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Magnetic sublattice interactions in UFe_4Al_8

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Previous neutron experiments on polycrystalline samples of UFe_4Al_8 have led to a series of conflicting proposals, including a spin-glass state, for the magnetic structure below the ordering temperature of $\sim$150 K. Our experiments on a stoichiometric single crystal show that the principal interaction is a commensurate antiferromagnetic ordering of the Fe sublattice, with a moment at 4.2 K of $1.08(2)\mu_B$ per Fe atom *in the basal plane* of the tetragonal structure. However, symmetry arguments suggest that the Fe sublattice has a weak ferromagnetic component also in the basal plane. Experiments in a magnetic field with polarized neutrons establish that the ferromagnetic U moment is $0.47(2)\mu_B$ per U atom. In a magnetic field applied in the [010] direction (basal plane) the Fe sublattice antiferromagnetism is aligned perpendicular to the field in the basal plane, i.e., in the direction [100]. Combining the neutron and magnetization results shows that the weak Fe ferromagnetic component in zero field is $\sim 0.3\mu_B$ so that the canting angle of the Fe moments is 16°. Relatively small fields are sufficient to cause a further canting of the Fe moments towards the field direction; for example at 4.6 T this canting is 25°. Polarized-neutron experiments in the paramagnetic state show that the Fe susceptibility is almost isotropic; however, the response of the U $5f$ electrons is much smaller along the **c** axis, so that it is the hybridization between the Fe $3d$ and U $5f$ electrons that gives rise to the measured anisotropy in this material. The weak ferromagnetism of the Fe sublattice, which may be a consequence of the interaction between the U $5f$ and Fe $3d$ electrons assures that the two sublattices develop their moments in the same manner. This is different from the situation in the RFe_4Al_8 (R=rare earth) compounds, in which the rare earth develops its full moment only at a lower temperature. [S0163-1829(97)01621-4]

I. INTRODUCTION

Compounds with the ordered $ThMn_{12}$ structure originally attracted attention because they were seen as possibly high-T_C materials for applications.[1] Examples are $SmFe_{10}Si_2$ in which the Curie temperature is 606 K and the uniaxial anisotropy is substantial. Of course, in these materials it is well understood that the high T_C arises from the Fe-Fe interactions, and the anisotropy from the rare-earth sublattice. Recently, more effort has been focused on compounds of the form $Nd_2Fe_{14}B$ and these are now used in applications. In the tetragonal body-centered $ThMn_{12}$ structure, space group $I4/mmm$ (No. 139) four different sets of equivalent positions are occupied. The thorium (actinide or rare earth) (M) atom occupies the $2a$ sites (000) and the transition-metal (Fe) atoms start by occupying a set of $8f$ sites. If the ratio of Fe to M is greater than 4 to 1 the remaining Fe atoms are distributed over a set of $8i$ and $8j$ positions in a manner that is determined by the heat of mixing between the M and p-electron atom (Al or Si). It is noteworthy that no compounds of composition MFe_{12}, in which all three eightfold sites are fully occupied by Fe, have been found. A recent

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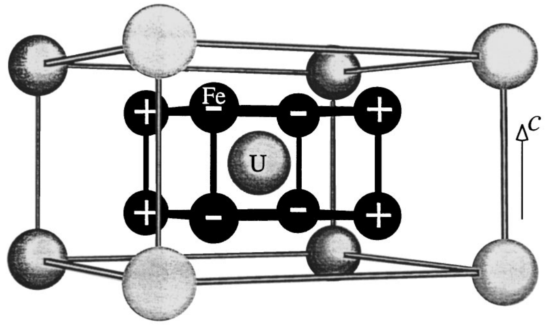

FIG. 1. Crystallographic unit cell of body-centered-tetragonal MFe_4Al_8. The space group is $I4/mmm$. The M atoms, in this case uranium, are situated at the origin and body-centered position. The Fe atoms are situated at the $8f$ positions (1/4 1/4 1/4) and symmetry related positions. For clarity the Al atoms are not shown. The G-type antiferromagnetism associated with the moments of the Fe atoms is shown as + and − signs. The experiments shown that the easy axis of magnetization is in the **ab** plane.

theoretical study of these high-Fe content compounds has proposed noncollinear structures and spin-glass behavior.[2]

A special situation exists for MFe_4Al_8. Here, at least ideally, the Fe atoms only occupy the $8f$ sublattice, and the Al atoms occupy the $8i$ and $8j$ positions. Moreover, the total occupation of the $8f$ sites by Fe (see Fig. 1) means that each M atom is surrounded by a parallelopiped of eight Fe atoms. In the interesting situation that a simple antiferromagnetic structure (as illustrated, for example, by the $+-$ signs in Fig. 1) is found on the Fe sublattice, the molecular field from the Fe sublattice at the M site is zero.[3] This aspect has been recognized for some time, and the separate sublattices of M and Fe allows a study of their interaction. Of course, such a low Fe content means that the ordering temperature drops substantially (to between 100 and 200 K for various compounds[3]) and, in addition, the compounds do not exhibit large ferromagnetic moment in small magnetic fields.

