



Contrasting Kondo behavior and resonant inverse photoemission spectra of CeTSi_3 and CeTGe_3 ($T = \text{Rh}$ and Ir)

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Abstract

CeTSi_3 and CeTGe_3 ($T = \text{Rh}$ and Ir) were investigated by measuring the magnetic susceptibility, specific heat, electrical resistivity, the resonant inverse photoemission (RIPES) and $M_{\text{IV,V}}$ X-ray absorption spectra (XAS). The germanides showed a very weak Kondo effect, but the silicides exhibited a negatively large Weiss temperature (≈ -130 K) and a $\ln T$ dependence of magnetic resistivity above 100 K, suggesting that they are Kondo-lattice compounds with a high Kondo temperature T_K (≈ 100 K). The Curie–Weiss law suggests that Ce atoms in these compounds remain close to $3+$ down to about 150 K in spite of their high T_K . Both RIPES and $M_{\text{IV,V}}$ XAS support their apparently stable valency. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: CeTX_3 ($T = \text{Rh}, \text{Ir}; X = \text{Si}, \text{Ge}$); Kondo-lattice compound; RIPES; X-ray absorption spectra

We have recently investigated four Ce compounds of the tetragonal BaNiSn_3 -type, CeTSi_3 and CeTGe_3 ($T = \text{Rh}$ and Ir) by measuring the low-temperature specific heat, magnetic susceptibility and electrical resistivity [1] and found that CeRhSi_3 and CeIrSi_3 are Kondo-lattice compounds with a very high Kondo temperature like CeFeGe_3 [2] and CeIr_2Ge_2 [3]. CeFeGe_3 appears to possess a stable valency at least down to about 150 K in spite of the very high Kondo temperature (more than 100 K) [2]. These two silicides, however, order magnetically at low temperatures contrary to the nonmagnetic ground state found in the latter germanides. On the other hand, CeRhGe_3 and CeIrGe_3 exhibit a very weak Kondo effect and a complex magnetic ground state.

Sample preparation and measurements of the specific heat, susceptibility and resistivity are reported in Ref. [1].

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The RIPES measurements were performed in an ultra-high vacuum chamber where the base pressure was about 5×10^{-11} Torr. Clean sample surfaces were obtained by scraping with a diamond file in vacuum every 10–40 min at 25 K. A thermal cathode-type electron gun was used for the excitation source. The kinetic energy, E_k , of the electron was calibrated by the electron energy analyzer. The resonant inverse photoemission (RIPES) was measured by a soft X-ray emission system, Rowland mounted-type spectrometer. The Fermi level position and an energy resolution of the system were determined by referring to the Fermi edge in the RIPES spectra of Au which was evaporated on the sample holder. The energy resolution of this system is 0.44 eV at $E_k = 90$ eV. The $M_{\text{IV,V}}$ X-ray absorption spectra (XAS) were measured at room temperature using the beamline BL-2C at Photon Factory, KEK. The spectra were obtained from the total electron yield measurements and normalized by the mirror current. The photon energy was calibrated by the photoelectron spectra of Au 4f-core level. The energy resolution is 0.3 eV at 900 eV.

The specific heat of CeRhSi₃ and CeIrSi₃ revealed a very sharp magnetic transition at 1.8 and 5.0 K, respectively, as shown in Fig. 1. We found from the calculated magnetic entropy displayed in the inset of Fig. 1 that the entropy gain at the magnetic transition attains only 12% and 23% of $R \ln 2$ for CeRhSi₃ and CeIrSi₃, respectively. This fact suggests that a strong Kondo compensation has occurred at these temperatures in both compounds. Their magnetic susceptibility data obey a Curie–Weiss law above about 150 K. The effective magnetic moment, μ_{eff} , is only slightly larger than that of the free Ce³⁺ ion, while the Weiss temperature, θ , is negatively very large (−128 K for CeRhSi₃ and −142 K for CeIrSi₃), as often found for intermediate-valence materials. Contrarily the trivalency of the Ce ions in these silicides seems to be well warranted by the Curie–Weiss law held above 150 K. The resistivity of these silicides revealed a maximum around 100 K in their magnetic part of resistivity, ρ_{mag} , which is obtained by subtracting the resistivity of La counterpart. Such a thermal variation of ρ_{mag} is interpreted as a result of the Kondo effect under the influence of a crystalline electric field (CEF). The peak temperature around 100 K may not necessarily be directly related to their T_K , but the assignment of about 100 K to T_K of these compounds seems to be reasonable from the discussions given above regarding their magnetic entropy and θ . Very similar behavior has been reported for CeFeGe₃ and CeIr₂Ge₂ [2,3].

The valence state of Ce can be easily verified by RIPES [4] or $M_{\text{IV,V}}$ XAS [5]. Fig. 2 shows the RIPES of CeRhSi₃ and CeIrSi₃ compared with those of CeFeGe₃ measured around 30 K, each of which show only a very small f^1 peak of an indicator of the intermediate-valence state of Ce ion. The stable valency in these compounds is also confirmed by $M_{\text{IV,V}}$ XAS, for which satellite peaks assigned to $3d^9 4f^1$ final states were barely seen. CeRhSi₃ and CeIrSi₃ are hence considered as a new type of Kondo-lattice compounds with a relatively stable Ce valency in spite of the high T_K , like CeFeGe₃ and CeIr₂Ge₂.

The specific heat of CeTGe₃ (T = Rh and Ir) revealed three magnetic transitions for each and the magnetic entropy of each compound reaches $R \ln 2$ below 20 K indicating a very weak Kondo effect in these germanides, contrary to the case of the silicide counterparts discussed above. Their magnetic susceptibility obeyed the Curie–Weiss law above 100 K. μ_{eff} and θ were found to be $2.53 \mu_B$ and −28 K for CeRhGe₃, $2.39 \mu_B$ and −21 K for CeIrGe₃. ρ_{mag} of these germanides is temperature independent below room temperature down to about 100 K where a rapid decrease occurs. This behavior implies that the CEF splitting between the ground doublet and the first excited one is about 100 K and that the spin-disorder resistivity due to the CEF is dominant over

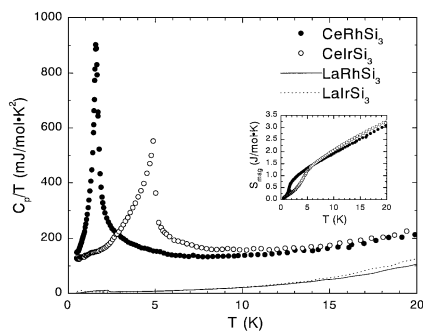


Fig. 1. The specific heat of CeRhSi₃, CeIrSi₃ and La counterparts in a plot C/T versus T . The inset shows the magnetic entropy of CeRhSi₃ and CeIrSi₃.

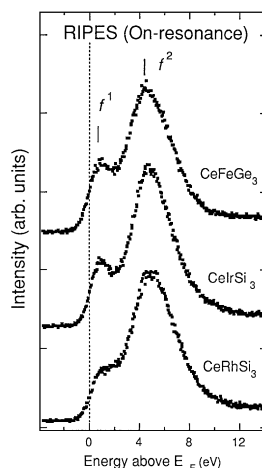


Fig. 2. RIPES spectra of CeRhSi₃, CeIrSi₃ and CeFeGe₃ taken around 30 K.

Kondo-type resistivity. The fact indicates a much lower T_K than that of the silicides, which is consistent with the above results of the specific heat and the small Weiss temperature.

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