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# A Mössbauer study of the interface in U/Fe multilayers

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## Abstract

U/Fe multilayers were fabricated with the expectation that the magnetic anisotropy behaviour (perpendicular magnetization of the Fe layers) seen in analogous multilayers of Ce and Fe would occur also in these multilayers. This behaviour has not been observed and it is suspected that the reason is connected with the nature of the interfaces between the U and Fe layers. In this investigation Mössbauer spectra of a standard U/Fe multilayer are compared to those of samples that may reproduce the conditions of the interface. The results show that samples of co-sputtered U and Fe simulate reasonably well the interface spectra and that the non-magnetic Fe seen in this material would inhibit magnetism induced in the U layers via the adjacent Fe layers and hence explain the absence of the expected magnetic anisotropy in U/Fe multilayers.

## 1. Introduction

Magnetic multilayers are fabricated from selected materials with layer thickness adjusted to optimize desired magnetic properties. In attempting to engineer these properties an extremely important influence is the magnetic anisotropy arising from the hybridizing of magnetic orbitals at the layer interfaces. In U/Fe multilayers it is expected that the U–Fe interaction would induce magnetization in the U atoms adjacent to the interface and that the magnetic anisotropy of these U moments could act to affect the magnetization of the Fe layers. Since the large orbital moment arising from the U(5f) shell and its coupling to the lattice have produced extremely large anisotropies in uranium compounds [1], it is hoped that this interface anisotropy could be a strong effect in these multilayers.

In this work the composition of the material of the U/Fe interface is studied.

An illustration of the sensitivity of interface magnetic anisotropy to interface conditions is seen in the partly analogous examples of the Ce/Fe and CeH<sub>2</sub>/Fe multilayer systems where the anisotropy of the Fe layers is always in-plane in Ce/Fe [2] but can be engineered to be normal to the layer plane in the CeH<sub>2</sub>/Fe system [3].

Measurements on both U/Fe and Ce/Fe multilayers by x-ray magnetic circular dichroism (XMCD) have shown induced f shell moments of  $\sim 0.1 \mu_B$  on U(5f) [4, 5] and Ce(4f) [6] atoms adjacent to the interface (and also Ce(5d) moments over a greater range from the interface).

Studies on U/Fe multilayers by Mössbauer spectroscopy (MS) and polarized neutron reflectivity (PNR) have shown no perpendicular magnetization of the Fe layers [7, 8].

It is the focus of this study to use Mössbauer spectroscopy to probe the nature of the Fe atoms in the interface region of U/Fe multilayers as this may hold the key to the interface anisotropy.

Mössbauer spectra (Fe<sup>57</sup>) of U/Fe [7] multilayers show a component of Fe that is not magnetically ordered down to 4.2 K. Comparison of the relative area of this component with Fe layer thickness shows that it arises from a region of  $\sim 10 \text{ \AA}$  at the interface of U and Fe layers [7].

In this study Mössbauer spectra of samples that may reproduce the conditions at the U/Fe interface were taken and compared with a standard [Fe(30 Å)/U(30 Å)]<sub>120</sub> multilayer. Spectra were also taken of the multilayer and these reference alloy samples in applied fields to study the magnetic behaviour of this interface material.

## 2. Experimental techniques and analysis

The multilayer sample and the reference alloy samples were deposited by sputtering on Kapton substrates which are transparent to the Fe<sup>57</sup> 14.4 keV Mössbauer radiation and are therefore suitable for study by transmission Mössbauer spectroscopy (TMS). The sputtering system comprises three DC magnetron guns with individual shutters in a UHV chamber. Prior to deposition the chamber was baked enabling a base pressure of  $\sim 10^{-10}$  mbar to be obtained. The multilayer

was fabricated by depositing a 50 Å Nb buffer layer on the Kapton and then alternately sputtering U and Fe layers with timed shutter sequence to form the  $[U(30 \text{ \AA})/Fe(30 \text{ \AA})]_{120}$  multilayer. A 50 Å Nb capping layer completed the sample growth to protect the sample when it is removed from the UHV environment. Layer thicknesses were measured by x-ray reflectivity (XRR). In separate experiments it was shown that the Mössbauer spectra taken in reflection by conversion electron Mössbauer spectroscopy (CEMS) from such Kapton based multilayers are similar to those fabricated on glass and sapphire substrates.

