Unraveling $5f$-$6d$ hybridization in uranium compounds via spin-resolved L-edge spectroscopy

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The multifaceted character of $5f$ electrons in actinide materials, from localized to itinerant and in between, together with their complex interactions with $6d$ and other conduction electron states, has thwarted efforts for fully understanding this class of compounds. While theoretical efforts abound, direct experimental probes of relevant electronic states and their hybridization are limited. Here we exploit the presence of sizable quadrupolar and dipolar contributions in the uranium L$_3$-edge X-ray absorption cross section to provide unique information on the extent of spin-polarized hybridization between $5f$ and $6d$ electronic states by means of X-ray magnetic circular dichroism. As a result, we show how this $5f$-$6d$ hybridization regulates the magnetism of each sublattice in UCu$_2$Si$_2$ and UMn$_2$Si$_2$ compounds, demonstrating the potentiality of this methodology to investigate a plethora of magnetic actinide compounds.
The extent of f-state electronic hybridization with valence/conduction band states is key in defining the physical properties of rare earths and actinide compounds. Materials that defy straightforward description in terms of either localized or itinerant electron behavior are a longstanding challenge to understanding novel electronic matter. The actinide materials are at the heart of this debate because the 5f electrons are intermediate between localized and delocalized. In those two limiting cases, well-defined methodologies exist that account for their magnetic behavior such as Hund’s Rules, crystal-field theory, and quenched angular momentum theory. In uranium generally failed. For example, the extent of 5f behavior in UTe1, UGe2, and the recent interpretation of hysteresis in their magnetic behavior such as Hund’s limit cases, well-defined methodologies exist that account for their electronic behavior. The actinide materials that defy straightforward description in terms of either bonding or non-resonant high-resolution X-ray spectroscopy techniques. It has recently been proposed to study the electronic structure of actinide compounds showing decisive results in some cases. However, a method to directly and selectively probe the electronic 5f, 6d states and their hybridization has not been available yet, which, in turn, is crucial for a comprehensive understanding of the unconventional mechanisms that regulate the physics of 5f electrons in actinides. In addition, with the intrinsic difficulties handling actinide elements due to their toxic and radioactive nature, theoretical work on actinide compounds has been much more extensive than experimental work. The need for a technique capable of directly probing the relevant electronic states, as well as testing theoretical predictions, is abundantly clear.

It is well known that 5f states of actinides can be directly probed by X-ray absorption spectroscopy at Mn5 (3d→5f) or Nd5 (4d→5f) transitions allowing determination of orbital and spin magnetic moment via X-ray magnetic circular dichroism (XMCD) sum rules. But information on the 6d states is absent in these measurements. By using L3,2-edges spectra instead, which involve 2p→6d and 2p→5f transitions in the dipolar and quadrupolar channels, respectively, information on both 5f and 6d states (and their hybridization) can in principle be obtained. However, experimental difficulties such as the inefficiency to produce circular polarization and the significantly low amplitude signals, ~1/50 when compared to the ones at Mn5,4,-edges, have thwarted efforts to perform careful L3,2 XMCD studies on uranium or any other actinide compound. We note that the use of magneto-optical sum rules to separately determine the orbital and spin magnetic moments from XMCD spectra as proposed by P. Carra et al. is a unique tool. However, these sum rules normally cannot be directly applied for the case of L-edges of rare earths and actinides due to the presence of 5f quadrupolar contributions in the XMCD spectra and the influence of the 5f-6d hybridization on the spatial extent of 6d orbitals. While some ways to overcome this have been proposed, it relies on the inclusion of spin-asymmetry parameters and the combination of the experimental data with theoretical calculations, in addition to the correct extraction of the 5f quadrupolar contribution from the spectra.

Here we report on XMCD in the uranium L3-edge (17.166 keV) for UT2Si2 (T = Cu, Mn) compounds. We demonstrate this technique offers a unique opportunity to directly probe the magnetism of actinides compounds and also address the question of electronic hybridization between 5f and 6d states. By analyzing the relative amplitudes of (sizable) quadrupolar and dipolar contributions to the XMCD signal at U-L3 edges we describe the individual content of 6d and 5f spin-polarized states as well as 6d-5f electronic hybridization in prototypical uranium compounds. The bulk-sensitivity of the method at this relatively high energy also allows investigating these electronic effects as a function of applied pressure.

