Can We Describe the Spectral Function of CeRh₃ by a Band Picture?

In a recent Letter, Weschke et al. [1] present a photoemission and bremsstrahlung isochromat spectroscopy (BIS) study of $CeRh_3$. Their measurements reveal, especially in the BIS spectrum, inconsistencies with the prediction of the single-impurity (SI) Anderson model. They claim that an analysis in the framework of the SI Anderson model is unsatisfactory whereas a local density approximation (LDA) calculation of the unoccupied density of states (DOS) "describes the experimental data remarkably well, in particular the position and shape of the $4f^1$ peak." That leads the authors to the conclusion that the $4f$ states have a bandlike character in strongly hybridized Ce systems.

The far-reaching consequences of this statement demand a careful verification of the experimental data. The authors' conclusion is largely based on the anomalous energy position of the $4f^1$ peak in the BIS spectrum (1.4 eV), higher than in any other Ce compound studied so far [2]. Since the energy calibration of BIS spectra is a notoriously delicate problem, we have decided to repeat the BIS measurement of $CeRh_3$ by using special precautions to obtain a reliable calibration. The Fermi level position was determined by measuring, under the same experimental conditions, the BIS spectrum of a copper film evaporated onto the $CeRh_3$ surface. With this procedure, the Fermi level position could be determined with an accuracy of ± 0.05 eV [Fig. 1(a)]. This energy calibration yields a $4f^1$ peak centered at $E = 1.0 \pm 0.05$ eV instead of 1.4 eV as proposed by Weschke et al. This is confirmed by inverse photoemission measurements in the UV range [Fig. 1(b)]. At an energy of 10.5 eV, the cross section of the $4f$ states is very weak [3]. The spectrum is quite similar to the calculated "d" DOS presented in Ref. [1]. This agreement shows that LDA gives a good description of the d states. At 33 eV, on the other hand, the spectrum is sensitive to the $4f$ states and exhibits a peak at 1 eV in perfect agreement with the high energy result. This value is close to those found in other strongly hybridized Ce compounds like $CeRu_2$ ($E = 0.8$ eV) [4]. It is unlikely that the shift of the $4f^1$ peak with respect to the position of Ref. [1] be due to segregation or oxidation effects because the sample was regularly cleaned and the measurements were carried out at 20 K, a temperature where diffusion is negligible. Then, if we agree with Weschke et al. that the simple SI Anderson model does not give a satisfactory description of the $4f$ spectral function in $CeRh_3$ (e.g., the temperature dependence of the photoemission spectrum and the absence of $4f^2$ structures observed in Ref. [1]) we think that it is not possible to conclude that a bandlike picture correctly describes the $4f$ states.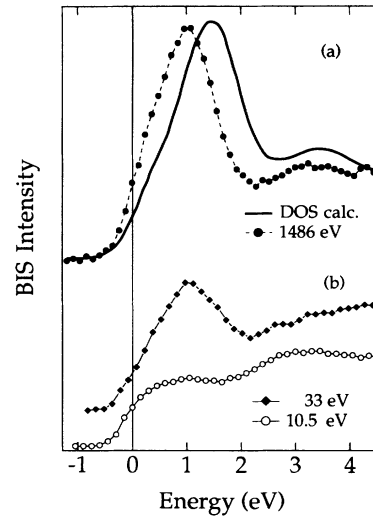 Therefore, the nature of the $4f$ states, itinerant or localized, in strongly hybridized systems remains an open question.

FIG. 1. (a) Dots: experimental spectrum of $CeRh_3$ with the calibration obtained from the Fermi step of a Cu film evaporated onto the sample; solid line: calculated spectrum from the LDA-DOS of Ref. [1]. (b) Inverse photoemission measurements in the UV range.

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