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Determination of the direction of the antiferromagnetic modulation below the low-temperature re-ordered regime in Fe_2MnSi

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Abstract. Neutron polarization analysis investigations have been made on an Fe_2MnSi single-crystal specimen at 36 K. Measurements carried out around the $\frac{1}{2}(111)$ and $\frac{1}{2}(113)$ peak positions have confirmed the previously assumed orthogonality between the ferromagnetic and antiferromagnetic directions below the re-ordering temperature T_R for $Fe_{3-x}Mn_xSi$ alloys.

The $Fe_{3-x}Mn_xSi$ system ($0 \leqslant x \leqslant 3$) has aroused much interest because of its magnetic phase diagram (see figure 1). When manganese atoms are added to Fe_3Si for $x < 0.7$ the magnetic phase is ferromagnetic (FM). Beyond this composition range however, i.e. $0.7 \leqslant x \leqslant 1.75$, a low-temperature FM phase with an antiferromagnetic (AFM) component is observed, confirmed by the appearance of additional superlattice reflections below the re-ordering temperature T_R. The additional low-temperature superlattice reflections have been observed for $\frac{1}{2}(hkl)$ with hkl all odd, with the $\frac{1}{2}(111)$ reflection absent (Ziebeck and Webster 1976, Yoon and Booth 1977). No neutron polarization analysis (PA) study has been carried out on $Fe_{3-x}Mn_xSi$ alloys before, but it has been established from unpolarized neutron powder diffraction data that the direction of the AFM wavevector is most probably along the [111] direction and that it is most likely to be longitudinal. No definite connection has been established between the FM and AFM directions, since neutron powder data alone cannot be used to interpret magnetic intensities in terms of unique spin directions. However, the structure has been modelled assuming they are orthogonal (Yoon and Booth 1977), i.e. the AFM component is directed along the [111] axis and the FM component is in the (111) plane. Yoon and Booth (1977) also mention that in the FM regime the nuclear and magnetic contributions could be separated by applying a magnetic field along the scattering vector (κ), but below T_R the magnetic contribution could not be extinguished in this way due to the possible canting of the magnetic moments. Miles *et al* (1991) on the other hand, for the FM regime between T_R and T_c, proposed that the spins on the Mn predominant sites were also canted at an angle to the FM direction and randomly distributed in direction. It was suggested that at T_R the canted spins lock into specific directions to give a net AFM component perpendicular to the FM axis.

In the application of a vector model to the internal fields in $Fe_{3-x}Mn_xSi$ alloys for $x > 0.75$, Niculescu *et al* (1983) used the composition dependence of the canting angle θ between the Mn moments on Mn predominant sites and the [111] axis. These θ values

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were then used to calculate the Fe and Mn fields at Fe predominant sites. The general requirements for the application of this vector model were that (1) the magnetic structure proposed by Yoon and Booth (1977) was adopted, (2) the internal fields were either parallel or antiparallel to the moments which generate them and (3) set core polarization, exchange interaction and hyperfine coupling constant values were used. For the Mn concentration $x = 0.75$, $\theta = 90°$, with all the moments being parallel and in the (111) plane. For $x = 1.0$, $\theta = 45°$. Other concentration-dependent values of θ are also given in the paper by Niculescu *et al* (1983).

