Profile of the U $5f$ magnetization in U/Fe multilayers

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Recent calculations, concerning the magnetism of uranium in the U/Fe multilayer system have described the spatial dependence of the $5f$ polarization that might be expected. We have used the x-ray resonant magnetic reflectivity technique to obtain the profile of the induced uranium magnetic moment for selected U/Fe multilayer samples. This study extends the use of x-ray magnetic scattering for induced moment systems to the $5f$ actinide metals. The spatial dependence of the U magnetization shows that the predominant fraction of the polarization is present at the interfacial boundaries, decaying rapidly towards the center of the uranium layer, in good agreement with predictions.

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I. INTRODUCTION

Magnetic multilayers exhibit a broad range of interesting phenomena, which have both technological and scientific importance. These properties are primarily driven by the electronic interactions at the multilayer interfaces. Our interest lies in the fundamental nature of interactions between the U $5f$ and Fe $3d$ electrons, particularly concerning the magnetism of uranium.

Theoretical predictions have been made regarding the polarization of uranium in U/Fe multilayer systems, based on both scalar and fully relativistic calculations. The approach adopted an exchange correlation potential treated in the generalized gradient approximation (GGA), which reproduced earlier theoretical predictions for the magnetism of the surface of alpha uranium. A model U(001)/Fe(110) supercell structure was proposed with lattice constants taken as the average between those for uranium and iron. The calculations revealed the importance of U-Fe electronic hybridization and predicted a spin moment on the uranium site of 0.92$\mu_B$, significantly larger and aligned opposite to that of the orbital moment, 0.16$\mu_B$. The total uranium moment was predicted to align antiparallel to the Fe moments and to decrease rapidly within just two atomic layers. We have employed the x-ray resonant magnetic scattering technique in reflection geometry to probe directly the spatial dependence of the U polarization.

Since the first experimental evidence for x-ray magnetic scattering, advances in x-ray sources and development of new materials have lead to a surge of scientific activity in this field. This has been aided by the discovery of large resonant enhancements of the magnetic scattering at the $L_2,3$ edges of the rare earth metals and the $M_{4,5}$ edges of the actinides. In multi-component systems, such as magnetic multilayers, resonant enhancements in the scattering factor can dramatically improve the chemical contrast between elements. Moreover, it is possible to detect strong magnetic dichroism at these resonant absorption edges with the employment of polarized x rays. X-ray resonant magnetic reflectivity (XRMR) combines the benefits of magnetic dichroism with structural information from the charge scattering so that it is possible to determine the spatial profile of the magnetization within the layers. It is a technique ideally suited to the investigation of the magnetism of uranium in U/Fe multilayers. The use of XRMR to investigate buried interfaces in magnetic nanostructures is well-documented,
certainly for the case of soft x rays, but has been less
exploited in the hard x-ray regime. At these energies, it
is most often the L edges of the rare-earth elements
which are of interest, but some progress has been made
in understanding the induced moment in 5d transition
metal systems also.

Preliminary measurements at the U M4 edge have re-
ported an induced magnetic moment on the U site and
later, the separation of spin and orbital components of
this moment, using x-ray magnetic circular dichroism
(XMCD). The total magnetic moments in this case are small (\( \sim 0.1 \mu_B \)). However, the element selectivity
and brightness of x rays at a synchrotron source, coupled
with the large resonances of the U M edges, allow the
moments to be easily detected. The XMCD technique
provides only an average of the U magnetization for the
whole multilayer and is not sensitive to the distribution of
the 5f polarization within the layer. The spatial depen-
dence of the induced U moment, determined by XRMR,
provides a unique insight into the extent of the U 5f, Fe
3d interaction.

II. X-RAY RESONANT MAGNETIC
REFLECTIVITY

X-ray reflectivity measurements have become stan-
dard practise for determining the structure of multi-
layers. Most commonly, a single wavelength measurement
is used and a calculation of the reflected intensity, based
on Parratt’s recursive method is employed to model
the layer thickness and roughness parameters. However,
in order to determine a profile of the magnetization of
a layer, particularly one whose polarization is strongly
thickness-dependent, it is important to determine more
precisely the interfacial structure. This can be achieved
by varying the electronic contrast of the respective ele-
ments; measuring the scattering intensity as a function
of energy through an absorption edge of one of the con-
stituent materials, in this case the M4 edge of uranium.

Once the multilayer structure has been determined it
is possible to measure the Q-dependent magnetic scatter-
ing, see Fig. 1. By employing circularly polarized
synchrotron radiation and applying a magnetic field at
the sample position, the magnetic signal is detected as
the difference in intensity of the elastic scattering when
either the helicity of the incoming x rays or the magnetic
field direction is reversed.