Although we have been studying a number of the RFe_4Al_8 compounds (with $R=$Lu, Dy, Ho, and Y) we shall confine our discussion in this paper to UFe_4Al_8, which has a special place. In other rare-earth materials there is clear evidence, principally from Mössbauer and neutron experiments, that the moments develop on the two sublattices at *different* temperatures.[4,5] In principle, since there is only one magnetic propagation vector for both sublattices, they must order at the same temperature; however, the growth of the ordered moments can be quite different as a function of temperature. The first report of neutron experiments[6] on UFe_4Al_8 showed only *one* transition temperature at ~ 150 K consistent with the Fe Mössbauer findings, and proposed a magnetic ordering on the Fe sublattice only with the spins of $1.6\mu_B$ at 4.2 K parallel to the **c** axis. Schäfer *et al.*[7] then proposed the same structure with $\sim 1\mu_B$ on the Fe sites, but with a ferromagnetic contribution of $0.8(1)\mu_B$ at the U site. In a subsequent paper[8] by the same group as Ref. 7, they reported neutron-diffraction measurements in a magnetic field, and these, together with ac and dc magnetic susceptibility results, led them to propose a highly unusual *spin-glass* state. They claimed to establish that moments existed on both the actinide and Fe sublattices—perhaps the cleanest experiment being the ^{237}Np Mössbauer signal appearing at the same temperature as the Fe hyperfine field in the isostructural $NpFe_4Al_8$ compound. Thus, it appeared that moments did exist on both actinide and Fe sublattices at the ordering temperature, but the low-temperature behavior was unusual. It should be remarked that the same authors had proposed a ''spin-glass'' state in their experiments on the isostructural compound $HoFe_4Al_8$.[9] The next experiments reported[10] on UFe_4Al_8 were those of magnetization in a high field, in which evidence for a strong anisotropy was found, and the authors proposed that the easy axes was in the basal tetragonal plane, in contradiction to the direction proposed from the neutron experiments. All of the above experiments were performed on polycrystalline samples, in which domain effects are known to play an important role, especially if anisotropy is present. Indeed in a related study, Felner, Nowik, and Seh[11] observed unusual behavior in the magnetic properties of the MFe_5Al_7 compounds (with the same structure) and concluded that much of it could be explained by domain effects without resort to any exotic spin-glass state.

Our interest in this material was such that we undertook the production of single crystals by the Czochralski technique. Magnetization measurements on these single crystals emphasized the unusual nature of the interactions between the sublattices. The easy axis was found to be $\langle 100\rangle$, with little signal induced when the field is applied along the **c** axis.[12] At 4.2 K an important remanence of $1.6\mu_B$/mole was found, and measurements of the magnetoresistance showed an unusual reversibility.[13] We report in this paper a series of neutron experiments to illuminate the behavior of the two sublattices in this interesting compound.

II. UNPOLARIZED NEUTRONS

A. Stoichiometry

In view of the uncertainties as to site occupation we undertook a four-circle neutron experiment on the crystal, which was approximately spherical with a volume of 11 mm^3, at Risǿ National Laboratory. A total of 478 reflections were measured at $\lambda = 1.027$ Å, which gave a set of 155 independent reflections after averaging equivalents. The results of the crystallographic least-squares refinement are shown in Table I.

The values for x for the $8i$ and $8j$ sites are in excellent agreement with those refined (from polycrystalline data) in Ref. 7. This refinement gives confidence that the sample is stoichiometric, with, in particular, the Fe and Al sublattices fully ordered. In addition, we know that the extinction from the crystal is appreciable, but is well characterized by this four-circle experiment. This is important in processing the polarized-beam data below. The minimum extinction coefficient was $y_{\mathrm{min}}=I_{\mathrm{obs}}/I_{\mathrm{calc}}=0.42$ and the average correction was $y_{\mathrm{av}}=0.88$.

B. Temperature dependence

To make a preliminary investigation of the magnetic ordering the crystal was examined at the four-circle diffractometer at Risǿ National Laboratory, Denmark, equipped with a closed-cycle helium cryostat ($T_{\mathrm{min}}\sim 20$ K). These measurements were later extended at the Siloë reactor, CEN-Grenoble, down to 4.2 K. Reciprocal-lattice scans in the low-

