AIP Journal of Applied Physics

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Citation: Journal of Applied Physics **39**, 461 (1968); doi: 10.1063/1.2163480
View online: http://dx.doi.org/10.1063/1.2163480
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JOURNAL OF APPLIED PHYSICS VOLUME 39, NUMBER 2 1 FEBRUARY 1968

Iron-Group Intermetallic Compounds

E. KRÉN AND J. S. KOUVEL, *Chairmen*

Neutron-Diffraction Study of $MnSn_2$*

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$MnSn_2$ undergoes an abrupt transition at ~74°K from an alternating-sheet antiferromagnetic structure $(+-+-)$ to a double-sheet structure $(++--)$. The propagation vector as well as the AF spin axis are directed along [110] in both structures. Neutron-intensity measurements on bulk samples confirm the sharpness of the transition as well as the hysteresis observed in the susceptibility and resistivity measurements of Kouvel and Hartelius. No change in the magnitude of the ordered AF moment is observed in passing through the transition. In the neighborhood of 90°K, however, weak satellite reflections appear, indicating the presence of a small modulated spin component. The extra reflections vanish abruptly at the transition and exhibit the same hysteresis shown by the main AF component. The diffraction data are consistent with a modulated spin of amplitude $\sim 0.5\ \mu_B$ oriented parallel to the AF spin axis [110] and propagated perpendicular to this direction [1$\bar{1}$0] with a wavelength of ~31 Å. This result is discussed in relation to the model of Kouvel and Jacobs.

INTRODUCTION

The intermetallic compound $MnSn_2$, crystallizes with the body-centered tetragonal $C16$ structure shown in Fig. 1. Each Mn atom is surrounded by a twisted cube of eight Sn atoms which connect it to its nearest neighbors (2.72 Å) along the c axis as well as to its second (4.65 Å) and third (5.39 Å) nearest neighbors.

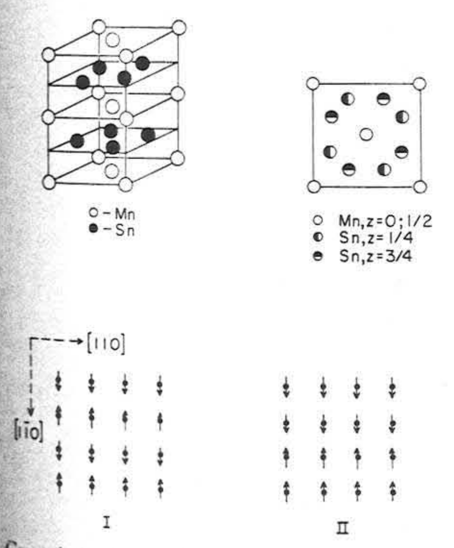

FIG. 1. Crystal structure and spin arrangement in $MnSn_2$. I and II refer to high- and low-temperature states, respectively.

As previously reported,[1] neutron-diffraction measurements show that $MnSn_2$ is antiferromagnetic below ~325°K with a simple structure consisting of ferromagnetic (110) sheets antiferromagnetically coupled to each other. The spin direction is perpendicular to the sheets. This structure is shown in Fig. 1 and labeled I. Measurements by Kouvel and Hartelius[2] of the susceptibility show a very abrupt drop at ~74°K with an accompanying hysteresis. The neutron data indicate a change in magnetic structure from an alternation of single ferromagnetic sheets to one in which pairs of

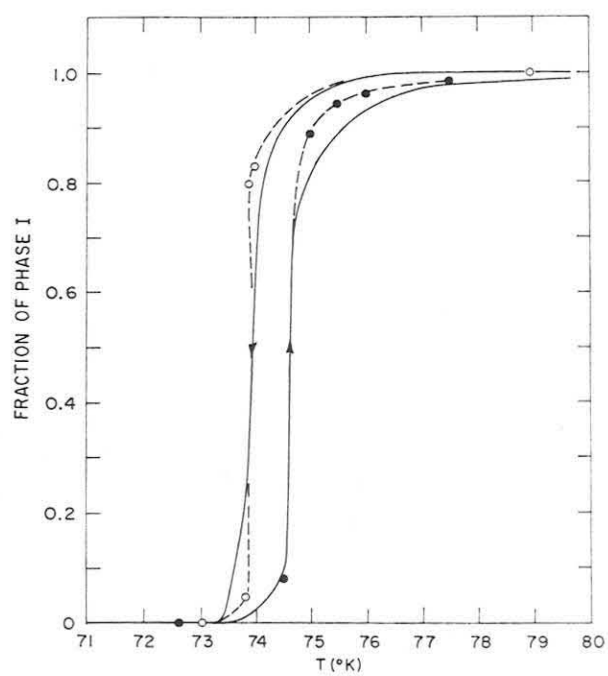

FIG. 2. Fraction, $\alpha(T)$, of state I in the region of the abrupt transition. The solid lines are obtained from susceptibility data (Ref. 2), with arrows indicating direction of temperature variation. The open (cooling) and closed (warming) circles are based on neutron intensities.