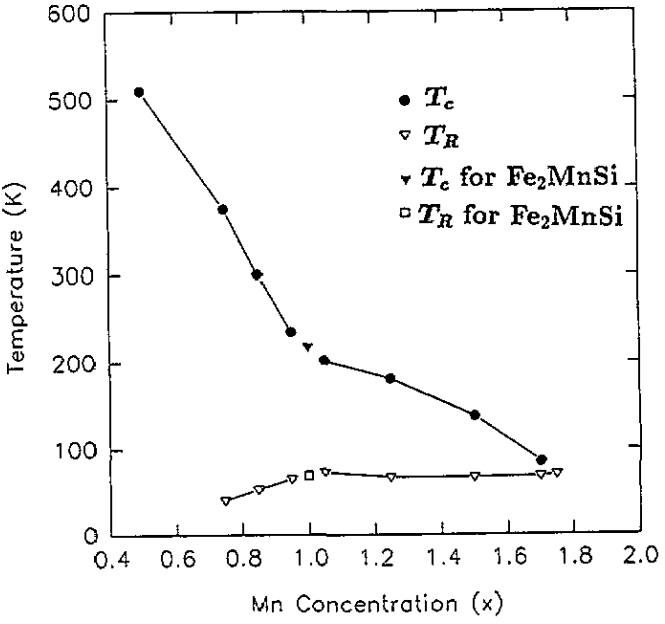

Figure 1. The magnetic phase diagram of the $Fe_{3-x}Mn_xSi$ alloy system ($0 \leqslant x \leqslant 3$), showing the variation of the Curie temperature (T_c) and the magnetic re-ordering temperature (T_R) with Mn composition (after Yoon and Booth 1977).

In order to understand the mechanism for the formation of this low-temperature phase we have undertaken polarized neutron diffuse scattering studies of an Fe_2MnSi single crystal, with a composition well inside the re-ordered phase (see figure 1). The full set of results and analysis of the diffuse scattering will be presented at a later date. Here we present the data from our single-crystal neutron PA measurements which confirm the assumed orthogonality of the magnetic structure in the re-ordered phase.

The crystal was mounted with its $[1\bar{1}0]$ axis vertical within a c-shaped permanent magnet in a vertical field of 7 kOe, so that the $\frac{1}{2}(111)$ and $\frac{1}{2}(113)$ AFM Bragg superlattice reflections were in the scattering plane. The neutron PA experiment was carried out on the LONGPOL spectrometer at the HIFAR Research Reactor (Lucas Heights, Australia). The polarization and analysis were vertical and the incident polarization could be flipped. The measurements were performed at 36 K in order to ensure that the specimen was in the re-ordered regime below T_R. For the LONGPOL spectrometer, the beam polarization used was 35% and the neutron wavelength was 3.6 Å.

The measurements were concentrated on the $\frac{1}{2}(113)$ Bragg peak from the AFM component of the low-temperature order in Fe_2MnSi. In the one-dimensional PA method

