Polarization of U 5*f* states in uranium multilayers

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X-ray magnetic circular dichroism (XMCD) measurements were performed at the U $M_{4,5}$ edges for U/Fe, U/Co, U/Ni, and U/Gd multilayers. In the case of the U/Fe system, a significant polarization of the U 5*f* electrons was found, but not for U/Co or U/Ni multilayers. We ascribe this to a hybridization of U 5*f* and Fe 3*d* states, which overlap in energy near the Fermi level. In the U/Gd system, the XMCD signals were small and yielded magnetic moments of order $0.02\mu_B$. The spatial extent of the U 5*f* polarization is different from that of the U/Fe system and is oscillatory, extending into the center of the U layers.

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I. INTRODUCTION

Magnetic multilayers have provided scientists and engineers alike with a number of new phenomena to study and exploit.[1,2] The possibility to juxtapose elements with very different electronic properties, to tune these properties and engineer devices in the nanometer regime, allows a direct manipulation of material properties on an atomic scale. Our interest lies in the fundamental electronic interactions that take place at the multilayer interfaces, specifically, concerning the magnitude of the polarization of the nonmagnetic layer and the spatial distribution of the induced magnetization.

The majority of magnetic multilayer systems to date have focused on 3*d*, 4*d*, and 5*d* transition metals. We have extended these investigations into the 5*f* actinides. The elements of the actinide series are characterized by a strong spin-orbit coupling and a localization of the 5*f* electronic band on moving from the light to the heavy elements. Uranium is a light actinide element and has an itinerant 5*f* character, analogous to the transition metals.[3] In order to determine the role of electronic hybridization in multilayers of uranium, we have studied systems with Fe, Co, Ni, and Gd layers. For the case of the transition metal [TM =Fe,Co,Ni] ferromagnets, it is the itinerant 3*d* electrons that are responsible for the magnetism. A consideration of their position and extent in energy with respect to the 5*f* states of uranium should reveal important information about the nature of the 3*d*-5*f* hybridization and thus the U polarization. An investigation of the induced U magnetization in the U/Gd system contrasts the nature of the atomiclike Gd 4*f*-5*f* U interactions with those of the transition metals.

The study of two component magnetic systems, such as multilayers, requires the use of element-sensitive techniques to fully characterize the magnetic properties. X-ray magnetic circular dichroism (XMCD) is well suited for systematic studies of induced ferromagnetism in thin multilayer samples.[4] The large resonant enhancements at the U $M_{4,5}$ edges, which involve electric-dipole transitions from the U $3d_{3/2}$ and U $3d_{5/2}$ levels to the 5*f* states, can be used to probe small magnetic moments.[5–10] Information concerning the magnitude of the total magnetic moment and the separation of this moment into spin and orbital contributions can be extracted using the magneto-optical sum rules.[11,12] A discussion of the sum rules applied to the absorption spectra of actinide ions has been presented by van der Laan and Thole.[13]

The present paper builds on discussions and results presented in a previous XMCD study of U/Fe multilayers.[14] An induced 5*f* magnetic moment of $\sim 0.1\mu_B$ was reported. A theoretical treatment of a model U(001)/Fe(110) supercell structure[15] predicted a moment on the U site, aligned antiparallel to the magnetization of the Fe layers. The induced polarization was predicted to decrease sharply away from the interface region. A recent x-ray resonant magnetic reflectivity study of the U/Fe system has supported these predictions.[16] The aim of this investigation is to prepare U multilayers with four ferromagnetic elements (Fe, Co, Ni, and Gd) and compare the XMCD signals from the U 5*f* states to better understand the role of electronic hybridization in these systems.

The outline of this paper is as follows, After a description of the experimental details, a comparison of the XMCD spectra at the U M_4 edge for the U/TM systems is presented. Then, a comparison is made between the U/Fe and U/Gd results, both their spectral form and the magnitudes of the observed effects. An XMCD study at the U M_4 and M_5 edges

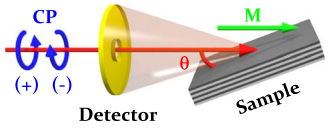

FIG. 1. (Color online) Schematic diagram of the experimental geometry. The helicity of the incident x rays was reversed between right (+) and left (−) circularly polarized states. **M** is the magnetization of the sample and θ is the angle between the sample and the incident beam.

for the U/Gd system is presented, and a profile of the induced U magnetization within the U layers is determined. Finally, we discuss possible explanations for the large variation in the magnitude of the polarization of the $5f$ states from one system to another.

