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LETTER TO THE EDITOR

Polarised neutron study of the magnetic ordering in the simple alloy YFe_2

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Abstract. The magnetic properties of YFe_2 were investigated using the polarised neutron technique. Flipping ratios were measured for the first five reflections of a powder. The magnetic intensity on the [220] reflection proved that the yttrium site carries a magnetic moment antiparallel to that of the iron site. The results were discussed with regard to different magnetic form factor curves to be used for yttrium. The best fit to the experimental data was achieved when assuming 80% spin and 20% orbital contribution, values of $\mu_Y = (0.67 \pm 0.04)\mu_B$ and $\mu_{Fe} = (1.77 \pm 0.08)\mu_B$ were found.

In order to develop a better understanding of the magnetic properties of alloy systems containing a rare earth and a transition metal, numerous studies have been undertaken of pseudo-binary compounds of the type $A(Fe_{1-x}B_x)$ with A ≡ Y, Zr, Gd, Ho and B ≡ Mn, Co, Al [1–3]. All of these crystallise in the cubic $MgCu_2$-structure, it being possible for the Fe atoms at the 16d positions to be partly substituted by Mn and Co without changing the crystallographic structure and therefore allowing the study of the 3d–3d interaction. Local environmental effects were studied by diluting the transition metal by the non-magnetic Al [4]. The influence of the rare earth atoms upon the magnetism of the 3d electrons of Fe was investigated in the series $A(Fe_{1-x}Al_x)_2$ with A ≡ Y, Gd, Dy, Ho [5]. In all cases the interpretation of the results were based on the assumption that the rare earth atoms in these Laves phases do not carry a magnetic moment. Susceptibility measurements of the binary compound YFe_2 have given a total magnetic moment of $\mu_{YFe_2} = 2.9\mu_B$. Noting the surprisingly low value of $\mu_{Fe} = 1.45\mu_B$ it was interpreted as arising solely from the Fe atoms [3].

Band structure calculations lead, for the first time, to the idea that the picture of a simple ferromagnetic arrangement of the Fe atoms in YFe_2 has to be replaced by a ferrimagnetic arrangement with the Y atoms carrying a moment antiparallel to that of the Fe site [6]. Further evidence for this idea was provided by experiments measuring the pressure dependence of the magnetisation and the pressure dependence of the hyperfine field as seen by nuclear magnetic resonance (NMR) [7].

However, as clear experimental proof is still lacking it was decided to search directly for any magnetic moment on the Y site by using polarised neutrons.

The experimental procedure will now be described. The sample of YFe_2 was prepared by arc-melting the stoichiometric amounts of the constituent metals with a small surplus of yttrium under argon atmosphere and annealing for one week at 840 °C. The ingot was

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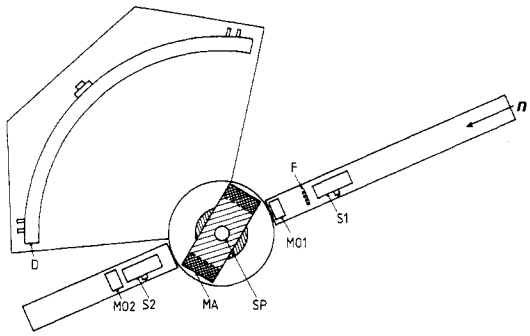

Figure 1. Plan view of the D1B powder diffractometer in polarised neutron mode. Key: S1, polariser; S2, analyser; F = π-coil flipper; MO1, and MO2, monitors; MA, electromagnet; SP, sample position; D, multidetector spanning 80° in 2θ.

powdered and then annealed for a further week at 840 °C. No weight loss occurred during the preparation.

The powder diffractomer D1B at the Institut Laue–Langevin (ILL) in Grenoble was used to measure the flipping ratios of five reflections in the range $20° < 2\theta < 100°$. Because of its high neutron flux at $\lambda = 2.52$ Å, this machine with its large multidetector spanning 80° in 2θ is normally used to search for magnetic peaks of very low intensity, for surface structure studies or for real-time kinetic experiments. For this experiment the arrangement of the apparatus was changed as depicted schematically in figure 1. A supermirror (S1) consisting of numerous layers of Co and Fe evaporated onto the glass substrate (covered with Gd and Ti layers to avoid reflection of the wrong spin state) was used as polariser [8]. Maximum intensity of polarised neutrons was found by translating and rotating the mirror relative to the incoming neutron beam. A partial loss of intensity had to be accepted as the neutron beam arriving from the three focusing monochromators in the primary beam has a height of about 8 cm at the supermirror position, whose height is for stability reasons limited to 5 cm. The final flux of polarised neutrons was of order 8×10^5 neutrons $cm^{-2}\,s^{-1}$. A π-coil (F) flipper was placed behind the mirror, the strength of the flipping and guide fields being optimised by using the 1 1 1 reflection of a Heusler-alloy single crystal. In order to determine the values of the initial polarisation P_i, and of the depolarisation D occurring as the beam passes through the magnetically imperfectly aligned sample and of the efficiency E of the flipper, several measurements were made in the direct beam. A second supermirror (S2) was placed behind the sample followed by a second monitor (MO2 in figure 1). Knowing the transmission of the sample and of the soft iron which was used to depolarise the beam, the absolute counting rates with the incident beam successively spin up, spin down, and depolarised (each measurement with and without the sample in the beam) allowed the exact evaluation of $P_i = 0.97$, $D = 0.25$ and $E = 0.99$. The sample of finely powdered YFe_2 was placed inside a cryostat with a small tail entering through the open pole pieces of an ordinary electromagnet (MA) producing a field of 1.1 T. Changing the spin state every 30 minutes the sample diffraction patterns were measured at 2 K for 6 hours in spin up and for 6 hours in spin down configuration.

