Abrupt Magnetic Transition in $MnSn_2$

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The magnetic susceptibility and the electrical resistivity of the intermetallic compound $MnSn_2$ are found to decrease precipitously as the temperature is lowered through 73°K. Both these abrupt changes exhibit a small temperature hysteresis and are highly suggestive of a first-order transition. From 73°K down to 4.2°K, the susceptibility is essentially constant; above 73°K, the susceptibility rises slowly to a maximum (at 86°K) and then decreases in a manner consistent with the Curie-Weiss relation. It is tentatively concluded that the abrupt transition at 73°K involves only a partial disordering of an antiferromagnetic state.

IN the course of studying the magnetic properties of the Mn-Sn system, we have discovered that the intermetallic compound $MnSn_2$ undergoes an unusually rapid transition at about 73°K. At this temperature, we observe a large, abrupt change in both its magnetic susceptibility and its electrical resistivity.

An ingot of this material was prepared by induction melting of electrolytic manganese and tin in an argon atmosphere. From this fairly brittle ingot, whose chemical analysis gave 68.5 at. % Sn, we were able to machine out a small cylindrical specimen for our magnetic measurements and a short ribbon for our electrical-resistivity experiments. Both these polycrystalline specimens were annealed for 4 days at 490°C, then furnace-cooled to room temperature. X-ray diffraction study of filings given the same heat treatment revealed a strong $MnSn_2$ pattern with some very weak lines attributable to β-Sn. The lattice parameters obtained for $MnSn_2$ ($a=6.66$ A, $c=5.43$ A) agree very closely with those previously reported for this ordered tetragonal ($C16$) structure.[1] The magnetization was measured in an apparatus described elsewhere[2] in various fields up to 10 koe at many different temperatures between 4.2° and 300°K. Measurements of resistivity, by standard potentiometric techniques, were carried out over the same temperature range.

Our magnetic measurements indicated the residual presence of a ferromagnetic ingredient in our specimen, which saturated in fields greater than 4 koe and had a Curie temperature of about 270°K. From subsequent work on other Mn-Sn alloys, we can identify this ferromagnetic ingredient as approximately 0.2% (by volume) of the ordered alloy Mn_2Sn. However, the dominant component of the magnetization of our specimen increased linearly with field and was easily extracted from the data. The volume susceptibilities χ_v, thus obtained for $MnSn_2$, are plotted vs temperature in Fig. 1. It is evident that as the temperature is raised from 4.2°K, the susceptibility remains essentially constant until, at about 73°K, it suddenly increases over twofold. It then continues to increase, but less rapidly, and reaches a maximum at about 86°K. Above this latter temperature, where χ_v is gradually decreasing, values for the reciprocal of χ_v are also plotted in Fig. 1. A straight line, shown dotted in the figure, fits all the χ_v^{-1} vs T points very well, except those around 270°K (whose deviation may be due to the increased susceptibility of the small amount of Mn_2Sn at its Curie point). Thus, above its susceptibility maximum, the $MnSn_2$ exhibits a classical paramagnetic behavior with a small, if not zero, paramagnetic Curie temperature. From the slope of the χ_v^{-1} vs T curve, we calculate a value of 4.53 μ_B for the effective paramagnetic moment $\{g\mu_B[S(S+1)]^{\frac{1}{2}}\}$ per Mn atom. Assuming $g=2$, we convert this to a $g\mu_BS$ value of 3.64 μ_B, which is quite reasonable for the atomic moment of Mn.

For a more detailed study of the abrupt change in susceptibility, we immersed the $MnSn_2$ specimen in a liquid-nitrogen bath whose pressure was slowly reduced and then slowly brought back to atmospheric. The results of quasistatic susceptibility measurements made during this temperature cycle between 77° and 68°K are shown in Fig. 2. Even though the temperature scale in this figure is considerably expanded, the large change in χ_v at about 73°K still appears almost discontinuous. Moreover, there is a temperature hysteresis of about

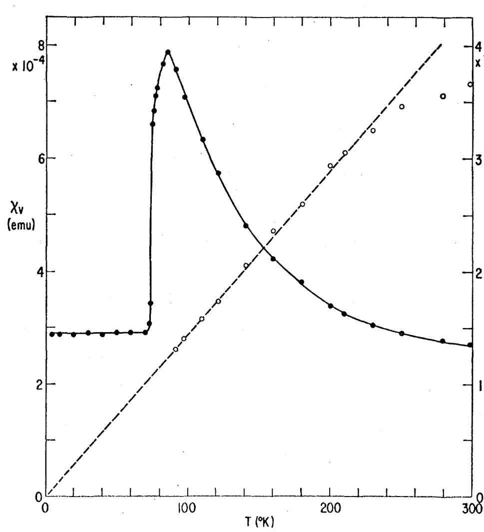

FIG. 1. Volume susceptibility of $MnSn_2$ (closed circles) and its reciprocal (open circles) vs temperature.

