

Magnetization density in $\text{UFe}_{10}\text{Si}_2$

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1998 J. Phys.: Condens. Matter 10 4071

(<http://iopscience.iop.org/0953-8984/10/18/016>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.209

The article was downloaded on 14/05/2010 at 13:07

Please note that [terms and conditions apply](#).

Magnetization density in $\text{UFe}_{10}\text{Si}_2$

J A Paixão†*, J C Waerenborgh‡, M S Rogalski§‡, A P Gonçalves‡, M Almeida‡, A Gukasov||, M Bonnet¶, J C Spirlet+ and J B Sousa§

† Departamento de Física, Faculdade Ciências e Tecnologia, Universidade de Coimbra, P-3000 Coimbra, Portugal

‡ Departamento de Química, Instituto de Tecnologia Nuclear, P-2686 Sacavém Codex, Portugal

§ IFIMUP, Faculdade de Ciências da Universidade do Porto, P-4150 Porto, Portugal

|| Laboratoire Léon-Brillouin (CEA-CNRS), Centre d'Études Saclay, F-91191 Gif-sur-Yvette, France

¶ Département de Recherche Fondamentale sur la Matière Condensée, SPSMS, F-38054 Grenoble Cédex 9, France

+ European Commission, Joint Research Centre, Institute for Transuranium Elements, Postfach 2340, D-76125 Karlsruhe, Germany

Received 28 August 1997, in final form 18 December 1997

Abstract. A neutron scattering study of the magnetization density distribution in $\text{UFe}_{10}\text{Si}_2$ is reported. Results are compared with the ^{57}Fe Mössbauer data obtained for the same sample. Polarized neutron scattering measurements show that the magnetization aligned parallel to an applied field of 2 T is mostly due to the Fe atoms. The values of the magnetic moment at the three crystallographic sites occupied by the transition metal atom are $1.67(3) \mu_B$ (8f), $2.30(3) \mu_B$ (8i) and $1.73(4) \mu_B$ (8j). A small magnetic moment ($\sim 0.3 \mu_B$) is measured at the U site, aligned ferromagnetically with the Fe moments. So small a value appears to be due to a partial cancellation of the individual orbital and spin magnetic moment contributions of the actinide atom. The values of the ^{57}Fe hyperfine magnetic field at the three crystallographic sites occupied by the Fe atoms change in the same order as the magnetic moments determined by neutron scattering, although the ratio B_{hf}/μ_{Fe} is not constant.

1. Introduction

There has been considerable interest in studying iron-rich ternary intermetallic compounds with the tetragonal ThMn_{12} type of structure owing to their relatively high Curie temperature, saturation magnetization and favourable magnetocrystalline anisotropy which could be of potential use in permanent magnet applications. $\text{UFe}_{10}\text{Si}_2$ was the first actinide compound of this family with a high concentration of a transition metal to be studied (Suski *et al* 1989, Berlureau *et al* 1989).

The first results on the magnetic properties of $\text{UFe}_{10}\text{Si}_2$, obtained from polycrystalline samples, showed that this compound is ferromagnetic below $T_C \approx 650$ K (Berlureau *et al* 1989) with a considerable uniaxial magnetic anisotropy at room temperature. The occurrence of a first-order magnetization process (FOMP) at $H \approx 3$ T was first reported by Andreev *et al* (1992a), and was shown to exist up to unexpectedly high temperatures of about $0.5 T_C$ (Andreev *et al* 1994).

* Author to whom any correspondence should be addressed.

‡ Present address: UCEH, Universidade do Algarve, Gambelas, P-8000 Faro, Portugal.

