Neutron Diffraction Studies of Chromium-Modified Mn_2Sb

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Existence of a ferrimagnetic to antiferromagnetic phase transition with decreasing temperature in Mn_2Sb modified with 2.3 weight percent chromium is confirmed by neutron diffraction.

IN an earlier note, Swoboda *et al.*[1] reported a transition in chromium-modified Mn_2Sb from a weak magnetic to a strong magnetic state with increasing temperature. It was suggested that the weak magnetic state is actually antiferromagnetic and that the weak magnetism arises from an MnSb precipitate. Here we report on neutron diffraction studies of this material, which confirm completely that the weak magnetic region is antiferromagnetic.

Measurements were made on a cylindrical single crystal of $Mn_{1.9}Cr_{0.1}Sb_{0.95}In_{0.05}$ with its tetragonal axis as the cylinder axis. The temperature dependence of the magnetization of this crystal is shown in Fig. 1, Curve *A*. The range of temperature over which the exchange inversion occurs (0° to 40°C) is broader than that described for the examples in reference 1. One possible explanation for this effect is that the chromium concentration is not uniform throughout the large crystal used for the neutron diffraction. A variation of only ±5% could account for the observed breadth.

Measurement of the (00*l*), (*h*00), and (*hh*0) reflections were made over the temperature range −60° to +60°C. At +60°C, neutron diffraction intensities are in agreement with published values for Mn_2Sb,[2] which has the ferrimagnetic structure shown in Fig. 2(a). At −60°C, strong (00*l*) reflections are observed for a magnetic unit cell having twice the *c* axis of the x-ray cell. Data for the (001), (00$\frac{3}{2}$), (002), (00$\frac{5}{2}$), and (110) reflections, where indexing is based on the x-ray cell, are shown in Table I. Appearance of the (00$\frac{3}{2}$) and (00$\frac{5}{2}$) reflections at −60°C and the fact that the (001) and (002) reflections are the same intensity at −60°C as at +60°C indicate that the low-temperature phase is antiferromagnetic. The (*h*00) reflections are quite weak, and no reflections for odd *h* are observed. Thus it appears that the ferromagnetic alignment within the Mn_I and Mn_{II} layers is the same as in Mn_2Sb, but that the Mn_{II}−Mn_{II} interlayer orientation is reversed in the antiferromagnetic structure. One or both of the Mn sites may contain chromium, but at present we have not identified the chromium position.

An antiferromagnetic structure compatible with the −60°C neutron diffraction data is shown in Fig. 2(b). If the moments in the two Mn_{II} layers adjacent to the

TABLE I. Neutron diffraction intensities (arbitrary units).

	A. $Mn_{1.9}Cr_{0.1}Sb_{0.95}In_{0.05}$[a]		*B.* Mn_2Sb[b]	
(*hkl*)	+60°C No external field	−60°C Antiferromagnetic	Room temp. Saturating field in (110) direction	No external field
(001)	197	197	855	251
(00$\frac{3}{2}$)	0	1000	0	0
(002)	314	314	950	314
(00$\frac{5}{2}$)	0	107	0	0
(110)	560	212	305	907

[a] The (00$\frac{3}{2}$) reflection at −60°C was arbitrarily assigned an intensity of 1000.
[b] The room temperature data for Mn_2Sb were taken from reference 2 and normalized to the intensity of the (002) reflection of $Mn_{1.9}Cr_{0.1}Sb_{0.95}In_{0.05}$. This procedure does not normalize the (110) reflection because of differences in crystal geometry.

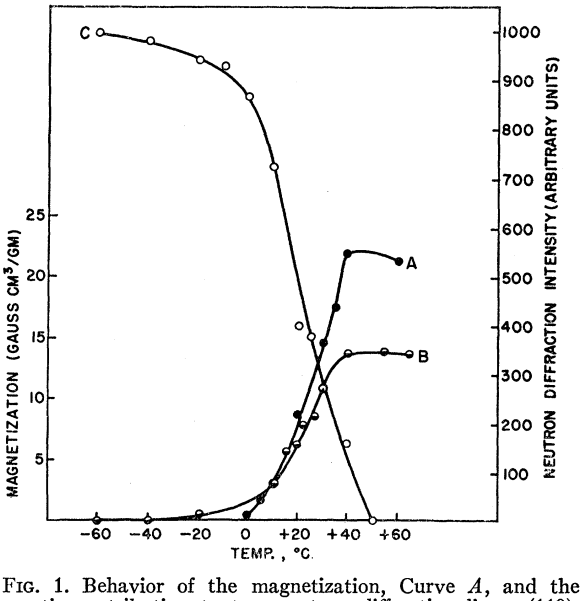

FIG. 1. Behavior of the magnetization, Curve *A*, and the magnetic contribution to two neutron diffraction lines, (110), Curve *B*, and (00$\frac{3}{2}$), Curve *C*, in the vicinity of the exchange inversion temperature.