The atomic scattering factor, \( F(E) \), can be written in
terms of a combination of charge, \( F_c(E) \), and magnetic,
\( F_m(E) \), structure factors:

\[
F(E) = (\hat{e}_r \cdot \hat{e}_i) F_c(E) - i(\hat{e}_r \times \hat{e}_i) F_m(E)
\]

where \( \hat{e}_i \) and \( \hat{e}_r \) are the unit polarization vectors of the
incident and scattered x rays, respectively. The charge
structure factor can be written as a summation over all
the atoms in the multilayer,

\[
F_c = \sum (f_0 + f_c(E) + i f''_c(E)) e^{iQ \cdot r}
\]

where \( f_0 \) is the tabulated atomic form factor, and \( f'_c(E) \)
and \( f''_c(E) \) are the real and imaginary parts of the com-
plex resonant anomalous scattering factor, respectively.
The resonant magnetic structure factor can be written in
a similar way, as a summation over the resonating mag-
netic atoms,

\[
F_m = \sum \hat{z}(f'_m(E) + i f''_m(E)) e^{iQ \cdot r}
\]

where \( f'_m(E) \) and \( f''_m(E) \) are the real and imaginary
parts of the resonant magnetic scattering factor, respec-
tively, and \( \hat{z} \) is the unit vector along the quantization axis
parallel to the local magnetic moment. The U M4 ab-
sorption edge represents the excitation of electrons from
the 3d_{3/2} to the 5f_{5/2} states, where electric dipole tran-
sitions provide the strongest contributions to the mag-
netic scattering. For dipole transitions, the resonant
magnetic scattering factors can be represented as,

\[
f'_m(E) + i f''_m(E) = \left( \frac{3}{4kr_e} \right) [F_{11}(E) - F_{1-1}(E)]
\]

where \( F_{LM} \) is determined by atomic properties and is
related to the strength of the resonance, \( r_e \) is the classical
electron radius and \( k \) is the wavevector.

The intensities observed in elastic scattering are re-
lated to the square of the atomic scattering factor, which
on inspection of equation 1 yields cross terms that rep-
resent the resonant magnetic-charge interference scatter-
ing.

\[
Q = k - k_i
\]

FIG. 1: Schematic diagram of the longitudinal geometry used
in the XRMR measurements. \( k_i \) and \( k_f \) are the wavevectors
of the incoming and outgoing circularly polarized x rays, re-
spectively. \( M \) is the magnetization of the sample.
\[ \sum |F^+|^2 - \sum |F^-|^2 = -2(\hat{k} + \hat{k}' \cos 2\theta) \cdot (F'_m F''_m + F'_c F''_c) \]  
\[ = I^+ - I^- \]  

The magnetic-charge interference can be accessed either by flipping the helicity of the incoming photons or by flipping the magnetic field. Conventionally, \( + \) represents right circularly polarized (RCP) x-rays and \( - \) represents left (LCP). In our case, we held the polarization constant and flipped the magnetic field. Equation (5) indicates that the magnetic-charge interference scattering is only sensitive to the component of the magnetization within the scattering plane, hence the magnetic field was applied along the direction defined by the sample plane and the scattering plane, see figure 11.

The scattered intensity was modeled by adapting Parratt’s recursion formula for nonmagnetic specular reflectivity from a multilayer. The complex amplitudes of the electric fields of the transmitted and reflected x-rays of both magnetic field states were included in the calculations. In the frame of reference of the incoming circularly polarized x-ray beam, the x-rays experience different refractive indices for each of the magnetic field directions, \( n^\pm = 1 - \delta^\pm + i\beta^\pm \).

\[ \delta^\pm = \left( \frac{2\pi n_0 r_e}{k^2} \right) (f_0 + f'_c(E) \pm f'_m(E) \cos \theta \cos \phi) \]  
\[ \beta^\pm = \left( \frac{2\pi n_0 r_e}{k^2} \right) (f''_c(E) \pm f''_m(E) \cos \theta \cos \phi) \]  

where \( n_0 \) is the number of atoms per unit volume. The imaginary parts of the charge and magnetic scattering factors are then modeled as a function of energy and a Kramers-Kronig transformation is used to relate the real and imaginary parts of the respective scattering factors.

