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Determination of spin and orbital magnetization in the ferromagnetic superconductor UCoGe

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The magnetism in the ferromagnetic superconductor UCoGe has been studied using a combination of magnetic Compton scattering, bulk magnetization, x-ray magnetic circular dichroism, and electronic structure calculations, in order to determine the spin and orbital moments. The experimentally observed total spin moment M_s was found to be $-0.24 \pm 0.05\mu_B$ at 5 T. By comparison with the total moment of $0.16 \pm 0.01\mu_B$, the orbital moment M_l was determined to be $0.40 \pm 0.05\mu_B$. The U and Co spin moments were determined to be antiparallel. We find that the U $5f$ electrons carry a spin moment of $\mathrm{U}_s \approx -0.30\mu_B$ and that there is a Co spin moment of $\mathrm{Co}_s \approx 0.06\mu_B$ induced via hybridization. The ratio $\mathrm{U}_l/\mathrm{U}_s$, of -1.3 ± 0.3, shows the U moment to be itinerant. In order to ensure an accurate description of the properties of $5f$ systems, and to provide a critical test of the theoretical approaches, it is clearly necessary to obtain experimental data for both the spin and orbital moments, rather than just the total magnetic moment. This can be achieved simply by measuring the spin moment with magnetic Compton scattering and comparing this to the total moment from bulk magnetization.

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UCoGe is one of a family of uranium compounds in which superconductivity and ferromagnetism coexist. This unconventional superconductivity was first observed under high pressure in UGe_2 [1], and more recently at ambient pressure in URhGe [2] and UCoGe [3]. Unlike conventional superconductivity, in these ferromagnetic superconductors, spin-triplet pairing is responsible, involving electrons with parallel spins. This means that ferromagnetic order is not antagonistic to the superconducting state, and indeed the pairing mechanism is considered to be mediated via ferromagnetic fluctuations.

In UCoGe, ferromagnetism and superconductivity have been shown to coexist using microscopic probes such as muon spin relaxation [4] and nuclear magnetic resonance [5]. It is considered to be a weak itinerant ferromagnet, with $T_C \approx 2.4$ K and an ordered magnetic moment between $0.07\mu_B$ and $0.18\mu_B$. The superconducting phase occurs below $\approx$0.5 K. When the superconducting transition is probed as a function of pressure, it is clear that superconductivity also occurs in the paramagnetic phase and the transition extrapolates to a ferromagnetic quantum critical point at the critical pressure [6]. Fundamental thermodynamic properties such as magnetization [7,8] and superconductivity [7,9,10] are highly anisotropic, and numerous experiments have shown the existence of the critical ferromagnetic fluctuations [10–12] thought to be necessary for the spin-triplet pairing.

There has been considerable impetus to understand the electronic structure and magnetism in $5f$ materials, including this series of superconducting ferromagnets, owing to the wide variety of ground state properties exhibited. Theoretical models are required to explain the properties of interactions and fluctuations, and a consequence of this is the need of direct knowledge of the spin and orbital moments. A unique situation can be formed where the spin orbit coupling is typically of a similar magnitude to the crystal field. The delicate balance between these can lead to different ground states in apparently similar compounds, depending on the degree of localization of the $5f$ electrons. For U, Hund's rules, which describe a local moment system, can be used to obtain the ratio of the orbital moment (U_l) and the spin moment (U_s). In a free ion the ratio is given by $\mathrm{U}_l/\mathrm{U}_s = -3.29$ for U^{4+} and $\mathrm{U}_l/\mathrm{U}_s = -2.56$ for U^{3+}, and values below these are then used to characterize the itinerancy of the $5f$ electrons [13].

In UGe_2 and UCoGe, defining a single parameter to characterize the degree of itinerancy is insufficient: It has been proposed that the $5f$ electrons simultaneously display both itinerant and localized behavior [14,15]. In the case of UGe_2, there is indeed significant evidence for this so-called electronic duality. The magnetic order is well described by localized electrons, and analysis of the magnetization was found to be consistent with U^{4+} [16]. It should be noted, however, that although polarized neutron diffraction (PND) [17] experiments revealed no evidence of any diffuse magnetization, the orbital to spin moment is reduced with respect to the free-atom value. However, the muon spin relaxation data also exhibit signatures of the presence of itinerant electrons, with a contribution to the moment estimated to be $0.02\mu_B$. The magnetoresistance and specific heat data also have the characteristics expected of itinerant electrons [18,19]. There have been several theoretical studies of the electronic structure and magnetism in UCoGe. These predict significant spin and orbital U $5f$ magnetic moments, of similar magnitudes, resulting in near cancellation