Recently, large $\text{UFe}_{10}\text{Si}_2$ single crystals were grown by the Czochralski method (Gonçalves *et al* 1994). Single-crystal magnetization measurements performed along the main crystallographic directions confirmed the uniaxial character of the magnetic anisotropy and determined c to be the easy axis (Estrela *et al* 1995). The saturation magnetization ($19.5 \mu_B/\text{f.u.}$ at 4.2 K) was found to be significantly larger than previously reported values obtained with aligned powders. Furthermore, the analysis of the FOMP through the simultaneous measurement of the longitudinal ($\parallel \mathbf{H}$) and transverse ($\perp \mathbf{H}$) components of the magnetization at different temperatures determined that this transition is due to a rotation of the total magnetization and allowed the calculation of the anisotropy constants $K_1 = 3.85 \text{ MJ m}^{-3}$, $K_2 = -1.6 \text{ MJ m}^{-3}$, $K_3 = 0.39 \text{ MJ m}^{-3}$. Comparing these values to those of the rare-earth analogue compounds suggests the existence of an important exchange interaction between the U and Fe atoms which could also explain the enhanced Curie temperature of $\text{UFe}_{10}\text{Si}_2$ as compared to that of $\text{YFe}_{10}\text{Si}_2$. Engelen and Buschow (1990) have studied the magneto-optical properties of a series of Fe-rich ThMn_{12} -type compounds, including $\text{YFe}_{10}\text{Si}_2$ and $\text{UFe}_{10}\text{Si}_2$. They have found an enhancement of the Kerr effect in $\text{UFe}_{10}\text{Si}_2$ that would imply a strong magnetic interaction between the 5f and 3d electrons in this material and a non-zero U magnetization at room temperature.

Although the magnetization curves of both $\text{UFe}_{10}\text{Si}_2$ and $\text{YFe}_{10}\text{Si}_2$ show that both compounds exhibit uniaxial anisotropy, the anisotropy energy of $\text{UFe}_{10}\text{Si}_2$ is much larger. The first anisotropy constant of $\text{YFe}_{10}\text{Si}_2$ is $K_1 = 1.4 \text{ MJ m}^{-3}$ (Andreev *et al* 1993), a third of the value measured for $\text{UFe}_{10}\text{Si}_2$. This fact and the existence of a field-induced magnetic transition in $\text{UFe}_{10}\text{Si}_2$ when the field is applied along the hard axis suggests the presence of a sizeable orbital moment at the U site. It is, however, difficult to determine the value of the magnetization of the U sublattice by comparison of the saturation magnetization of the two compounds. First, there are no measurements reported on single crystals for $\text{YFe}_{10}\text{Si}_2$, and the values determined from aligned powders may differ substantially from those measured on single crystals. For $\text{YFe}_{10}\text{Si}_2$ the value of the saturation magnetization at 4.5 K measured on oriented powders is $18.2 \mu_B/\text{f.u.}$. Neutron diffraction data on $\text{YFe}_{10}\text{Si}_2$ (Lin *et al* 1991) determined a total moment of $21.1 \mu_B/\text{f.u.}$ but no such measurements have been reported for $\text{UFe}_{10}\text{Si}_2$ up to now. Assuming that the same average value of the hyperfine field of $12.7 \text{ T}/\mu_B$ holds for both compounds, the iron contribution to the saturation magnetization in $\text{UFe}_{10}\text{Si}_2$ is $18.1 \mu_B/\text{f.u.}$, and a uranium moment of $1.4 \mu_B$ was estimated by Estrela *et al* (1995) from their single-crystal magnetization data. A smaller value of $0.5 \mu_B/\text{f.u.}$ had been estimated by Andreev *et al* (1992a) from their powder measurements. However, the use of an average hyperfine field based on the assumption of a linear relationship between the hyperfine field (B_{hf}) and the magnetic moment (μ_{Fe}) at the Fe sites to determine the value of the U moment is questionable (see below). Therefore, neutron diffraction data are needed to determine the value of the individual moments in $\text{UFe}_{10}\text{Si}_2$. From the point of view of theory, the magnetization density distribution in $\text{UFe}_{10}\text{Si}_2$ was recently addressed by Matar *et al* (1994) who have reported spin-polarized band-structure calculations in the local spin-density approximation for the ThMn_{12} compounds $\text{UFe}_{10}\text{Si}_2$, $\text{UCo}_{10}\text{Si}_2$ and $\text{YFe}_{10}\text{Si}_2$. Their calculations predict an antiparallel spin alignment for U and Fe.