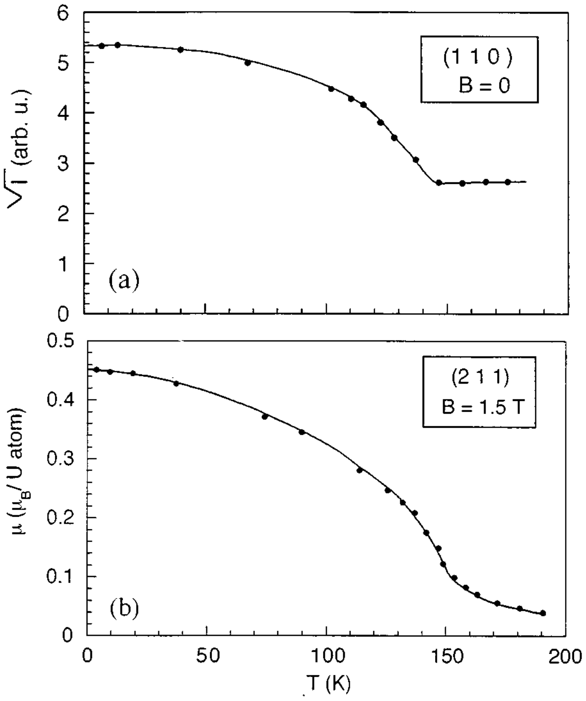

FIG. 2. (a) Evolution of the square-root of the intensity of the (110) reflection as function of temperature. The *increase* at low temperature above the nuclear intensity is proportional to the ordered magnetic moment. This reflection has a dominant contribution from the Fe sublattice. (b) Temperature dependence of the magnetization of the U sublattice in an applied magnetic field of 1.5 T//[010]. The moment of the U atoms was determined from the flipping ratio of the (211) Bragg reflection, which has no contribution from the Fe sublattice.

Q region were performed along selected high-symmetry directions of the first Brillouin zone around several reciprocal lattice points. No new antiferromagnetic (AF) peaks were observed, but rather an increase of intensity in certain nuclear peaks [Fig. 2(a)]. One of the difficulties with the magnetic structure refinements of previous studies[6–8] has been the small number of peaks that were observed to change intensity below the ordering temperature of 150 K. We have had the same difficulty with a single crystal. Only the reflections (110), (310), and (330) have small enough nuclear structure factors that the small additional magnetic intensity below the ordering temperature may be measured (Table II) without the use of special techniques involving polarization analysis of the scattered neutrons. These additional intensities are consistent with the model developed in this paper from using polarized neutrons, but they are not, by themselves, sufficient to determine the individual moments in zero field.

These experiments show that the sublattice of Fe spins does indeed have the so-called G-type antiferromagnetic (AF) order (see Fig. 1) and that $T_N = 145(3)$ K. However, the direction of the moment appears to be most probably *in the basal plane*, which is consistent with the magnetization measurements.[10,12]

C. Magnetic structure and wave vector

The U atoms, being at the origin of the unit cell, contribute to all reflections.

The four Fe atoms in a primitive unit cell may be chosen as

$$1,\left(\frac{1}{4}\,\frac{1}{4}\,\frac{1}{4}\right);\quad 2,\left(\frac{3}{4}\,\frac{3}{4}\,\frac{1}{4}\right);\quad 3,\left(\frac{1}{4}\,\frac{3}{4}\,\frac{1}{4}\right);\quad 4,\left(\frac{3}{4}\,\frac{1}{4}\,\frac{1}{4}\right).$$

The other four sites are derived from the body-centering translation. If the magnetic moment vectors on these four sites are μ_1, μ_2, μ_3, and μ_4 their magnetic structure factors are

$$(\mu_1+\mu_2+\mu_3+\mu_4)$$

reflections with H, K, and L all even,

$$(\mu_1+\mu_2-\mu_3-\mu_4)$$

for reflections with H and K odd, L even,

$$(\mu_1-\mu_2+\mu_3-\mu_4)$$

for reflections with H and L odd, K even,

$$(\mu_1-\mu_2-\mu_3+\mu_4)$$

for reflections with K and L odd, H even.

The AF intensities are found on reflections with H and K odd, L even indicating that the antiferromagnetic vector is given by the sum $(\mu_1+\mu_2-\mu_3-\mu_4)$. This is the so-called G-type AF structure[7] shown in Fig. 1. The ferromagnetic (F) component is of course the sum of all moments. It is important to remember that even for those reflections sensitive to the AF contribution from the Fe sublattice there will be a ferromagnetic contribution from the uranium moments.

TABLE I. Results of the crystallographic least-squares refinement of the four-circle neutron data of UFe_4Al_8. The equivalent isotropic Debye-Waller coefficient B_{eq} is calculated as $B_{eq} = (8\pi^2/3)\Sigma_i\Sigma_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$, where $\mathbf{U}_{ij}$ is the anisotropic displacement tensor. The room-temperature lattice parameters are $a = 8.740(1)$ Å; $c = 5.036(1)$ Å.

	x	y	z	B_{eq} (Å^2)	Occupancy
U (2a)	0	0	0	0.26(5)	1.0(fixed)
Fe (8f)	1/4	1/4	1/4	0.37(3)	1.027(10)
Al (8i)	0.3440(2)	0	0	0.39(6)	1.008(14)
Al (8j)	0.2805(2)	1/2	0	0.28(6)	1.016(10)
$R = 2.18\%$	$\chi^2 = 3.7$				