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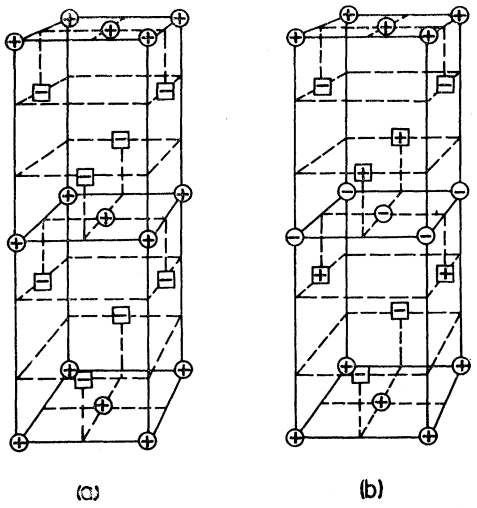

FIG. 2. (a) Magnetic structure of Mn_2Sb. Layers of Mn_I atoms (circles) are aligned antiparallel to layers of Mn_{II} atoms (squares). Above $-20°C$, the atomic moments of Mn_2Sb are parallel to the c axis. Between $-20°C$ and $-40°C$, the moments become perpendicular to the c axis (reference 2). (b) Proposed antiferromagnetic structure below the exchange inversion temperature for chromium-modified Mn_2Sb. The moments are perpendicular to the c axis.

same Mn_I layer were not antiparallel to the moments in the Mn_I layer, the $(00\frac{3}{2})$ reflection would be weaker than the $(00\frac{5}{2})$ reflection, but the $(00\frac{3}{2})$ reflection is observed to be 9 times stronger than the $(00\frac{5}{2})$ reflection (see Table I). Thus, the $+-+-+-+$ arrangement of the Mn_I and Mn_{II} layers shown in Fig. 2(b) must be chosen over a $++---++$ arrangement. Swoboda *et al.*[1] have suggested that the transition from ferrimagnetism to antiferromagnetism arises because of competition between nearly equal exchange interactions of opposite sign. We suggest that the exchange interactions in question are between the two Mn_{II} layers that do not have an Mn_I layer interposed between them. A change in sign of the net exchange between these two layers is sufficient to transform the ferromagnetic structure of Fig. 2(a) to the antiferromagnetic structure of Fig. 2(b).

Jaep[3] has pointed out that this exchange inversion transition must be first order. Kittel[4] has proposed a theory that introduces explicitly the effects of strain dependence on isotropic exchange. If the value of the lattice parameter at which an interlattice exchange interaction changes sign happens to occur within the range of normal thermal contraction, then a first order ferromagnetic-antiferromagnetic phase transition will occur. If we may adapt this theory to chromium-modified Mn_2Sb, it appears that the principal role of the chromium is to reduce the lattice dimension of Mn_2Sb just sufficiently to allow this critical dimension to occur at accessible temperatures.

It has already been established that the magnetocrystalline anisotropy in the ferrimagnetic temperature range is similar to that of Mn_2Sb.[1] The question now arises: Does the anisotropy continue to be similar to that of Mn_2Sb below the exchange inversion transition or does the anisotropy also change at the transition? This question can be answered by observation of the temperature dependence of the (110) and $(00\frac{3}{2})$ intensities.

Comparison of the (110) lines above the Curie temperature and at $-60°C$ shows that the temperature compensated intensities are the same, a result compatible with the proposed AF structure. The magnetic contribution to the (110) reflection at intermediate temperatures, obtained by subtracting off the nuclear part, and the intensity of the $(00\frac{3}{2})$ reflection, which is entirely magnetic, are shown as functions of temperature in in Fig. 1. It is apparent that the (110) intensity is proportional to the saturation moment which, in turn, is inversely proportional to the $(00\frac{3}{2})$ intensity. Because of the $\sin^2\alpha$ dependence between the scattering vector and the magnetic moment, these results indicate that the magnetic moments rotate into the basal plane as the sample is cooled through the exchange inversion transition. If the temperature dependence of the anisotropy had remained similar to that of Mn_2Sb, the moments would not rotate into the basal plane until near $-25°C$, the temperature at which the anisotropy in Mn_2Sb changes sign. Therefore, we conclude that the magnetocrystalline anisotropy in the antiferromagnetic region is negative with the moments perpendicular to the tetragonal axis even in the temperature region where the anisotropy of Mn_2Sb is positive.

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