II. EXPERIMENTAL DETAILS

The fabrication and the characterization of the structural and magnetic properties of uranium multilayer systems have been reported previously.[17–20] The growth of U/TM multilayers is characterized by relatively poor interfaces, exhibiting rough interdiffused regions of 10–15 Å thick and in the case of the U/Ni system, up to 30 Å.[21] The structure of uranium in these multilayers is orthorhombic $\alpha-$U, but it is not well crystallized. The transition metals exhibit their expected growth orientations: [110] for bcc Fe, [00.1] for hexagonal Co, and [111] for fcc Ni. The situation in U/Gd multilayers is different. The interfacial region is considerably smaller (<5 Å) and there is little evidence of intermixing.[19] The Gd layers grow in an hcp crystal structure with the (00.2) planes oriented along the growth direction. However, the uranium now forms a hcp-U structure ([00.1] orientation along the growth direction), a phase which has not been observed in uranium bulk metal.[19]

The XMCD measurements were carried out on the ID12 beamline[22] at the European Synchrotron Radiation Facility in Grenoble, France. An electromagnetic-permanent magnet helical undulator was used to provide a circularly polarized x-ray beam. At the energies of the U M_4 (3728 eV) and U M_5 (3552 eV) edges, the degree of polarization is 45% and 35%, respectively. X-ray absorption near-edge spectra (XANES) were measured in a backscattering geometry with an incident angle $\theta=15°$ (see Fig. 1). A magnetic field of 1 T, large enough to saturate the magnetic layers, was applied parallel to the incident beam direction, using a superconducting cryomagnet. The sample temperature was 10 K for all of the measurements. To obtain the XMCD signal, the XANES spectra were collected while reversing the helicity of the circularly polarized incident beam at each energy. Experimental artifacts were eliminated by repeating the measurements with the magnetic field applied antiparallel to the incident x rays.

A Si photodiode detector was used to measure the absorption spectra in fluorescence yield. The XMCD spectra were determined as the difference in the XANES signal for opposite helicities of the incident x-ray beam, normalized to the

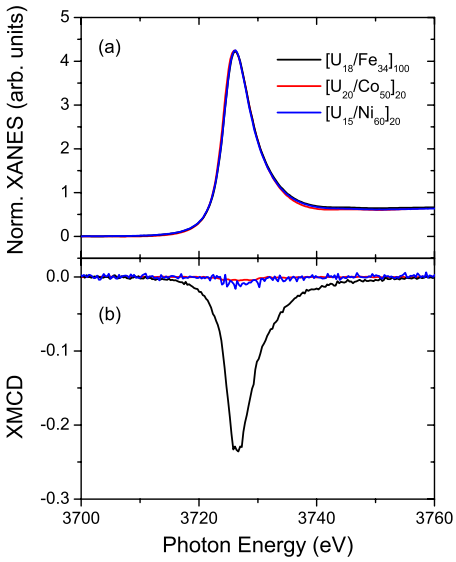

FIG. 2. (Color online) Normalized (a) XANES and (b) XMCD spectra measured at the M_4 edge of U at 10 K in an applied field of 1 T. Selected samples of U/Fe, U/Co, and U/Ni multilayers are presented for comparison.

average of the XANES signals. In order to extract the correct magnitude of the XMCD signal, the XMCD spectra were corrected for the energy-dependent degree of circular polarization. In the fluorescence yield detection mode, self-absorption effects are well known to reduce dramatically the observed signals.[23] However, since the spin-orbit coupling is large at the U $M_{4,5}$ edges, it is possible to correct for these effects, taking into account the multilayer structure,[19] the angle of incidence of the x-ray beam, and the solid angle of the detector.[24,25] Due to the energy separation of the U $M_{4,5}$ edges, data were collected in separate scans. Hence, to display the two edges simultaneously, the M_5 edge jump was normalized from 0 to 1 (several eV before the absorption edge to several eV after) and the M_4 edge from 1 to 2. Finally, the spectra were scaled according to the relative occupation numbers for the spin-orbit-split core levels for the M_5 ($3d_{5/2}$, six electrons) and M_4 ($3d_{3/2}$, four electrons) edges, 1:2/3.