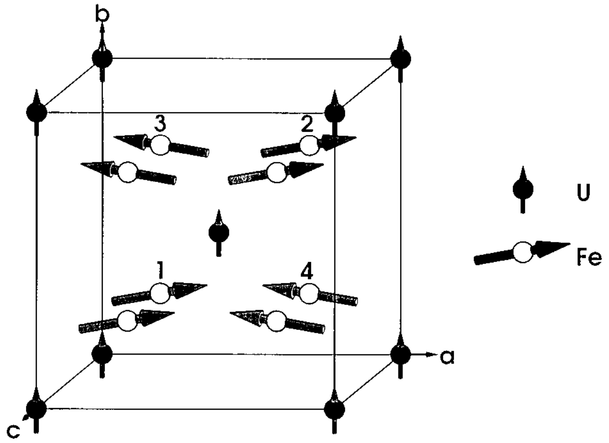

FIG. 3. Magnetic structure of UFe_4Al_8 in an applied field of 4.6 T as derived from the analysis of the polarized-neutron data. The solid points are the uranium atoms, with the open points the Fe atoms. The sizes of the arrows are an indication of the sizes of the moments. Note that this is a different perspective compared to Fig. 1. The angle ϕ discussed in the text is the angle between the spin on atom 1 and the **a** axis.

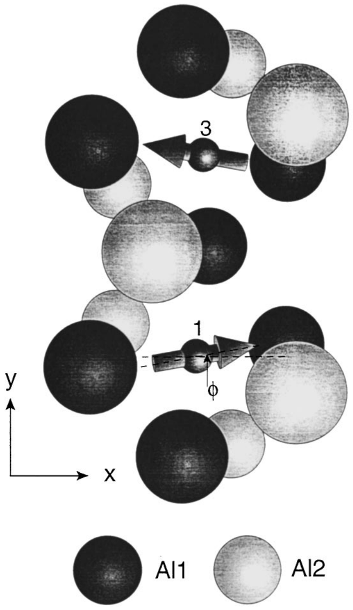

FIG. 4. Schematic drawing to show the arrangement of aluminum atoms around the Fe atoms. Note that the rotation of the two-fold axis discussed in the text implies that Fe 1 and 3 are equivalent only when the spins rotate from ϕ in Fe 1 to (180-ϕ) at Fe 3.

The magnetic structure deduced from our measurements is shown in Fig. 3. The symmetry elements that relate the Fe atoms in the unit cell play an essential role in establishing the magnetic structure. The symmetry element relating atoms 1 at (1/4 1/4 1/4) and 2 at (3/4 3/4 1/4) (and equivalently 3 and 4) is a diad axis parallel to **c** passing through the U atom at the body-centered position. Since the action of the diad axis is to rotate the components of the moment perpendicular to it by 180°, and we find that μ_1 and μ_2 are parallel, this symmetry element must be combined with the time inversion operator when defining the magnetic space group. Similarly, atoms 1 and 3 (and equivalently 2 and 4) are related by a screw diad parallel to **b** passing through the centers of both atoms. If this diad is combined with time inversion it ensures that the **b** (parallel to the diad) components of μ_1 and μ_3 are reversed, whereas the **a** (perpendicular) components are parallel. If this is the case the ferromagnetic component is parallel to **a** and the AF component parallel to **b**. If the diad is not combined with time inversion then the reverse will be true. It is in fact the latter situation that is found in the experiment and shown in Fig. 3. Similar arguments can be used to discuss the screw diad parallel to **a** which relates atoms 1 and 4 (and equivalently 2 and 3).

The magnetic wave vector is $\mathbf{q}=[000]$. The contribution from the U atoms is ferromagnetic. However, the above symmetry considerations show that the Fe sublattice may have *both* an antiferro- as well as a ferromagnetic contribution. It is this aspect that has not been recognized in the past when these compounds have been considered. For example the remanance observed with a single crystal is 1.6 μ_B/mole, and this was ascribed to the U moment. Our unpolarized measurements show that a major part of the Fe magnetism is antiferromagnetically aligned, but a small Fe ferromagnetic component is *not* excluded by symmetry.

The symmetry does not predetermine the direction of the moments in the **ab** plane. The angle ϕ that the Fe moment

TABLE II. Nuclear and magnetic intensities of the low-Q reflections measured with unpolarized neutrons. The magnetic intensity was estimated by subtracting the room-temperature intensities from the intensities measured at 4.2 K. The model used to calculate the magnetic intensities is that derived from the analysis of the polarized-neutron data. All intensities are given in barns (10^{-24} cm^2). y is the extinction correction for the nuclear intensities at $\lambda=1.53$ Å, such that $I_N=y\times|F_N|^2$, where F_N is the nuclear structure factor calculated with the parameters given in Table I. The coherent nuclear-scattering amplitudes used for the calculation are $b_U=0.8417$, $b_{Fe}=0.954$ and $b_{Al}=0.3499$ (all in units of 10^{-12} cm).