* Research performed under the auspices of the U.S. Atomic Energy Commission.

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[2] J. S. Kouvel and C. C. Hartelius, Phys. Rev. **123**, 124 (1961).

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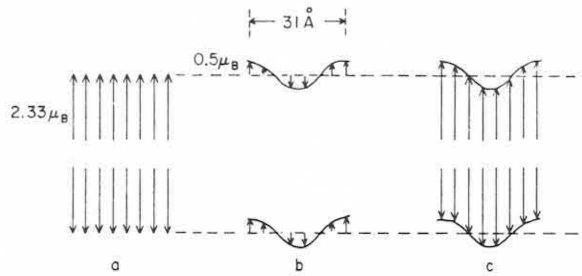

FIG. 3. Modulated antiferromagnetic moments in state I of $MnSn_2$.

ferromagnetic sheets are reversed, i.e., $+-+-$ to $++--$.[3] The structure is labeled II in Fig. 1. Both the sharpness of the transition as well as the hysteresis are confirmed by the neutron measurements. Figure 2 shows the fraction, $\alpha(T)$, of state I as a function of temperature as deduced from both the neutron and susceptibility data. The solid line is based on the magnetic measurements,[2] for which

$$\alpha(T)=(\chi_T-\chi_{II})/(\chi_I-\chi_{II}),$$

where χ_I and χ_{II} refer to the extrapolated susceptibilities of the pure states I and II at the temperature T based on the limiting slopes in the neighborhood of the transition. χ_T is the measured susceptibility at temperature T. The points, on the other hand, are from the neutron data. Based on the neutron data, the ordered Mn moment just above and just below the abrupt transition is 2.33 μ_B and remains essentially constant down to 4.2°K.

MODULATED SPIN COMPONENT

In the vicinity of 90°K, several additional weak reflections were observed which could be indexed as satellites of the (110) and (200) nuclear peaks. This assignment predicted a relatively strong (000) satellite in the direction of forward scattering. With increased resolution this low-angle reflection was indeed observed at the expected position. The satellite reflections increased in intensity as the temperature was lowered and disappeared abruptly at the 74°K transition, exhibiting the same hysteresis as the main antiferromagnetic structure.

A fit of a sinusoidal modulation to the observed positions of the satellite reflections yielded a temperature-independent wavelength of $\sim$31 Å (equivalent to $\sim$6.7 at. spacings) and a propagation vector in a [110] direction. Relative intensities of the satellite reflections indicated that the modulation was of the form $\mu=\mu_0 \cos \boldsymbol{\tau}\cdot\mathbf{r}$ with $\boldsymbol{\mu}$ perpendicular to the propagation vector $\boldsymbol{\tau}$. Normalization of the intensities to the nuclear scattering gave the amplitude of the modulation, μ_0, as approximately 0.5 μ_B.

DISCUSSION

Since the antiferromagnetic component and the modulated component are observed in independent sets of reflections, the experiment does not indicate whether the modulated spin is parallel or perpendicular to the antiferromagnetic spin axis. In the former case the results are summarized in Fig. 3. The average moment per site exhibits the spacial correlation shown in (c) of Fig. 3, where oppositely directed arrows refer to the two antiferromagnetic sublattices. The mean sublattice moment [Fig. 3(a)] appears in the antiferromagnetic Bragg reflections, while the oscillatory part [Fig. 3(b)] is observed in the satellite reflections. This picture is consistent with the model of Kouvel and Jacobs[4] if we regard the low moments in Fig. 3(c) to be produced by thermal fluctuations in magnitude. These moments can be identified with the "paramagnetic" moments of their model which are partially aligned by a weak exchange field. Application of an external magnetic field strong enough to overcome the weak exchange field would be expected, on the basis of moment values obtained from our neutron data, to produce a magnetization of approximately 1 μ_B/at., in agreement with the predictions and high-field measurements of Kouvel and Jacobs. Still higher values would of course be obtained in fields high enough to reverse the remainder of the spins.

[3] The present description of the magnetic structure of state II is formally equivalent to that given in Ref. 1. The wavelength quoted there should be divided by 2π.

[4] J. S. Kouvel and I. S. Jacobs, J. Appl. Phys. **39**, 467 (1968).

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