[1] H. Nowotny and K. Schubert, Z. Metallk. **37**, 17 (1946).
[2] J. S. Kouvel, C. D. Graham, and J. J. Becker, J. Appl. Phys. **29**, 518 (1958).

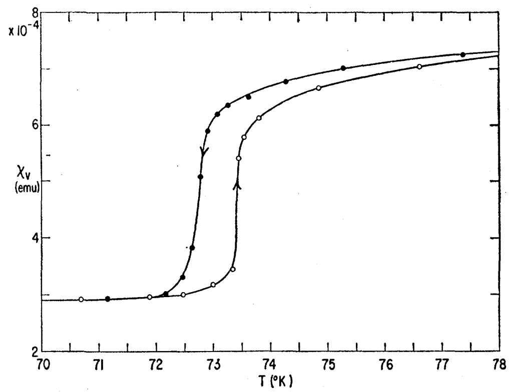

FIG. 2. Volume susceptibility of $MnSn_2$ for decreasing and increasing temperature (closed and open circles, respectively).

0.7°K in this susceptibility change, which is not inherent in our apparatus and must therefore be attributed to $MnSn_2$. Both the abruptness of this susceptibility change and its temperature hysteresis are highly suggestive of a first-order transition. Such a transition would be accompanied by a latent heat and a discontinuous variation of the atomic spacings, but measurements of these effects have not yet been made.

The electrical resistivities of our $MnSn_2$ ribbon specimen are plotted vs temperature in Fig. 3, and it is clear that the resistivity, also, changes abruptly at about 73°K. Although it is not indicated in the figure, this resistivity anomaly was found to have a temperature hysteresis similar in magnitude to that observed in the susceptibility. The sharp decrease in resistivity when the temperature is lowered below 73°K appears to signify a sudden development of magnetic order. Hence, the corresponding drop in susceptibility at 73°K, as well as its constant value at lower temperatures, may be taken as more specific evidence that $MnSn_2$ is in an ordered antiferromagnetic state in this low-temperature range. However, the susceptibility maximum at 86°K, seen in Fig. 1, would suggest that the antiferromagnetic order in this material is not completely destroyed by the abrupt transition at 73°K. The persistence of temperature hysteresis in the susceptibility well above 73°K, as shown in Fig. 2, may be another indication of some residual magnetic order between the temperatures of the abrupt transition and the susceptibility maximum.

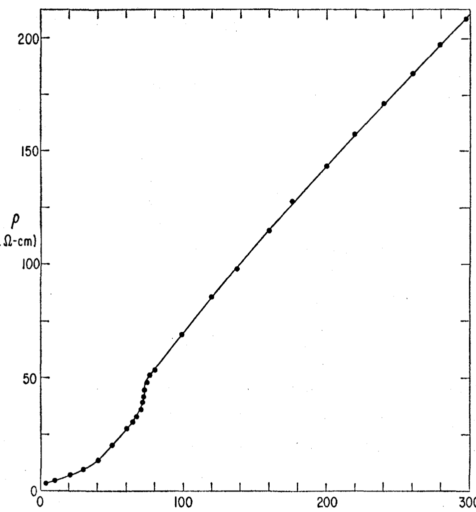

FIG. 3. Electrical resistivity of $MnSn_2$ vs temperature.

The possibility inferred from our results that $MnSn_2$ is an antiferromagnet at low temperatures contradicts Guillaud's report of ferromagnetism for alloys of approximately this composition,[3] but the specimens of this earlier work probably contained some ferromagnetic Mn_2Sn, as our experience with these materials would indicate. It should be noted that $FeSn_2$, which is isomorphous with $MnSn_2$, was recently reported[4] to be an antiferromagnet with a Néel temperature (susceptibility maximum) of about 380°K. However, there is no evidence from these susceptibility measurements above room temperature, or from our own recent measurements of resistivity down to 4.2°K, that $FeSn_2$ undergoes an abrupt transition of the kind we have encountered in $MnSn_2$. The determination of the spin configurations in $MnSn_2$ and $FeSn_2$ by neutron diffraction is being attempted at present.

[3] C. Guillaud, thesis, University of Strasbourg, 1943 (unpublished).

[4] K. Kanematsu, K. Yasukochi, and T. Ohoyama, J. Phys. Soc. Japan **15**, 2358 (1960).