Orientation of ground-state orbital in CeCoIn\textsubscript{5} and CeRhIn\textsubscript{5}

M. Sundermann,\textsuperscript{1,2} A. Amorese,\textsuperscript{1,2} F. Strigari,\textsuperscript{1,2} M. W. Haverkort,\textsuperscript{3} L. H. Tjeng,\textsuperscript{2} M. Moretti Sala,\textsuperscript{4,†} H. Yavas,\textsuperscript{5,†} E. D. Bauer,\textsuperscript{6} P. F. S. Rosa,\textsuperscript{6} J. D. Thompson,\textsuperscript{6} and A. Severing\textsuperscript{1,2}

\textsuperscript{1}Institute of Physics II, University of Cologne, Zülpicher Straße 77, 50937 Cologne, Germany
\textsuperscript{2}Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Straße 40, 01187 Dresden, Germany
\textsuperscript{3}Institute for Theoretical Physics, Heidelberg University, Philosophenweg 19, 69120 Heidelberg, Germany
\textsuperscript{4}European Synchrotron Radiation Facility, 71 Avenue des Martyrs, CS40220, F-38043 Grenoble Cedex 9, France
\textsuperscript{5}PETRA III, Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, 22607 Hamburg, Germany
\textsuperscript{6}Los Alamos National Laboratory, New Mexico 87545, USA

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We present core level non-resonant inelastic x-ray scattering (NIXS) data of the heavy fermion compounds CeCoIn\textsubscript{5} and CeRhIn\textsubscript{5} measured at the Ce N\textsubscript{4,5}-edges. The higher than dipole transitions in NIXS allow determining the orientation of the Γ\textsubscript{7} crystal-field ground-state orbital within the unit cell. The crystal-field parameters of the CeMIn\textsubscript{5} compounds and related substitution phase diagrams have been investigated in great detail in the past; however, whether the ground-state wavefunction is the Γ\textsubscript{7}\textsuperscript{+} (x\textsuperscript{2} − y\textsuperscript{2}) or Γ\textsubscript{7} (xy orientation) remained undetermined. We show that the Γ\textsubscript{7}\textsuperscript{+} doublet with lobes along the (110) direction forms the ground state in CeCoIn\textsubscript{5} and CeRhIn\textsubscript{5}. A comparison is made to the results of existing DFT+DMFT calculations.

I. INTRODUCTION

At high temperature, heavy-fermion materials are described by decoupled localized f electrons and conduction electron bands. Upon cooling, the localized f electrons start to interact with the conduction electrons (cf-hybridization) and become partially delocalized. The resulting entangled fluid consists of heavy quasiparticles (hybridization) and become partially delocalized. The resulting entangled fluid consists of heavy quasiparticles with masses up to three orders of magnitude larger than the free electron mass. These quasiparticles may undergo magnetic or superconducting transitions. In the Doniach phase diagram temperature T versus exchange interaction J\textsubscript{f}, magnetic order prevails for small J\textsubscript{f} whereas a non-magnetic Kondo singlet state forms for large J\textsubscript{f}. Between these two regimes quantum critical behaviour occurs which is often accompanied by a superconducting dome that hides a quantum critical point (QCP).\textsuperscript{1,2} Understanding how these quasiparticles, that have atomic-like as well as itinerant character, give rise to these ground states is a challenging question in condensed-matter physics, and the answer to this question will provide predictive understanding of these quantum states of matter.\textsuperscript{3}

The tetragonal compounds CeMIn\textsubscript{5} (M = Co, Rh, Ir) are heavy fermion compounds that display different ground states for different transition metal ions; for M = Co and Ir the ground state is superconducting (T\textsubscript{c} = 2.3 and 0.4 K) and for M = Rh it is antiferromagnetic (T\textsubscript{N} = 3.8 K).\textsuperscript{4} High-quality CeMIn\textsubscript{5} crystals can be grown, making this family suitable for determining the parameter that drives the different ground states. Within the above mentioned extended Doniach phase diagram, CeRhIn\textsubscript{5} is on the weak side of hybridization, CeCoIn\textsubscript{5} close to the QCP and CeIrIn\textsubscript{5} is on the side of stronger cf-hybridization, i.e. superconductivity goes along with stronger cf-hybridization. Although there are strong indications for localization (Rh) and delocalization (Co,Ir) in, e.g., the size of the Fermi surface,\textsuperscript{5,12} it is not possible to detect the differences in f occupations. They are so subtle that they are below the detection limit.\textsuperscript{13}