HKL	I_{obs} (300 K)	I_{obs} (4.2 K)	y (1.53 Å)	I_N (calc)	I_{obs} I(4.2 K)-I(300 K)	I_M (calc)
(110)	0.72(3)	2.97(3)	0.97	0.44	2.25(4)	2.66
(310)	2.94(3)	4.32(5)	0.90	2.79	1.38(6)	1.13
(200)	12.3(6)	12.5(6)	0.49	13.0	0.02(8)	0.05
(220)	15.8(6)	16.2(6)	0.50	16.82	0.4(7)	0.26
(330)	6.78(5)	7.96(2)	0.82	6.68	1.18(6)	1.16
(101)	0.42(1)	0.46(1)	0.98	0.40	0.04(2)	0.20

TABLE III. Individual site susceptibilities as determined from polarized-neutron experiments at 155 K. The magnetization was induced by an applied magnetic field of 4.6 T (**H**∥[001]) and 5.0 T (**H**∥[100]). The fourth column (neutron total) is simply $\chi_U+4\chi_{Fe}$ and the last column gives the bulk susceptibility per mole of UFe_4Al_8 at nominally the same temperature measured on an oriented small single-crystal using a superconducting quantum interference device magnetometer.

$10^3\chi(\mu_B/T)$	U	Fe	Neutron total	Bulk
$H\parallel[001]$	6.4(3)	40.3(6)	168(5)	115(2)
$H\parallel[100]$	52(4)	68.9(16)	328(10)	225(14)

makes with the **a** axis (see Figs. 3 and 4) must be determined by experiment. The ferromagnetic and antiferromagnetic components of the Fe moment are given by μ sinϕ and μ cosϕ, respectively.

III. POLARIZED NEUTRONS

A. Paramagnetic state

To determine the individual sublattice susceptibilities in the paramagnetic state we have performed polarized-neutron experiments with a magnetic field applied first parallel to the [001] and then the [100] axis. Since certain reflections have magnetic contributions from the U sublattice only, and others from both sublattices, the individual sublattice susceptibilities may be deduced provided that the crystal structure and Fe and U form factors are known. For the form factors we have used the well-known metallic iron form factor,[14] and for the U we have used the standard U^{3+} form factor. This latter point is addressed in more detail below; the approximation used here is sufficiently accurate to give a reliable extrapolation to $Q=0$, which gives the susceptibility. The results of these measurements are given in Table III. It is clear that the actinide atom is the source of the high magnetic anisotropy of this system, the induced moment at $T=155$ K with the field applied parallel to the easy axis (**a**) being about an order of magnitude greater than that measured when the field is applied along the hard axis (**c**). The iron atom is easily polarized even when the field is applied along the hard axis, with a ratio of 1.7 between the susceptibilities measured along the **a** and **c** axis.

B. The ordered state with applied field

To establish the relationship between the two sublattices below the ordering temperature we have performed an experiment on the D3 polarized-neutron spectrometer at the ILL. The neutron wavelength used was 0.843 Å, which is sufficiently short that the extinction corrections are quite small and can be made with certainty.

The polarized-beam technique, of course, is ideal for looking at ferromagnets or signals from paramagnets in a field, as in Sec. III A above. In this case the general feature is that all moments are *collinear* with the applied field, which is also the direction of polarization of the incident neutrons. In the case of UFe_4Al_8 we expect the U contribution and any *ferromagnetic* Fe contribution to be parallel to the applied field, but the majority of the Fe moment orders *antiferromagnetically*. The situation is then much more complex. Since the ordering wave vector is $\mathbf{q}=[000]$ there is overlap between the magnetic and nuclear reflections. Before continuing to evaluate the magnetic structure factors we need to consider possible domain configurations, of which there are two:

(i) Tetragonal domains are those for which the spin directions are all rotated by $\pi/2$. For example, if the moment is aligned along **a**, it may equally well be aligned along **b**;

(ii) 180° domains are those in which all spins are reversed in direction;

In zero-field domains (i) and (ii) should be equally populated. In an applied field within the basal plane it is clear that for $H>H_{\text{critical}}$ a single domain is formed. From Ref. 12, $H_{\text{critical}}\sim1.5$ T.

Another more subtle point is to understand the preference for a certain ϕ angle as drawn in Fig. 4. It would seem at first glance that changing the directions of the Fe moments on atoms 1 and 3 in the figure would lead to the same configuration. That is by changing ϕ to $(180-\phi)$. To understand the preference for a particular value of ϕ, reference should be made to similar experiments on the weak ferromagnet NiF_2.[15] Briefly, in the $ThMn_{12}$ structure the Fe have site symmetry $2/m$ and the twofold axis is parallel to a ⟨110⟩ axis. If one considers the Fe atom at (1/4 1/4 1/4) (Fe atom 1 in Fig. 3) the local twofold axis is [110], but for the atom at (1/4 3/4 1/4) (Fe atom 3 in Fig. 3) it is [1,−1,0], etc. That is, there is a local rotation of symmetry by $\pi/2$ about the tetragonal **c** axis every time a translation of $a/2$ or $b/2$ is made. If the field is then applied along the direction [100] the *local* field around the two Fe spins + and − in Fig. 3 is different. The application of the symmetry operators shows that if the crystal-field term favors the twofold axis as the spin direction, and the exchange field favors antiparallel coupling, then a weak ferromagnetic structure will result. Thus the local symmetry operators assure an optimum value for ϕ. Put another way, it is the local coordination of the Al atoms around the Fe 1 site in Fig. 3 that rotates by $\pi/2$ on translating from Fe 1 to Fe 3. This is shown more directly by Fig. 4 in which we have attempted to show the three-dimensional configuration of Al atoms around the Fe atoms 1 and 3 in Fig. 3. Clearly, values of ϕ and $(180-\phi)$ are *different* with respect to the local environment.

