



Electronic structure of LaTe and CeTe



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ABSTRACT

We report a comparative study of the electronic structure of the compounds LaTe and CeTe, both of which crystallize in the rock salt structure. LaTe is a paramagnetic metal while CeTe is known to exhibit anomalous Kondo-like transport behaviour and undergoes a transition to a complex magnetically ordered state at low temperature ($T_N = 2.2$ K). We carry out hard X-ray photoelectron spectroscopy (HAXPES) of the core-levels and valence band of LaTe and CeTe at $T = 20$ K, in order to characterize their intrinsic electronic structure, and to address the role of Kondo effect on the electronic structure of CeTe. The bulk sensitive core level HAXPES spectra show evidence of screened features in the La 3d and Ce 3d states mixed with plasmon features. From a careful analysis of the Te, La and Ce derived core levels, we separate out the respective origins of the satellites and show that CeTe indeed exhibits definitive but weak f^0 and f^2 satellites due to Kondo screening, in addition to the main f^1 peak. The comparison of the valence band spectra of CeTe obtained using HAXPES and soft X-ray PES clearly identifies the Ce 4f derived features. Resonant photoelectron spectroscopy across the Ce 3d – 4f threshold confirms the Ce 4f¹ final state at the Fermi level, corresponding to the tail of the Kondo resonance feature which occurs above the Fermi level, while the Ce 4f⁰ final state feature is observed at a binding energy of 2.4 eV. The 4f⁰ and 4f¹ final states show giant resonances compared to the off-resonant spectra. However, in contrast to typical Kondo systems, the tail of the Ce 4f¹ Kondo resonance at the Fermi level is relatively suppressed compared to the Ce 4f⁰ feature, which exhibits an unusually strong resonance enhancement. The results are indicative of a weakened Kondo effect which favours the magnetically ordered ground state in CeTe.

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1. Introduction

The rare-earth monochalcogenides RX (R = rare-earth, such as La, Ce, Sm, Tm; X = S, Se, Te) have been extensively studied experimentally and theoretically due to the variety of properties they exhibit and because they crystallize in the simple rock salt structure [1–4]. Well-known examples include SmS, which shows a pressure-induced metal-insulator transition, and TmSe which shows an anomalous valence fluctuating state. These properties are considered to originate from the role of strong electron–electron correlations of the 4f-electrons vis-a-vis the hybridization between the 4f and conduction electrons. The Ce-monochalcogenides (CeX, X = S, Se and Te) are regarded as the simplest of these systems

because the Ce ion is expected to be in the trivalent state with one occupied 4f electron ($\text{Ce}^{3+} \equiv 4f^1$).

In many Ce-based systems, the Kondo effect is known to compete with the Rudermann–Kittel–Kasuya–Yoshida (RKKY) interaction, leading to a non-magnetic ground state with a Kondo/Abrikosov–Suhl resonance at the Fermi level, or a magnetically ordered ground state, respectively [5–10]. The CeX series is known to exhibit unusual properties [5–9]. CeTe shows evidence for a dense Kondo lattice in the resistivity, which is quite complex as it shows a weak maximum at $T_m = 2.7$ K and a sharp drop at $T_N = 2.2$ K [9,11]. So it is difficult to quantitatively estimate T_K of CeTe, but it can be roughly estimated as $3T_m \sim 8$ K, based on the work of Bickers, Cox and Wilkins [12]. The smaller crystal field splitting in CeTe compared to CeS and CeSe obstructs the observation of the $\ln T$ term at low temperatures, but an indication is observed in $d\rho/dT$ [9]. A weakly logarithmic behavior is seen upto ~ 30 K where it has another maximum associated with the crystal field splitting Δ i.e. the splitting between the Γ_7 doublet and Γ_8 quartet [9]. At very low

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temperatures, the series CeX exhibits the unusual type-II antiferromagnetic order indicative of a dominantly RKKY interaction [9,10]. Among the series, CeTe exhibits the lowest $T_N = 2.2$ K while CeS has the highest $T_N = 8.4$ K [11]. The magnetic moments are also anomalous with a $\mu_{\text{eff}} = 0.30\mu_B$ for CeTe, and $\mu_{\text{eff}} = 0.57\mu_B$ for CeS, while the expected value for the Γ_7 doublet is $\mu_{\text{eff}} = 0.71\mu_B$. These differences are attributed to the crystal-field splitting which is estimated to be ~ 30 K for CeTe and ~ 140 K for CeS [11]. Most interestingly, an angle-resolved photoelectron spectroscopy (ARPES) study of the LaX and CeX series showed that CeTe was unique in the series. The authors showed that CeX can be considered to show the electronic structure of the isostructural LaX series with an added feature due to the localized 4f-level, but for CeTe, an additional weakly dispersive feature at 2.4 eV binding energy was observed and attributed to ligand states hybridizing with the 4f-level i.e. $p-f$ mixing [13].

In this study, we carry out comparative electronic structure measurements of LaTe and CeTe using a combination of hard X-ray and soft X-ray photoelectron spectroscopy (HAXPES and SXPES), X-ray absorption spectroscopy (XAS), as well as Resonant PES (Res-PES). HAXPES studies have played an important role in the study of the bulk-sensitive intrinsic electronic structure of solids due to the large inelastic mean free paths of photoemitted electrons with high kinetic energies [14–19]. However, there is no report to date of a very low T_K Kondo system using the combination of HAXPES and Ce 3d–4f Res-PES. Core-level HAXPES spectra of LaTe and CeTe show evidence of screened features in the La 3d and Ce 3d states mixed with plasmon features. From a careful analysis of the Te and Ce derived core levels, we show that Ce 3d spectra of CeTe indeed exhibits definitive but weak f^0 and f^2 satellites due to Kondo screening, in addition to the main f^1 peak. The comparison of the valence band spectra of CeTe obtained using HAXPES and SXPES clearly identifies the Ce 4f derived features. XAS and Res-PES across the Ce 3d–4f threshold reveals giant resonances of the Ce 4f¹ final state at the Fermi level, and of the Ce 4f⁰ final state observed at a binding energy of 2.4 eV. In contrast to typical Kondo systems, the Ce 4f¹ feature at the Fermi level (which corresponds to the tail of the Kondo resonance above the Fermi level) is relatively suppressed, while the Ce 4f⁰ feature exhibits an unusually strong resonance. The behaviour of the tail of the Kondo resonance at the Fermi level indicates a weakened Kondo effect and favours the magnetically ordered ground state in CeTe.