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of the total moment. They all also predict a Co spin moment. In the case of Refs. [20,21], this is parallel to the net U moment, but antiparallel for Ref. [22]. However, when discussing the underlying electronic structure, it is vital to consider that the Co moment is in all cases *antiparallel* to the U spin moment (and parallel to the U orbital moment). The apparent flipping with respect to the U total moment arises simply because the U moment is taken to be parallel to whichever is larger out of its spin and orbital contributions: In Refs. [20,21], $U_l \geqslant U_s$, but in the calculations of Ref. [22], $U_l \leqslant U_s$. All these calculations predict a much larger total magnetic moment than is measured experimentally. To explain this discrepancy, Diviš [22] suggested the Co moments are not collinear, giving rise to a smaller net moment, as observed in UNiAl [23], however, the degree of canting required would have to be $\approx 20^\circ$ and seems unlikely to be the case due to the highly anisotropic magnetization measurements. Alternatively, this discrepancy could arise from the reduction of the bulk moments due to the presence of strong magnetic fluctuations associated with the proximity of the ferromagnetic critical point. Furthermore, in contradiction to the theoretical predictions, analysis of experimental PND data suggested that the U and Co spin moments are in fact parallel [24]. This will be discussed in the light of our results later here.

In this Rapid Communication we report work combining magnetic Compton scattering, magnetization, x-ray magnetic circular dichroism (XMCD) experiments, and *ab initio* electronic structure calculations, and are able to resolve the ground state magnetic configuration of UCoGe. We have determined the site specific spin and orbital contributions to the magnetization. The XMCD measurements confirm a Co spin moment, antiparallel to the U spin moment. It is clear that it is important to be able to resolve both spin and orbital moments, rather than just the total moment when addressing the electronic structure of the actinides, and our approach is ideal for such studies.

UCoGe belongs to the family of ternary compounds U*TX*, with *T* a transition metal and *X* a *p*-electron atom. It crystallizes into the orthorhombic *Pnma* space group. The U atoms arrange themselves in zigzag chains along the *a* axis [Fig. 1(a)] and each U atom has only two U nearest neighbors at a distance of 0.35 nm, characteristic of the critical region between localized and itinerant 5*f*-electron behavior (Hill limit) [25]. The degree of 5*f* localization is down to two things: the direct overlap of corresponding 5*f* wave functions on neighboring atoms governed by the Hill limits and also on the 5*f*-6*d* hybridization with ligand states.

The 1.08 g single crystal used in this experiment was cut from the sample used in Ref. [11], which was grown by the Czochralski technique followed by a predefined annealing procedure [26]. A small piece had a residual resistance ratio (RRR) of 4. The bulk sample used for magnetic Compton and XMCD was characterized using dc magnetization and ac susceptibility. Arrott plots showed a ferromagnetic transition of 2.4 K and the onset of the superconducting transition is seen at 0.6 K from ac susceptibility measurements. The extrapolated value of the upper critical field obtained with the field along the *c* axis are coincident with previous work on a sample with a RRR of 30 [27], demonstrating that even for such a large sample the fundamental properties of UCoGe remain. Indeed, the anisotropic fluctuations seen in this large sample [11] are also observed in much smaller samples [12].

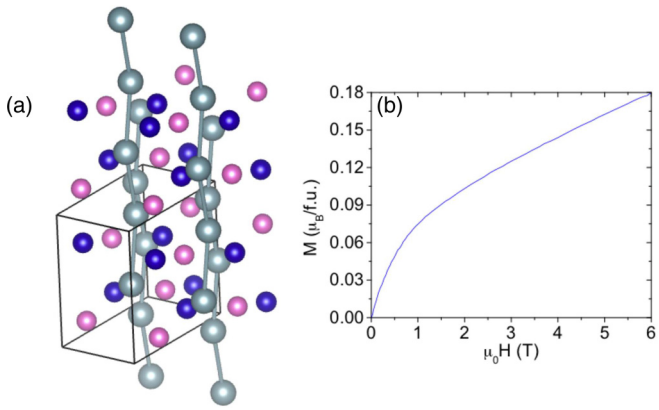

FIG. 1. (Color online) (a) The crystal structure of UCoGe. The gray atoms are U atoms showing the zigzag alignment, dark blue are Co, and purple are Ge. The box outlines the unit cell. (b) Bulk magnetization of UCoGe at 1.8 K along the *c* axis, measured with a superconducting quantum interference device (SQUID) which includes both the spin and orbital moments.