The sequence of layers in the mimic samples were—Kapton, 50 Å Nb buffer layer, 2000 Å co-sputtered U + Fe, 50 Å Nb cap. Two such samples were prepared, with atomic ratios of U:Fe of 1:1 and 1:2 respectively.

In addition a sample of the alloy  $UFe_2$  was studied. This comprised 98 mg of powdered alloy mixed with 74 mg of inert BN filler to ensure sufficient sample volume for even coverage of the sample holder.

Mössbauer spectra were taken with the  $Co^{57}$  in Rh source ( $\sim 50$  mCi) at room temperature and the samples in a He flow cryostat (zero field spectra) or in a He bath cryostat (applied field spectra). Isomer shift values are quoted relative to a  $25 \mu\text{m}$   $\alpha$ -Fe foil at room temperature.

Applied magnetic fields were produced by a 10 T superconducting magnet comprising a Helmholtz pair of windings. In the geometry used the gamma-ray beam is directed between the Helmholtz coils normal to the direction of the magnetic field.

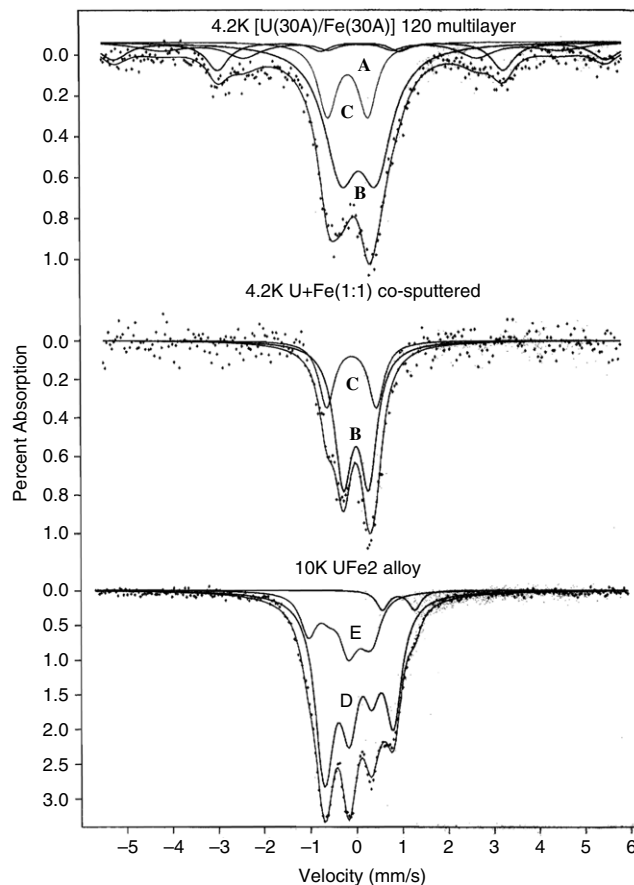
### 3. Results and analysis

#### 3.1. Identification of the interface material

Mössbauer spectra of the  $[U(30 \text{ \AA})/Fe(30 \text{ \AA})]_{120}$  multilayer and the co-sputtered U + Fe 1:1 reference alloy sample taken at 4.2 K are shown in figure 1 with the spectrum of the  $UFe_2$  alloy taken at 10 K. Fitting parameters for these spectra are listed in table 1.

The spectrum of the multilayer can be seen to contain a magnetic sextet with broadened lines (component A) and a non-magnetic part that is fitted to two doublet components (components B and C). The sextet part of the spectrum originates from the iron layers of the multilayer and the non-magnetic part from the U/Fe interfaces. The focus of this study is to account for just the non-magnetic part of this spectrum. The lower two spectra are different attempts to compare materials that might constitute the interface material with the interface signal from the multilayer.