A light-polarization analysis of soft X-ray absorption spectra shows that the crystal-field wavefunction of the ground state correlates with the ground state properties in the temperature-transition metal (substitution) phase diagram of CeCoIn\textsubscript{5} - CeRhIn\textsubscript{5} - CeRh\textsubscript{1−δ}Ir\textsubscript{δ}In\textsubscript{5} - CeIrIn\textsubscript{5}; orbitals more compressed in the tetragonal ab-plane favour an antiferromagnetic ground state as for CeRhIn\textsubscript{5} and the Rh rich compounds with δ ≤ 0.2. The compounds with more elongated orbitals along the c axis, however, have superconducting ground states (CeCoIn\textsubscript{5}, CeIrIn\textsubscript{5} and also the Ir rich compounds with δ ≥ 0.7).\textsuperscript{14,15} The obvious conclusion is that the more pronounced extension of the ground state orbitals in the direction of quantization (crystallographic c direction) promotes stronger hybridization in c direction since superconductivity goes along with stronger hybridization. This is supported by combined local density approximation plus dynamical mean field theory (LDA+DMFT) calculations by Shim \textit{et al.} who find for CeIrIn\textsubscript{5} the strongest hybridization with the out-of-plane In(2) ions (see unit cell in Fig. 1 (a)). It was also shown that the suppression of superconductivity in CeCo(In\textsubscript{1−δ}Sn\textsubscript{δ})\textsubscript{5} by about 3% of Sn is due to a homogeneous increase of hybridization in the tetragonal ab plane since the Sn ions go preferably to the In(1) sites.\textsuperscript{17} Accordingly, we found that here the hybridization with In(1) ions plays a decisive role; the 4f ground state orbital extends increasingly in the plane as the Sn content is increased.\textsuperscript{18}

Hence, the ground state wavefunction is a very sensitive probe for quantifying hybridization. Haule \textit{et al.} obtained a 4f Weiss field hybridization function for CeMIn\textsubscript{5} based on realistic lattice parameters using density functional theory plus dynamical mean field (DFT+DMFT) calculations which they have decomposed into crystal-field components.\textsuperscript{19} Here our goal is to verify that the crystal-field components that were extracted in these cal-
culations are in agreement with reality.

The tetragonal point symmetry of Ce in CeMIn$_5$ splits the Ce Hund’s rule ground state into three Kramers doublets, two $\Gamma_7$ doublets $\Gamma_{7}^{+/-} = |\alpha| \pm |\pm 5/2| \pm \sqrt{1-\alpha^2}|\mp 3/2|$ and $\Gamma_{7}^{-/+} = \sqrt{1-\alpha^2}|\pm 5/2| - / + |\alpha| \mp |3/2|$, and one $\Gamma_6 = |\mp 1/2|$. We write $+/ -$ or $-/-$ because the sign has not yet been determined, and this is the scope of the present manuscript. $\Gamma_6$ as a pure $J_z$ state has full rotational symmetry around the quantization axis $c$ but the mixed states have lobes with fourfold rotational symmetry. The magnitude of $\alpha$ describes the extension of the $\Gamma_7^{+/-}$ orbitals in $c$ direction whereby the sign in the wavefunction determines how the orbitals are oriented within the unit cell; with the lobes along [100] ($\Gamma_7^+: x^2 - y^2$) or with the lobes along [110] ($\Gamma_7^−: xy$). Figure 1 (b) shows the scenario of a $\Gamma_7^−$ ground state and $\Gamma_7^+$ excited state for several $\alpha^2$ values. Figure 1 (c) shows the same for a $\Gamma_7^+$ ground and excited $\Gamma_7^−$-state.