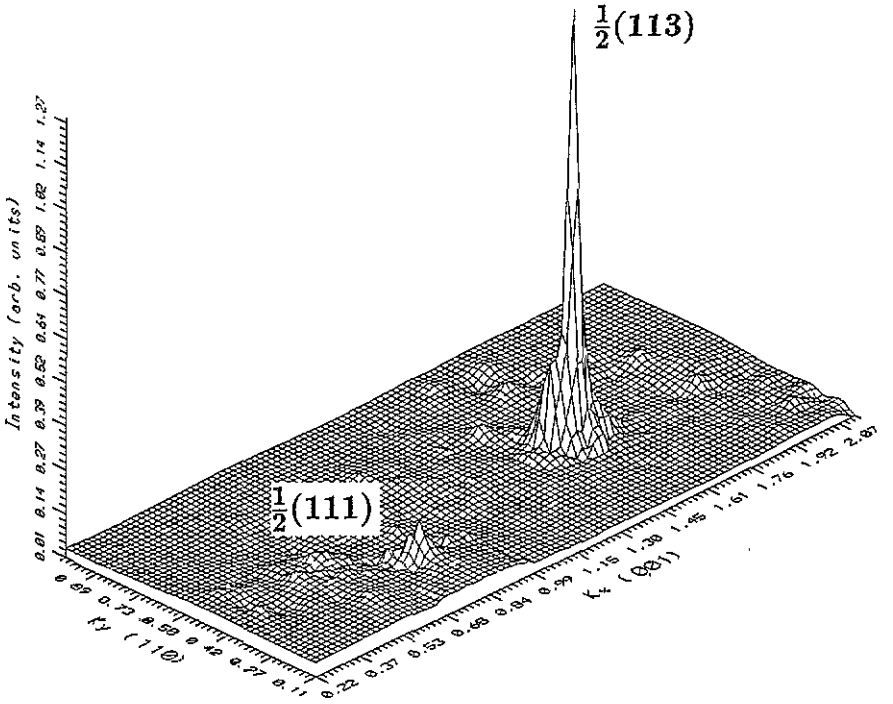

Figure 2. The SF scattering profile of the $\frac{1}{2}(113)$ AFM Bragg peak.

of Moon *et al* (1969) the PA of coherent Bragg scattering implies that the nuclear peaks will always involve non-spin-flip scattering and the magnetic peaks will involve a mixture of spin-flip (SF) and non-spin-flip (NSF) scattering, depending on the orientation of the atomic moments and the neutron polarization direction ($\boldsymbol{P}$) relative to $\boldsymbol{\kappa}$. For the geometry in which $\boldsymbol{P}$ is perpendicular to $\boldsymbol{\kappa}$ (as in our experimental set-up), i.e. $\boldsymbol{P}\cdot\boldsymbol{\kappa} = 0$, the magnetic scattering will be either SF, NSF or a mixture, depending on the direction of the atomic moments. In this method only a single component of the scattered polarization is analysed. However, the existence of other non-centrosymmetric structures also needs to be considered. Experimentally, this would involve three-dimensional PA to determine all three components of the polarization of both the incident and scattered neutrons. Such experiments have been carried out using the zero-field neutron polarimeter CRYOPAD at ILL (Tasset *et al* 1988). However, from the results of our PA measurements we are still able to confidently exclude, for instance, the occurrence of chiral structures, as will be described below.

Since the $\frac{1}{2}(113)$ Bragg peak is of entirely magnetic origin and for simplicity considering only the magnetic terms the final polarization (from Blume's (1963) general formula and for an elastically scattered beam) is given by

$$\boldsymbol{P}'\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right) = -\left(\frac{e\gamma}{\hbar c}\right)^2 [(\boldsymbol{M}_\perp^*(\boldsymbol{\kappa})\cdot\boldsymbol{M}_\perp(\boldsymbol{\kappa}))\boldsymbol{P} - (\boldsymbol{M}_\perp^*(\boldsymbol{\kappa})\cdot\boldsymbol{P})\boldsymbol{M}_\perp(\boldsymbol{\kappa}) - \boldsymbol{M}_\perp^*(\boldsymbol{\kappa})(\boldsymbol{M}_\perp(\boldsymbol{\kappa})\cdot\boldsymbol{P}) + \mathrm{i}(\boldsymbol{M}_\perp^*(\boldsymbol{\kappa})\times\boldsymbol{M}_\perp(\boldsymbol{\kappa}))] \quad (1)$$

where $\boldsymbol{P}'$ is the polarization of the scattered neutrons, $\boldsymbol{P}$ is the incident beam polarization, $\boldsymbol{M}_\perp$ is the magnetic structure factor perpendicular to $\boldsymbol{\kappa}$ of the specimen and $\mathrm{d}\sigma/\mathrm{d}\Omega$ is the specimen cross-section.

The last term in equation (1) turns the polarization towards the scattering vector. But with $M_\perp$ complex (i.e. $M_\perp = a + ib$) and considering the incident polarization to be in the plane perpendicular to κ, the final polarization, according to Nunez *et al* (1992), is at an angle ϵ_κ to κ with