**Results**

5f/6d hybridization in UCu2Si2 and UMn2Si2. We have selected UCu2Si2 and UMn2Si2 compounds to benchmark this method since their family of intermetallics (UT2X2 (1:2:2) with T = transition metal and X = Si or Ge) presents a wide variety of electronic and magnetic properties, often times argued to be due to the relationship between magnetism and the hybridization effects among uranium 5f and 3d/4d/5d states. While much attention has been given to UCu2Si2, only a few studies on UMn2Si2 have been reported so far. The UCu2Si2 compound is reported to show dual itinerant/localized character of the 5f electrons being both a strongly anisotropic ferromagnet, with ordering temperature of 103 K, and exhibiting the Kondo lattice behavior. In this picture the ferromagnetic order comes from the part of the U 5f highly localized electrons giving rise to the high Tc value. On the other hand, UMn2Si2 orders ferromagnetically below 377 K for the Mn sublattice and below about 100 K for the sublattice. The electronic structure of this series is characterized by the relative energy positions of the d states of a given transition metal atom compared with those of the uranium 5f states. While in the Mn-compound the 3d and 5f states are closer in energy and the f-d hybridization is considered to be the largest among the family of 1:2:2 silicates, in the Cu compound the 3d and 5f states are most distant and therefore the hybridization is almost negligible.

Uranium L3 edge (17.166 keV) X-ray absorption near edge structure (XANES) and XMCD measurements on UCu2Si2 and UMn2Si2 compounds are presented in Fig. 1a, b. Two well-defined peaks are clearly present in both compounds. Similarly to rare earths, we relate these spectral features in the XMCD signal to a dipolar (6d) contribution (in the high-energy peak) and overlapping quadrupolar (5f) and dipolar (6d) contributions as a result of 5f/6d hybridization (low energy peak; see Fig. 1b). This interpretation is also supported by first principle calculations of the XMCD spectra which reproduce well the experimental features and differences between the compounds, as shown in Fig. 1c–e. The theoretical spectra in Fig. 1d,e show that while the quadrupolar contribution (green curve) which essentially probes 5f orbitals presents only one peak, two peaks are observed due to the dipolar term (blue curve). While the first dipolar peak at low energy arises from the sizable 5f-6d hybridization, the second peak comes exclusively from 6d unoccupied orbitals. (See Supplementary Note 3 for calculations of the density of states using the LDA + U approach). The large 5f-quadrupolar term at the L3 edge relies on the fact that the X-ray photon wavelength (l=0.722 Å in this case) is comparable to the size of the electron orbit. In other words, the high energy of the actinides L-edges (16–25 keV) enhances the quadrupolar contribution from the transition Hamiltonian operator when compared to lower energy L-edges of rare earths, which presents a crucial role on the appearance of a quadrupolar contribution. In addition, considering that the spin–orbit coupling and presence of a sizeable orbital moment plays a crucial role on the appearance of a quadrupolar contribution, we can envision that all magnetic actinide materials may present sizeable quadrupolar contributions in their L3-edge XMCD spectra. Even materials...
the UCu2Si2. Interestingly, the amplitude ratio between the two XMCD peaks is opposite for the two compounds. As supported by the calculations of the former being much less hybridized than the latter in agreement with the theoretical predictions. The difference observed on XMCD experimental data behavior is discussed in the main text. The individual contributions to the theoretical XMCD spectra are shown in for the ratio between the intensities of quadrupolar and dipolar intensities is very well reproduced in the simulated spectra and the explanation for this.

such as Curium with 5f electronic configuration (without orbital moment) could show some quadrupolar contributions if a charge transfer between the 5f and 6d is promoted in the system. This would be in a similar fashion to what happens to 4f europium materials when subjected to applied pressures, in strong difference to 4f configuration EuO3 which already shows a strong quadrupolar contribution at the L3 at ambient pressure due to its large orbital moment and spin–orbit coupling. It is also noteworthy that while the 5f component in the XMCD spectrum is a contribution mainly from the orbital moment, the other terms (6d and 6d-5f) involve more evenly both orbital and spin moments. The availability of a thorough theoretical description of sum rules analysis for this case that could be easily applied to the experimental data without relying on first principles calculations would be an important asset in order to carefully determine the spin and orbital moments of all the contributions in the spectra. Probing the pure 6d, pure 5f, and hybridized 6d-5f terms through dipolar and quadrupolar contributions to the L3-edge XMCD spectrum makes this a unique tool to directly probe both 5f and 6d orbitals and their hybridization in the same experiment.

