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A neutron scattering study of the magnetic excitations in a triangular itinerant antiferromagnet, Mn_3Sn

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Abstract. The intermetallic compound Mn_3Sn has a triangular arrangement of spins below the Néel point at 420 K. A second magnetic transition occurs at 230 K. A proper screw structure is developed along the c-axis, with a period of $12c$. The occurrence of this transition appears to depend on the degree of annealing of the sample. We have investigated the spin dispersion relations in this intermetallic compound as a function of temperature. The dispersion relations appear to be characteristic of an antiferromagnet in the commensurate phase and change drastically in the low-temperature phase. The presence of steep spin-wave excitations and other features indicate a close similarity to itinerant antiferromagnets.

1. Introduction

Investigation of spin-wave dispersion relations in antiferromagnets is of interest since the cross section of the excitations can give much information concerning the magnetic and electronic properties of the material. The dispersion relations of a localized antiferromagnet are relatively well understood, and they differ sharply from those of the itinerant type, for which theory is still being developed. A recent theoretical study of itinerant magnetism in the case of Mn_3Sn, using spin-density-functional theory, appears to offer the possibility of detailed comparison with experimental results [1].

Phenomenologically, it has been suggested that certain features of the dispersion curves for neutron scattering offer the best criteria for distinguishing these types. We have studied the dispersion relations in the special case of an antiferromagnet, Mn_3Sn, in which the excitations change their characteristics qualitatively between room temperature and 8 K.

The intermetallic compound Mn_3Sn has a hexagonal crystal structure $P6_3/mmc$ corresponding to group 194 of the International Tables. The lattice parameters [2] at room temperature are: $a = b = 5.665$ Å, $c = 4.5310$ Å, $\alpha = \beta = 90°$, $\gamma = 120°$. We have measured them at 10 K, obtaining $a = b = 5.6493$ Å, $c = 4.5175$ Å. There are six Mn atoms in the unit cell in the positions 6h and two Sn atoms in the positions 2c.

A free position parameter allowed by the structure refers to the distance between neighbouring atoms in the same plane, denoted by $a[1/2 + 3(x - 5/6)]$. Tomiyoshi finds $x = 5/6 + 0.0055$. The basal plane projection of four unit cells is shown in figure 1.

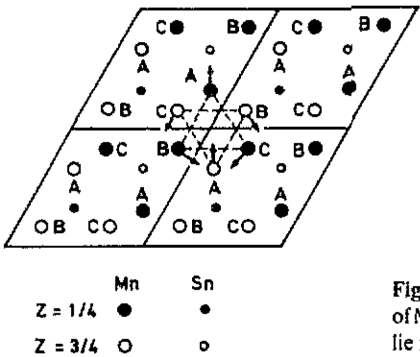

Figure 1. Crystallographic and magnetic structure of Mn_3Sn. Projection on the basal plane. The spins lie in the basal plane.

Mn_3Sn is an antiferromagnet with a Néel temperature of $T_N = 420$ K. There are three magnetic sublattices labelled A, B, C in figure 1, and the vector sum of the moments in each is zero. Tomiyoshi concludes on the basis of polarized neutron measurements that the orientation of the spins is as shown in figure 1 [2, 3].

A further magnetic transition occurs at 230 K [4]. A relative rotation of the triangular arrangements produces a proper screw propagating along the c-axis with a period of $12c$, independent of the temperature. Certain quenched samples do not undergo this transition.

2. Experimental details

Single crystals were produced by the Bridgman technique. The composition was about $Mn_{3.2}Sn$. One of them was quenched from 990 °C and the other was annealed carefully for 50 hours at 850 °C and cooled at ten degrees per hour to room temperature. They were sealed in aluminium cans and maintained with the appropriate scattering plane horizontal in a Displex refrigerator on the spectrometers. The Bragg reflections were broad, indicating a large mosaic spread, sometimes up to 30′. Neutron inelastic scattering measurements were performed on triple-axis spectrometers at the reactor Orphée, Laboratoire Léon Brillouin. Monochromators and analysers of pyrolytic graphite set at the (002) reflection were used. To start with, the horizontal collimations were 90′, 120′, 58′ and 58′ in each part of the spectrometer, and were varied during the measurements to maximize the signal-to-noise ratio. (For instance, in figure 3, later, values of 30′, 40′, 20′, 20′ were used.)

The measurements were performed with fixed incident or outgoing wave-vectors of different values, usually at 2.662 $Å^{-1}$ in combination with graphite filters appropriately placed to eliminate higher-energy neutrons. Constant-E or constant-Q scans were used when appropriate. The magnetic excitations were measured around the Bragg peaks (1, 1, 0) (1, 0, 0) and (3, 0, 0).

The peak positions obtained from these scans were plotted as a function of ξ, the reduced wave-vector. The dispersion relations for the quenched specimen and annealed specimens are identical and are shown in figure 2. An interesting feature of the results is the gap in the energy at $q = 0$.

