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LETTER TO THE EDITOR

Longitudinal fluctuations in an isotropic antiferromagnet

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Abstract. The neutron scattering cross section below the Néel temperature in $RbMnF_3$ has been studied with the aid of neutron spin polarisation analysis. In addition to the spin-wave scattering, a small central component was observed and found to be longitudinal in character. This longitudinal scattering is quasi-elastic, with an intensity that decreases with increasing wavevector and with decreasing temperature below T_N.

In this Letter we report measurements of the longitudinal susceptibility in the nearly isotropic $n = 3$ antiferromagnet, $RbMnF_3$. The behaviour of the longitudinal susceptibility in isotropic systems is an unresolved problem in statistical mechanics because the predicted behaviour has not been observed experimentally. Although mean-field theory predicts that the longitudinal static susceptibility, $\chi_L(0)$, is finite below T_N, spin-wave and renormalisation group theories predict $\chi_L(0)$ diverges at all temperatures below T_N because of spin-wave fluctuations (Holstein and Primakoff 1940, Herring and Kittel 1951, Brézin *et al* 1973, Brézin and Wallace 1973). For three-dimensional systems the wavevector dependence of the longitudinal susceptibility, $\chi_L(q)$, is calculated to diverge as $1/q$ in zero field, while the spin-wave or transverse susceptibility, $\chi_T(q)$, diverges as $1/q^2$ (Kawasaki and Mori 1961, 1962, Kawasaki 1967, Mazenko 1976). The frequency dependence of the longitudinal susceptibility has been predicted to be a three-peaked structure with inelastic peaks at the spin-wave frequencies and a central peak (Vaks *et al* 1968), a two-peaked structure with no central peak (Villain 1971) and a quasi-elastic peak (Mazenko 1976).

Experimentally, the behaviour of the spin fluctuations in several isotropic ferromagnets has been studied using neutron scattering techniques, and attempts have been made to distinguish the longitudinal component. The most successful were the measurements of Mitchell *et al* (1984) who used polarisation analysis to separate the longitudinal scattering in $Pd_{0.9}Fe_{0.1}$, and found the longitudinal fluctuations to be quasi-elastic under all conditions. However, they did not find evidence to support a divergent wavevector dependence of $\chi_L(0)$ below T_C, although this may be because the magnetic field applied to produce the single ferromagnetic domain destroyed the isotropic symmetry and

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possibly the divergent contribution. Because of this we have chosen to use an isotropic antiferromagnet when the field required to keep the neutrons polarised is not necessarily coupled to the order parameter.

The sample was mounted with the (11 −2) axis vertical in a vertical-field cryomagnet, on the triple-axis spectrometer IN12 at the Institut Laue–Langevin. The incident beam was collimated by reflection from a pyrolitic graphite monochromator and in the polarised beam experiments a supermirror bender was used to polarise the beam. A vertically curved Heusler crystal was used for energy and spin analysis of the scattered neutrons. Tuned DC coil flippers were placed before and after the sample to enable a separate measurement to be made of the neutron spin-dependent cross sections as described by Moon *et al* (1969).

The experiments were performed with the wavevector of the scattered neutrons held fixed with $K_f = 1.5$ Å^{-1} with collimation 30′–30′–30′–30′, and the vertical divergence was ≃0.5°. The energy resolution was ≃0.024 THz (FWHM) for isotropic elastic scattering, and the in-plane wavevector resolution ≃0.01 Å^{-1} (FWHM). The polarisation efficiency of the spectrometer was measured by comparing the ratio of elastic scattering at $(-0.5, -0.5, -0.5)2\pi/a$ measured in the spin-flip and non-spin-flip channels. (The magnetic Bragg scattering at (−0.5, −0.5, −0.5) is expected to be entirely spin-flip.) The flipping ratio was found to vary with temperature within the range 8 to 27.

