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SPIN CORRELATIONS IN GAMMA MANGANESE

by B. C. G. HAYWOOD, R. D. LOWDE, M. W. STRINGFELLOW et W. B. WAEBER

Atomic Energy Research Establishment, Harwell, England

Résumé. — La diffusion inélastique des neutrons par des monocristaux des alliages de Mn antiferromagnétiques contenant 27 et 9 % de nickel et se trouvant dans la phase γ , a été mesurée employant un spectromètre à trois axes. On trouve des magnons d'une largeur de raie naturelle considérable. Qualitativement on révèle certaines particularités d'un comportement d'antiferromagnétique à électrons itinérants.

Abstract. — The inelastic scattering of neutrons by single crystals of antiferromagnetic γ -phase alloys of manganese containing 27 and 9 % nickel has been surveyed using a triple-axis spectrometer. Magnons having a considerable natural line-breadth are found. Qualitatively, a number of features of the behaviour of itinerant-electron antiferromagnets are evident.

Face-centred tetragonal γ -manganese is of considerable interest, in that it is an antiferromagnetic transition metal with a fairly substantial moment — $2.4 \mu_B$ — and by all accounts a quite simple magnetic structure consisting of alternating 001 ferromagnetic sheets. It is, in fact, a member of a series of like substances that can be fabricated as alloys of the transition elements with electron-to-atom ratios over the region bracketing the values for Mn and Fe. A number of studies have indicated that the entropy, specific heat, resistivity and magnetic susceptibility of γ -Mn are all inconsistent with the idea of localised moments [1].

There is a growing literature on the fundamental theory of an itinerant-electron antiferromagnet, much of which has, however, concentrated on the picture presented by chromium, which is not exactly the same as for manganese. The essential idea is that a self-consistent exchange field lowers the symmetry of the spin system, alters the Brillouin zone, and by coupling together different branches of the band structure produces an energy gap across the Fermi surface. In chromium the interaction is slight, and the gap small, as we know from the detailed analysis of Asano and Yamashita [2]; on the contrary, their subsequent investigation of Mn with Wakoh [3] and the work of Fletcher [4] have shown that the antiferromagnetism of γ -Mn can be understood as arising from a strong self-consistent interaction repelling related levels to the extent of 3-4 eV.

In the hope of throwing light on this situation we have measured the magnetic spin correlation, up to $\hbar\omega$'s of 50 MeV, by means of neutron inelastic scattering. The nature of the scattering phenomena to be expected with such materials has been examined theoretically by Sokoloff [5] and by Liu [6]. Details have not yet been worked out for manganese, however, and of at least phenomenological importance is Kouvel's discussion [7] of the spin-wave dispersion law for a material of the same structure with the Heisenberg Hamiltonian. Kouvel finds the usual antiferromagnetic spin wave « gap » at zero wave vector \mathbf{q} ; but with appropriate parameters the dispersion curve can rise to a maximum and then descend, on the lines where h and k are integers of differing parity, to another gap which can be larger or smaller than that at $q = 0$.

It is difficult to obtain satisfactory crystals of the pure material. For this reason we, like other workers,

have had to employ alloys, though of composition as close to 100 % Mn as possible. We have been able to make progress by adding carbon to the metallic contents. The carbon, it is believed, dissolves interstitially and leaves the transition-shell electron behaviour essentially unaffected; at the same time it stabilises the fcc phase, and has enabled us to obtain single crystals of composition $Mn_{85}Ni_9C_6$ which, if the carbon can truly be ignored, should adequately stand in for MnNi crystals of composition $Mn_{90}Ni_{10}$. These we compare with crystals of higher nickel content up to $Mn_{73}Ni_{27}$, which last can be grown carbon-free.

The question of residual short-range order in these alloys has been much studied; it requires a discussion that we do not have space to reproduce here. The analysis of Wells and Smith [8], however, enables us to show that the degree of short-range order in our