In order to clarify the individual contributions of the actinide and transition metal sublattices to the magnetic properties of this compound, a combined Mössbauer and neutron diffraction study on the *same* single crystal as was used for the magnetization study by Estrela *et al* (1995) was performed. Neutron diffraction results are reported in this work and the estimated iron moments compared with the hyperfine-field values obtained from low-temperature Mössbauer data (Waerenborgh *et al* 1997). The use of the same sample in both techniques is emphasized, since it has been observed for other ThMn_{12} compounds that

the site distribution of the transition metal and p element may depend on the preparative conditions and on the subsequent heat treatments, and that small variations in the atomic arrangement can result in significant differences in the physical properties (Gonçalves *et al* 1998). It is tempting to correlate the magnetic hyperfine field estimated from Mössbauer spectroscopy and the local iron moments deduced from neutron diffraction data. Considering that in some cases a linear relationship between B_{hf} and μ_{Fe} is found (e.g. for the binary Y–Fe compounds; Sinnemann *et al* (1989), Denissen *et al* (1990)), the magnitudes of the Fe moments have sometimes been tentatively deduced from the measured hyperfine fields in the absence of neutron measurements (Andreev *et al* 1992b, Estrela *et al* 1995). In this work we compare how the Mössbauer hyperfine fields measured in $UFe_{10}Si_2$ correlate with the magnetic moments derived from neutron diffraction for the three sites occupied by the Fe atoms, so that the uncertainties involved in the above approximation may be appreciated.

Table 1. Results of the least-squares structure refinement of $UFe_{10}Si_2$ from the unpolarized neutron diffraction data.

Site	x	y	z	B_{eq} (\AA^2)	%Fe
2a	0	0	0	0.84(2)	0
8f	1/4	1/4	1/4	0.74(2)	0.61(2)
8i	0.355 75(15)	0	0	0.93(1)	0.96(2)
8j	0.282 36(18)	1/2	0	0.92(2)	0.93(3)

$a = b = 8.3729(5)$ \AA , $c = 4.7269(4)$ \AA
 $R = 4.1\%$, 167 reflections, 18 variables

2. Neutron scattering

2.1. Unpolarized neutron diffraction

In view of the uncertainties as to site occupation and also as to characterizing the extinction of the $UFe_{10}Si_2$ single crystal, an unpolarized neutron diffraction experiment at room temperature was performed at Siloé, CENG, on the DN4 four-circle diffractometer. This instrument is equipped with a curved Cu(200) monochromator reflecting neutrons with wavelength 1.184 \AA . The $\lambda/2$ contamination of the neutron beam was estimated to be $\approx 0.5\%$. The crystal examined was grown by the Czochralski method and is from the same batch as that used for the x-ray diffraction and magnetization measurements reported by Estrela *et al* (1995). The sample is a small cylinder with a height of 3 mm and a diameter of 2 mm, with the growth direction approximately parallel to the c -axis. The intensities were measured by means of ω - 2θ scans up to $(\sin\theta)/\lambda = 0.70$ \AA^{-1} . The minimum and maximum scan width were 1.25° and 2.0° . During the data collection the intensities of 1161 Bragg reflections were measured. Equivalent reflections were averaged resulting in a set of 175 independent reflections, with an internal agreement factor between equivalent reflections, after averaging, of $R_{\text{int}}(F^2) = 3.6\%$. The structure refinement was performed using a locally modified version of the least-squares program SFLSQ included in the CCSL library (Brown and Matthewman 1987). Atomic positions, isotropic thermal displacement parameters, an extinction correction and site occupancies were refined. The extinction model included in the least-squares program is that of Becker and Coppens (1974). We have found that extinction is small in our crystal, the maximum intensity

loss due to extinction effects being $\Delta I/I_{\max} = 21\%$. Suitable constraints were applied to the site occupancies to ensure that the nominal stoichiometry was preserved during the least-squares cycles. As $\text{UFe}_{10}\text{Si}_2$ is already magnetically ordered at room temperature, the intensities of the weak reflections at low Q ($=4\pi \sin \theta/\lambda$) are likely to obtain a significant magnetic scattering contribution. Therefore, and in order to obtain unbiased values for the structural parameters, we have decided to reject these weak reflections subject to the criteria $I < 0.1I_{\max}$ and $(\sin \theta)/\lambda < 0.4 \text{ \AA}^{-1}$. The results of the least-squares refinement are shown in table 1. We have found, in agreement with previous x-ray studies and also with the Mössbauer data (Waerenborgh *et al* 1997), that the 8i sites are almost exclusively occupied by the transition metal atoms. The 8j and 8f sites are shared by Fe and Si atoms but there is a clear preference for the Fe atoms to occupy the 8j sites.

