Surface-bulk differences in a Kondo system, CeB$_6$

Kalobaran Maiti,$^1$ Nishaina Sahadev,$^1$ Khadiza Ali,$^1$ Swapnil Patil,$^1$ Deepnarayan Biswas,$^1$
Sangeeta Thakur,$^1$ and Geetha Balakrishnan$^2$

$^1$ Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Colaba, Mumbai - 400 005, India.

$^2$ Department of Physics, University of Warwick, Coventry, CV4 7AL, UK.

Kondo effect appears due to the antiferromagnetic coupling (Kondo coupling) between the local magnetic moment and conduction electrons leading to a non-magnetic phase. In the strong coupling regime, the conduction electrons exhibit a sharp feature at the Fermi level at low temperatures, called Kondo resonance feature. The intensity of the feature decreases with the increase in temperature and vanishes at temperatures sufficiently higher than the Kondo temperatures. This phenomena has extensively been studied during past few decades. The puzzle that has been bothering for long is the observation of strong Kondo resonance feature even at temperatures significantly higher than the Kondo temperature.[1, 2, 3] Here, we studied the evolution of Kondo resonance feature in a typical dense Kondo system, CeB$_6$ employing high resolution x-ray photoemission spectroscopy. High quality single crystalline sample was prepared by mirror furnace. The photoemission measurements were carried out at P09 beam line, PETRA III Hamburg, Germany with 5947.9 eV photon energy at an energy resolution of 150 meV and monochromatic Al K$\alpha$ ($h\nu = 1486.6$ eV) laboratory source at TIFR, Mumbai at an energy resolution of 350 meV. The experiment temperature was varied using an open cycle helium cryostat. The sample was fractured in the ultra high vacuum chamber before each measurement.

![Intensity vs Binding Energy](image1.png)

Figure 1: (a) Ce 3d core level spectra at 40 K collected using Al K$\alpha$ radiations and hard x-rays. Valence band spectra at 40 K and 200 K using (b) Al K$\alpha$ radiations and (c) hard x-rays.

In Fig. 1(a), we show the Ce 3d core level spectra obtained at both the photon energies. Each of the core level spectra exhibits multiple features; the most intense features appear around 887 eV and 905 eV binding energies along with several weak features in the lower binding energy region. The intense features can be attributed to the spin-orbit split photoemission transition, $|3d^1\,4f^1\rangle \rightarrow$
known as poorly screened feature, where the 3d core hole is not screened in the final state of photoemission. The weak features at lower binding energies appear due to the well screened final states (transition: \(|3d^4f^1 \rightarrow |3d^4f^2 \mathcal{C}\rangle\), \(\mathcal{C}\) is a hole in the conduction band), where a conduction electron screens the core hole positive charge created by photoemission.[4]

In addition to these features, a sharp feature is observed around 918 eV binding energy in the Al K\(\alpha\) spectrum. This feature is often attributed to the photoemission signal corresponding to \(|3d^4f^0 \rightarrow \text{final state}\), where the \(f\)-electron formed an entangled state with the conduction electrons. Thus, the presence of this feature provides an evidence of the Kondo resonance in the core level spectra.[2, 3, 4, 5] Interestingly, the same feature appears at much lower binding energy (~ 915.5 eV) in the hard x-ray spectrum collected at the same temperature, and the feature becomes weaker and broader. Such a scenario indicates lower Kondo temperature in the corresponding electronic structure. Since the photoemission technique becomes significantly bulk sensitive at hard x-ray energy regime,[6] the above observation suggests a lower Kondo temperature in the bulk while the surface has higher Kondo temperature.

In order to verify this, we have probed the valence band spectra using both the photon energies - the experimental data are shown in Fig. 1(b) and 1(c). The spectra in 1(b) corresponding to the Al K\(\alpha\) energy exhibit two distinct features around 0.5 eV and 2.3 eV binding energies corresponding to the Kondo resonance peak and the lower Hubbard band.[3] The Kondo resonance peak becomes weaker at higher temperature indicating typical Kondo behavior of this feature. The same scenario is observed in Fig. 1(c), where the relatively more bulk sensitive spectra is shown. Clearly, the temperature dependence in this later case is significantly weaker than that observed in Fig. 1(b) consistent with the observations in the core level spectra.

In summary, we studied the Kondo resonance feature in CeB\(_6\) using 1486.6 eV and 5947.9 eV photon energies. While the Kondo behavior has been observed in both valence band and core level spectra, the temperature dependence of the spectral features is significantly stronger in the surface electronic structure compared to the bulk. These results indicate an enhancement of the Kondo temperature at the surface that can explain the observation of large Kondo features in the high resolution photoemission measurements, which is a significantly surface sensitive at these energies. Theoretical studies are required to understand this phenomena in more detail.

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References