It can be seen directly from comparison of the two lower spectra to the non-magnetic part of the multilayer spectrum that whereas the non-magnetic doublet component of the co-sputtered spectrum is reasonably similar to the non-magnetic part of the multilayer spectrum, the spectrum of  $UFe_2$ , which orders magnetically at 167 K [9], albeit with a small hyperfine field, does not even qualitatively resemble the central part of the multilayer spectrum. This dissimilarity is made more quantitative by comparing the fitting parameters of components



**Figure 1.** Low temperature Mössbauer spectra of, from top to bottom, the  $[U(30 \text{ \AA})/Fe(30 \text{ \AA})]_{120}$  multilayer, the U:Fe (1:1) co-sputtered sample and the powder  $UFe_2$  sample. Comparison of the spectra shows that the co-sputtered spectrum is similar to the non-magnetic component of the multilayer spectrum but the spectrum of  $UFe_2$  is not similar.

B and C of the multilayer spectrum with components D and E of the  $UFe_2$  spectrum. (The spectrum of  $UFe_2$  was fitted with the crystallographic 3:1 Fe site occupation but required the addition of a 4% impurity phase for a reasonable fit.) On this comparison alone the presence of the alloy  $UFe_2$  can be ruled out as anything but a small minority material present in the interface of U/Fe multilayers.

Comparison of the parameters fitting the non-magnetic part of the multilayer spectrum with those fitting the spectrum of the co-sputtered spectrum requires closer scrutiny. In this comparison care must be taken that the contribution of component A that lies under components B and C in the multilayer spectrum is correctly represented. A problem with the spectrum seen in figure 1 is that component A is not well defined. Accordingly in the total fit to this spectrum component A is represented by the two listed sextets that are taken from well defined fits of U/Fe multilayers of similar Fe thickness at 4.2 K. The parameters of component A are not varied and thus the final fit varies parameters of components B and C only. The parameters of the non-magnetic components B and C for the multilayer and for the co-sputtered spectra show sufficient similarity to indicate that the material produced by co-sputtering is similar to that found at the interfaces of U/Fe

**Table 1.** Fitting parameters for all Mössbauer spectra shown in figures 1 and 2. Components A, B, C, D and E correspond to the profiles identified in the figures. In the column of relative intensity the value for component A of the multilayer spectra are labelled not included (NI). Uncertainty values on the parameters of components B and C are  $\sim \pm 0.04 \text{ mm s}^{-1}$  for isomer shift and quadrupole interaction values,  $\sim \pm 0.3 \text{ T}$  for the total magnetic field in the applied field spectra and  $\sim \pm 10\%$  for the relative intensities.

Sample	Temperature (K)	Applied field (T)	Component	Isomer shift ( $\text{mm s}^{-1}$ )	Quadrupole interaction ( $\text{mm s}^{-1}$ )	Magnetic field (T)	Relative intensity (%)
[U(30 Å)/Fe(30 Å)] <sub>120</sub> multilayer	4.2	0	A	0.11	0.00	33.1	NI
				0.11	0.00	26.9	NI
			B	0.06	0.76	0	74
			C	-0.16	0.87	0	26
Fe + U(1:1)	4.2	0	B	0.04	0.66	0	78
			C	-0.22	0.85	0	22
UFe <sub>2</sub>	10	0	D	-0.07	-0.49	4.3	75
			E	-0.27	-0.57	3.1	25
[U(30 Å)/Fe(30 Å)] <sub>120</sub> multilayer	4.2	9.0	A	0.11	0.00	24.1	NI
				0.11	0.00	17.9	NI
			B	-0.05	-0.04	9.3	68
Fe + U(1:1)	4.2	8.0	B	-0.04	-0.02	8.2	71
			C	0.05	-0.36	2.7	29

multilayers. It is seen that all components show considerable line broadening which indicates a range of isomer shift and quadrupole splitting values which are thought to represent a range of site environments arising from an amorphous or poorly crystalline environment.

### 3.2. Magnetic nature of the interfaces

In all spectra down to 4.2 K it is observed that the interface material is not magnetically ordered. This can arise from paramagnetic iron atoms (atoms with a magnetic moment) above the ordering temperature for the material or alternatively from diamagnetic iron atoms (atoms with no magnetic moment). Mössbauer spectra taken in applied fields should be able to distinguish between these cases. In an applied field  $B_A$  the fitted field  $B_T$  in the Mössbauer spectrum is given by

$$B_T = B_A + B_{\text{hf}}$$

where  $B_{\text{hf}}$  is the internal hyperfine field generated by the iron atom at the nucleus. In a diamagnetic atom  $B_{\text{hf}} = 0$  and thus  $B_T = B_A$  while for a paramagnetic atom  $B_T$  is given by the above sum. In the conditions of low temperature (4.2 K) and high applied fields ( $> 8 \text{ T}$ ) any atomic moments can be assumed to be aligned with the applied field and the addition becomes an algebraic sum.