A fit of the trend of the experimental values to a quadratic expression gave $(\hbar\omega)^2 = 932.32q^2 + 1.00$ for the $(1 + \xi, 1 + \xi, 0)$ direction and $(\hbar\omega)^2 = 502.55q^2 + 1.00$ for the

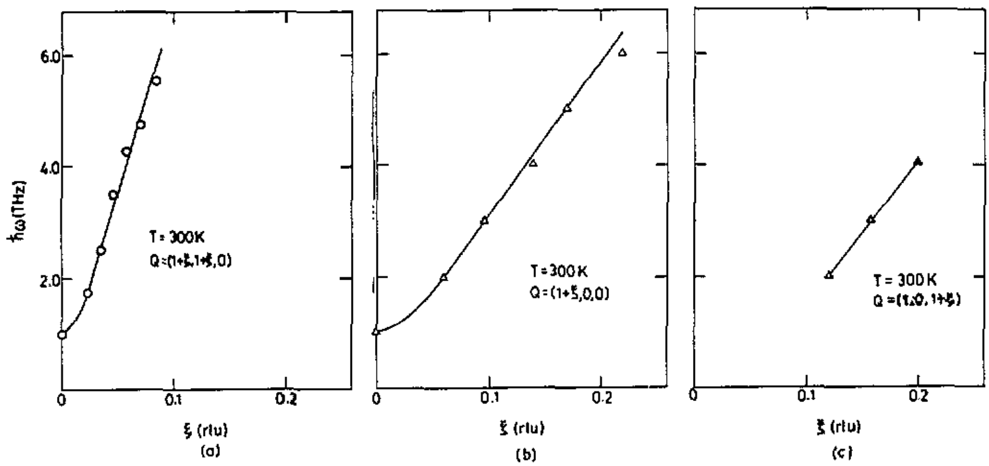

Figure 2. Dispersion relations in Mn_3Sn at room temperature: (*a*) direction $(1 + \xi, 1 + \xi, 0)$; (*b*) direction $(1 + \xi, 0, 0)$. The full curves correspond to the fits described in the text. (*c*) Dispersion relations in annealed Mn_3Sn at room temperature, direction $(1, 0, 1 + \xi)$.

$(1 + \xi, 0, 0)$ direction. The energy is in THz and the reciprocal wave-vectors in $Å^{-1}$. In terms of the reduced wave-vector, $q = 2\pi\xi/d$ where d is the interplanar distance, or $q = 2.2182\xi$ in the $(1 + \xi, 1 + \xi, 0)$ direction and 1.2807ξ in the $(1 + \xi, 0, 0)$ direction. The estimated errors correspond to ± 0.1 THz in energy and to $\pm 0.8 \times 10^{-2}$ $Å^{-1}$ in q.

In the case of the annealed specimen, it was possible to measure the dispersion in the $(1, 0, 1 + \xi)$ direction, although the peaks were rather large and ill-defined.

3. Low-temperature results

Further measurements were made on the annealed specimen at different temperatures. The temperature was reduced, and elastic scans on the positions of the satellites around the Bragg peaks (1, 0, 0) and (1, 0, 1) showed that the propagation vector is along the c-axis and has a value of $\tau = 0.09c^*$. The positions of the satellites were nearly independent of temperature. The screw structure occurred in all the specimens, independently of the heat treatment, and not only in the annealed specimen as found by Tomiyoshi previously. This difference may be due to differences in composition or to other factors. In addition, the relative intensities of the satellites and the main peak were much smaller than those observed by him. In the case of the quenched specimen, a comparison of the relative intensities of these peaks, $(1, 0, -\delta)$, $(1, 0, 0)$ and $(1, 0, +\delta)$, where δ is about 0.1 $Å^{-1}$, gave a smaller moment on the manganese atom.

The inelastic scattering was measured at 8 K, with mostly constant-E scans through (1, 0, 0) and (1, 1, 0), and occasionally at (3, 0, 0). Typical features of the results are discussed with examples of experimentally observed scans.

The reasonable assumption that the normalized spectral weight function describing the dynamics in the expression for neutron inelastic scattering in this magnetic system has a Lorentzian form was made [5, 6]. The data were fitted to a set of Lorentzians taking the resolution into consideration. In certain cases, such fits were not satisfactory and best estimates were used.