III. RESULTS AND DISCUSSION

This section is divided into three parts. The first compares the XMCD spectra measured at the U M_4 edge for the transition metal multilayers. The second compares the U $M_{4,5}$ XMCD spectra for U/Gd and U/Fe samples. The third part deals with the induced U $5f$ magnetization in U/Gd multilayers and its dependence on the U layer thickness.

A. U/TM multilayers

Figure 2 shows the normalized XANES (upper panel) and the XMCD signal (lower panel) through the U M_4 edge for the three transition-metal multilayers: U/Fe, U/Co, and U/Ni. Since the major XMCD signal is at this M_4 edge, which involves transitions into the partially occupied j

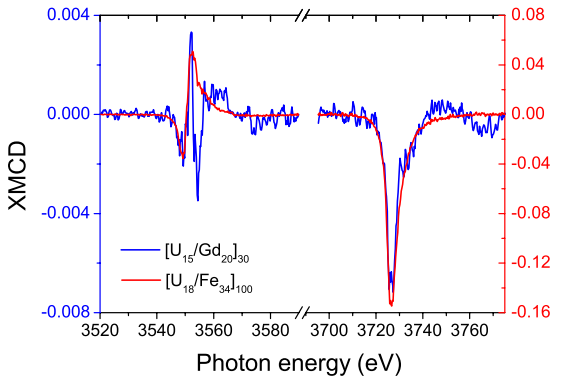

FIG. 3. (Color online) Normalized XMCD spectra measured at the U $M_{4,5}$ edges of U at 10 K in an applied field of 1 T. The detected signals from example U/Fe and U/Gd multilayers are presented to compare the magnitude and shape of the dichroic spectra.

$=5/2$ state, the strength of the signal at the U M_4 edge is proportional to the induced moment of the U $5f$ states.[7] We thus see that, although the XANES signals for all three multilayers are similar, the XMCD signals are substantially different. In particular, the moment on the U atoms in the U/Fe system[14] is significantly larger than in U/Co and U/Ni multilayers, and we shall discuss possible reasons for this in the final section.

B. Comparison of 3*d*-5*f* and 4*f*-5*f* hybridizations

As shown in Fig. 3, the U/Fe spectral shape at the U M_5 edge is rather similar to that found in the heavy-fermion compounds UPt_3 and UBe_{13},[8] but markedly different from that found in UFe_2 (Ref. 6) or in UAs/Co multilayers.[10] It should be remembered that in the latter multilayers, the UAs layers are spontaneously ferromagnetic, whereas in the U/Fe multilayers discussed here, the polarization within the U layers is a consequence of the U $5f/3d$ Fe hybridization. Yaresko *et al.*[9] as well as earlier Shishidou *et al.*[7] have stressed that the exact form of the M_5 spectral feature in the XMCD will depend on the band structure and the exact states present at the Fermi level E_F. The U M_5 dichroism results primarily from transitions to the $j=7/2$ states, although there are also matrix elements involving the $j=5/2$ states.

The U M_4 edge spectral features for the U/Gd and U/Fe samples are similar, whereas the XMCD signals at the M_5 edge are significantly different. Below the energy of the M_5 edge, a dispersive shape is observed for the U/Fe sample (this shape is independent of U or Fe thickness[14]), which is clearly replicated for the U/Gd sample; however, above this threshold, the U/Gd spectrum is more complex. The multipeak structure observed for the U/Gd multilayers has not been reported in experimental data for any other uranium system. Simulations of the dichroic spectra at the U M_4 and M_5 edges have been reported previously[26] for various electronic configurations within an octahedral crystal field. Moreover, for a $5f^2$ configuration and for small crystal fields, the spectral shape at the M_5 edge resembles our experimental data for the U/Gd multilayers presented in Figs. 3 and 4.

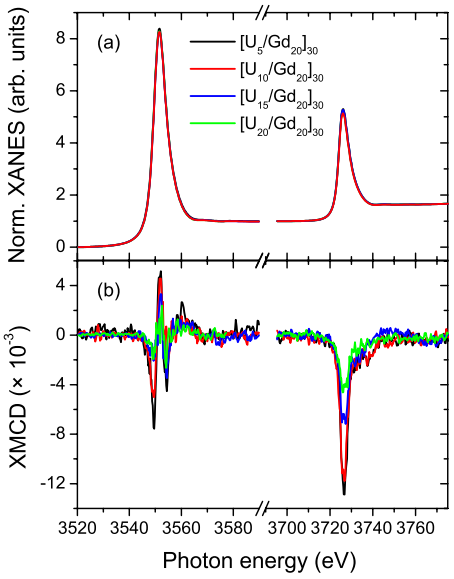

FIG. 4. (Color online) Normalized (a) XANES and (b) XMCD detected at the $M_{4,5}$ edges of U in U/Gd multilayers at 10 K in an applied field of 1 T. Spectra are presented from a series of multilayers with constant Gd layer thickness (20 Å) and increasing U thickness.