The crystal-field potential of the CeMIn$_5$ has been determined withinelastic neutron scattering (INS) and the ground state wavefunctions were studied in greater detail with linear polarized soft x-ray absorption spectroscopy. Hence, the crystal-field energy splittings, the sequence of states $(\Gamma_{7}^{+/-}, \Gamma_{7}^{+/-}, \Gamma_6)$ and also the magnitude of the $\alpha$-values are known (0.36, 0.62, 0.5 for Co, Rh and Ir). Only the sign of the wavefunction remains unknown because it cannot be determined with any of these dipole-selection-rule based spectroscopies. We, therefore, set up an experiment to determine the sign of the ground-state wave function in the CeMIn$_5$ compounds in order to find out which one of the two scenarios in Fig. 1 (a) applies.

II. METHOD

We performed a core level non-resonant inelastic x-ray scattering (NIXS) experiment at the Ce $N_{1,5}$-edges (4d $\rightarrow$ 4f). It has been shown previously that this method is able to detect anisotropies with higher than twofold rotational symmetry. In the following, we briefly recap the principles of NIXS, a photon-in photon-out technique with hard x-rays ($E_{\text{in}} \approx 10$ keV). Because of the high incident energies, NIXS is bulk sensitive and allows one to reach large momentum transfers $|q|$ of the order of 10 Å$^{-1}$ when measuring in back scattering geometry. At such large momentum transfers, the transition operator in the scattering function $S(q^2\omega)$ can no longer be truncated after the dipole term. As a result, higher order scattering terms contribute to the scattering intensity. For a $d \rightarrow f$ transition at about 10 Å$^{-1}$, octupole (rank $k=3$) and triakontadipole ($k=5$) terms dominate the scattering intensity whereas the dipole part ($k=1$) is less prominent. Accordingly, the directional dependence of the scattering function in a single crystal experiments follows multipole selection rules, in analogy to the dipole selection rules in a linearly polarized x-ray absorption experiment. Thus single crystal NIXS yields information not only about the orbital occupation but also the sign of the wavefunction that distinguishes the $xy$ and $x^2 - y^2$ orientations of a $\Gamma_7$.

III. EXPERIMENT

CeCoIn$_5$ and CeRhIn$_5$ single crystals were grown using the standard In-flux technique. CeCoIn$_5$ crystals are plate-like with the [001] direction perpendicular to the plate, whereas CeRhIn$_5$ crystals are more three-dimensional. All samples were aligned by Laue diffraction before the experiment. For each compound two samples were cut, one with a (100) and a second one with a (110) surface so that specular geometry could be realized in the experiment.

The experiments were performed at the NIXS end stations of two beamlines, ID20 at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France and the Max-Planck P01 at PETRA III/DESY in Hamburg, Germany. CeCoIn$_5$ was measured at ID20. The ID20 setup was as follows: the incident energy is scanned using a double Si(111) monochromator and the energy of the scattered intensity was set to $E_{\text{final}}=9690$ eV. The scattered intensity was analyzed by three columns of three Si(660) analyzers (in total nine)
at in-plane scattering angles of $2\Theta = 140^\circ, 146^\circ$, and $153^\circ$, corresponding to an averaged momentum transfer of $|\vec{q}| = 9.4 \pm 0.15 \text{ Å}^{-1}$. CeRhIn$_5$ was measured at P01 with a slightly different set up; the resolution was better and the scattering geometry was vertical so that the polarization vector of the incoming light was vertical to the scattering plane. At P01 the incident energy was selected with a Si(311) double monochromator and twelve Si(660) 1 m radius spherically bent crystal analyzers were arranged in 3 x 4 array as shown in Fig. 2 of Ref. 26 so that the fixed final energy was the same as at ID20. The analyzers were positioned at scattering angles of $2\theta \approx 150^\circ, 155^\circ$, and $160^\circ$ which provide an averaged momentum transfer of $|\vec{q}| = 9.6 \pm 0.1 \text{ Å}^{-1}$. At both beamlines the scattered beam was detected by a position sensitive custom-made detector (MAXIPIX at ID20, LAMBDA at P01), based on a Medipix3 chip detector. The elastic line was consistently measured and a pixel-wise calibration yields instrumental energy resolutions of $\approx 1.3 \text{ eV}$ at ID20 and $\approx 0.7 \text{ eV}$ full width at half maximum (FWHM) at P01. The improved resolution costs about a factor of five in intensity. Two samples of CeCoIn$_5$ and CeRhIn$_5$ were measured, one with a (100) and one with a (110) surface so that specular geometry for $\vec{q} \parallel [001]$ and $\vec{q} \parallel [110]$ could be realized.