In a Compton scattering experiment, the one-dimensional (1D) projection of the electron momentum density distribution is obtained via measurement of the energy distribution of high energy x rays scattered from the sample being studied. A monochromatic x-ray beam is used, and at a defined scattering angle the scattered photons have an energy spectrum that is directly related to the sample's electron momentum distribution via the Klein-Nishina cross section [28]. The Compton profile is defined as a 1D projection (onto the scattering vector) of the electron momentum distribution $n(\mathbf{p})$ [29], where the z direction is taken parallel to the scattering vector,

$$J(p_z) = \iint n(\mathbf{p}) dp_x dp_y. \tag{1}$$

If the incident beam is circularly polarized, the scattering cross section contains a spin-dependent term [30]. In principle, the spin dependence may be isolated by either flipping the direction of magnetization or the photon helicity parallel and antiparallel with respect to the scattering vector. Either method results in a *magnetic* Compton profile (MCP) $J_{\mathrm{mag}}(p_z)$ that is only sensitive to the net spin moment of the sample, and is defined as the 1D projection of the spin-polarized electron momentum density,

$$J_{\mathrm{mag}}(p_z) = \iint [n^{\uparrow}(\mathbf{p}) - n^{\downarrow}(\mathbf{p})] dp_x dp_y. \tag{2}$$

Here, $n^{\uparrow}(\mathbf{p})$ and $n^{\downarrow}(\mathbf{p})$ are the momentum densities of the majority and minority spin bands. The integrated area of this MCP provides the total spin moment per formula unit of the sample. The orbital moment is not observed [31], and its value can be determined simply by comparison with a bulk magnetization measurement. Since the MCP is the difference between two measured Compton profiles, components arising from spin-paired electrons cancel, as do most sources of systematic error. The high x-ray energies used in the experiments mean that the bulk electronic structure is measured. Crucially, the incoherent nature of Compton scattering means that all local and itinerant contributions to the spin moment are observed [32,33].

The scattering signal obtained is proportional to the Compton profiles defined in Eqs. (1) and (2). The spin moment may then be determined using the flipping ratio R of the integrated magnetic and charge measurements, where

$$R \propto \frac{\int J_{\mathrm{mag}}(p_z)dp_z}{\int J(p_z)dp_z}. \tag{3}$$

The spin moment can be obtained quantitatively from the experimental data simply as it is proportional to the measured flipping ratio [34]. It is determined via comparison with a reference measurement, made in the same experimental setup, of the flipping ratio for a sample with a known spin moment. In our experiment, we used Ni, for which the spin moment ($0.56\mu_B$) is well established.

The MCPs presented here were measured on beam line ID15 at the ESRF. An Oxford Instruments Spectromag cryomagnet allowed measurements at 5 T at 1.5 K. The energy spectrum of the scattered flux was measured using a 13-element Ge detector at a mean scattering angle of 172°. The incident energy of 90 keV and scattering angle of 172° resulted in a resolution of 0.44 a.u. of momentum (where 1 a.u. $= 1.99 \times 10^{-24}$ kg m s^{-1}). The magnetic signal was isolated by flipping the magnetic field applied to the sample. The data were corrected for energy-dependent detector efficiency, sample absorption, and the relativistic scattering cross section. The XMCD experiment was performed on I06 at Diamond Light Source. The vector superconducting magnet provides a sample environment down to 1.4 K in a magnetic field of 6 T. The branchline is fed by an APPLE-II undulator with an energy range between 100 and 1300 eV. All XMCD measurements were performed with a fluorescence detector.

The MCP of UCoGe measured in a field of 5 T at 1.5 K is shown in Fig. 2. The total spin moment M_s was determined to be $-0.24 \pm 0.05\mu_B$. Using a direct comparison of the bulk magnetization which is shown in Fig. 1(b) and gives the measured total magnetic moment as $0.16 \pm 0.01\mu_B$, the orbital moment is then determined to be $0.40 \pm 0.05\mu_B$. The magnetization data were obtained at 1.8 K, but little change is expected at 1.5 K in a 5 T applied field [35] and would not affect our orbital moment value. The contribution to the MCP from electrons associated with specific atoms are generally experimentally distinguishable, allowing the identification of site specific moments. However, the electron momentum distribution of U 5f and Co 3d are essentially (within experimental error) indistinguishable.