Firstly, the sample was cooled in zero applied magnetic field, when the Néel temperature was measured as 83 K from the temperature dependence of the intensity of the (−0.5, −0.5, −0.5) magnetic Bragg reflection. If there is a random arrangement of antiferromagnetic domains the scattering below T_N is proportional to $\frac{2}{3}(\chi_L(q, \omega) + 2\chi_T(q, \omega))$, so both the longitudinal and transverse susceptibilities should be observed. Constant-wavevector scans at temperatures below T_N showed two well resolved peaks at non-zero energies, which are identified as the two inelastic spin-wave peaks, together with a small component centred on zero frequency. The temperature and wavevector dependences of the spin waves behaved as expected, while the central component had an energy width comparable with the resolution of the instrument and an intensity that decreased with decreasing temperature and increasing wavevector from (−0.5, −0.5, −0.5).

Secondly, the sample was cooled in an applied magnetic field of 1 T in an attempt to pole the sample into a single domain. The spins tend to align along the crystallographic (111) direction and will flop into a plane perpendicular to the applied magnetic field (the critical field above which a spin-flopped phase occurs in $RbMnF_3$ is 0.27 T (Chaddhra and Seehra 1982)). With a (11 −2) axis vertical there is only one (111) direction perpendicular to the applied field, so a single-domain specimen should be produced causing the intensity of the (−0.5, −0.5, −0.5) reflection to drop dramatically. Only a slight reduction in intensity was observed which, together with the intensity of the spin waves, was consistent with the spins being perpendicular to the applied field but only partially aligned in the (11 −2) plane. This could occur because the (111) anisotropy field is only about 4.5 Oe. With this domain structure the total scattering is proportional to $\frac{1}{3}(\chi_L(q, \omega) + 5\chi_T(q, \omega))$, while the spin-flip component is proportional to $\frac{1}{3}(\chi_L(q, \omega) + 2\chi_T(q, \omega))$ and the non-spin-flip component is proportional to $\chi_T(q, \omega)$. It was found that removal of the field below T_N resulted in a random domain structure and so a field of 1 T was maintained during the measurements.

Constant-wavevector scans were measured using full polarisation analysis in the partially oriented domain structure at the temperatures 81, 78, 70, 40 and 1.5 K (background scans) for wavevectors $\boldsymbol{Q} = (-0.5 - \zeta, -0.5 - \zeta, -0.5 - \zeta)\ 2\pi/a$ where $\zeta =$