2.2. Polarized neutron scattering

The polarized neutron measurements were performed at the LLB, Saclay, on the 5C1 polarized neutron diffractometer. A Heusler alloy single crystal was used to monochromatize and simultaneously polarize the incoming neutron beam. A wavelength of 0.83 \AA was used and an Er foil was inserted in the incident beam to reduce $\lambda/2$ contamination. The residual $\lambda/2$ contamination is estimated to be less than 0.05%. A Meissner–Majorana cryo-flipper with efficiency close to 100% was used to invert the polarization of the neutron beam. The sample was cooled to 4.2 K in a field of 2 T and was kept at these values of temperature and field during the data collection. A total of 543 flipping ratios on the $(hk0)$ layer were measured up to a value of $(\sin \theta)/\lambda = 0.725 \text{ \AA}^{-1}$.

A minimum of three and typically six equivalent reflections were measured not only to improve the statistics but also to check the extinction correction and to detect a possible contamination by multiple scattering of weak reflections. Flipping ratios of symmetry-equivalent reflections were in general found to agree within the estimated errors bars. After averaging the equivalents, a set of 32 unique reflections was obtained. The magnetic structure factors were determined from the flipping ratios using the standard Cambridge procedure (the program SORGAM, CCSL library). Corrections for extinction, incomplete polarization of the beam and $\lambda/2$ contamination were included. A description of the details involved can be found in Paixão *et al* (1992). The magnetic structure factors were analysed by least-squares fitting to determine the individual magnetic moments at the 2a, 8i, 8f and 8j sites and also to obtain the maps representing the magnetization density distribution in the unit cell.

3. Results

The measured magnetic structure factors were fitted to a simple model where the magnetization density in the unit cell is described as a superposition of spherical magnetization densities located at the sites occupied by the actinide and transition metal atoms. The magnetic structure factors, $F_M(hkl)$, are the Fourier components of the magnetization density distribution in the crystal, and can be calculated in this model from

$$F_M(hkl) = \sum_{j=1}^n \mu_j s_j f_j(Q) e^{2\pi i(hx_j + ky_j + lz_j)} e^{-W_j}.$$

In this expression the sum extends over the n magnetic sites, μ_j is the moment of the atoms at the j -site, s_j is the site occupation and W_j the Debye–Waller factor. The function $f(Q)$ is the magnetic form factor, which is the Fourier transform of the magnetization density

Table 2. Atomic moments (in μ_B) in $UFe_{10}Si_2$ derived from the analysis of the polarized neutron diffraction data. The results obtained using two models (I and II) are shown. In I the U atom has a fixed $C_2 = \mu_L/\mu$ value of 1.64 whereas in II the orbital moment of the U atom is included as a refinable parameter.

Site	I	II
U (2a), μ	0.48(3)	0.26(7)
U (2a), μ_L	0.79(5)	1.05(8)
Fe (8f)	1.70(3)	1.67(3)
Fe (8i)	2.25(3)	2.30(3)
Fe (8j)	1.72(4)	1.73(4)
R	6.4	5.4
χ^2	15.7	10.5

associated with a single atom. The uranium magnetic form factor was calculated in the dipole approximation (Marshall and Lovesey 1971):

$$\mu f(Q) = \mu(\langle j_0 \rangle + C_2 \langle j_2 \rangle) \quad C_2 = \frac{\mu_L}{\mu}$$

where the C_2 -coefficient is the ratio between the orbital (μ_L) and total (μ) moments of the actinide atom. For a U^{3+} ion in an intermediate-coupling configuration corresponding to the spectroscopic g -value, $C_2 = 1.64$. We used the $\langle j_0 \rangle$ and $\langle j_2 \rangle$ functions calculated by Desclaux and Freeman (1978) for the U^{3+} state. For the Fe atoms we have used the Fe form factor measured by Shull and Yamada (1962) for the element metal. The results of the least-squares fits are shown in table 2. Two fits (I and II) were performed. In I the value of the C_2 -coefficient was fixed to the value 1.64 whereas in II the ratio of the orbital and total moments of the U atom was allowed to vary. The results show that the magnetic moment carried by the actinide atom is small, indeed much smaller than the full