Figures 2(*a*)–(*c*) show the spectra obtained for the spin-up and the spin-down state as well as the resulting difference spectrum $I^{\uparrow} - I^{\downarrow}$. The fact that the positions of the Bragg peaks are slightly shifted relative to the true position arises from the fact that the

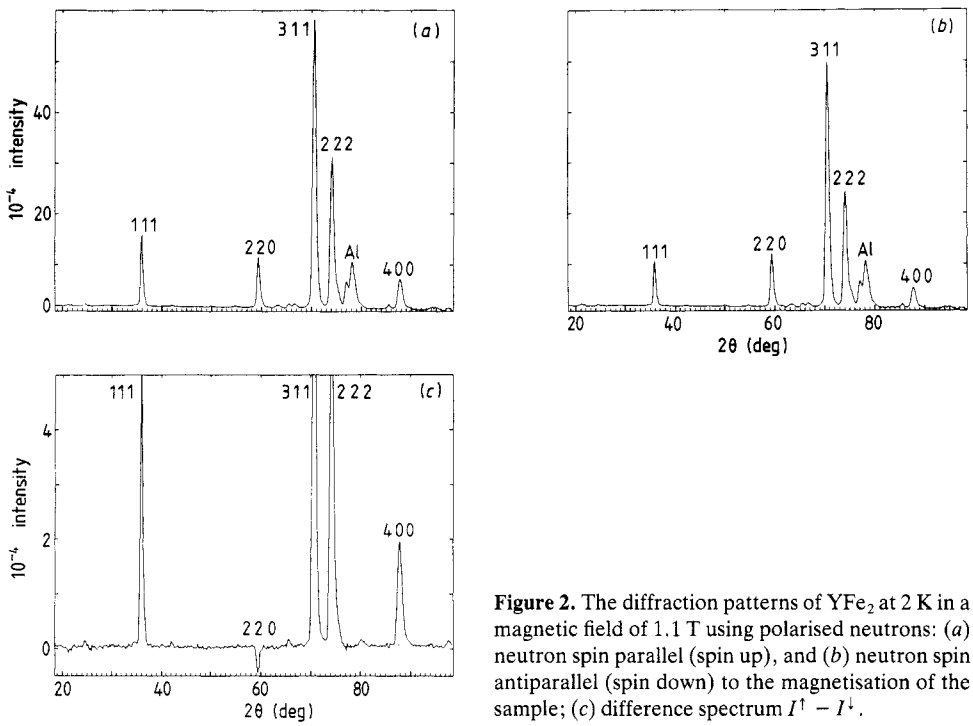

Figure 2. The diffraction patterns of YFe_2 at 2 K in a magnetic field of 1.1 T using polarised neutrons: (*a*) neutron spin parallel (spin up), and (*b*) neutron spin antiparallel (spin down) to the magnetisation of the sample; (*c*) difference spectrum $I^{\uparrow} - I^{\downarrow}$.

curvature of the supermirror bends the neutron beam away from the ideal sample position. An additional measurement with the normal set-up (unpolarised neutrons) was made to confirm the lattice constant of YFe_2, $a = 7.35$ Å.

Using the ABFFIT [9] program developed at the ILL, the five Bragg peaks of YFe_2 were integrated for the spin-up and spin-down measurements and from these the flipping ratio $R = I^{\uparrow}/I^{\downarrow}$ of each reflection was obtained. This ratio is given by

$$R = (N^2 + M^2 + 2P_iDNM)/(N^2 + M^2 - 2P_iDeNM) \tag{1}$$

where N and M stand for the nuclear and the magnetic structure factors respectively, $e = 2E - 1$ and P_i and D are as defined earlier. With $\gamma = N/M$, this transforms to [10]:

$$\gamma = \frac{1}{R-1}\{P_iD(Re+1) \pm [(P_iD)^2(Re+1)^2 - (R-1)^2]^{1/2}\}. \tag{2}$$