To separate the contribution of site specific U and Co moments, electronic structure calculations have been performed in the local spin density approximation (LSDA) using the spin-polarized relativistic KKR (SPR-KKR) package [36]. The obtained electronic structure and magnetic moments are consistent with previous results [21]. The spin-resolved electron momentum density, and hence the MCP, can be calculated directly from the electronic structure, enabling comparison with our experimental profile to give detailed information about the underlying electronic structure and magnetic moments [37–39]. The total spin and orbital moments obtained ($-0.71\mu_B$ and $1.21\mu_B$, respectively) from the calculation are both a factor of 3 larger than the experimental values. The calculated spin moment has been scaled to the experimental value, as the LSDA calculations do not take into account spin fluctuations [40] which are expected to reduce the moment [39], and the resultant fit of the calculation to the MCP is shown in Fig. 2. Scaling the contributing moments by the same proportion suggests that the U 5f electrons carry a spin moment of $U_s \approx -0.30\mu_B$ and that there is a Co spin moment of $Co_s \approx 0.06\mu_B$. From this, the U orbit/spin ratio is deduced to be -1.3 ± 0.3, showing that the U 5f electrons are highly itinerant in UCoGe. It is assumed that the predicted individual U and Co moment contributions scale by the same proportion as the total moments: This seems plausible, given that the total spin and orbital moments both scale by the same factor. Previous work on the $NbFe_2$ system [39], where spin fluctuations are thought to be responsible for the reduced experimental spin moment, were able to demonstrate that the different spin contributions did follow the total moment and that the electronic structure appeared to be unaffected. Even if we drop this assumption, because there is a nonzero Co spin moment which is antiparallel to the U spin moment (from XMCD data, which are discussed below), the U_s value must be greater than $-0.24\mu_B$, which means that the U orbit/spin ratio is certainly less than -1.6 ± 0.3. This low value, when compared to the free-atom value, suggests strong 5f-5f overlap and strong 5f-3d hybridization. It is also shown that the Co spin moment obtained from MCS is a moderately large $0.06\mu_B$ induced by strong hybridization with the U, suggesting that the Co orbitals play a significant role in the delocalization of the U electrons.

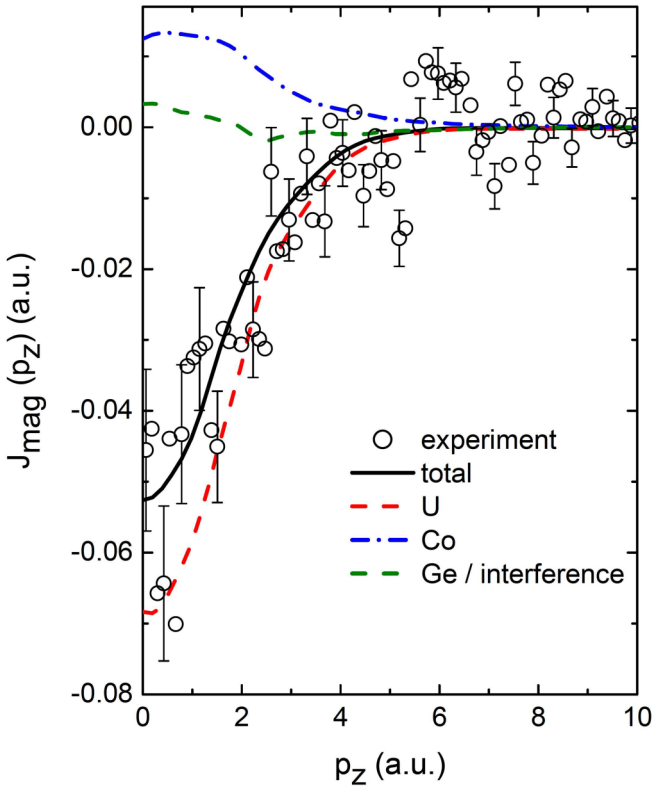

FIG. 2. (Color online) MCP of UCoGe along the c axis taken at 5 T, shown with spin density predicted by Korringa-Kohn-Rostoker (KKR) calculations normalized to the spin moment of $-0.24 \pm 0.05\mu_B$ and decomposed into the site projected profiles.

