



Resonant inverse photoemission study of $\text{CeCoGe}_{3-x}\text{Si}_x$

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Abstract

We have measured the resonant inverse photoemission spectra (RIPES) of $\text{CeCoGe}_{3-x}\text{Si}_x$ ($0 \leq x \leq 3$) as functions of composition and temperature. The strong composition dependence of 4f-state just above the Fermi level (E_F) shows that the localized 4f electrons in the compound become itinerant due to the enhanced Kondo effect. Temperature dependence of the f^1 -peak indicates a transition from the paramagnetic state at room temperature to a coherent Fermi-liquid regime at low temperatures. The most remarkable variation of the spectra has been observed above 200 K with increasing temperature in CeCoSi_3 which has a very high Kondo temperature T_K of about 900 K. This suggests that the change in the 4f-electronic state of CeCoSi_3 may be scaled by the coherence transition temperature rather than T_K . © 2000 Elsevier Science B.V. All rights reserved.

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The nature of $\text{CeCoGe}_{3-x}\text{Si}_x$ series of compound has been investigated by Eom et al. [1,2]. Substitution of silicon for germanium produces the normal chemical pressure effect and which reduces the unit-cell volume by about 10% from CeCoGe_3 to CeCoSi_3 . The coupling constant J between the local moments and the conduction electrons is thereby enhanced as the silicon concentration x increases and the antiferromagnetism in CeCoGe_3 is suppressed around $x = 1.2$ by the enhanced Kondo effect. The overall behavior of this system is qualitatively understood within Doniach's magnetic phase diagram [1,2]. In this work we study the crossover from the coherent Fermi liquid to paramagnetic state of $\text{CeCoGe}_{3-x}\text{Si}_x$ by means of RIPES at the Ce $N_{4,5}$ edge, which is a very useful method to probe the 4f-electronic structure in unoccupied states (for example Refs. [2–4]). One of the advantages of this technique is a high sensitivity to the variation of the 4f-occupancy, n_f , that is, the RIPES effectively detects the dependence of the 4f-state on material and temperature. We have measured the RIPE spectra of $\text{CeCoGe}_{3-x}\text{Si}_x$ and found a remarkable reconstruction of the 4f-electronic struc-

ture just above E_F with changing the composition and temperature.

Measurements were performed in an ultrahigh vacuum chamber where the base pressure is always under 2×10^{-10} Torr. Clean sample surfaces were obtained by scraping with a diamond file in the vacuum every ~ 60 min at the measurement temperature. The RIPES was measured by the soft X-ray emission system [5]. The E_F position was determined by referring to the Fermi-edge in the IPES spectra of Au which was evaporated on the sample holder. Strictly speaking, we must take the surface contribution in the RIPES into consideration, but it matters little in this discussion. The preparation and characterization of the polycrystalline samples have been presented elsewhere [1,2].

The RIPE spectra of CeCoSi_3 are displayed as a function of temperature up to the 285 K in Fig. 1. Continuous and striking reduction of the f^1 peak has been observed as temperature rises. We define the ratio of the f peaks as $r_f = I[f^2]/(I[f^1] + I[f^2])$ which is a good indicator of the n_f . Here, $I[f^n]$ represents the integrated intensities of the f^n peak. If the interference of the resonant processes having the f^1 and f^2 final states of RIPE process is disregarded, r_f is equivalent to the n_f . The r_f 's of the several compounds of $\text{CeCoGe}_{3-x}\text{Si}_x$ are plotted against temperature in Fig. 2(a). First, the r_f at 25 K increases

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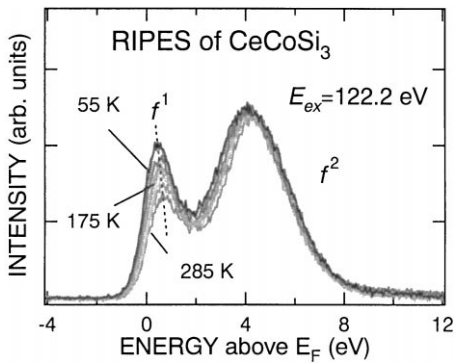


Fig. 1. The RIPES spectra of CeCoSi₃ as a function of the temperature. The spectral intensities are normalized by f² peak intensities.