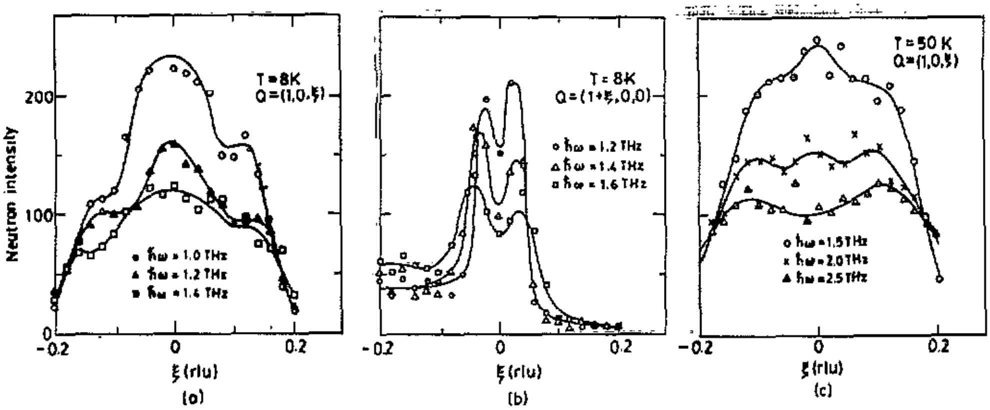

Figure 3. Typical neutron groups in Mn_3Sn at 8 K (*a*) in the direction (1, 0, ξ); (*b*) in the direction (1 + ξ, 0, 0), for energies of 1.2, 1.4 and 1.6 THz; and (*c*) at 50 K for 1.5, 2.0 and 2.5 THz.

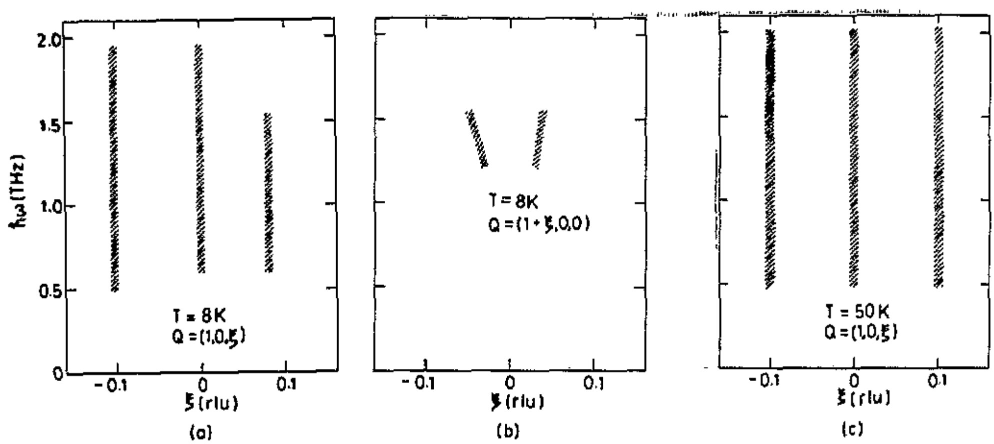

Figure 4. Dispersion relations in annealed Mn_3Sn at 8 K, in the helical phase: (*a*) direction (1, 0, ξ); (*b*) direction (1 + ξ, 0, 0); (*c*) direction (1, 0, ξ) at 50 K. The hatched lines indicate the location of the peaks of the neutron groups.

The neutron groups measured for constant-*E* scans at 8 K along (1, 0, ξ), in general three in number, are shown in figure 3(*a*). In figure 3(*b*), neutron groups in the direction (1 + ξ, 0, 0) are shown. In this case, they peak at smaller values of *q* and only two are observed for each energy. The intensity decreases at higher energies, making measurements above about 2 THz difficult. At 50 K, the qualitative features remain unchanged, but the neutron groups appear to have a nearly symmetrical three-peak structure, as shown in figure 3(*c*). Such groups are not observed above the temperature of the second magnetic transition, at 230 K, and are therefore unlikely to be due to phonons.

The peak positions obtained from the fits are shown in figure 4 for the three cases. The locations of the peak positions are indicated by hatched areas, taking the width into consideration. The widths are larger than they would be if they were limited by the resolution.

It is immediately evident that the excitation spectrum is qualitatively different from that observed for a localized antiferromagnet, and corresponds to that measured for itinerant materials, such as Mn_3Si and Cr [5, 6]. Steeply rising excitations are observed along the $(1, 0, \xi)$ direction at $\xi = (1, 0, -0.1)$ $(1, 0, 0)$ and $(1, 0, 0.1)$. There are also chimneys of excitations outside these values. In our experiments, no clear cones of excitations arising from the satellite points were seen. The slope is extremely high. These features, as well as the general absence of a simple dispersion curve, emphasize the resemblance to the excitations observed in itinerant materials.

4. Discussion

The magnetic excitations in Mn_3Sn at room temperature correspond to well-developed spin waves with a gap at zero wave-vector due to anisotropy of about 1.0 THz.

The magnetic behaviour of Mn_3Sn is very sensitive to strain and to the presence of impurities, which are often found to suppress the screw antiferromagnetic phase at low temperatures. The conditions of preparation of the specimens used here, which had compositions of around $Mn_{3.2}Sn$, favoured the production of this phase in all cases, although with a reduced manganese moment.

At low temperatures, dispersion relations reminiscent of itinerant antiferromagnets were measured, with a wealth of detail due to several branches. Excitations at wave-vectors corresponding to the satellite and central Bragg positions were observed. The slope of these excitations was very high. Measurements at higher energies and with possibly higher resolution are necessary to resolve the details of the dispersion relations.

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