in the correlated f-electron system.

Optical spectroscopy is a powerful bulk technique to investigate charge dynamics and band structure of materials as it probes both free carriers and interband excitations. In particular, it yields direct information about the formation of energy gaps. Optical spectroscopy measurements have been performed on many HF and IV compounds with different hybridization strengths. It is indeed found that the spectral features in optical response, including the relation between the hybridization gap and the Kondo temperature, for heavy fermion and IV compounds were well explained by the PAM.[3] In this work, we report our systematic optical spectroscopy study of $Yb_{1-x}Lu_xAl_3$ with $x = 0$, 0.25, 0.5, 0.75, and 1. In contrast to the expectation from the change of T_K, we find that the spectral structure associated with the hybridization effect shifts monotonically to lower energy scale with Lu substitution. We attribute the effect to the disruption of the Kondo lattice periodicity by the random substitution of Yb by Lu, though the Kondo lattice effect is still visible up to a very high Lu concentration.

2. Experimental techniques

$Yb_{1-x}Lu_xAl_3$ single crystals with nominal compositions of $x = 0$, 0.25, 0.5, 0.75, and 1 were grown by self-flux method in a procedure the same as that described in Ref. [15]. The excess aluminium was removed by a centrifugal machine. The actual x values determined by the energy dispersion x-ray spectrum (EDX) on different samples in the same batch are listed in Table 1.

Table 1. The nominal composition and the actual composition determined by EDX of the $Yb_{1-x}Lu_xAl_3$ compounds.

x	Nominal composition	Actual composition
0	$YbAl_3$	$YbAl_3$
0.25	$Yb_{0.75}Lu_{0.25}Al_3$	$Yb_{0.68}Lu_{0.32}Al_3$
0.5	$Yb_{0.5}Lu_{0.5}Al_3$	$Yb_{0.45}Lu_{0.55}Al_3$
0.75	$Yb_{0.25}Lu_{0.75}Al_3$	$Yb_{0.24}Lu_{0.76}Al_3$
1	$LuAl_3$	$LuAl_3$

The temperature dependent resistivity $\rho(T)$ was measured by standard four leads method in a Quantum Design physical properties measurement system (PPMS). The magnetic susceptibility $\chi(T)$ was measured by a Quantum Design magnetic property measurement system (MPMS) under a magnetic field of 0.5–2 T. The optical reflectance measurements were conducted on the Bruker IFS 80v and 113v spectrometers in a frequency range from 40 cm^{-1} to 25000 cm^{-1}. The *in situ* gold and aluminium overcoating technique was used to obtain the absolute reflectivity $R(\omega)$. The real part of the conductivity spectrum $\sigma_1(\omega)$ was obtained through the Kramers–Kronig transformation of $R(\omega)$. A Hagen–Rubens relation was used in the low-frequency extrapolation, and the x-ray atomic scattering function was employed in the high-energy extrapolation.

3. Results and discussion

Figure 1 shows the temperature dependent resistivity $\rho(T)$ of $Yb_{1-x}Lu_xAl_3$. The compounds have rather low resistivities. For undoped $YbAl_3$, $\rho(T)$ shows a strong temperature dependence. The resistivity at 2 K is about 40 times smaller than that at room temperature. The low temperature resistivity shows T^2-dependence roughly below 80 K. With Lu doping, the resistivity at high temperature drops systematically, but overall $\rho(T)$ shows weaker temperature dependence. As a result, the low temperature resistivity of the Lu substituted samples can be even higher than that of undoped $YbAl_3$. The results are consistent with the earlier reports.[19,20]

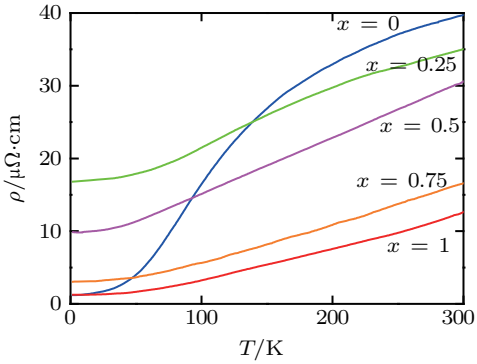


Fig. 1. (color online) Temperature dependence of the electrical resistivity of $Yb_{1-x}Lu_xAl_3$.