2. Experimental details

High-quality single crystals of LaTe and CeTe used in the present study were grown by the Bridgman method using a tungsten heater furnace. X-ray diffraction measurements confirmed the structure and the lattice constants of the crystals were in good agreement with the reported values. The residual resistivity ratio for both LaTe and CeTe was about 20 and this is about one order of magnitude larger than that reported in the literature, indicating the high quality of our samples [11]. HAXPES measurements were carried out at beamline BL19XU, SPring-8 using an incident photon energy of $h\nu = 7950$ eV and a spectrometer [20,21] equipped with a R-4000-10kV VG-Scienta analyzer. The total energy resolution was set to 225 meV for the HAXPES measurements. The soft X-ray PES was performed at BL17SU, at SPring-8 using a Gamdata-Scienta SES2002 hemispherical electron analyzer set to the total energy resolution of 200 meV. XAS was recorded in the total electron yield mode. All measurements were carried out at a sample temperature of $T = 20$ K obtained using a flowing liquid He cryostat. The measurements were carried out in a vacuum below 4×10^{-8} Pa and clean sample surfaces were obtained by cleaving. For the HAXPES measurements, the cleaving was done in the preparation chamber at room temperature and the sample was transferred immediately

into the analysis chamber and cooled, while for the SXPES, XAS and Res-PES measurements, the cleaving was carried out *in-situ* at $T = 20$ K. The spectra were calibrated using the Fermi level (E_F) measured from a gold film evaporated onto the sample holder.

3. Results and discussions

Fig. 1(a) and (b) shows the Te 2s HAXPES ($h\nu = 7950$ eV) core level spectra of LaTe and CeTe, respectively, measured at $T = 20$ K. The spectra show a clean single peak at 4940.0 eV binding energy for both LaTe and CeTe. This suggests that the charge transfer from La and Ce to Te is quite similar and thus, the La and Ce core levels maybe expected to be trivalent. In addition, for both the compounds, we see two weak satellites at higher binding energies. From a least-squares curve fitting analysis, the satellite positions were determined to be ~ 14 eV and ~ 28 eV higher binding energies compared to the main peak. These satellites are assigned to plasmon excitations of LaTe and CeTe, as we see very similar satellites in the La 3p and Ce 3p, as discussed in the following.

Fig. 2(a) shows the La 3p_{3/2} core level spectrum of LaTe and Fig. 2(b) shows the Ce 3p_{3/2} core level spectrum of CeTe measured at $T = 20$ K and using an incident photon energy of $h\nu = 7950$ eV. The La 3p_{3/2} shows a main peak at 1128.6 eV binding energy corresponding to La³⁺, followed by satellites at ~ 14 eV, ~ 28 eV and ~ 42 eV from the main peak. From a least-squares curve fitting analysis, the satellite positions could be determined accurately. Since the energy shifts from the main peak match the satellites for the Te 2s spectrum discussed in Fig. 1, we assign them to plasmon excitations of LaTe. For CeTe, the Ce 3p_{3/2} shows a main peak at 1186.0 eV binding energy typical of Ce³⁺, followed by satellites again at ~ 14 eV, ~ 28 eV and ~ 42 eV from the main peak, as determined by a least squares fit, and are thus assigned to plasmons. The only difference for La 3p_{3/2} and Ce 3p_{3/2} core levels compared to the Te 2s

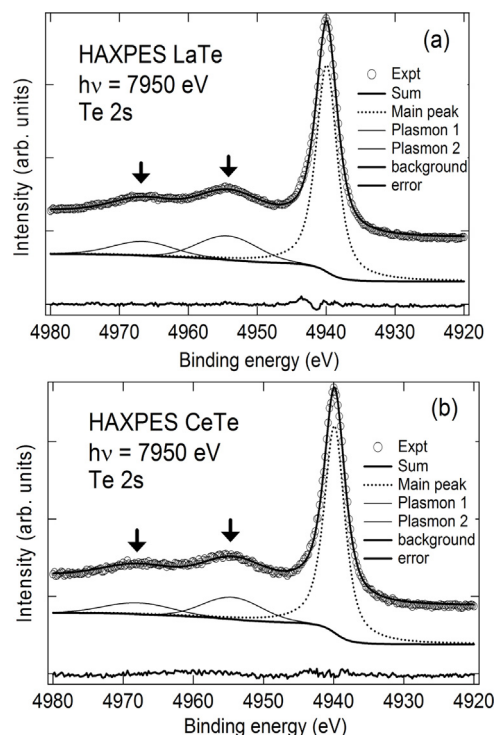


Fig. 1. (a) The Te 2s HAXPES core level spectrum of LaTe shows a main peak (binding energy 4940.0 eV) and plasmon satellites at ~ 14 eV and ~ 28 eV from the main peak. (b) The Te 2s HAXPES core level spectrum of CeTe also shows a main peak at the same binding energy as LaTe, and also plasmon satellites at nearly the same energies as LaTe.

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