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URANIUM FORM FACTORS IN SELECTED UTX COMPOUNDS*

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We present a study of uranium magnetic form factors in the UCoAl, UPtAl, UNiGa and UNiAl intermetallic compounds, all crystallizing in the hexagonal ZrNiAl-type structure. Our study is based on polarized neutron diffraction experiments. The $-\mu_L/\mu_S$ ratio determined from our data is reduced compared to the U^{3+} free-ion value for all the studied compounds, indicating delocalization of the 5*f*-electron states.

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1. Introduction

Existence and size of the uranium orbital moment in intermetallic compounds, in which the 5*f*-electron states are rather itinerant, are principal issues of the electron structure and electronic properties of these materials. Orbital moment is a vital ingredient in the mechanism responsible for a huge magnetic anisotropy observed even in very weak itinerant ferromagnets like UNi_2 or antiferromagnets, *e.g.* UNiAl. Delocalization of the 5*f*-electron states presumably involves reduction of the $-\mu_L/\mu_S$ ratio from the value expected for the U^{3+} free ion. Closer inspection of U-moment studies on

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different materials reveals that the reduction of the orbital moment is mainly involved in this effect.

We have studied the uranium magnetic form factors in the UPtAl, UCoAl, UNiGa and UNiAl compounds, all crystallizing in the hexagonal ZrNiAl-type structure. All these compounds, irrespective of the ground state which is ferromagnetic (UPtAl), antiferromagnetic (UNiAl, UNiGa) or paramagnetic (UCoAl), exhibit strong uniaxial magnetocrystalline anisotropy with magnetic moments aligned along the hexagonal c axis in the ordered state [1]. Our measurements have been performed in magnetic fields high enough to achieve ferromagnetic ordering in the studied materials, except for UNiAl. The maximum field was too small to induce a transition into the ferromagnetic state in UNiAl. Using the method of flipping ratios, only ferromagnetic component of the moments are measured.

2. Experimental

The polarized neutron diffraction experiments have been performed on the 5C1 diffractometer in LLB Saclay (UNiGa, UNiAl) and the D3 diffractometer in ILL Grenoble (UCoAl, UPtAl) at the wavelength of 0.85 Å, in the case of UCoAl also at 0.51 Å. The flipping ratios have been measured in magnetic field of 4 T (UNiGa and UNiAl), 8 T (UCoAl) and 9.6 T (UPtAl) at following temperatures: 10 K (UNiGa), 23 K (UNiAl), 2 K (UCoAl and UPtAl). Samples with a shape of a flat plate (typically $5 \times 3 \times 0.8$ mm) have been used. To refine the crystal structure parameters and the extinction correction, diffraction experiments on the D15 and D10 diffractometers in ILL have been performed.

3. Results and discussion

The measured flipping ratios R (for a noncentrosymmetric structure, R is given by a quite complex expression, see *e.g.* [2]) have been analyzed by two different approaches. In the first one, we reconstruct the magnetization density maps using the maximum entropy method, which makes no assumption on the magnetization distribution within the unit cell. Then we fit the magnetic structure factors calculated from these maps to an atomic model, in which the magnetic moments described by a given magnetic form factor are located on atomic sites. The magnetic structure factor is then given as:

$$F_{\mathrm{M}}(Q) = \sum_{\mathrm{at}} G_{\mathrm{at}}(Q) \mathrm{e}^{-W_{\mathrm{at}}} \mu_{\mathrm{at}} f_{\mathrm{at}}(Q)\,. \tag{1}$$

The summation goes over all atoms which carry a magnetic moment μ described by a form factor f. Q is the scattering vector, W is the Debye–Waller factor and G is given as

$$G(Q) = \sum_j \mathrm{e}^{iQr_j}\,, \tag{2}$$

where we sum over all equivalent positions of a given atom. The magnetic form factor of uranium has been taken as

$$\mu_\mathrm{U} f_\mathrm{U} = \mu_\mathrm{S}\langle j_0\rangle + \mu_\mathrm{L}(\langle j_0\rangle + \langle j_2\rangle)\,, \tag{3}$$

where $\langle j_n\rangle$ are radial integrals tabulated for individual ions. As an example, the form-factor curve of UNiGa is shown in Fig 1.