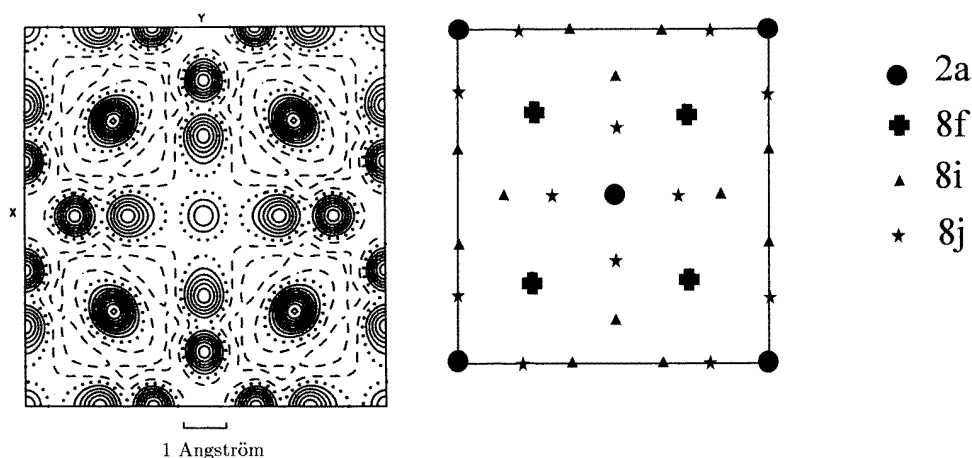


Figure 1. A magnetization density map of $UFe_{10}Si_2$ showing the projection of the magnetization density distribution on the a - b plane reconstructed by Fourier inversion of the polarized neutron scattering data. Solid and dashed contours correspond to positive and negative values of the density, respectively. The zero-density contour is shown as a dotted line. Contour step: $0.25 \mu_B \text{ \AA}^2$.

moment of the ion in either f^2 (U^{4+}) or f^3 (U^{3+}) configurations. This ‘suppression’ of the magnetic moment might arise from hybridization of the extended f electrons with the conduction band, a case in which one would expect the C_2 -coefficient to increase with respect to the free-ion value. According to Hund’s rules, μ_L and μ_S for the U atom have opposite signs, but μ_L is larger than μ_S for both f^2 and f^3 configurations. A partial quenching of the orbital moment due to hybridization between the $5f$ and the conduction electron states will reduce mainly the μ_L -component, bringing the ratio close to -1 and increasing the $C_2 = 1/(1 + \mu_L/\mu_S)$ value. In support of this view, releasing the coefficient C_2 in refinement II gives a better fit to the data with a larger C_2 -value ($C_2 = 4.0$) than refinement I ($C_2 = 1.64$). Although an extra parameter was introduced in refinement II, the decrease of five units in χ^2 when C_2 is released is statistically significant. Unfortunately, two low- Q reflections ((110) and (220)) that would be very sensitive to the value of the C_2 -coefficient could not be used in the refinements because they have small nuclear structure factors, and hence their magnetic structure factors could not be determined reliably. The whole moment in the unit cell calculated from the results of fit II is $19.6 \mu_B$, which is in very good agreement with the measured magnetization along the c -axis, implying that a possible contribution from a polarization of the conduction electrons is small. A more direct insight into the magnetization density distribution in the unit cell can be obtained by inverse Fourier transforming the magnetic structure factors (table 3) from reciprocal to direct space. As data could only be measured on the ($hk0$) layer, it is not feasible to reconstruct a 3D map of the magnetization density distribution. However, there is good resolution for mapping a projection of the magnetization density perpendicular to the applied magnetic field, i.e. along the c -axis. The Fourier map showing the projection of the magnetization density distribution on the tetragonal basal plane of $UFe_{10}Si_2$ is depicted in figure 1. In order to reduce series-termination effects, the density shown is an average of the point density on a cube of 0.2 \AA side. The map clearly confirms the results of the least-squares analysis, showing that most of the magnetic moment is located at the Fe sites, and only a small magnetic moment is present at the U site.