Without detailed calculations of the dichroism at the U/Gd interface, it would not be appropriate to designate a $5f^2$ configuration, but the similarity of the spectral features suggests that these M_5 edge spectra could be used to distinguish between theoretical models.

The size of the XMCD signal obtained for the U/Gd sample is more than an order of magnitude smaller than that for U/Fe (see Fig. 3). This indicates that the Fe($3d$)/U($5f$) hybridization mechanism is more strongly polarizing than the Gd($4f$)-U($5f$) or Gd($4f$)-Gd($5d$)-($6d$)U-($5f$)U routes; the former would be due to an indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY)-type mechanism, whereas the latter represents a polarization from the Gd conduction electrons, which carry a magnetic moment of only $\sim 0.5\mu_B$. Both mechanisms would result in a uranium magnetization considerably smaller than that induced from the $2.2\mu_B$ of the Fe $3d$ states, as observed experimentally.

C. U/Gd multilayers

Figure 4 shows the fluorescence (upper panel) and dichroism (lower panel) for a series of U/Gd multilayers, all of which have a constant Gd thickness, but varying U thickness from 5 to 20 Å. The XMCD spectral shapes for all samples are very similar (as was found in the U/Fe system[14]). Although the signal is much smaller in the case of the U/Gd multilayers, the good structural quality,[19] as well as the sensitivity of the XMCD technique, implies that quantitative results can still be obtained.

A feature of the U/Gd series shown in Fig. 4 is that the larger the uranium layer thickness, the smaller the XMCD signal becomes. This is a similar situation as found in the work on U/Fe multilayers[14] and allows a qualitative profile

TABLE I. Branching ratio (B), spin-orbit expectation per hole, the induced U $5f$ orbital, spin and total magnetic moments, and the ratio of the orbital to the spin magnetic moment are presented for a number of selected U/Gd multilayer samples. These values were deduced from XMCD measurements made at 10 K and 1 T, assuming $\langle T_Z \rangle = 0$. The correction Δ to the spin-orbit expectation per hole is negligible for transitions at the U $M_{4,5}$ edges (Ref. 27). The precision on the spin and orbital magnetic moments is strongly dependent on the errors derived in the application and validity of the sum rules ($\sim 15\%$), whereas the ratio of these is not.

Sample	B	$\frac{\langle w^{110} \rangle - \Delta}{n_h}$	Configuration	μ_L (U^{5f}) (μ_B)	μ_S (U^{5f}) (μ_B)	μ (U^{5f}) (μ_B)	$\frac{\mu_L}{\mu_S} \pm 0.06$ (μ_B)
$[U_5/Gd_{20}]_{30}$	0.678	−0.195	$5f^{2.5}$	0.035	−0.058	−0.024	−0.60
$[U_{10}/Gd_{20}]_{30}$	0.676	−0.190	$5f^{2.5}$	0.039	−0.057	−0.018	−0.68
$[U_{15}/Gd_{20}]_{30}$	0.673	−0.182	$5f^{2.5}$	0.023	−0.032	−0.009	−0.73
$[U_{20}/Gd_{20}]_{30}$	0.673	−0.183	$5f^{2.5}$	0.016	−0.025	−0.010	−0.62
α-U[a]	0.686	−0.215					

[a]Determined from electron-energy-loss spectroscopy measurements at the U $N_{4,5}$ edges (Ref. 28).

of the U $5f$ magnetization to be deduced. To do this, we assume that the interface properties of all the multilayers are similar and that the dipole operator $\langle T_Z \rangle = 0$. These assumptions stem from discussions of the U/Fe multilayer system.[14] If $\langle T_Z \rangle$ was of a significant magnitude, then a change in the shapes of the XMCD spectra at both U M_4 and M_5 edges as a function of uranium thickness would be expected. This effect is not observed in Fig. 4. Moreover, since $\langle T_Z \rangle$ was taken to be negligible for the U/Fe system, where the total induced moment was of order $0.1\mu_B$, for the U/Gd system, where the U magnetic moment is some ten times smaller, this assumption is even more robust.