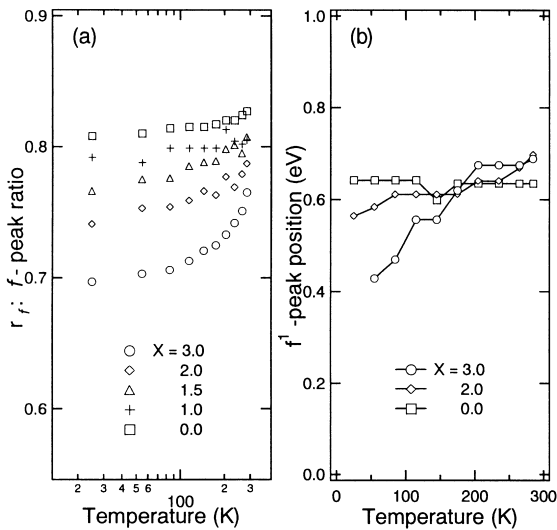


Fig. 2. (a) The f-peak ratio: r_f as function of x which plotted against temperature. (b) The f-peak positions of $x = 0.0, 2.0, 3.0$ which plotted against temperature.

with x . This fact indicates that the localized 4f electron in CeCoGe₃ becomes itinerant due to the enhanced Kondo effect as x increases. Next, the r_f s except for $x = 3.0$ slightly rises with temperature up to 285 K. The relatively constant r_f s of $x = 1.0$ and 0.0 reflect the stable 4f electron numbers $n_f \sim 1$, that is, the paramagnetic state with an effective magnetic moment close to the value of Ce³⁺. On the other hand, r_f of $x = 3.0$, for which the extremely high T_K of the order of 900 K has been reported [1,2], shows a dramatic rise above ~ 200 K. It should be noted that the onset of the rise of the r_f sets in at a sufficiently lower temperature than the single-impurity T_K contrary to the prediction in an impurity approach [6] and the long-range coherence among the local singlets seems to be already destroyed at room temperature.

The coherent dense Kondo state in CeCoSi₃ is strongly suppressed over the $T_{\text{coh}} \sim 200$ K. This result gives a direct evidence for the basic energy scaling by T_{coh} which controls the dense Kondo system. This T_{coh} value appears to correspond to the temperature where the maximum value of magnetic susceptibility is found ($T_{\text{max}}^{\chi} = 230$ K) [1]. The paramagnetic state with localized magnetic moments is sustained far below T_K . On the other hand, attention should be paid to the fact that the T_{coh} of CeCoSe_{3-x}Si_x is much smaller than the single-impurity T_K though in the case of CeRu₂(Ge_{1-x}Si_x)₂, the T_{coh} is comparable to the T_K [7]. This strong material dependence of T_{coh} suggests that the relationship between T_{coh} and T_K is not fixed and straightforward even in the spin-fluctuation systems with a large J .

Although the clear shift of the f¹ peak position to the higher-energy side with increasing the temperature, especially in CeCoSi₃ was observed as shown in Fig. 2(b), the cause for this is not fully settled at the moment and information about the band structure of CeCoGe_{3-x}Si_x or the density of state near E_F around the coherence line would be of great importance for our understanding.

The results of the RIPES of the CeCoGe_{3-x}Si_x ($0 \leq x \leq 3$) have been herein presented as a functions of silicon concentration x and temperature. The localized 4f electron in CeCoGe₃ becomes itinerant due to the enhanced Kondo effect as x increases and the f¹-peak in the spectra of $x = 2.0$ and 3.0 dramatically diminishes as temperature rises above about 200 K. This fact suggests that a transition from the paramagnetic state to the coherent fermi-liquid state occurs far below the T_K , so that the 4f-properties are scaled by the characteristic coherence transition temperature T_{coh} rather than T_K . It should be finally remarked that the energy shift of the f¹ peak with x shown in Fig. 1 does not seem to scale with either T_{coh} or T_K as the impurity Anderson model predicts. This point should be more carefully examined both in experimental and theoretical work in future.

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