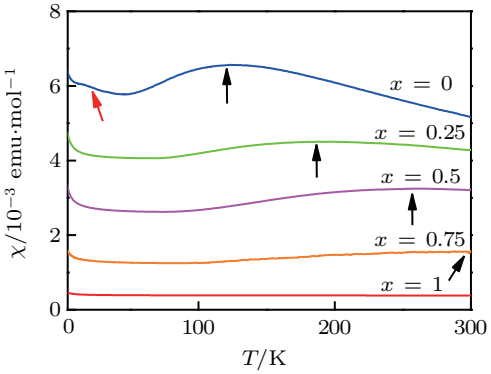


Fig. 2. (color online) $\chi(T)$ versus T of bulk samples $Yb_{1-x}Lu_xAl_3$ with different Lu contents. The black arrow indicates the temperature T_{max} where the magnetic susceptibility $\chi(T)$ exhibits a broad peak. The red arrow indicates a weak peak-like feature at low temperature for $YbAl_3$.

The temperature dependent magnetic susceptibility of $Yb_{1-x}Lu_xAl_3$ is shown in Fig. 2, which is also in good agreement with the earlier reports.[14,15] A broad maximum centered at $T_{max} \sim 125$ K is observed for undoped $YbAl_3$, which corresponds to the energy scale of a Kondo temperature T_K

(about 4–6 T_{max}) of the order 500 K.[14–18] By increasing the Lu concentration, the overall magnitude of χ monotonically decreases and the T_{max} shifts to higher temperature, indicating higher T_K for the Lu substituted samples in terms of earlier studies. Additionally, $\chi(T)$ increases below 40 K and exhibits another weak peak-like feature as indicated by the red arrow for undoped $YbAl_3$. This feature was demonstrated to be intrinsic for $YbAl_3$ and related to the low temperature energy scale of a fully coherent state below $T_{coh} \sim 40$ K.[15] It is very sensitive to alloy disorder and could be suppressed for $x \geq 0.05$.[15] In our measurement, the feature cannot be resolved in Lu substituted samples.