Table 1 shows the values of R determined from the integration for every reflection together with the corresponding values of γ calculated from equation 2. Using the atom positions as published in [11] for YFe_2 and the coherent scattering lengths b_{Fe} and b_Y as published in [12], the nuclear structure factors are known and the magnetic structure factors M_{exp} (table 1, column 5) were obtained from $M_{exp} = N/\gamma$. Knowing the nuclear and magnetic structure factors, the sign of the magnetic moments relative to the positive z direction of the applied field can be easily determined from

$$I^{\uparrow} - I^{\downarrow} = K\,2P_iD(1+e)NM$$

where K is a scale factor including the multiplicity of the reflection as well as geometrical factors of the experimental set up. For the 111 reflection, $I^{\uparrow} - I^{\downarrow}$ is positive (figure

Table 1. Comparison between the magnetic structure factors M_{exp} derived from the measured flipping ratios $R = I^{\uparrow}/I^{\downarrow}$ and M_{cal} calculated for the ferrimagnetic model with $\mu_{Fe} = 1.77\mu_B$ and $\mu_Y = 0.67\mu_B$.

hkl	R	γ	Nuclear structure factor	M_{exp}	M_{cal}
1 1 1	1.61 ± 0.01	1.16 ± 0.1	$8b_{Fe}-4\sqrt{2}b_Y$	3.71 ± 0.4	3.84
2 2 0	1.049 ± 0.003	0.051 ± 0.003	$-8b_Y$	0.312 ± 0.02	0.312
3 1 1	1.189 ± 0.005	0.188 ± 0.005	$-8b_{Fe}4\sqrt{2}b_Y$	2.24 ± 0.06	2.18
2 2 2	1.28 ± 0.02	0.27 ± 0.011	$16b_{Fe}$	4.21 ± 0.35	4.42
4 0 0	1.42 ± 0.02	0.436 ± 0.025	$16b_{Fe}-8b_Y$	3.89 ± 0.23	3.84

2(*c*)), and as N is positive (table 1, column 4), M has to be positive as well. For the 2 2 0 reflection, which is exclusively determined by the Y-site, $Y^{\uparrow} - I^{\downarrow}$ is negative, $N = -8b_Y$ and $M = -8\mu_Y f \times 0.27$ where f is the magnetic form factor of Y. It follows that the magnetic moment μ_F has to be antiparallel to the positive z direction. The other reflections were analysed accordingly.

The most important consequence of the values of M thus determined is that it can now be definitively stated that a significant magnetic moment exists on the Y site, oriented antiparallel to the magnetic spin of the Fe site.

Trying to interpret the data quantitatively, one is confronted with the problem of choosing the magnetic form factor curves to be used from iron and yttrium. It seems reasonable to use the values as published for pure iron in [13], or the nearly identical ones found in [14] for $LuFe_2$ to account for the iron magnetic moment. The only measured form factor curve for yttrium is the one of metallic paramagnetic Y, for which an atypical fast drop of the curve at low sin θ/λ values followed by an increase was found; this appears to be a characteristic of metals crystallising in a HCP structure with three electrons in s and d states [15, 16]. It would therefore be inaccurate to use this value for YFe_2.

The free-atom form factors as calculated in [17], using Hartree–Fock wave functions, were fitted using analytical approximations in [18]. The coefficients necessary to evaluate the magnetic form factors $\langle J_L \rangle$ as a function of sin θ/λ are available in tabulated form. From the 2 2 0 reflection, and using form factors assuming firstly 100% spin and secondly 100% orbital contribution, the magnetic moment of yttrium was determined as lying in the range $(0.4–0.8)\mu_B$. Fitting the complete data set one finds, however, that models involving less than 50% spin contribution fail. The best fit was achieved when assuming 80% spin and 20% orbital contribution. Table 1 shows the calculated data in comparison to the measured ones. The values for the magnetic moments were $\mu_{Fe} = (1.77 \pm 0.08)\mu_B$ and $\mu_Y = (0.67 \pm 0.04)\mu_B$. The errors reflect only the experimental ones. The value of μ_Y depends largely, of course, on the chosen form factor curve.

In view of the limited data set and the experimental errors involved, a proper determination of the form factor curve of yttrium in YFe_2 is unjustified. However, a form factor curve having a minimum at sin $\theta/\lambda \simeq 0.225$ followed by a recovery would describe the experimental data better than the conventional form factor curve used in the present analysis.

A possible sublattice disorder caused by a non-stoichiometric sample, where some of the Y atoms are replaced by Fe, will not alter the main findings. Firstly, the nuclear

intensities show that the sample is stoichiometric within 1.5% and such a disorder was not observed [19] in samples prepared in the same way as the one used in the present study. Secondly, assuming the additional Fe to be distributed on the Y sites (thereby occupying 3% of these sites) with moments aligned antiparallel to the moments on the Fe sites, it is not possible to account for the observed moment of $0.67\mu_B$ on the Y sites. Therefore it can be concluded that in YFe_2, the Y atoms have a moment of $0.67\mu_B$ which is antiparallel to the moment on the Fe atoms.

As experiments on the related compounds $ZrFe_2$, $HfFe_2$ and $Y(Fe_{1-x}Co_x)_2$ are under way, the important consequences of these results relating to the magnetism in rare earth transition metal alloys will be presented in due course.

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