The site occupation factors deduced from neutron diffraction (table 1) are consistent with the Mössbauer data obtained for the same sample (Waerenborgh *et al* 1997). As discussed in Waerenborgh *et al* (1997), two fits of the Mössbauer spectrum at 5 K may be performed. The fitting constants may be chosen such that the estimated distribution of the Fe atoms among the 8i, 8j and 8f sites is found to be in agreement with either the site occupation factors in table 1 or with those obtained from the refinement of the single-crystal x-ray diffraction data (Estrela *et al* 1995). The neutron data support the assumption made in the analysis of the spectra obtained below $T_C = 650 \text{ K}$ that the linewidths of the 8f sextet were 0.9 times the corresponding linewidths of the other sextets (Waerenborgh *et al* 1997).

The B_{hf} -values estimated from the Mössbauer spectrum obtained at 5 K, 24.7 T, 22.8 T and 20.6 T for the 8i, 8j and 8f sites, respectively (Waerenborgh *et al* 1997), decrease in the same order ($8i > 8j > 8f$) as the corresponding magnetic moments deduced from the neutron diffraction data (table 2). However, different ratios B_{hf}/μ_{Fe} are found for the different crystallographic sites: $12.7 \text{ T}/\mu_B$ (8f), $13.0 \text{ T}/\mu_B$ (8j) and $10.7 \text{ T}/\mu_B$ (8i), from which an average value of $11.8 \text{ T}/\mu_B$ may be deduced. Ratios of these two quantities, estimated from experimental data, have often been published. Values ranging from 14.5 to $15.5 \text{ T}/\mu_B$ based on neutron diffraction and Mössbauer data were reported by binary Y–Fe compounds, $Y_2Fe_{14}B$ and $YFe_{10}V_2$ (Sinnemann *et al* 1989, Denissen *et al* 1990). An average moment of $2.11 \mu_B/(\text{Fe atom})$ and an average B_{hf} of $26.85 \text{ T}/(\text{Fe atom})$ were estimated for $YFe_{10}Si_2$ from neutron powder diffraction (Lin *et al* 1991) and Mössbauer effect studies (Andreev *et al* 1992b), respectively. A lower average ratio of $12.7 \text{ T}/\mu_B$ is

therefore found for this compound and, as in the case of $UFe_{10}Si_2$ a significant spread of values is observed for the individual crystallographic sites: $15.3 \text{ T}/\mu_B$ (8f), $11.5 \text{ T}/\mu_B$ (8j) and $12.3 \text{ T}/\mu_B$ (8i). These ratios however should be treated with caution since the samples of $YFe_{10}Si_2$ measured by the two techniques were not obtained from the same material and, furthermore, the crystallographic site distributions of Fe and Si published by Lin *et al* (1991) and Andreev *et al* (1992b) are significantly different.

Table 3. Observed magnetic scattering amplitudes ($\mu_B/\text{unit cell}$) derived from the measured polarized neutron flipping ratios of the Bragg reflections and their calculated values from the best least-squares fit (model II, table 2)