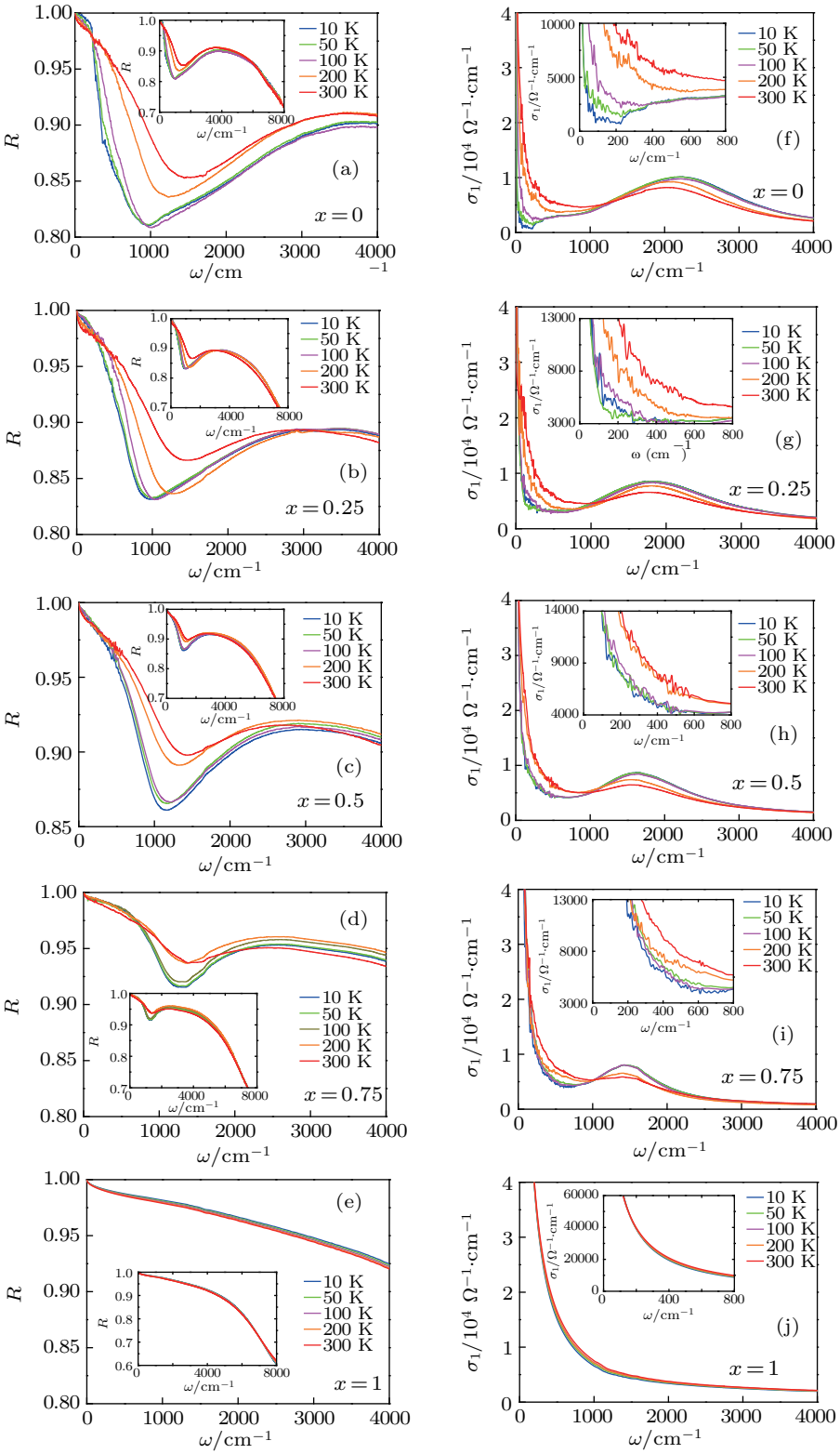


Fig. 3. (color online) (a)–(e) Optical reflectance spectra of $Yb_{1-x}Lu_xAl_3$ ($x = 0$, 0.25, 0.5, 0.75, and 1) single crystals at five different temperatures. The insets show the spectra up to 8000 cm^{-1}. (f)–(j) The real part of optical conductivity $\sigma_1(\omega)$ of $Yb_{1-x}Lu_xAl_3$ obtained through the Kramers–Kronig transformation. The insets show the expanded $\sigma_1(\omega)$ spectra in the low energy region.

Figure 3 shows the optical spectra of $Yb_{1-x}Lu_xAl_3$. The left panels are reflectance spectra measured at different temperatures and the right panels are calculated conductivity spectra. Let us first concentrate on the spectral features of the undoped $YbAl_3$ (top panels). Consistent with early results,[21] the frequency dependent reflectivity and optical conductivity of pure $YbAl_3$ at different temperatures show typical heavy fermion-like behaviors. $R(\omega)$ is suppressed below 4000 cm^{-1}, leading to a broad dip, which becomes more pronounced with decreasing T. Correspondingly, the conductivity $\sigma_1(\omega)$ shows a narrow Drude component at very low frequency and a pronounced mid-infrared peak near 2000 cm^{-1}. These are generic structures for HF or IV systems. What is specific to $YbAl_3$ is the observation of further depletion below 300 cm^{-1} in $\sigma_1(\omega)$ at the lowest temperature. This peculiar feature was also observed in earlier work on $YbAl_3$ by Okamura *et al.*[21] It was emphasized that this peculiar feature is not merely a tail of the mid-IR peak but related to the hybridization effect.[21–23] Okamura *et al.* proposed that this low energy structure comes from the excitation across the indirect hybridization energy gap while the mid-infrared peak at high energy represents the excitation across the direct hybridization energy gap.[21]