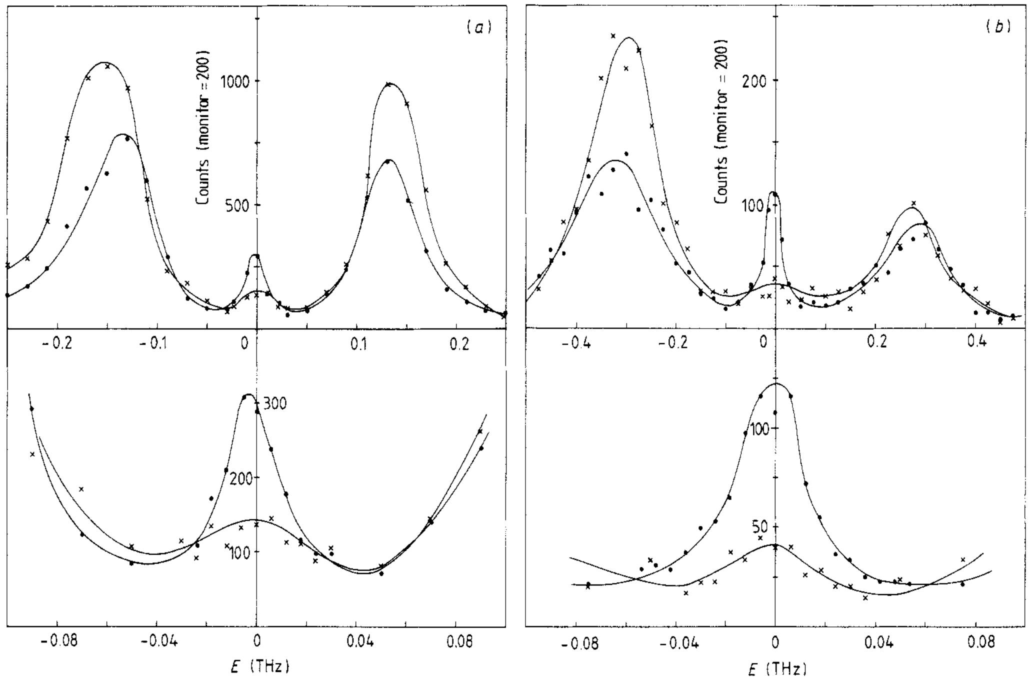

Figure 1. Constant-wavevector scans at $T_N - 5$ K for (*a*) $Q = (-0.52, -0.52, -0.52)\ 2\pi/a$ and (*b*) $Q = (-0.54, -0.54, -0.54)\ 2\pi/a$. (Applied field, 1 T; ●, spin-flip scattering; ×, non-spin-flip scattering.)

0.02–0.04. Figure 1 gives the results obtained at 78 K for $\zeta = 0.02$ and $\zeta = 0.04$, and shows the well resolved spin waves and the central component. It can be seen that while the spin waves are more intense in the non-spin-flip channel compared with the spin-flip channel, the central component is significantly smaller. This suggests that the central component is composed largely of the longitudinal fluctuation scattering. Figure 2 gives the results obtained at 70 and 1.5 K for $\zeta = 0.02$. A comparison between data for $\zeta = 0.02$ at temperatures of 78 and 70 K shows that while the difference in the intensity of the spin waves between the non-spin-flip and spin-flip channels is nominally the same, the difference in the intensity of the central component between the two channels is considerably reduced. This shows that the intensity of the central component is decreasing away from T_N and so results from critical fluctuations. At 1.5 K, the central component is the same in the non-spin-flip and spin-flip channels and much smaller, presumably resulting largely from elastic nuclear isotopic and spin incoherent scattering.

The intensity of the longitudinal fluctuation scattering was estimated by subtracting the non-spin-flip scattering centred on zero energy from the spin-flip scattering, for each wavevector and temperature considered, and making allowance for the flipping ratio. The results are summarised in figure 3.

Our results are surprisingly different from those of Tucciarone *et al* (1971) made at T_N, which showed a quasi-elastic peak of approximately the same intensity as the spin waves. Our results indicate a much weaker central component that has an energy width comparable with the instrumental resolution and is definitely longitudinal in character. Figure 3 shows that for temperatures just below T_N, $\chi_L(q)$ increases rapidly with decreas-