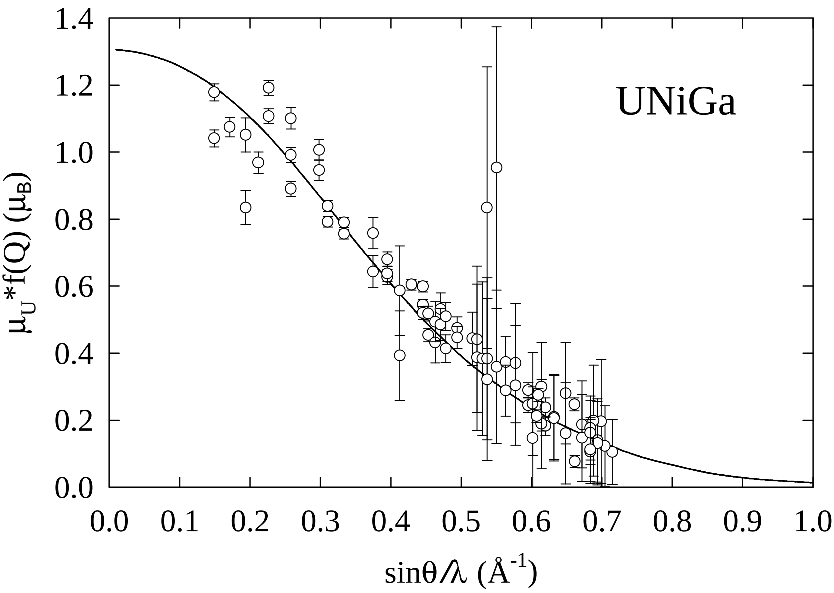

Fig. 1. Uranium form-factor curve of UNiGa; the points represent values calculated from the reconstructed density map after subtraction of all non-uranium contributions, the line represents the form factor calculated by equation (1) using the parameters given in Table I.

The other approach represents the direct refinement of the measured flipping ratios, independently on the maximum entropy results. We assume the atomic model as in the previous treatment.

In all the studied compounds, the main magnetic contribution comes from the uranium atoms. Additionally, we observe small induced magnetic moments on the transition metal atoms. These moments in relation to magnetocrystalline anisotropy are discussed in separate papers (see *e.g.* [2]). The resulting uranium orbital and spin magnetic moments are summarized in Table I. The results obtained by the two different methods are generally in a good agreement. Data given here have been obtained assuming $\langle j_n \rangle$ functions of the U^{3+} ion. Assuming U^{4+}, we have obtained the same values of the total moments and the same agreement with the experimental data, and cannot thus make any conclusions about the uranium valence.

TABLE I

The uranium orbital and spin moments in the studied compounds.

compound		μ_{total}(U) (μ_B/atom)	μ_L (μ_B/atom)	$-\mu_S$ (μ_B/atom)	$-\mu_L/\mu_S$
UNiGa	a	1.26(4)	2.73(4)	1.47(5)	1.86(7)
	b	1.30(4)	2.63(4)	1.33(5)	1.98(9)
UNiAl	a	0.136(4)	0.29(1)	0.15(1)	1.93(10)
	b	0.128(4)	0.29(1)	0.16(1)	1.81(10)
UCoAl	a	0.428(3)	0.73(1)	0.30(1)	2.43(5)
	b	0.412(5)	0.73(1)	0.32(1)	2.28(6)
UPtAl	a	1.18(5)	2.54(3)	1.36(6)	1.86(5)
	b	1.16(6)	2.73(4)	1.57(6)	1.74(6)

a — fit of equation (1) to maximum entropy results
b — refinement of flipping ratios

The ratio $-\mu_L/\mu_S$ determined by our analysis is in all the cases reduced compared to the U^{3+} free-ion value of 2.57. This result indicates delocalization of the uranium $5f$-electron states in these compounds. The reduction is similar to that reported for isostructural URhAl [3]. Surprisingly, the smallest reduction is found for UCoAl, which is considered as one of the UTX compounds with rather delocalized $5f$ electrons [1]. We shall note, however, that both μ_L and μ_S are in this case strongly reduced with respect to free-ion values.

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