For the samples with different Lu substitutions, the optical measurement still shows metallic frequency and temperature dependence: the $R(\omega)$ approaches to unit at zero frequency and low frequency $R(\omega)$ increases as the temperature decreases. The observations are in accordance with their resistivity results. Similar to the pure $YbAl_3$, the reflectance spectra of the samples containing Yb display a broad dip below 2000 cm^{-1}, but the features become less pronounced with increasing Lu content. On the other hand, the temperature dependent $R(\omega)$ spectrum of $LuAl_3$ exhibits a simple metallic behavior, which is in a strong contrast to that of other Yb-based compounds and also consistent with the fact that Lu^{3+} has a non-magnetic electronic configuration. The results indicate that the dip feature originates from the hybridization effect between conduction electrons and localized 4f electrons. For all $Yb_{1-x}Lu_xAl_3$ samples, the reflectivity at various temperature almost overlaps above 8000 cm^{-1} (as shown in the insets), which suggests that the electronic structure of $Yb_{1-x}Lu_xAl_3$ is relatively stable against temperature variation at high energy region.

Corresponding to the spectral structures in $R(\omega)$, the real part of conductivity $\sigma_1(\omega)$ of $Yb_{1-x}Lu_xAl_3$ shown in the right panels of Fig. 3 exhibits some generic features. The $\sigma_1(\omega)$ spectra have narrow Drude-like components related to the renormalized heavy quasiparticles at low frequency and an eminent peak in the mid-infrared region. With decreasing temperature, the Drude components become further narrowed and suppressed, and the mid-infrared peak is enhanced. Compared with undoped $YbAl_3$, Lu substitutions result in following changes. First, the peculiar low energy depletion below 300 cm^{-1} at the lowest temperature in $\sigma_1(\omega)$ is quickly suppressed. This can be seen in the expanded plots in the insets. The suppression feature vanishes in the Lu substituted samples. Second, the narrowing and further suppression of the Drude components upon lowering temperature become less noticeable with increasing Lu concentration. Third, the mid-infrared peak shifts to lower energy and its spectral weight is reduced. On the other hand, the Yb-free $LuAl_3$ compound shows an usual Drude like response with an absence of mid-infrared peak.

To quantitatively investigate the spectral changes in the Lu-substituted $YbAl_3$, we use a Drude–Lorentz model to decompose the $\sigma_1(\omega)$ into different components of the electronic excitations. The dielectric function of the Drude–Lorentz model can be expressed as

$$\varepsilon(\omega) = \varepsilon_\infty - \sum_s \frac{\omega_{ps}^2}{\omega^2 + i\omega/\tau_{Ds}} + \sum_j \frac{S_j^2}{\omega_j^2 - \omega^2 - i\omega/\tau_j}, \quad (1)$$

where ε_∞ is the dielectric constant at high energy; the last two terms describe the Drude and Lorentz parts, representing the contributions from conduction electrons and interband transitions, respectively.

We find that the low-frequency $\sigma_1(\omega)$ spectra at various temperature for the Lu-doped compounds can be approximately reproduced by using two Drude components and one Lorentz peak. As an example, we show the spectrum of $Yb_{0.75}Lu_{0.25}Al_3$ at 10 K together with Drude–Lorentz fits in Fig. 4(a). However, for undoped $YbAl_3$, an extra Lorentz component centered near 500 cm^{-1} is needed to fit the further suppression feature developed at the lowest temperature as plotted in Fig. 4(b). The overall plasma frequency can be extracted from the fit by $\omega_p^2 = \omega_{p1}^2 + \omega_{p2}^2$, which is an important quantity proportional to n/m^*, where n is the density of free carriers and m^* is the effective mass of quasiparticles. Figures 4(c) and 4(d) show the extracted values of the temperature dependent ω_p and mid-infrared peak positions for $Yb_{1-x}Lu_xAl_3$ samples in detail. Apparently, ω_p decreases substantially with decreasing temperature for undoped $YbAl_3$, reflecting the rapid enhancement of the quasiparticle effective mass at low temperature. The decrease of ω_p upon lowering temperature becomes less pronounced in the Lu-substituted samples. On the other hand, the overall plasma frequency increases significantly with increasing Lu concentration. The smaller ω_p in Yb rich samples can be attributed to the effective mass enhancement due to enhanced Kondo screening effect at low temperature. The missing spectral weight of the Drude component is mainly transferred to the mid-infrared Lorentz peak, which is clearly presented in the right panels of Fig. 4. This indicates