h	k	l	$(\sin \theta)/\lambda$	$F_M(\text{obs})$	$F_M(\text{calc})$
2	0	0	0.119	-0.109(4)	-0.106
4	0	0	0.239	11.7(2)	11.973
6	0	0	0.358	4.16(5)	3.950
8	0	0	0.478	4.17(8)	4.578
12	0	0	0.717	0.41(5)	0.456
3	1	0	0.189	0.92(3)	0.774
5	1	0	0.304	1.75(2)	1.614
7	1	0	0.422	-3.51(3)	-3.596
9	1	0	0.541	0.58(3)	0.586
4	2	0	0.267	-9.2(2)	-9.297
6	2	0	0.378	1.55(2)	1.601
8	2	0	0.492	-1.21(2)	-1.235
12	2	0	0.726	-0.22(5)	-0.224
5	3	0	0.348	4.24(7)	4.251
7	3	0	0.455	-1.36(3)	-1.314
9	3	0	0.567	1.16(3)	1.214
4	4	0	0.338	0.53(4)	0.854
6	4	0	0.431	-1.31(2)	-1.240
8	4	0	0.534	1.13(3)	1.184
10	4	0	0.643	-0.68(3)	-0.564
5	5	0	0.422	3.4(1)	3.316
7	5	0	0.514	-0.55(8)	-0.606
9	5	0	0.615	1.00(4)	0.945
11	5	0	0.722	0.46(7)	0.274
6	6	0	0.507	2.40(5)	2.270
8	6	0	0.597	0.1(3)	0.233
10	6	0	0.696	0.33(5)	0.272
7	7	0	0.591	-1.8(1)	-1.629
8	8	0	0.676	0.83(6)	0.655

The main contribution to B_{hf} , the Fermi contact term arising from the polarization of the 1s, 2s and 3s core states by the 3d electrons, is proportional to the 3d magnetic moment of the central atom ($-11.3 \text{ T}/\mu_B$ according to calculations from Coehoorn *et al* 1991). However, the contribution due to orbital moments and the Fermi contact term of the 4s valence electrons are also significant (Coehoorn *et al* 1991). These contributions are partly determined by interactions with the nearest-neighbour atoms: (i) changes of the total 4s electron density depending on the relative importance of s bonding or charge delocalization and (ii) polarization of the 4s band by the magnetic moments on neighbouring atoms, often called the 'transferred hyperfine-field' contribution. Differences in the isomer shifts arising from a variation of electron densities at the Fe nuclei at the three crystallographic sites of $UFe_{10}Si_2$ suggest that at least the 4s-electron contributions to the contact term should

not be the same. If the differences in these contributions are significant, it would not be surprising that the B_{hf}/μ_{Fe} ratio is not constant even for Fe atoms in the same compound but at different crystallographic sites, as already reported (e.g. Coehoorn *et al* 1991) for other Fe-containing intermetallics.

4. Conclusion

The polarized neutron data show that the actinide atom has a small magnetic moment not larger than $0.3 \mu_B$ ferromagnetically coupled to the Fe local moments of $1.67(3) \mu_B$ (8f), $1.73(4) \mu_B$ (8j) and $2.30(3) \mu_B$ (8i). The value of B_{hf} estimated from the Mössbauer data also increases in the order $8f < 8j < 8i$ but the ratios B_{hf}/μ_{Fe} deduced for each site are significantly different. Although the valence contribution to B_{hf} may scale with the core contribution, giving in some cases a linear relationship between B_{hf} and μ_{Fe} , as was found for the binary Y–Fe compounds (Sinnemann *et al* 1989, Denissen *et al* 1990), our results show that in general, as far as the ^{57}Fe Mössbauer effect is concerned, only qualitative information about μ_{Fe} may be expected from the values of B_{hf} .

Although the best estimate for the U magnetic moment gives a rather small value ($0.26(7) \mu_B$), the occurrence of a type-I FOMP and the analysis of the magnetization curve along the hard axis (Estrela *et al* 1995) suggest that there is an important exchange interaction between the U and Fe atoms. Our results support the picture in which the low value of the U moment results from a partial cancellation of the orbital and spin contributions, which are antiparallel and of similar magnitude ($\sim 1 \mu_B$). A similar situation was observed for UFe_2 (Wulff *et al* 1989) for which an almost exact cancellation of the orbital and spin moments was found from measurements made of the polarized neutron diffraction. It is interesting to note that the strong hybridization between the f and d electrons significantly lowers the T_C -value of UFe_2 (165 K) compared to that of the rare-earth analogues (~ 600 K). Contrasting behaviour is found for $\text{UFe}_{10}\text{Si}_2$, which has a Curie temperature higher than $\text{YFe}_{10}\text{Si}_2$ ($T_C = 540$ K) and any of the isostructural rare-earth compounds (the highest value is $T_C = 610$ K for $\text{GdFe}_{10}\text{Si}_2$). It is also worth mentioning that we have recently examined, using neutrons, the isostructural compound UFe_4Al_8 (Paixão *et al* 1997), a non-collinear ferrimagnet where the actinide and the transition metal sublattices order simultaneously at 145 K. The ordered moment of the uranium atom at 4 K is $0.47(2) \mu_B$, comparable with the value obtained in this work for $\text{UFe}_{10}\text{Si}_2$. However, the ratio of the orbital and spin moments of the uranium atom measured using polarized neutrons for UFe_4Al_8 is $-\mu_L/\mu_S = 1.9(1)$, indicating a smaller f–d hybridization than those observed for $\text{UFe}_{10}\text{Si}_2$ and UFe_2 .