To analyze the polarized-beam data it is convenient to divide the reflections in two subsets: (a) ‘‘ferromagnetic reflections’’ (reflections with H or K even) that are sensitive only to the component of the magnetization aligned parallel to the applied field; and (b) ‘‘antiferromagnetic reflections,’’ those that, in addition to a contribution from the U moment, are also sensitive to the antiferromagnetic component of the Fe moments and therefore have a magnetic structure factor that is *not* parallel to the field. This last subset comprises the reflections with H and K odd and L even.

For the ‘‘ferromagnetic reflections’’ (i.e., those that are not sensitive to the Fe AF component) a standard procedure can be used to extract the magnetic structure factors from the measured flipping ratios, after due corrections for extinction, imperfect polarization, and $\lambda/2$ contamination of the neutron beam. The analysis of the subset of ‘‘antiferromagnetic reflections’’ is more complicated, because the direction of the magnetic structure factors is not known *a priori*. It depends

TABLE IV. Magnetic moments of the U and Fe sublattices derived from the least-squares refinements. (I) refinement on the structure factors of the ‘‘ferromagnetic’’ reflections; (II) refinement on the flipping ratios using the whole data set. C_2 is also a parameter in these refinements (see text); its value is 2.1(1). The number of reflections and parameters is given, and the final row gives the R factors which give a measure of the goodness of fit.

Moment (μ_B)	(I)	(II)
μ_U	0.50(3)	0.47(1)
μ_{Fe}(par.)	0.50(1)	0.47(2)
μ_{Fe}(perp.)		0.99(2)
Reflections	56	251
Parameters	3	4
R	10.9%	2.13%

on the relative magnitudes of the antiferromagnetic and ferromagnetic components of the Fe atoms. A similar situation occurs when the structure is noncentrosymmetric in which case it is also not possible to extract the magnetic structure factors from the flipping ratios.[16] In such cases, a magnetic model has to be fitted directly to the measured flipping ratios. In all calculations the magnetic form factor of the U atom was calculated in the dipole approximation, $f_U = \langle j_0 \rangle + C_2 \langle j_2 \rangle$, where the coefficient $C_2 = \mu_L / \mu$ is the ratio between the orbital and total moment of the U atom. The $\langle j_{0,2} \rangle$ functions are derived from the radial $5f$ density distribution calculated by Desclaux and Freeman.[17]

The subset of ‘‘ferromagnetic’’ reflections was analyzed in a least-squares refinement of the magnetic structure factors that includes as variables the value of the magnetic moment at the U and Fe sites and the coefficient C_2 of the U form factor. The results of the least-squares fit are shown in Table IV, column 1. The residual error between the observed and calculated magnetic structure factors is $R = 10\%$. This relatively high value may be a result of imperfect corrections for extinction, multiple-scattering effects on some of the weaker reflections, and the assumption of spherical magnetization densities when processing the data. The total magnetization derived from the refinement is $2.50(6)\mu_B$ per mole, in excellent agreement with the value of the bulk magnetization. As expected, a model including only a moment at the Fe or U site cannot fit the data at all. The major magnetic contribution to these reflections comes from the Fe moments because there are four Fe atoms to only one U atom. In Fig. 5 we have plotted the effective moment derived from this set of reflections as a function of $\sin\theta/\lambda$. Since the moments are on both sites, the observed results do not fall on a single $f(Q)$, but the observed and calculated are represented by open circles and filled triangles, respectively. They are in good agreement and fall near a conventional Fe form factor (solid curve).

We then proceeded to an analysis of the full data set. All measured flipping ratios were included in this refinement (251 observations), with exception of two weak reflections (101) and (002) that are very sensitive to multiple scattering and second-order contamination. The model allows the Fe moments to tilt towards the direction of the applied magnetic field, while preserving the mm' symmetry of the magnetic structure. We have obtained a good fit to the data with a tilt angle of 25.4(3)° at 4.6 T with a final residual factor $R = 2.13\%$ (Table IV, column II). A list of the measured and calculated flipping ratios of the low-angle ‘‘antiferromagnetic reflections’’ is shown in Table V. The magnitude of the Fe moment $1.10(2)\mu_B$, obtained by adding the two components in quadrature, is in good agreement with the value expected from the measured Mössbauer hyperfine field at the Fe nuclei.[8] Also an important consistency check of the refinement of the flipping ratios is that the values of the U moment and the component of the Fe moment aligned with the field agree with the results of the previous refinement of the ferromagnetic magnetic structure factors.