that the Kondo lattice effects are still important for describing the optical response in the Lu substituted samples. Assuming that the low frequency Drude components spectral weight is contributed by the renormalized quasiparticles after taking account of the hybridization effect, and the sum of the mid-infrared component and the low frequency Drude components is related to the total spectral weight contributed by the unrenormalized quasiparticles, then we can roughly estimate the enhancement of the quasiparticle effective mass relative to the band mass from the ratio of those spectral weights. We obtain $m^*/m_B = 35$, 5.4, 3.1, and 2 for $x = 0$, 0.25, 0.5, and 0.75 at 10 K, respectively.

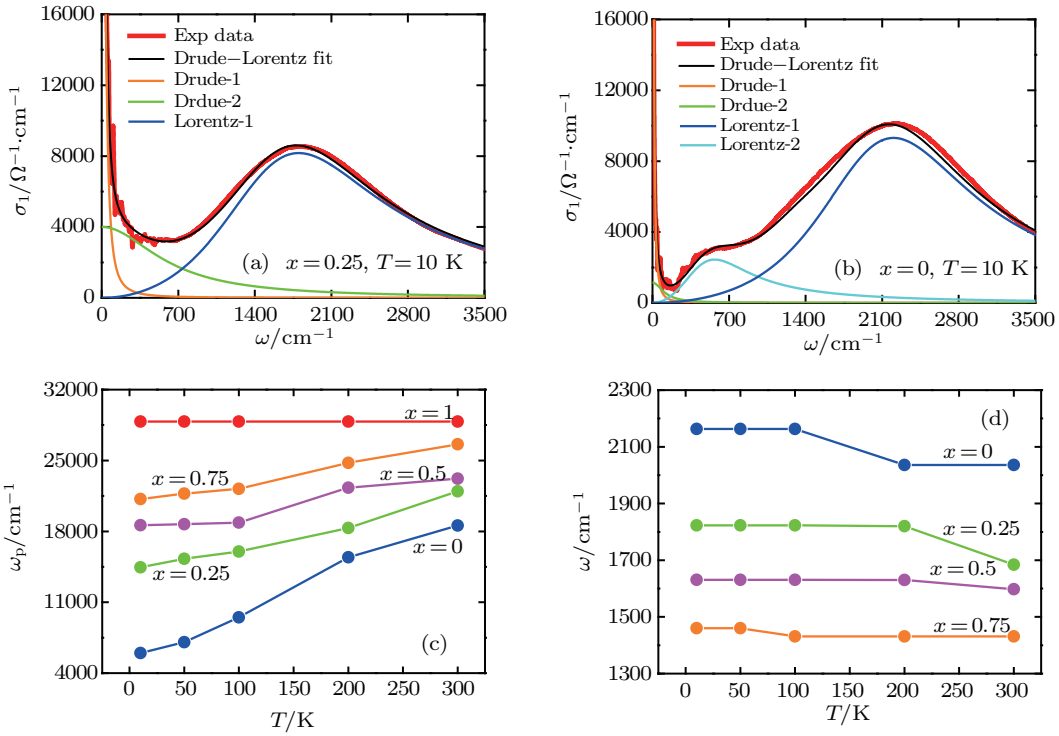


Fig. 4. (color online) The experimental $\sigma_1(\omega)$ together with fit to the Drude–Lorentz model of (a) $Yb_{0.75}Lu_{0.25}Al_3$ and (b) $YbAl_3$ at 10 K, respectively. (c) Temperature and doping dependent plasma frequency ω_p of $Yb_{1-x}Lu_xAl_3$. (c) Temperature and doping dependent central frequency of the mid-infrared Lorentz peak of $Yb_{1-x}Lu_xAl_3$ except for $LuAl_3$.