The spectra that test the magnetic nature of the interface iron atoms are shown in figure 2. In this figure the spectra at 4.2 K and in large applied magnetic fields are shown for the multilayer and for the co-sputtered sample. In these spectra the applied magnetic fields are in the plane of the layers and normal to the Mössbauer gamma-ray beam. In the case of the multilayer the applied field was 9.0 T while that applied to the co-sputtered sample was 8.0 T. The fitting parameters for these applied field runs are listed in table 1.

In the fit to the multilayer sample in 9 T applied field component A was defined by the parameters of the zero field

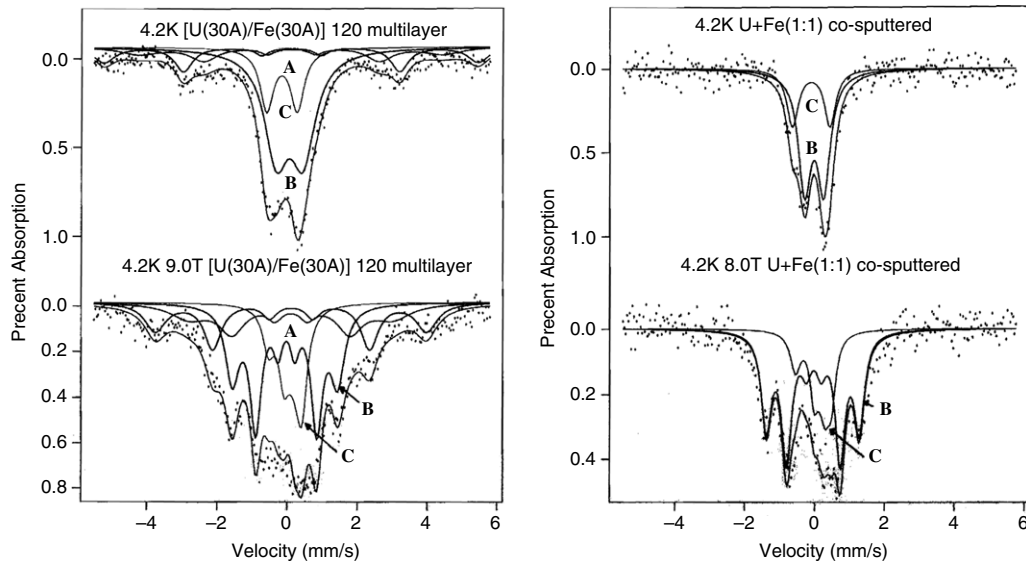
run except that the hyperfine fields were changed to take account of the 9 T applied field.

The comparison involved in this work, however, concerns the other components of the fits that arise from the iron at the interfaces of the multilayer and that in the co-sputtered sample. These fits again show that for both the multilayer and co-sputtered sample two components are required (components B and C).

In the applied field spectra for both samples, the components of greater area (component B) show magnetic splitting closely corresponding to the applied field. In the multilayer sample component B shows a total field of 9.3 T with an applied field of 9.0 T and in the co-sputtered sample component B shows a total field of 8.2 T in an applied field of 8.0 T. From the above discussion this would arise from iron atoms with zero magnetic moment.

The component of minor area (component C) appears to have hyperfine fields of  $B_{\text{hf}} = 9.0 - 2.8 = 6.2 \text{ T}$  in the multilayer sample and  $B_{\text{hf}} = 8.0 - 2.7 = 5.3 \text{ T}$  in the co-sputtered sample. Interpreting such a hyperfine field in terms of the moment on such iron atoms in a metallic sample is not straightforward but from the equivalence of 11.0 T for one Bohr magneton that applies in non-metallic cases, the moments in these paramagnetic iron atoms would be of the order of  $0.5 \mu_B$ . The area ratios (component B: component C) are seen to be 68:32 for diamagnetic: paramagnetic iron atoms in the multilayer sample and 71:29 in the co-sputtered sample.