$$\cos\epsilon_\kappa = 2\kappa \cdot (a \times b)/(a^2 + b^2). \tag{2}$$

In our one-dimensional polarization and analysis experiment components of the scattered neutron spin perpendicular to the ferromagnetic direction would precess about that direction and be indistinguishable from a partially depolarized beam. However, the neutron spins scattered from the $\frac{1}{2}(113)$ reflection are completely reversed, as observed in the SF and NSF scattering, obtained from the low-temperature neutron polarization analysis measurements on the Fe_2MnSi crystal shown in figures 2 and 3. The predominant peak is clearly the $\frac{1}{2}(113)$ and some diffuse intensity can be seen near the $\frac{1}{2}(111)$ position. The results indicate that the scattered beam is fully polarized along a direction opposite to the initial polarization. There is no additional component of the $\frac{1}{2}(113)$ scattering. If there were a chiral component contributing to the $\frac{1}{2}(113)$ and similar peaks the polarization would not be *completely* reversed. Thus the last term in equation (1) is zero in this case.

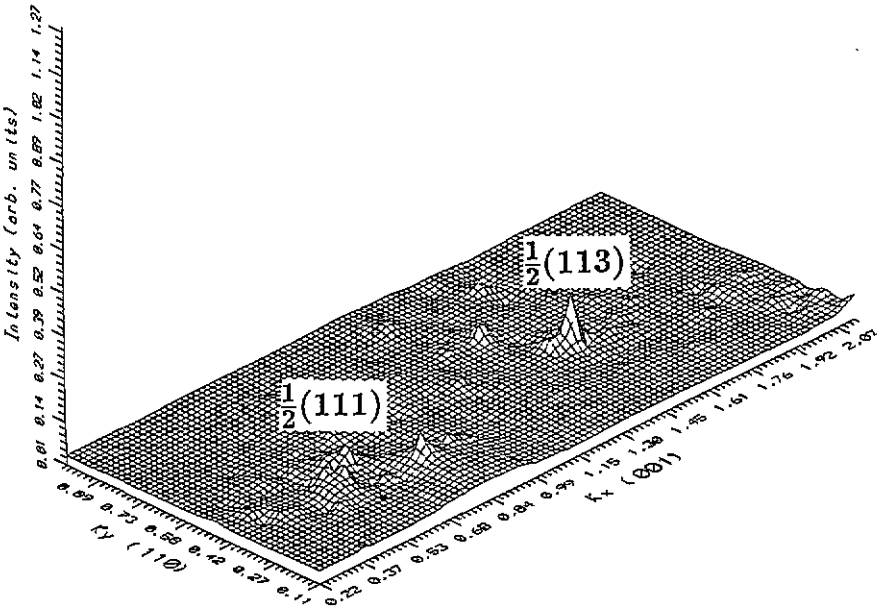

Figure 3. The NSF scattering profile of the $\frac{1}{2}(113)$ AFM Bragg peak.

Two conditions arise for AFM Bragg peaks which are entirely of magnetic character.

(i) Contributions arise only from components of the magnetic moment which are perpendicular to κ, and from the terms remaining in equation (1).

(ii) Neutron spin-flip scattering occurs only from components of the moments perpendicular to P.

From condition (i) above we can postulate that the absence of a $\frac{1}{2}(111)$ peak means either that the $\frac{1}{2}(111)$ in the $(1\bar{1}0)$ plane is not the direction of the AFM modulation or that, if it is, the modulation must be longitudinal, i.e. there are no components of the moment

which are perpendicular to [111]. It can be reasonably assumed that $\frac{1}{2}(111)$ is the AFM modulation magnitude and direction as all the AFM peaks in neutron powder patterns can be indexed at positions which are nuclear fundamental reciprocal lattice vectors $\pm\frac{1}{2}(111)$.

The fact that nearly all of the scattering is with SF (figures 2 and 3) means that all the components with moment modulation are perpendicular to $\boldsymbol{P}$ (condition (ii)). The direction of $\boldsymbol{P}$ being vertical, along the FM direction, implies that all the components of the AFM modulation must be perpendicular to the FM direction. This further indicates that the AFM modulation, in this experimental set-up, is only along those [111] directions which are perpendicular to the FM direction.

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