In fact, the Kondo interaction can cause the flat band from 4f electrons to hybridize with different dispersive bands from conduction electrons, which therefore leads to the formation of multiple hybridization energy gaps near E_F with different energy scales.[24,25] We speculate that the doping could have different effects on different conduction electron bands. Then it is possible that a small doping concentration of Lu can effectively suppress the hybridization gap with smaller energy scale but leave the mid-infrared peak essentially unaffected. Okamura *et al.*[21] suggested that the peculiar low energy gap-like suppression at very low temperature was related to the indirect hybridization energy gap while the mid-infrared peak was caused by the direct hybridization energy gap. In our opinion, they both reflect direct hybridization energy gaps and are due to the hybridization of 4f electrons with different conduction electron bands, since both of them can be clearly detected by optics. Indirect interband transition across an energy gap requires assistance from either impurity or other bosonic excitations and generally has rather weak structure. Based on the above analysis, the sensitivity of the peculiar small gap feature to disorders can be understood by the multiple gaps scenario.

The hybridization effect is perhaps the most important characteristic feature in heavy fermion systems and IV compounds. According to the PAM, the hybridization energy gap is related to the Kondo temperature and the bandwidth of conduction electrons by $\Delta_{dir} \sim \sqrt{T_K W}$.[3,14,17,18,26] This relation has been testified in a number of HF and IV compounds, notably in the IV system of $YbIn_{1-x}Ag_xCu_4$.[26] It deserves to remark that the mid-infrared peak arising from the hybridization effect in $YbInCu_4$, which is centered near 1800 cm^{-1}, is observed only at low temperature. This is because $YbInCu_4$ experiences a first-order valence state phase transition at 40 K with Kondo temperature decreasing abruptly from over 400 K in the low-T phase to 25 K in the high-T phase. As a result, the mid-infrared peak associated with the hybridization effect suddenly becomes invisible above the phase transition due to the very small Kondo temperature.[27] For the present Yb-based samples, the mid-infrared Lorentz peaks are present at high temperature and become more pronounced with decreasing temperature. Since the Kondo temperature T_K in $Yb_{1-x}Lu_xAl_3$ becomes higher upon Lu substitution, the energy scale of the hybridization gap is expected to increase with increasing x in

the system according to the PAM. Surprisingly, its central position gradually shifts to lower frequency with increasing x, which is just opposite to the theoretical expectation. It deserves to remark that the bandwidth of the conduction electrons should not change appreciably, since the cubic lattice parameters of $YbAl_3$ ($a = 4.203$ Å) and $LuAl_3$ ($a = 4.190$ Å) differ by only 0.3%. Then, the decrease of the hybridization energy gap upon Lu substitution must have a different origin.

It is important to note that the substitution of each Yb^{3+} by Lu^{3+} removes a magnetic moment at the Yb site. With increasing Lu concentration, the periodical magnetic lattice structure formed by localized 4f electrons is more significantly disrupted. Experimentally, the mid-infrared peak exhibits a sizable shift towards lower energies and decreases in intensity with Lu substitution, and eventually becomes invisible in pure $LuAl_3$. These results highlight the importance of the Kondo lattice effects for the Lu-substituted $YbAl_3$, for which the coherent lattice periodicity plays an essential role. Although random substitution of Yb by Lu will inevitably disrupt the periodical magnetic lattice, the presence of the hybridization gap feature implies that the local Kondo lattice effect is still present in $Yb_{1-x}Lu_xAl_3$ up to high Lu concentration. The deviation from the prediction by the PAM suggests that a more realistic model taking account of randomly disrupted Kondo lattice is required to explain the effect and to reconcile the different experimental observations.

4. Summary

In summary, we present a systematic optical spectroscopy study of $Yb_{1-x}Lu_xAl_3$ single crystals with $x = 0$, 0.25, 0.5, 0.75, and 1. We find substantial increase of the free carrier spectral weight and less pronounced plasma frequency reduction upon lowering temperature with increasing Lu substitution. The peculiar low energy gap feature formed in undoped $YbAl_3$ at very low temperature is quickly removed by the Lu substitution, meanwhile the mid-infrared peak shifts to lower energy and its spectral weight is reduced. These results suggest a weakening of Kondo coupling or hybridization effect between the conduction electrons and localized rare-earth 4f electrons, which is opposite to the expectation based on the further increase of Kondo temperature in the Lu substituted compounds. These results highlight the importance of the lattice periodicity of rare earth elements for understanding the Kondo lattice phenomena.

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