Commonly, the optical parameters are well known, such as for the transition metal L edges, and only a token number of energies need be sampled to well describe the energy dependence of the scattered intensity. Another approach is to measure the fluorescence and calculate the XMCD, where the XMCD absorption coefficient is related to the imaginary part of the magnetic scattering factor,

\[ \mu_m(E) = - \left( \frac{8\pi n_0 r_e}{k} \right) (\hat{k} \cdot \hat{\mathbf{z}}) f''_m(E) \]  

However, significant self absorption effects can be present in fluorescence measurements. The corrections for these effects then presuppose a knowledge of the structure. In our case the optical constants are not well known and the resonance of the U M4 edge is large. Also, previous measurements on the U/Fe system\textsuperscript{18,19,20} have indicated that the structure of the U/Fe multilayer interfaces cannot be modeled simply. In this investigation we have used a double Lorentzian squared line-shape to model the imaginary part of the scattering factor and an arctan function to model the non-resonant photoelectric absorption. A total of 17 energies were used to precisely track the scattered intensity as a function of energy.

The layer was divided into slices along the z-direction, approximately one atomic plane in thickness (\( \sim 2.5\) Å). The interfacial structure was then modeled by varying the relative densities of the uranium and iron to give a profile of both the U and Fe densities through the multilayer. The calculation of the charge scattering was fitted to the experimental data simultaneously for two different multilayers at all energies. Several parameters were held constant from sample to sample. An important simplification included using identical interfacial regions for each bilayer for each sample.

In order to determine a profile of the induced U magnetization a coefficient was applied to the magnetic scattering factors for each slice of the bilayer that contained some uranium density. The coefficients are proportional to the magnetic moment per uranium atom, hence, by fitting these values to the magnetic-charge interference scattering it is possible to model the spatial dependence of the induced U 5f polarization along the growth direction.

### III. EXPERIMENTAL DETAILS

The samples were prepared by dc magnetron sputtering in a UHV loadlocked growth chamber, operating at a base pressure of \( 5 \times 10^{-10}\) mbar. The multilayers were grown on 50 Å thick niobium buffer layers deposited onto single-crystal sapphire plates. The multilayers were sputtered at a growth rate of \( \sim 1\) Å/s in an argon atmosphere of \( 5 \times 10^{-3}\) mbar. The samples were protected from oxidation by a 50 Å Nb capping layer. The structural and bulk magnetic properties have been reported previously\textsuperscript{20,21}.

The XRMR measurements were carried out at the XMaS beamline (BM28) at the ESRF in Grenoble. This beamline is situated on a bending magnet section of the synchrotron, where the optics and experimental hutch set-up have been designed for the study of x-ray magnetic scattering and the photon flux has been optimized at energies in the vicinity of the U M edges. A complete description of the beamline optics and experimental capabilities has been reported\textsuperscript{22}. The sample views the x-ray beam on orbit, so that the incident flux is linearly polarized. A 90% rate of circular polarization was achieved, by employing a diamond (111), quarter-wave, phase-plate\textsuperscript{23}. In order to preserve as much flux as possible, necessary at the relatively low energies of the uranium M edges, the flight paths of the incident and scattered x rays were
under high vacuum.

The samples were mounted on copper stubs and attached to a magnet assembly, consisting of water-cooled pole pieces, generating an applied magnetic field of 0.1 T. This field was large enough to saturate the iron moments, but small enough to be flipped rapidly. The pole pieces were arranged so that they could provide a field aligned parallel to the scattering plane. The magnet was fixed on a precision sample mount on an 11 circle Huber diffractometer and a Bicron detector was mounted on the 2θ arm. All experiments were performed at room temperature, as the previous studies showed a significant dichroic arm. The measurements were carried out on two U/Fe samples, SN71, [U9/Fe34]30 and SN76, [U27/Fe57]20, whose nominal layer thicknesses were determined by x-ray reflectivity. XRMR measurements were made across the U M4 edge (3728 eV) at room temperature, for circularly polarized x rays in an applied field of 0.1 T. The field was flipped in the following sequence: + − − +, at each point. Data were collected for 17 energies spanning 20 eV below the M4 edge to 20 eV above, providing a mesh of the x-ray reflectivity and magnetic-charge interference scattering (defined as $\Gamma^+ - \Gamma^-$) as a function of Q and energy.

**IV. RESULTS**

Fig. 2 shows the experimental x-ray resonant reflectivity data and calculated intensities for sample SN71. The model reproduces well the fine detail contained between the Bragg peaks and the changes in the reflectivity with energy. A particular feature is the broadening of the Bragg peaks at the resonant energy. Fig. 3 shows the real (a) and imaginary (b) parts of the resonant factor is its Kramers-Kronig transform. The insert of (b) is a comparison of the modeled imaginary scattering factor and the fluorescence, reported previously.