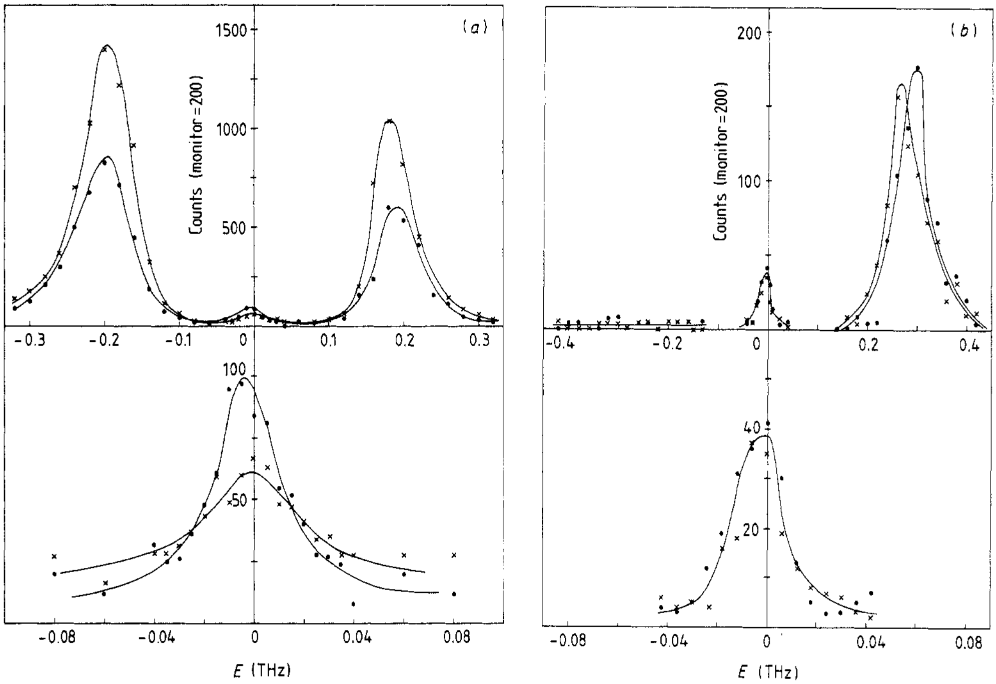

Figure 2. Constant-wavevector scans at (*a*) $T_N - 13$ K and (*b*) $T_N - 81.5$ K for $Q = (-0.52, -0.52, -0.52)\,2\pi/a$. (Applied field, 1 T; ●, spin-flip scattering; ×, non-spin-flip scattering.)

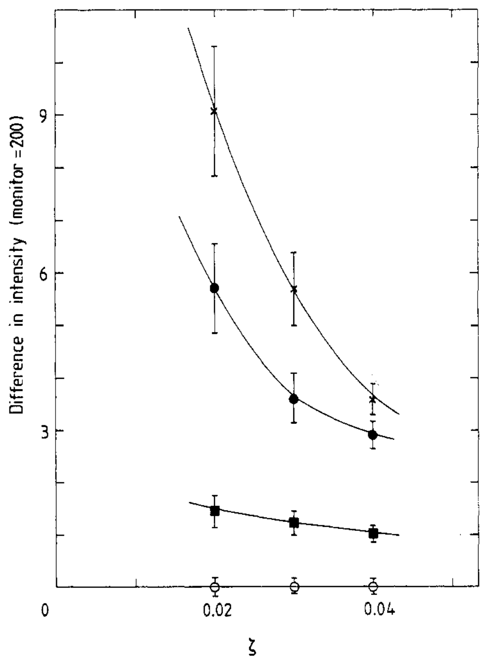

Figure 3. The wavevector dependence of the integrated intensity of the longitudinal fluctuation scattering for $Q = (-0.5 - \zeta, \; -0.5 - \zeta, \; -0.5 - \zeta)\,2\pi/a$. (Applied field, 1 T; ×, $T_N - 2$ K; ●, $T_N - 5$ K; ■, $T_N - 13$ K; and ○, $T_N - 43$ K.)

ing wavevector q, but because the data close to $q = 0$ are limited it is impossible to determine the analytic form.

The results can be compared with those of Mitchell *et al* (1984) obtained on a ferromagnet, who also found the longitudinal susceptibility to be quasi-elastic, but with an energy width $\simeq 0.25$ THz, which decreased with decreasing wavevector. In our experiments the scattering was much narrower in energy, and nominally independent of wavevector, although the width decreases slightly with decreasing temperature.

In conclusion, we have observed the spin fluctuations in $RbMnF_3$ below T_N using neutron scattering techniques both in an unpolarised specimen and in a partially oriented domain structure. Using neutron spin polarisation analysis we have determined the longitudinal component in the scattering cross section and find it to be quasi-elastic with an energy width comparable with the instrumental resolution. We were unable to determine whether $\chi_L(q)$ was divergent as q tends to zero, and hope to perform further experiments with a smaller applied field to produce a truly single-domain sample in the near future.

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