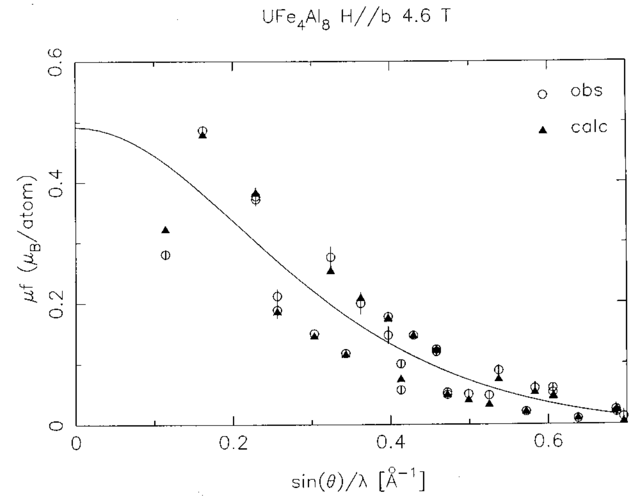

FIG. 5. Magnetic-scattering amplitude of a subset of reflections with H, K, and L all even plotted on a Fe form-factor curve. These reflections have contributions from both U and Fe ferromagnetic components. If the U moment was zero the observed amplitudes (circles) should follow the Fe form factor, shown as a solid curve. The calculated values (triangles) include the U moment derived from the least-squares refinement of the flipping ratios.

C. Uranium moment—temperature dependence and form factor

Once the sublattice interactions are understood in this compound, we may focus on the temperature dependence of the uranium sublattice. In fact the uranium ferromagnetic moment contributes to the (110) reflection, see Fig. 2(a), but its contribution is only ~25% of the nuclear (110) intensity. The large increase of a factor of ~5 (Table II) in the intensity of the (110) reflection comes from the antiferromagnetic Fe component. Instead we show in Fig. 2(b) the growth of the uranium moment by selecting a reflection such as (211), which has H and K of different parity, and L odd, and this senses the U sublattice only. The polarized-beam technique may then be used to observe the change of the U ferromagnetic component with temperature. A field of 1.5 T is used for these measurements, which is the minimum that can be used with the superconducting magnet on D3 otherwise the incident neutrons become depolarized. In the paramagnetic state, $T > 155$ K, we observe the induced moment, but when comparing Figs. 2(a) and 2(b) it is clear that the development of magnetic order with temperature on the two sites is very similar.

TABLE V. Observed and calculated flipping ratios of the low-angle subset of ‘‘antiferromagnetic’’ reflections *H* and *K* odd, and *L* even.

HKL	R_{obs}	R_{calc}	*HKL*	R_{obs}	R_{calc}
$\bar{1}10$	2.35(7)	2.428	$\bar{5}1\bar{2}$	1.130(9)	1.097
110	2.72(9)	2.693	$51\bar{2}$	1.061(9)	1.007
$\bar{3}10$	0.569(5)	0.588	512	1.120(10)	1.055
310	0.590(16)	0.577	$\bar{5}32$	1.409(9)	1.353
$\bar{1}30$	0.626(3)	0.615	$53\bar{2}$	1.476(10)	1.400
130	0.507(2)	0.519	532	1.455(10)	1.377
$\bar{1}12$	1.240(7)	1.209	$1\bar{1}4$	1.36(5)	1.320
$11\bar{2}$	1.073(6)	1.062	114	1.32(5)	1.341
112	1.168(7)	1.169	$\bar{1}34$	1.19(3)	1.161
$\bar{3}12$	1.531(8)	1.584	134	1.08(3)	1.067
$3\bar{1}\bar{2}$	1.872(10)	1.901	$3\bar{1}\bar{4}$	1.205(2)	1.192
312	1.743(9)	1.774	$\bar{3}1\bar{4}$	1.155(2)	1.108
$\bar{1}32$	1.316(10)	1.357	712	0.691(2)	0.679
132	1.613(13)	1.586	$71\bar{2}$	0.667(2)	0.722
$5\bar{1}0$	1.985(11)	1.894	334	1.167(8)	1.143
510	1.952(14)	1.918	$3\bar{3}\bar{4}$	1.170(11)	1.133
530	0.764(4)	0.752	514	1.23(2)	1.183
$\bar{3}30$	2.273(10)	2.216	$11\bar{4}$	1.356(6)	1.320
330	2.521(12)	2.426	114	1.324(9)	1.341

We show in Fig. 6 the deduced magnetic moment as a function of sinθ/λ for those reflections that have a contribution from the U moment only. One of the early motivations for performing these experiments was the question of whether the U form factor, $f(Q)$, would show a similar effect to that found[18] in UFe$_2$, in which the partial cancellation of the spin and orbital contributions results in a $f(Q)$ for uranium that has a maximum at sin$\theta/\lambda \sim 0.2$ Å^{-1} rather than at 0. Although there is a spread in the $f(Q)$ for the first few reflections that is unusual in these kind of measurements, there appears no evidence for anything else than fluctuations around a conventional form factor. These ‘‘deviations’’ in Fig. 6 may be caused by multiple-scattering effects. A more interesting possibility is that they might be caused by nonspherical contributions from the Fe spin distribution, but to determine this requires more experiments. A better measure of the U form factor is obtained from the refinement of the C_2 coefficient in the dipole approximation. This we find to be 2.1(1). Using the relationship $\mu_L/\mu_S = C_2/(1-C_2)$, where μ_L and μ_S are the orbital and spin moments, respectively, on the uranium site, we find $\mu_L/\mu_S = -1.9(1)$. The values of μ_L/μ_S for $5f^2$ and $5f^3$ are -3.32 and -2.56, respectively. Thus, the $|\mu_L/\mu_S|$ ratio in UFe$_4$Al$_8$ is close to that found in URhAl,[16] and the hybridization between the U $5f$ and Fe $3d$ electrons is not sufficient to cause the reduction in the orbital moment similar to that found in UFe$_2$ and which results in $\mu_L/\mu_S \sim -1$.