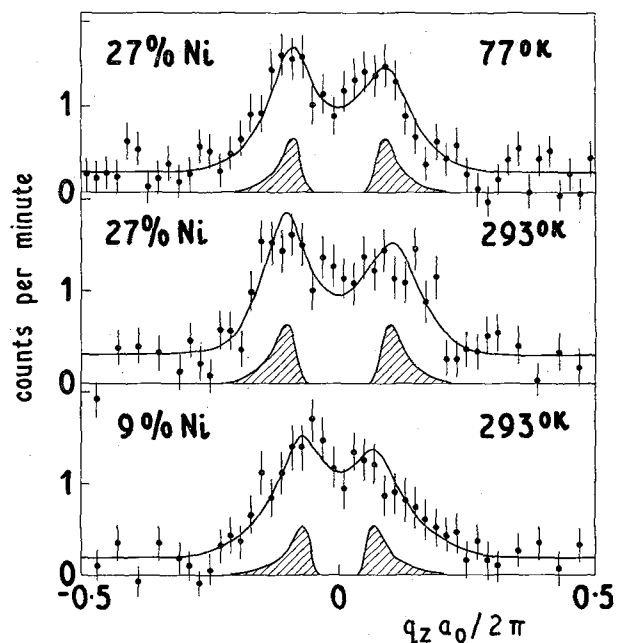


FIG. 1. — Surveys from $-q_{max}$ to q_{max} of the neutron scattering at a constant energy transfer of 35 MeV. The background count of 2.4 min^{-1} has been subtracted. Small hatched peaks indicate the magnon lines derived by assuming that there is an intrinsic Gaussian spread of A in the energy $\sqrt{(25 + A^2 q^2)}$; the combined effect of instrumental resolution and this spread is shown by the solid lines.

crystals was insignificant, affecting the composition even of near-neighbour co-ordination shells by less than 5%.

Measurements were made with our triple-axis spectrometer at the PLUTO reactor. Figure 1 shows surveys across the Brillouin zone, taken at a constant energy transfer of 35 MeV; two peaks are resolved. For one possible domain orientation the traverse would be from $(1, 1, -\frac{1}{2})$ to $(1, 1, \frac{1}{2})$, but for the other two orientations it would be of the type $(1, -\frac{1}{2}, 1)$ to $(1, \frac{1}{2}, 1)$. We have attempted to correct the data for the resolution function of the instrument with the results shown. The quality of fit obtained is seen to be satisfactory, and a well-defined but nevertheless heavily broadened excitation is revealed. The experimental points are not consistent with a simple Gaussian spread in q at constant $\hbar\omega$, but do support the idea of a Gaussian spread in the magnetic interaction parameter.

A survey along the energy axis at zero momentum transfer is shown in figure 2, and exhibits a well-

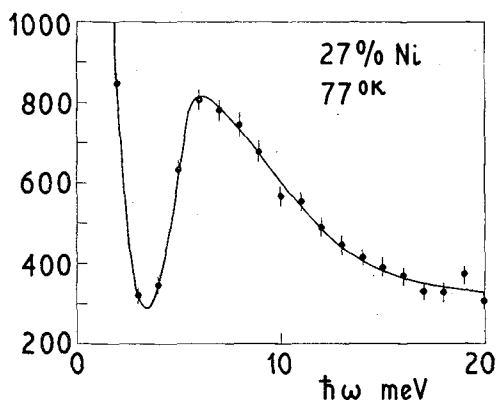


FIG. 2. — A survey at $q = 0$, showing the spin wave energy gap.

resolved gap. Figure 3, which displays a range of measurements, demonstrates that our broad distribution of intensity marks out a well-defined and highly dispersive path in $\hbar\omega, q$ space.

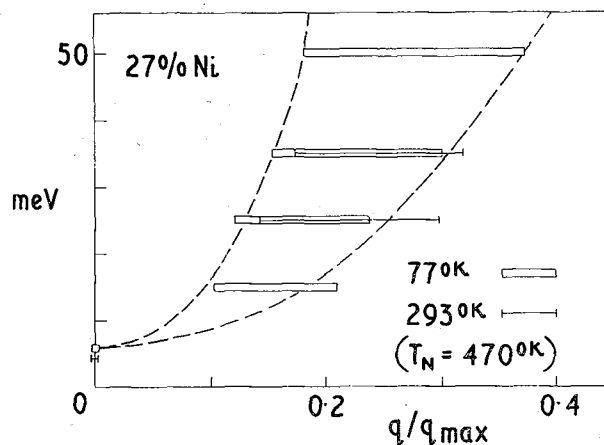


FIG. 3. — A plot of the (constant-energy) surveys, indicating by bars the observed resolution-corrected full widths corresponding to the half-height values of A (defined at Fig. 1). Broken lines show the trend.

Because of uncertainty about the magnetic domain orientations it will require further work to determine which of Kouvel's « gaps » we have found (if, indeed, either is appropriate to γ -Mn). The trend in figure 3 could be taken to suggest that it is the type where h and k have different parity. Actually the great breadths of the peaks, the large excitation energies involved (the fast-rising part of our dispersion law has a slope of approximately half what is found for chromium [9]) and the slight temperature dependence are all characteristics of the itinerant-electron antiferromagnet discussed by Liu [6].

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