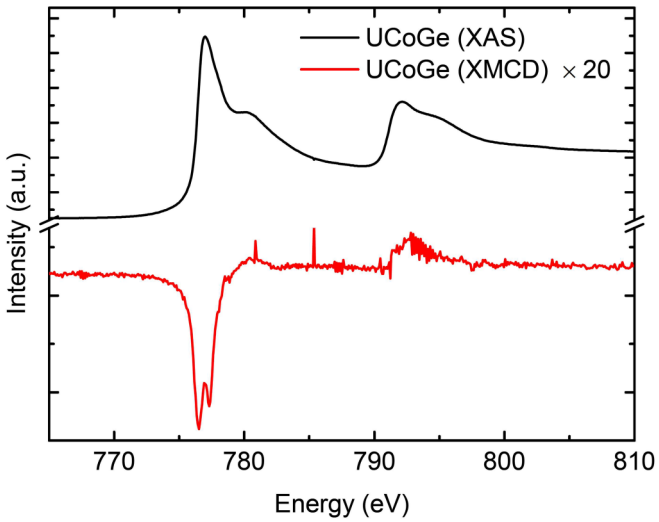

FIG. 3. (Color online) XAS and XMCD signal of UCoGe at 1.5 K and 6 T showing the Co dichroic signal.

To confirm the antiparallel alignment of the U and Co moment, we have used XMCD obtained from absorption spectroscopy (XAS) to study the magnetization at specific elemental edges. Figure 3 shows a typical XAS and XMCD signal below the ferromagnetic transition where dichroism at the Co L_3 and L_2 edge was observed. Using $EuCoO_3$ as a standard Co reference [41], the valence state of UCoGe is Co^{3+} and the Co moment is aligned with the field. This confirms the result of the antiparallel alignment observed with MCP. One complication is that the positions of the Co L_3 and the U N_4 edge overlap strongly. Indeed, the difference in binding energy is only 0.2 eV. However, we do not anticipate a significant dichroic signal at the U N_4 edge since any dichroism at the U N_5 edge was too small to be observed in our experiment. (The dichroism from the N edge may be an order of magnitude smaller than that from M-edge transitions [42].) We have not attempted a quantitative analysis from the XMCD because of the overlap. A very recent study of XMCD at the U M edges complements our experimental observations [43]. For an applied field of 5 T, their orbital moment ($\approx 0.3\mu_B$) is similar to ours, but their spin moment ($\approx -0.14\mu_B$) is smaller. This then leads to a higher U orbit/spin ratio of $\approx -2.3 \pm 0.3$. The origin of this discrepancy is not clear. However, it is possible that it arises from the analysis required to obtain the spin moment from XMCD, which is more difficult in $5f$ systems than for the orbital moment (for example, see Ref. [44]). As discussed above, our U spin moment is greater than $-0.24\mu_B$, and hence our orbit/spin ratio is less than -1.7 ± 0.3.

In order to progress the theoretical description of these U-based superconductors, experimental measurements of spin and orbital magnetic moments are required. For materials where the total moments are small but arise from the cancellation of the spin and orbital moments, a measurement providing their individual contributions is crucial. A recent study using PND was published, with a number of significant findings [24]. First, in contrast with the various theoretical studies, the authors' analysis determined the U $5f$ and Co $3d$ spin moments to be aligned in parallel, rather than antiparallel. Second, the relative contributions to the magnetization density changed as the applied magnetic field was increased, with the Co spin moment being enhanced relative to the U moment at 12 T compared to 3 T. Taking their derived U spin and orbital moments gives orbital/spin ratios of -3.6 ± 1.5 (3 T) and -2.9 ± 1.6 (3 T), which are somewhat larger than our value. However, the total magnetic moments determined from the PND data were significantly less than the total bulk magnetization. This discrepancy was ascribed to the existence of an itinerant moment which could not be attributed to either the U or Co sites.

In summary, we have used magnetic Compton scattering, XMCD, and magnetization measurements to characterize a bulk sample of UCoGe, and to clarify the properties of the site specific moments. It has been shown clearly that the U and Co moment are aligned antiparallel. Moreover, UCoGe is not composed of two large opposing orbital and spin moments, but instead consists of two opposing fairly weak spin and orbital moments. The magnitudes of the individual moments are indicative of a strongly delocalized electron system, with the delocalization mechanism being a strong overlap between U $5f$ and Co $3d$ electrons which consequently result in a non-negligible Co moment. By use of XMCD experiments, the alignment of the moments are determined to be in agreement with *ab initio* calculations, but in contrast with PND measurements. Most of the total moment comes from the orbital contribution, but the majority of the spin moment comes from the U $5f$ electrons with a small non-negligible contribution from the Co $3d$ electrons. Magnetic Compton scattering in combination with standard magnetization is a powerful probe to separate spin and orbital moments, and could be pertinent to the iridate pyrochlore systems. This work highlights the importance of determining not only the total moment, but also the spin and orbital contributions.

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