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Angle-Resolved Photoemission from the Valence-Bands of Ce(111)

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Introduction

Rare earth elements are characterized by their partially filled 4*f*-shell. Even in solid-state crystals, the 4*f* electrons are highly localized and basically preserve atomic many-body states. The 4*f* state is not stable. The 4*f* electrons exhibit pronounced valence fluctuations. A typical example of a valence fluctuating system is metallic Ce, which has been intensively studied especially because of its unique isostructural (fcc to fcc) γ-α phase transition with increasing temperature. We report the results of angle-resolved (ARPES) and resonant photoemission of epitaxial thin films of Ce grown on W(110). The evolution of γ→α→γ phase transition of Ce is observed with increasing Ce coverage and the 5*d*6*s* valence-band structures of γ-like Ce film are determined.

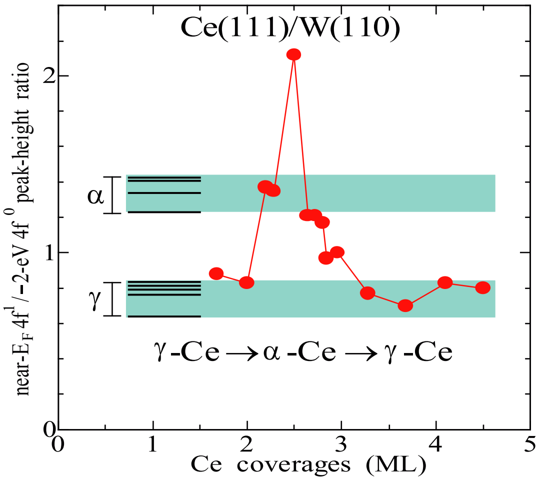
Experimental

ARPES spectra were measured using a VSW. The energy resolution was 50-100 meV. Ce films deposited on the clean W(110) surface showed sharp hexagonal LEED patterns corresponding to the formation of well-ordered Ce(111) surfaces. No traces of any contaminations were found by AES. The amounts of deposits were determined from the Auger-peak-intensity ratio $I_{Ce(82eV)}/I_{W(169eV)}$.

Results and Discussion

We measured the valence-band ARPES spectra at normal emission with $h\nu$=122 eV, exploiting a giant resonant enhancement of the 4*f* photoionization cross section as a function of Ce coverage. At this $h\nu$=122 eV, the contributions of the Ce 4*f* electrons dominate the spectra, whereas non-4*f* contributions to the Ce spectra are negligible. The observed spectra reveal a characteristic double-peaked structure: a well-screened $4f^1$ final-state feature near E_F and a poorly-screened $4f^0$ final-state feature at about 2 eV below E_F.

Figure shows the Ce-coverage dependence of the height ratio of the near-E_F $4f^1$ peak to the −2-eV $4f^0$ peak. It is found that the $4f^1/4f^0$ ratio is changed with increasing Ce coverage: the ratio is ~0.8 below ~2 ML, increases to ~2.1 (max.) between ~2 ML and ~3 ML, and then decreases to the initial value of ~0.8 beyond ~3 ML. The $4f^1/4f^0$ ratio has been reported to be ~0.8 for γ-Ce and ~1.2-1.4 for α-Ce. With increased coupling of 4*f* electrons to valence-band electrons, screening becomes more likely and the intensity of the screened $4f^1$ emission feature increases at the expense of the unscreened $4f^0$ feature, *and vice versa*. The changes in relative intensity of the two peaks with increasing coverage are similar to those for the γ→α (lower coverage) or α→γ (higher coverage) transition. Therefore, it can be concluded that the Ce film grown on W(110) is γ-like below ~2 ML, becomes α-like around ~2.5 ML, and then finally goes back to γ-like above ~3.5 ML.



We measured off-normal emission spectra of Ce(111)/W(110) in the valence-band region measured as a function of the emission angle θ_e, and determined the E-versus-$k_{//}$ dispersion [$E(k_{//})$] for Ce(111) (not shown here).

Our data demonstrated importance of using well-ordered and atomically clean Ce(111) epitaxial films grown on a well-ordered and atomically clean W(110) surface for carrying out Ce photoemission experiments. Recent studies claimed that all attempts to prepare a monocrystalline film of α-Ce metal have failed and the surface of α-Ce should be γ-like, since surface effects occur in α-Ce, leading to a γ-like surface layer. However, our experiments revealed that that is not necessarily right.

The surface is expected to represent an important perturbation of the electronic structure of Ce metal, such as substantially reduced hybridization strength at the surface. Ce ordered overlayers in various thickness grown on appropriate substrates may tune the hybridization strength in a controlled way. Thus, the surface environment is expected to throw new light on the investigation of the new physical properties of Kondo systems. Carefully prepared and characterized single-crystal Ce metal surfaces can open the door for studies of ambitious experiments, instead of considering the surface only as a nuisance.

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