Temperature dependence of the 5f and 6d orbital contributions. In addition to this orbital-selective probe of the spin-dependent empty density of states using XMCD at the U-L3 edge, it is also worth commenting on the element selectivity of the technique which allowed us to determine that the net uranium moment for both compounds seem to have comparable magnitudes. This is surprising considering the lack of other examples of a magnetic moment on uranium at ambient temperature, as we observed here for the UMn2Si2. Since a previous neutron scattering study shows ordering of the uranium sublattice only below 100 K, the sizeable XMCD signal in the U atoms at 300 K should be induced by f-d hybridization with the magnetically ordered Mn sublattice. This observation of induced quadrupolar contribution is quite unusual especially when compared to the case of rare earths. In 4f systems an induced 5d moment would not lead to an induced 4f moment since these orbitals would not strongly hybridize considering the localized nature of the 4f orbitals. Here, on the other hand, the 5f-6d orbitals hybridize strongly so an induced 6d moment can also induce a 5f moment. In this scenario we would expect that at low temperatures (T<100 K) the XMCD signal should increase due to the magnetic ordering of the uranium sublattice. Indeed, as shown in Fig. 2a–c.
the uranium XMCD signal is overall enhanced at low temperature. Besides this overall increase, the ratio between the intensities of the low energy and higher energy peak is also modified with the temperature reduction. It is clear from the experimental data that the amplitude of the high-energy peak, which is purely due to the 6d electron (dipolar) contribution, increases much less than the low energy peak, which has both 5f (quadrupolar) and 5f-6d (hybridized) contributions, when we compare the XMCD data for the UMn$_2$Si$_2$ compound at temperatures of 300 K and 22 K. The experimental XMCD data with the respective statistical error bars for $T =$ 300 K and $T =$ 22 K are shown in b, c. The red lines are guides for the eyes to facilitate the visualization of the disproportional changes on the amplitude of each peak (dipolar and quadrupolar) when the temperature is reduced from 300 K to 22 K. Experimental error bars for each energy point are defined as the standard deviation (s.d.) between the multiple averaged spectra.

In summary, we demonstrate that U-L$_3$ XMCD measurements provide a unique measure of both 5f and 6d orbitals, as well as their hybridization, through dipolar and quadrupolar contributions to the XMCD spectra. Since this hybridization is key in determining the physical properties of a vast number of actinide compounds this methodology is bound to improve our understanding of these correlated electron systems. To demonstrate the potentiality of our methodology we have studied the magnetic properties of UCu$_2$Si$_2$ and UMn$_2$Si$_2$ compounds. The

![Fig. 2](image-url) Temperature dependence of the X-ray Magnetic Circular Dichroism (XMCD) contributions. a) XMCD uranium L$_3$ edge for the UMn$_2$Si$_2$ compound at temperatures of 300 K and 22 K. The experimental XMCD data with the respective statistical error bars for $T =$ 300 K and $T =$ 22 K are shown in b, c. The red lines are guides for the eyes to facilitate the visualization of the disproportional changes on the amplitude of each peak (dipolar and quadrupolar) when the temperature is reduced from 300 K to 22 K. Experimental error bars for each energy point are defined as the standard deviation (s.d.) between the multiple averaged spectra.

![Fig. 3](image-url) Isolated probe of the 5f contribution. Uranium M$_4$-edge X-ray Magnetic Circular Dichroism (XMCD) spectra measured for the UMn$_2$Si$_2$ compound at temperatures of 14 K and 300 K. The area of the XMCD spectra is proportional to the magnetic moment solely of the 5f orbitals. By the ratio between the area of the two XMCD spectra we estimate that the uranium 5f magnetic moment increases by a factor 2.2, in good agreement with the results obtained at uranium L$_3$ edge (see main text). Note that due to the direct (3d–5f) transition in the dipolar channel at the M$_{4,5}$ the signal amplitude here is a factor of 50 higher than at the L$_3$ edge data as shown in Fig. 2.

Moreover, these temperature dependent results illustrate the relevance of the 5f-6d hybridization for the magnetic properties of this class of systems. This is also supported by the projected density of states simulations provided in the Supplementary Fig. 2. The 5f-6d hybridization result in the magnetically ordered Mn 3d to induce a moment at both 5f and 6d uranium orbitals through the overlapped 3d-6d bands. This leads to our observation of dipolar and quadrupolar contributions to the XMCD signal of the uranium sublattice at room temperature in the UMn$_2$Si$_2$ compound. This 5f-6d hybridization acts in the opposite direction in the UCu$_2$Si$_2$ compound by mediating an induced magnetic moment at the Cu sublattice. This fact is also supported by the observation of a XMCD signal at K-edge of Cu with comparable amplitude to the signal of the Mn (Supplementary Fig. 3). We note that this observation is not a proof that Cu ions are magnetic, but indicate that a considerable amount of uranium 5f orbital magnetic moment is transferred to the 4p-3d Cu orbitals.
interpretation of XMCD experiments at uranium L₃ edge, together with its temperature dependence, allowed us to demonstrate that the 5f-6d hybridization is the most relevant parameter to determine the magnetic properties of these materials. This hybridization is responsible to induce a magnetic moment at room temperature at the uranium site for the materials. This hybridization is responsible to induce a magnetic parameter to determine the magnetic properties of these theoretical models. It is noteworthy that the methodology analysis, this methodology provide a unique route to validate assignment of the quadrupolar/dipolar contributions can be directly tested through temperature dependent experiments and analysis, this methodology provide a unique route to validate theoretical models. It is noteworthy that the methodology developed here, using penetrating high-energy X-rays, can also be applied to study the evolution of f-d hybridization under applied pressure providing a unique tool for tuning electron correlations. The ability to directly probe experimentally the 5f-6d hybridization should guide effort to understand the magnetic and electronic structures that govern a vast number of actinide compounds with yet bewildering physical properties.