A spin-orbit sum rule[27] for $d \rightarrow f$ transitions has been proposed, which relates the expectation value of the $5f$ spin-orbit operator, $\langle w^{110} \rangle$ (angular part), and the number of $5f$ holes (n_h^{5f}) to the branching ratio B. The branching ratio describes the proportion of electrons excited into each of the spin-orbit-split empty states. This can be determined experimentally as,

$$B = A_{5/2}/(A_{5/2} + A_{3/2}), \tag{1}$$

where $A_{5/2}$ is the integrated area under the M_5 absorption edge peak and $A_{3/2}$ is the M_4 edge peak area. Table I shows the branching ratios and $\langle w^{110} \rangle / n_h^{5f}$ values for a series of U/Gd multilayers. These are essentially constant from sample to sample, 0.675(3) and −0.188(7), respectively, and are very close to values for α-U (Ref. 28) and those reported for the U/Fe multilayer system.[14] Theoretical calculations of $\langle w^{110} \rangle / n_h^{5f}$ versus the $5f$ band filling, for the actinide series of elements[27] have been made, considering LS, jj, and intermediate coupling schemes. Our values for uranium in U/Gd multilayers, assuming a $5f^{2.5}$ occupation, fall close to the LS coupling curve.

Since the number of electrons in the $5f$ band is equivalent to the sum of the electrons in the $j=5/2$ and $j=7/2$ angular-momentum levels and $\langle w^{110} \rangle = n_{7/2} - 4/3 n_{5/2}$, it is possible to calculate the respective occupations. For a $5f^{2.5}$ configuration, taking $\langle w^{110} \rangle / n_h^{5f} = -0.188$, $n_{5/2} = 2.01$ and $n_{7/2} = 0.49$.

A $5f$ electron count of ~ 2.5 is consistent with theoretical and experimental evidences presented in Ref. 3 and was used to calculate the spin, orbit, and total induced U $5f$ magnetic moments, assuming $\langle T_Z \rangle = 0$. These values are obtained by applying the magneto-optical sum rules[11,12] and are given in Table I. The ratios of the orbital to spin magnetic moment are also presented and these are similar to those determined for U/Fe multilayers[14] ($\langle T_Z \rangle = 0$).

Using the values of the U spin, orbital, and total magnetic moment given in Table I for the four U/Gd multilayers, it is possible to approximate the profile of the magnetization within the $[U_{20}/Gd_{20}]_{20}$ multilayer, assuming that the profile is symmetric and that the interfaces are sharp.[19] A further assumption (explained in Fig. 9 of Ref. 14) is that the moment determined from the $[U_5/Gd_{20}]_{20}$ sample is representative of the first 5 Å of U in any U/Gd multilayer, the $[U_5/Gd_{20}]_{20}$ describes the next 5 Å and so on. Figure 5 shows the resulting profiles of the spin (upper panel—blue), orbital (middle panel—green), and total magnetic moment (lower panel—red) in a U/Gd multilayer with U layers of 20 Å thick. Note that the spin moment dominates because of the assumption of $\langle T_Z \rangle = 0$, but an oscillatory function would be found for any value of $\langle T_Z \rangle$. Theory also suggests that for a small induced magnetization within the U layers, the value of the dipole operator $\langle T_Z \rangle$ is likely to be small or zero.[15] The overall U $5f$ moment induced in the U/Gd multilayers is small (no greater than $0.02\mu_B$), but it does have a damped oscillatory form, as shown by the solid line drawn through the data, with a repeat distance of ~ 10 Å. This is in marked contrast to the situation found in U/Fe multilayers, by using the XMCD technique,[14] or the more direct determination performed by x-ray resonant magnetic reflectivity.[16] In the U/Fe case, the polarization of the $5f$ states falls rapidly with the distance from the bcc Fe atoms, and there is no significant evidence for any oscillatory behavior of the U $5f$ polarization.