We used the full multiplet code Quanty$^{35}$ for simulating the NIXS data. A Gaussian broadening of 1.3 eV accounts for the instrumental resolution of the ID20 data (CeCoIn$_5$) and of 0.7 eV for the P01 spectra (CeRhIn$_5$). An additional Lorentzian broadening of 0.4 eV FWHM accounts for life-time effects. The atomic parameters were taken from the Cowan code,$^{36}$ whereby the Hartree-Fock values of the Slater integrals were reduced to about 60% for the 4f-4f and to about 80% for the 4d-4f Coulomb interactions to reproduce the energy distribution of the multiplet excitations of the Ce $N_{4,5}$-edges. This reduction accounts for configuration interaction processes not included in the Hartree-Fock scheme.$^{37}$

IV. RESULTS

Figure 2 shows NIXS data (dots) at the Ce $N_{4,5}$-edges of CeRhIn$_5$ (a) and CeCoIn$_5$ (b) plus simulations (lines) for two scattering directions, $\vec{q} \parallel [100]$ (blue) and $\vec{q} \parallel [110]$ (green). The CeRhIn$_5$ data have more structure than the CeCoIn$_5$ data since they were taken with better resolution, but the overall shape of the spectra looks very similar and represents the multipole scattering expected for the Ce $N_{4,5}$-edges.$^{23–26,30}$

We discuss first the simulations in order to show what directional effect to expect due to the in-plane anisotropy of the $\Gamma_7$ orbital. The simulations in Fig. 2 have been performed for ground states with $\Gamma_7$ (xy) orientation,

![Figure 2](image_url)

**FIG. 2.** (color online) Non resonant inelastic x-ray scattering (NIXS) spectra of CeCoIn$_5$ (left) and CeRhIn$_5$ (right) at the Ce $N_{4,5}$-edges for the two crystallographic directions $\vec{q} \parallel [100]$ blue and $\vec{q} \parallel [110]$ green at low temperatures with different resolutions; dots data and lines simulations for a $\Gamma_7$ state (xy orientation) (see text).

![Figure 3](image_url)

**FIG. 3.** (color online) Simulated difference spectra $I_{\vec{q}[110]} - I_{\vec{q}[100]}$ (dichroism) for $\alpha^2$ values of 0 (1) and 0.5, and of 0.38 for CeRhIn$_5$ and 0.13 for CeCoIn$_5$; the solid lines account for an energy resolution of 0.7 eV, the dotted line of 1.3 eV.
taking into account the respective $J_z$ admixtures ($\alpha^2$ values) and resolutions. The directional dependence shows clearly below 105 eV where the scattering for $\vec{q} \parallel [110]$ (blue) is stronger than for $\vec{q} \parallel [110]$ (green), i.e. the simulations show blue over green. At 105 eV the two intensities cross and at further increasing energy transfers green is stronger than blue. Especially at about 108 eV the scattering for $\vec{q} \parallel [110]$ (green) is stronger, and at the falling tale of the edge at about 112 eV both intensities cross again.