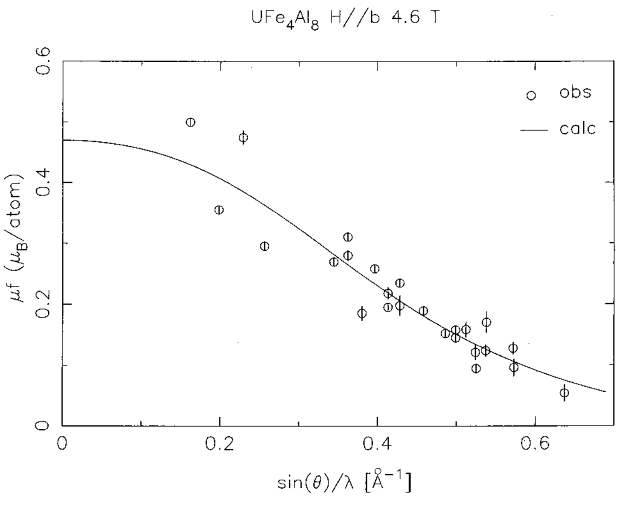

FIG. 6. Uranium form factor derived from the flipping ratios measured at 4.2 K with an applied magnetic field of 4.62 T//[010]. The reflections shown have *H* and *K* of different parity (and *L* odd) and are sensitive only to the U moment. The solid curve represents the U^{3+} form factor calculated with the dipole approximation.

IV. DISCUSSION

An important result of our experiments on a single crystal is that they prove beyond all doubt that UFe$_4$Al$_8$ is *not* a spin glass. The results of Gal *et al.*[8] can be readily understood by realizing that in a powder sample there is considerable motion of the particles when a field is applied and the material is anisotropic.[19] No doubt the same applies to the material HoFe$_4$Al$_8$.[9] However, Gal *et al.*[8] were correct in stating that the sublattice interactions are unusual in this compound. The AF part of the Fe moment orders ($\sim 1\mu_B$ per Fe) such that the spins lie always in the basal plane of the tetragonal structure. There is little anisotropy within this basal plane—as already suggested by magnetization measurements.[12] The uranium sublattice also orders, but ferromagnetically, with the U moment again within the basal plane. Symmetry considerations show that the Fe moments can have a small ferromagnetic component, i.e., the Fe spins are canted as a result of the exchange field established by the U moments.

This canting in zero field is small, only 16° (see Fig. 3) but it gives rise to a ferromagnetic component of $0.3\mu_B$ per Fe. The remanence deduced from magnetization measurements[12] may be understood as arising from *both* U and Fe contributions. In a field applied in the basal plane this canting angle rapidly increases, so that by 4.6 T the measurements show that it is 25°. This increase of the canting angle accounts for the large high-field susceptibility.

Measurements just above T_C of the individual site susceptibilities show that it is the U moment which provide the major contribution to the *bulk* anisotropy. A magnetic field in the hard axis [001] induces a sizable moment on the Fe site, but little response on the U site. Of course, this situation is common in actinide compounds, where the actinide ion itself provides the major contribution to the magnetic anisotropy.

The Fe and U are clearly closely coupled; for example in rare-earth alloys of the same composition the rare earths develop their full magnetic moments only at low temperature (typically 20–40 K), a process primarily driven by the hybridization of the rare-earth $5d$ electrons with the Fe $3d$ electrons, rather than a direct $4f$-$3d$ interaction.[1,3] In UFe_4Al_8 the temperature dependence of the magnetization is similar for both sublattices; no doubt as a consequence of the molecular fields from the weak ferromagnetism of the Fe sublattice.

Our measurements have clarified a long-standing problem of characterizing the magnetic structure of UFe_4Al_8. However, they have also raised a number of questions. For example, in their measurements on the magnetoresistance Bonfait *et al.*[13] interpreted the results in terms of unusual domain configurations. Our measurements suggest that this model for the domain configurations is probably correct. However, in writing $\rho_\parallel$ and $\rho_\perp$ in terms of a current flow either parallel or perpendicular to the *ferromagnetic* component our measurements suggest this is a great simplification. In fact, when the current is perpendicular (or parallel) to the *ferromagnetic* U component it is clearly parallel (or perpendicular) to the principal *antiferromagnetic* component of the Fe sublattice. Although no theoretical treatment of the magnetoresistance results has yet appeared, it will be important to bear these points in mind in developing any theory.

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