IV. CONCLUSIONS

We have reported in this paper a series of XMCD measurements on U multilayers in which the U layers are sys-

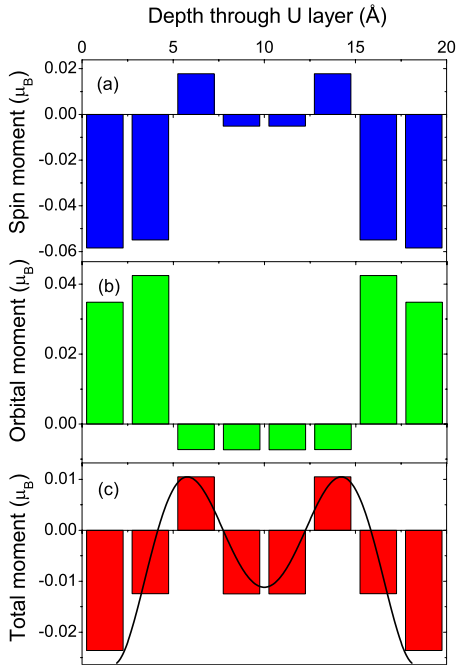

FIG. 5. (Color online) Profiles of the induced U 5*f* (a) spin, (b) orbital, and (c) total magnetic moment are presented for a model U_{20}/Gd_{20} multilayer. A valence state of 2.5 and $\langle T_Z \rangle = 0$ are assumed. The solid black line in panel (c) is a guide for the eyes, showing the damped oscillatory behavior.

tematically interleaved with ferromagnetic elements: Fe, Co, Ni, and Gd. There are two main conclusions.

(1) In the U/TM systems, significant polarization of the U 5*f* electrons occurs only for U/Fe and not for U/Co or U/Ni. A theoretical consideration of these results is clearly greatly complicated by the poor quality of the interfaces in the U/TM series.[16,19] The only theoretical investigation[15] has chosen an ideal interface of U(001)/Fe(110), which is unfortunately unrealistic, and there are no predictions for U/Co or U/Ni. However, we may draw certain ideas from this paper, as well as from earlier work on the U Laves phases UFe_2, UCo_2, and UNi_2 by Johansson and co-workers.[29–31] The crucial aspect is that for significant polarization of the U 5*f* states to occur there must be appreciable overlap between the U 5*f* states and the TM 3*d* states. The U 5*f* states are certainly near the Fermi level (E_F), so the main question is the position and width of the 3*d* states. In the study of the Laves phases,[29–31] the 3*d* states are at E_F for Fe and progressively move to lower E (as well as narrowing in energy) for Co and Ni. In UNi_2, there is no moment associated with the Ni 3*d* states,[30] which is clearly not the case here, as Ni is ferromagnetic in the U/Ni multilayers. However, the general feature that the 3*d* states move to lower energy and become narrower as more 3*d* electrons are present will certainly be respected. For the case of the U/Gd system, it is likely that the U 5*f* and heavily screened Gd 4*f* bands are separated in energy and that any U polarization would be due to a hybridization between the U 5*f* states and the Gd 5*d* conduction electrons, which carry a small ($\sim 0.5\mu_B$) magnetic moment, or due to an indirect RKKY-type, oscillatory interaction. Repeating the calculations performed on the U/Fe system[15] for U/Co, U/Ni, and U/Gd multilayers could provide interesting further insight into why the polarization varies so dramatically from one ferromagnet to the next.

(2) Whereas in the U/Fe multilayers, the overwhelming conclusion is that the U 5*f* polarization occurs only when the U atoms are adjacent to the ferromagnetic bcc Fe,[16] the situation in the U/Gd multilayers is different. In this case, we find a very small, but certainly nonzero, oscillatory polarization of the U 5*f* moments through the U layer. In terms of polarization induced in the U 5*f* by the Gd ferromagnetism, it is appropriate to recall the situation in Gd/Y multilayers,[32] one of the earliest multilayers studied. Here, the oscillations in the Y 4*d* conduction-electron states have a repeat distance of ~20 Å, whereas the periodicity in Fig. 5 in the U 5*f* states in U/Gd multilayers is ~10 Å. These periodicities are related to the band structures of the nonmagnetic species,[33] in our case U and Y in Gd/Y multilayers. An alternative is to compare with polarizations in predominantly *f* electron systems. Ce/Fe multilayers have been studied in a variety of forms.[34,35] In the measurements of these systems the 4*f* polarization has a periodicity of about 10 Å, much the same as that shown in Fig. 5.

We are not aware of any calculations for a U/Gd multilayer, so this polarization of the U 5*f* states, although small, remains a challenge for calculations. Of particular interest is whether such a polarization is dependent on the form of the uranium layers, recalling that in the U/Gd multilayers, unlike the orthorhombic α-U form found in U/TM multilayers, the structure of the U layers is a different hcp U.[19]

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