In Figure 3, we compare the directional dependence $I_\parallel \{ [110] \} - I_\parallel \{ [100] \}$ (dichroism) for several $\alpha^2$ values. For $\alpha^2 = 0$ or 1 the dichroism is zero because in this case the $\Gamma_7$ state is a pure $J_z$ state and rotational invariant; for $\alpha^2 = 0.5$ the dichroism is largest. For CeRhIn$_5$ ($\alpha^2 = 0.38$) the mixing factor $\alpha^2$ is closer to 0.5 than for CeCoIn$_5$ ($\alpha^2 = 0.13$) so that the expected dichroism is larger in the former. For a $\Gamma^+_7$ ground state the intensities are simply opposite so that the present experiment yields an either positive or negative result; a positive (negative) dichroism at a certain energy transfers yields the sign of the wavefunction, i.e. the interpretation of the data is straightforward.

We now start comparing data and simulation of CeRhIn$_5$ in Fig. 2(a). The dichroism of the pre-edge, the crossing of intensities at 105 eV, the stronger scattering for $\vec{q} \parallel [110]$ at 108 eV, and also the dichroism and crossing of intensities at about 112 eV are very well reproduced with the $\Gamma_7$ simulation.

For CeCoIn$_5$ the effect is weaker as expected. According to the simulations in Fig. 3 the dichroism should amount to only about 60% of the dichroism of CeRhIn$_5$ (compare red and green solid lines). In addition, in the CeCoIn$_5$ experiment the resolution was worse so that the dichroism is more washed out (compare dotted green line in Fig. 3) and the magnitude $|\vec{q}|$ of the momentum transfer is slightly smaller than in the set-up at P01 for CeRhIn$_5$. Nevertheless, the crossing of intensities and the stronger scattering for $\vec{q} \parallel [110]$ at 108 eV strongly suggests that also here the $\Gamma_7^-$ is the ground state. Especially here, the fact that we are dealing with an either-or experiment helps the interpretation.

V. DISCUSSION

Figure 4 summarizes what we know about the crystal-field splittings of the $J = 5/2$ multiplet of CeMIn$_5$. The splittings and $\alpha^2$ values are taken from inelastic neutron scattering and x-ray absorption as published in Ref. 20–22. NIXS experiments on CeCoIn$_5$ and CeRhIn$_5$ add the information that the $\Gamma_7^-$ is the ground state for both compounds, i.e. the lobes of the ground state are along the crystallographic (110) direction. These $\Gamma_7^-$ ground state orbitals extend more in $z$-direction than the $\Gamma_7^+$ at about 6-7 meV. The $\Gamma_7^+$, especially for CeCoIn$_5$, extend mostly in the $xy$ plane so that they do not point to the out-of-plane In2 ions as suggested by Haule et al.\(^{19}\)

![FIG. 4. (color online) Crystal-field-field splittings of $J = 5/2$ multiplet of CeCoIn$_5$, CeRhIn$_5$ and for completeness of CeIrIn$_5$ as adapted from Refs. 20–22. For $M = Co$ and Rh the sign of the wavefunction is now determined which has been taken into account when drawing the $f^2$ charge densities of the respective states.]
a maximum transition temperature very close to that of CeCoIn$_5$ and also changes the Fermi surface from small too large as in CeCoIn$_5$. We do not know if the $f$-orbital configuration of CeRhIn$_5$ at these pressures is the same as that of CeCoIn$_5$, but this is an interesting possibility that merits study.

VI. SUMMARY

In $f$-based materials, the shape of the crystal-field wavefunctions ultimately determines the origin of anisotropic hybridization in these materials and their ground state. Here, we show that the ground state of CeMIn$_5$ ($M = $ Co, Rh) is a $\Gamma_7 = |\alpha| / [\pm 5/2 - \sqrt{1 - \alpha^2}] \mp 3/2$ doublet with lobes $\Gamma_7^-$ pointing toward the 110 direction, i.e., the lobes have $xy$ character. Though careful DFT+DMFT calculations shed light on these materials, the crystal-field scheme obtained is different from our experimental one. Our work settles the question on the orientation of $f$-orbitals in the doublet ground state of CeMIn$_5$ and will stimulate theoretical developments that take into account the actual wavefunctions.

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