Methods
XMCD experiments. Uranium L₃ XMCD measurements were performed at the D04-DXAS beamline of the Brazilian Synchrotron Light Laboratory (LNLS). To cover the energy range around 17.166 keV (L₃ edge) we used the (311) and (333) reflections of the bent Si crystal in the dispersive geometry25. In this scheme the energy resolution of 1.3 eV at the L₃ edge was set mainly by the area detector used and not by the intrinsic resolution of the high order Bragg reflection. Circularly polarized X-rays were selected by a slit above the orbit plane where the photon flux intensity was 1/3 of the central beam produced by the bending magnet, giving us a degree of circular light of ~54% at the L₃ edge26. XMCD spectra where obtained from the difference between normalized XANES spectra measured in transmission geometry with opposite directions of the 1 T applied magnetic field. Uranium M₄-edge XMCD measurements were performed at the 4-ID-D beamline of the Advanced Photon Source (APS/Argonne)31 using a 50 µm phase-retarding optics to convert the linear polarization of undulator radiation to circular. M₄-edge XMCD spectra were obtained in an equivalent procedure as described above, but switching the helicity of the incoming X-rays in addition to switching the direction of the 0.1 T applied magnetic field which was enough to magnetize the sample. While at the U-L₃ edge data were collected on powders in transmission geometry at T = 300 and 22 K for UMn₂Si₂ and T = 300 K for UCu₂Si₂ at the M₄-edge the data were collected in fluorescence mode. The XMCD signals were corrected for each degree of circular polarization.

XMCD/XANES calculations. XANES and XMCD ab initio simulations were performed using the full multiple scattering approach implemented in the FDMNES code32 including spin–orbit coupling and a 3.0 eV core hole broadening. The atomic potentials obtained from DFT calculations were used as basis and FDMNES code was employed only to include the absorption transition matrix elements to determine the most reliable XANES/XMCD simulations and to avoid difficulties regarding to the Fermi level determination which is critical to XMCD. Since the magnetic moment of both compounds are strongly anisotropic, our simulations were done for the easy magnetization axis of the crystal structure while the experiments were done in a powder sample.

Density functional theory calculations. Density functional theory ab initio calculations were performed using the Wien2k implementation of the full-potential linearized augmented plane-wave method with a double-covering scheme and the rotationally invariant local density approximation LDA + U function with U = 1.25 eV and including spin–orbit coupling33 for the easy axis magnetization directions as reported in the literature34,35. More detailed parameters for these simulations have already been reported in the literature25. Although the density functional theory methods are normally not applicable when mixed/flucluating valence is present, for the systems discussed in this manuscript, however, DFT predictions are indeed validated by experiments which are consistent with a predominant single valence state for uranium in these compounds as strongly supported by the experimental evidence.

Sample growth. Polycrystalline samples of UMn₂Si₂ and UCu₂Si₂ were prepared by arc melting the high purity elements (U = 99.9%, Mn, Cu and Si = 99.9999%) in argon atmosphere followed by an annealing at 800 °C for 5 days in an evacuated quartz ampoule. The X-ray diffraction pattern confirms the single-phase formation of the compounds. The evaluated lattice parameters and unit cell volume obtained using a Rietveld profile fit are shown in the Supplementary Note 1.

Data availability. All relevant data supporting the findings of this study are available from the corresponding author on request. Experimental L₃-edge XMCD data for UMn₂Si₂ at ambient and low temperature are provided in Supplementary Tables 2 and 3.

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**Author contributions**

N.M.S.-N. proposed this research. R.D.d.R. and F.G.G. prepared the samples. R.D.d.R., L.S.I.V., and N.M.S.-N. performed U-L3 edge experiments. R.D.d.R., C.A.E., J.C.L., and D.H. performed the M4 edge experiments. R.D.d.R., L.S.I.V., Y.J., and N.M.S.-N. performed the simulations. R.D.d.R., J.C.L., Y.J., D.H., and N.M.S.-N. interpreted the results. R.D.d.R., D.H., and N.M.S.-N. wrote the manuscript. All authors contributed discussing the results and helping to write the final version of the manuscript.

**Additional information**

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