Acknowledgments

This work is part of a project financed by JNICT under contract Praxis/3/3.1/FIS/29/94. J A Paixão and A P Gonçalves acknowledge the support provided under the Large Installation Programme of the EU to allow the performance of the neutron scattering experiments at Siloé and Saclay.

References

- Andreev A V, Bartashevich M I, Aruga Katori H and Goto T 1994 *J. Alloys Compounds* **216** 221
- Andreev A V, Suski W and Baranov N V 1992a *J. Alloys Compounds* **187** 293
- Andreev A V, Suski W, Vagizov F G and Drulis H 1993 *Physica B* **186–188** 730
- Andreev A V, Vagizov F G, Suski W and Drulis H 1992b *J. Alloys Compounds* **187** 401

- Becker P J and Coppens P 1974 *Acta Crystallogr. A* **30** 129
- Berlureau T, Chevalier B, Fournes L and Etourneau J 1989 *Mater. Lett.* **9** 21
- Brown P J and Matthewman J C 1987 The Cambridge Crystallography Subroutine Library—Mark 4 users' manual
Rutherford Appleton Laboratory Internal Report RAL-93-009
- Coehoorn R, Denissen C J M and Eppenga R 1991 *J. Appl. Phys.* **69** 6222
- Denissen C J M, Coehoorn R and Buschow K H J 1990 *J. Magn. Magn. Mater.* **69** 6222
- Desclaux J P and Freeman A J 1978 *J. Magn. Magn. Mater.* **8** 119
- Engelen P P J and Buschow K H J 1990 *J. Magn. Magn. Mater.* **84** 47
- Estrela P, Godinho M, Gonçalves A P, Almeida M and Spirlet J C 1995 *J. Alloys Compounds* **230** 35
- Gonçalves A P, Almeida M, Walker C T, Ray J and Spirlet J C 1994 *Mater. Lett.* **19** 13
- Lin C, Li G Z, Liu Z X, Jiang H W, Wan Z, Yang J L, Zhang B S and Ding Y F 1991 *J. Appl. Phys.* **70** 6543
- Marshall M and Lovesey S W 1971 *Theory of Thermal Neutron Scattering* (Oxford: Clarendon)
- Matar S F, Chevalier B and Etourneau J 1994 *J. Magn. Magn. Mater.* **137** 293
- Paixão J A, Lander G H, Brown P J, Nakotte H, de Boer F R and Brück E 1992 *J. Phys.: Condens. Matter* **4** 829
- Paixão J A, Lebech B, Gonçalves A P, Brown P J, Lander G H, Burllet P, Delapalme A and Spirlet J C 1997 *Phys. Rev. B* **52** 14370
- Shull C G and Yamada Y 1962 *J. Phys. Soc. Japan* **17** 1
- Sinnemann Th, Erdmann K, Rosenberg M and Buschow K H J 1989 *Hyperfine Interact.* **50** 675
- Suski W, Baran A and Mydlarz T 1989 *Phys. Lett.* **136A** 89
- Waerenborgh J C, Rogalski M S, Gonçalves A P, Sousa J B and Almeida M 1997 *Solid State Commun.* **104** 271
- Wulff M, Lander G H, Lebech B and Delapalme A 1989 *Phys. Rev. B* **39** 4719