The applied field spectra indicate that the interface material in U/Fe multilayers and that formed by co-deposition (1:1 atomic ratio) are both well fitted with a major proportion of diamagnetic iron atoms and a minor proportion of iron atoms with small ( $\sim 0.5 \mu_B$ ) magnetic moment. The second co-sputtered sample with atomic ratio U:Fe of 1:2 showed similar spectra to that of the 1:1 sample at 4.2 K in zero field. This suggests that the configuration of the iron atoms in the interface material is not strongly affected by the ratio of abundance U:Fe but is formed by the overall nature of the material. The U:Fe (1:2) sample was not studied in applied fields.



**Figure 2.** Mössbauer spectra, taken at 4.2 K, showing the effect of applied field on (left) the U/Fe multilayer sample (in applied field  $B_A = 9.0$  T) and (right) the co-sputtered sample (in applied field  $B_A = 8.0$  T). Analysis of these spectra yields components of diamagnetic (component B) and paramagnetic (component C) iron for both the co-sputtered sample and for the non-magnetic component of the multilayer.

A possible, but unlikely, interpretation of the observed total field  $B_T = 9.0$  T in the major area component of the multilayer sample would be that in the applied field  $B_A = 9.0$  T, a hyperfine field of  $B_{hf} = 18.0$  T is induced in the majority iron atoms giving a total field of  $B_T = 18.0 - 9.0 = 9.0$  T. However in the co-sputtered sample the induced hyperfine field would need to be  $B_{hf} = 16.0$  T and in a spectrum in applied field  $B_A = 6.0$  T (not shown) the  $B_T = 6.0$  T would require in this case an induced hyperfine field  $B_{hf} = 12.0$  T. At such large fields at a temperature as low as 4.2 K any induced hyperfine field would be expected to be saturated rather than proportional to the applied field—thus we are confident that in the major area component (component B—corresponding to the major fraction of atoms) the equality of applied and total fields reflects diamagnetic iron atoms.

In table 1 it is seen that the quadrupole interactions for components B and C in the applied field spectra are considerably reduced from the values seen in the zero field spectra. This effect can be understood by appreciating that in conditions in which the magnetic hyperfine interaction is appreciably larger than the quadrupole interaction, this latter quadrupole interaction can be treated as a perturbation whose effect is seen as  $\frac{1}{2} eQV_{zz} \cdot \frac{1}{2}(3 \cos^2 \theta - 1)$  where  $\theta$  is the angle between the magnetic field direction and the principal axis of the electric field gradient  $V_{zz}$ . A sum ( $\sin \theta$  weighted) over a random distribution of  $V_{zz}$  directions results in an overall value of zero for the quadrupole interaction. While the values of table 1 do not show zero values for the quadrupole interaction in the applied field spectra the reduced values support a situation of a range of  $V_{zz}$  directions to the applied magnetic field consistent with an amorphous or poorly crystalline environment.

#### 4. Discussion

The inference from the data and analysis presented above is that the interface material is very similar if not identical to that made by co-sputtering of U and Fe atoms. Magnetically this material, while not completely diamagnetic, contains only a minority of Fe atoms with moments much smaller ( $\sim 0.5 \mu_B$ ) than those in bulk iron ( $2.2 \mu_B$ ).

Diffraction measurements on U/Fe multilayers with layer thicknesses less than about 20 Å did not show any crystalline nature [7].

The metallic solubility of U and Fe is negligible thus no crystalline phase containing a mixture of U and Fe atoms can exist and the possibility of the interface containing the metallic compound  $UFe_2$  has been excluded from the results above.

It thus appears as if the interfaces in U/Fe multilayers are composed mainly of an amorphous mix of U and Fe atoms—the majority of atoms not carrying a magnetic moment and the minority with a small ( $\sim 0.5 \mu_B$ ) moment.

The nature of this interface in U/Fe also accounts for the absence of the magnetic anisotropy that gives rise to perpendicular anisotropy. It thus appears that the inability of the U + Fe to grow sharp, crystalline interfaces compromises the hoped for magnetic anisotropy in this seemingly promising system.

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