

Determining the local low-energy excitations in the Kondo semimetal CeRu₄Sn₆ using resonant inelastic x-ray scattering

Andrea Amorese,^{1,2} Kurt Kummer,³ Nicholas B. Brookes,³ Oliver Stockert,² Devashibhai T. Adroja,⁴ André M. Strydom,⁵ Andrey Sidorenko,⁶ Hannes Winkler,⁶ Diego A. Zocco,⁶ Andrey Prokofiev,⁶ Silke Paschen,⁶ Maurits W. Haverkort,⁷ Liu Hao Tjeng,² and Andrea Severing^{1,2}

¹*Institute of Physics II, University of Cologne, Zùlpicher StraÙe 77, 50937 Cologne, Germany*

²*Max Planck Institute for Chemical Physics of Solids, Nöthnitzer StraÙe 40, 01187 Dresden, Germany*

³*European Synchrotron Radiation Facility, 71 Avenue des Martyrs, CS40220, F-38043 Grenoble Cedex 9, France*

⁴*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot Oxon OX11 0QX, United Kingdom*

⁵*Highly Correlated Matter Research Group, Physics Department, University of Johannesburg, P.O. Box 524, Auckland Park 2006, South Africa*

⁶*Institute of Solid State Physics, Vienna University of Technology, Wiedner HauptstraÙe 8-10, 1040 Vienna, Austria*

⁷*Institute for Theoretical Physics, Heidelberg University, Philosophenweg 19, 69120 Heidelberg, Germany*

(Dated: June 14, 2018)

We have investigated the local low-energy excitations in CeRu₄Sn₆, a material discussed recently in the framework of strongly correlated Weyl semimetals, by means of Ce *M*₅ resonant inelastic x-ray scattering (RIXS). The availability of both ²F_{5/2} and ²F_{7/2} excitations of the Ce 4*f*¹ configuration in the spectra allows for the determination of the crystal-electric field parameters that explain quantitatively the temperature dependence and anisotropy of the magnetic susceptibility. The absence of an azimuthal dependence in the spectra indicates that all crystal-electric field states are close to being rotational symmetric. We show further that the non-negligible impact of the \tilde{A}_6^0 parameter on the ground state of CeRu₄Sn₆ leads to a reduction of the magnetic moment due to multiplet intermixing. The RIXS results are consistent with inelastic neutron scattering (INS) data and are compared to the predictions from *ab-initio* based electronic structure calculations.

In several Ce compounds the localized 4*f* electrons hybridize with the conduction electrons (*cf*-hybridization) so that hybridization gaps can form and give rise to Kondo insulating, semiconducting or semimetallic ground states.¹ These materials are presently the focus of interest due to the proposal that the combination of strong spin-orbit coupling, bands of opposite parity (4*f* and 5*d*), plus the hybridization induced gap should give rise to strongly correlated non-trivial topological phases.²⁻⁶ CeRu₄Sn₆ is a tetragonal, non-centrosymmetric (*I42m*)⁷ compound. Its electrical resistivity increases as temperature decreases which has been attributed to the opening of a hybridization gap of the order of 100 K.⁸⁻¹² The absence of magnetic order down to 50 mK¹³ and the non-integer valence of 3.08^{14,15} confirm the importance of strong *cf*-hybridization. Recently, band structure calculations in the LDA+Gutzwiller scheme have suggested that CeRu₄Sn₆ is a correlated Weyl semimetal,¹⁶ a conjecture that remains to be tested experimentally, especially since the non-centrosymmetric crystal structure complicates the prediction for gap openings after a band inversion.

To understand the properties of CeRu₄Sn₆ and to assess the reliability of the theoretical predictions we need to know not only the ground state but also the low-energy excitations of this system. The linear dichroism (LD) in soft x-ray absorption (XAS) and the direction dependence in non-resonant inelastic scattering (NIXS) have shown that the crystal-electric field (CEF) ground state symmetry must be the Γ_6 ¹⁴ in agreement with magne-

tization measurements.¹⁰ However, there is so far no information about the CEF level scheme, i.e. about the energy splittings ΔE_1 and ΔE_2 and the mixing factor α of the excited CEF states. The present resonant inelastic x-ray scattering (RIXS) study aims at giving a full description of the CEF level scheme of CeRu₄Sn₆.

In an ionic model the trivalent (4*f*¹) configuration of Ce is split by the effect of spin-orbit interaction (≈ 280 meV) in two multiplets, ²F_{5/2} and ²F_{7/2}, with 6-fold ($J_z = \{-\frac{5}{2}; \dots; +\frac{5}{2}\}$) and 8-fold degeneracy ($J_z = \{-\frac{7}{2}; \dots; +\frac{7}{2}\}$). This degeneracy is further reduced by the interaction with the surrounding ions in the crystal and can be modeled with an effective CEF potential, written as a sum of (renormalized) spherical harmonics

$$C_k^m = \sqrt{\frac{4\pi}{2k+1}} Y_k^m.$$

$$V_{CEF}(r, \theta, \Phi) = \sum_{k=0}^{\infty} \sum_{m=-k}^k A_k^m r^k C_k^m(\theta, \Phi).$$

The expectation values $\langle r^k \rangle$ cannot be calculated *ab-initio* and are usually included in the phenomenological CEF parameters $\tilde{A}_k^m = A_k^m \langle r^k \rangle$ that must be determined experimentally. Five independent parameters \tilde{A}_2^0 , \tilde{A}_4^0 , $\tilde{A}_4^{\pm 4}$, \tilde{A}_6^0 and $\tilde{A}_6^{\pm 4}$ fully describe the CEF problem for a Ce³⁺ ion with tetragonal point symmetry as in CeRu₄Sn₆. Non-zero $\tilde{A}_4^{\pm 4}$ and $\tilde{A}_6^{\pm 4}$ mix the J_z states according to $\Delta J_z = 4$, i.e. $J_z = \pm \frac{3}{2}$ and $\mp \frac{5}{2}$, and $J_z = \pm \frac{1}{2}$ and $\mp \frac{7}{2}$, respectively. The intermixing of the two J multiplets ²F_{5/2} and ²F_{7/2} is usually negligible and the impact