The magnetic-charge interference scattering, $(\Gamma^+ - \Gamma^-)$, was calculated separately for each sample. A real and imaginary magnetic scattering factor was modeled for each sample. Fig. 4 shows the magnetic difference across the first four Bragg peaks in the vicinity of the U M4 edge for sample SN71, compared to the fitted calculation. Higher order Bragg peaks did not yield a measurable magnetic effect. The experimental data and the calculated intensities have been scaled by the theoretical Q-dependence of the reflected intensity.

The imaginary part of the magnetic scattering factor used to model the magnetic-charge interference scattering for sample SN71 is shown in Fig. 5 (b) and the real part (a) is its Kramers-Kronig transform. The insert of
The results presented for each energy have been offset by 20 cts/mon for clarity. The agreement between calculated and experimental data is good for both structural and magnetic data. The scattering factors of Fig. 3 determined from the fitted calculations of the charge scattering, are similar in shape and magnitude to those measured across the U $M_4$ edge in other uranium systems. The good agreement between the fluorescence measurements, corrected for self absorption effects, and the imaginary part of the resonant scattering factor, shown in Fig. 5 (b), may be attributed to the large number of energies sampled. This also supports the comparison of the XMCD signals shown in the insert of Fig. 5 (b).

Fig. 5 (b) shows a comparison between the XMCD signal determined from the imaginary part of the magnetic scattering factor (normalized to the imaginary part of the charge scattering factor) and XMCD data (normalized to the fluorescence) measured on the same sample at room temperature and in an applied field of 1T.

V. DISCUSSION

The agreement between calculated and experimental data is good for both structural and magnetic data. The scattering factors of Fig. 3 determined from the fitted calculations of the charge scattering, are similar in shape and magnitude to those measured across the U $M_4$ edge in other uranium systems. The good agreement between the fluorescence measurements, corrected for self absorption effects, and the imaginary part of the resonant scattering factor, shown in Fig. 5 (b), may be attributed to the large number of energies sampled. This also supports the comparison of the XMCD signals shown in the insert of Fig. 5 (b).

The relative densities of the uranium and iron within a bilayer are shown in panels (a) and (c) of Fig. 6 for samples SN71 and SN76, respectively. The density profiles show extended regions on either side of the central iron and uranium layers, which consist of a mixture of iron and uranium atoms. This result is consistent with the models proposed in earlier studies, using the Mössbauer technique and suggests that the non-magnetic ”dead” layer could be a result of the alloying of the Fe atoms in the interfaces, labeled as amorphous Fe in Fig. 6. These alloy-type regions account for effects of interfacial roughness and interdiffusion that can dramatically alter the profile of the magnetization.

The magnetic-charge scattering was calculated by assigning a magnetization to each slice of the bilayer containing uranium. The calculations were fitted to the experimental data without restrictions on the shape or symmetry of the profile. The resultant profiles of the induced magnetization within the uranium component of the multilayers are shown in panels (b) and (d) of Fig. 6 and are scaled to the relative densities of uranium in each slice. It is clear that the polarization occurs mainly when the uranium atoms are close to the central iron layers, which contain the magnetic bcc component. Furthermore, the magnetization of the uranium falls off very rapidly away from the central iron layers and is in the same direction at each side of the interface.

Calculations of the polarization of uranium in U/Fe multilayers used a model system, which consisted of a sharp interface region and a lattice-matched superstructure. A moment of about $\mu_B$ was predicted, but was found to be considerably smaller ($\sim 0.1\mu_B$) in XMCD studies. This can be attributed to differences between the idealized model calculation and the real multilayers. The spatial dependence of the induced magnetic moment was predicted to fall away very quickly from the
maximum value, so that within two atomic planes ($\sim 5\text{Å}$) it is almost zero. In this respect the calculations and experimental data are in good agreement.

VI. CONCLUSIONS

Energy dependent x-ray resonant reflectivity at the U $M_4$ edge has been used to determine the detailed structure of U/Fe multilayers. Interfacial regions are present, containing a uranium-iron alloy. The fitted charge scattering factors shown in Fig. 3 are similar to those found in other uranium systems$^{24}$ and are in close agreement with self absorption corrected fluorescence data$^{12}$.

The magnetic-charge interference scattering has been used to determine the profile of the uranium magnetization. Agreement between calculation and experiment could only be achieved with the introduction of extended interdiffused regions at the interfacial boundaries. The U polarization is predominantly at the low uranium concentration end of the interface, in close proximity to the bcc iron and decays rapidly as a function of depth towards the centre of the uranium layer. This is in qualitative agreement with theoretical calculations$^2$, emphasizing the importance of the $3d$-$5f$ hybridization for the induced magnetization of uranium in U/Fe multilayers.

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