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PARAMAGNETIC SCATTERING OF NEUTRONS FROM KMnF$_3$
IN THE SHORT RANGE ORDERED REGION

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Résumé. — Nous avons étudié les spectres en énergie des neutrons lentis diffusés par KMnF$_3$, à plusieurs températures au-dessus de la température de Néel, dans le domaine, $T_N < T < 300$ °K, où l'ordre à courte distance entre les spins n'est pas négligeable. Les angles de diffusion, $\theta$, auxquels ces spectres sont étudiés, sont tels qu'ils explorent le pic dû à l'ordre à courte distance, centré à $Q_0\left(= \frac{4 \pi}{\lambda} \sin \frac{\theta}{2}\right) = 1.3$ Å$^{-1}$. Nos résultats expérimentaux indiquent la présence d'excitations collectives magnétiques, dans ce domaine de températures. Nous avons analysé ces résultats dans le cadre des théories actuelles. Nous avons aussi essayé d'ajuster nos résultats avec les calculs basés sur deux modèles théoriques, (1) le modèle des magnons amortis, et (2) l'approximation gaussienne modifiée.

Abstract. — The energy spectra of cold neutrons scattered by KMnF$_3$ has been studied above the Néel point, $T_N$, at various temperatures in the temperature region, $T_N < T < 300$ °K, in which short range order between spins is not negligible. The scattering angles, $\theta$, at which these energy distributions have been obtained, have been chosen so as to scan the short range order peak centred around, $Q_0\left(= \frac{4 \pi}{\lambda} \sin \frac{\theta}{2}\right) = 1.3$ Å$^{-1}$. The experimental data indicates the presence of collective excitations of magnetic origin in KMnF$_3$, at these temperatures. The experimental results have been examined in the light of existing theories. An attempt has also been made to obtain a quantitative fit to our results, of calculations based on two of the theoretical models, (1) a damped magnon model and (2) a Gaussian approximation.

The presence of collective excitations (paramagnons) in short-range ordered paramagnets has been indicated by neutron scattering [1] and light scattering experiments [2] for temperatures, $T$, well above the transition temperatures. However the magnetic systems studied in these experiments are anisotropic and the presence of paramagnons in Heisenberg (isotropic) spin systems is still subject to some doubt. The present work was undertaken with a hope to resolve this uncertainty. This paper describes neutron paramagnetic scattering measurements made on polycrystalline KMnF$_3$, KMnF$_3$ is an antiferromagnet having a perovskite structure, the magnetic ions forming a simple cubic structure. Its Néel temperature $T_N$, is at 88 °K and its magnetic anisotropy is negligible in the region of temperature studied here.

The inelastic scattering measurements were made, using a rotating crystal (time-of-flight) spectrometer. The incident neutron energy, $E_0$, was 4.7 meV ($\lambda = 4.18$ Å) and the resolution was about 0.35 meV at the incident energy.

The time-of-flight distributions of the scattered neutrons were studied at 135 °K ($\geq 1.5$ $T_N$), 170 °K ($\geq 2T_N$), and at 300 °K ($\geq 3.4$ $T_N$). The diffraction patterns for KMnF$_3$ were also obtained at these temperatures. The magnetic structure factor $S(Q)$ obtained from these patterns is shown in figure 1, as a function of the momentum transfer, $Q$. The time-of-flight ($t$) spectra of neutrons scattered at angles, $\theta$, equal to 15°, 25°, 51° and 75° (corresponding to values of $Q_0\left(= \frac{4 \pi}{\lambda} \sin \theta/2\right)$ of 0.39, 0.65, 1.30 and 1.84 Å$^{-1}$), obtained at 135 °K and 170 °K, are shown in figure 2. The spectra obtained for the three lower values of $Q_0$ (except the one obtained at $T = 170$ °K, $Q_0 = 1.3$ Å$^{-1}$) indicate the presence of a quasi-elastic peak. In the spectra for $Q_0 = 0.39$ and 0.65 Å$^{-1}$, there are also distinct inelastic peaks (corresponding to energy transfers, $E$, in the region 15 meV to 40 meV), the origin of which is not clear to us at present. At $Q_0 = 1.84$ Å$^{-1}$ both distributions indicate the possible presence of a quasi-elastic peak and an inelastic peak.

Van Hove [3] has shown that the magnetic scattering cross-section $\frac{d^2 \sigma}{dE d\Omega}$ is related to $S(Q, \omega)$, the Fourier transform of the space-time correlation function between spins. It is simpler to deal with the relaxation function [4] $R(Q, \omega)$, equal to

$$S(Q, \omega) = \frac{\hbar \omega}{(1 - \exp(-\hbar \omega/k_B T))^{-1}}.$$  

Here $\hbar \omega = (-E)$ is the energy loss of the neutron. It has been shown by de Gennes [5] that when no short

![Figure 1](http://dx.doi.org/10.1051/jphyscol:19711210)
range order exists between spins \((T \geq T_\text{N})\) and for \(Qb > \pi\) (where \(b\) is a lattice constant), (1) \(R(Q, \omega)\) can be well approximated to a Gaussian function in \(\omega\), with a width proportional to the second moment, \(<E^2>\), of the neutron energy distribution, and (2) \(<E^2> \propto \sum Z_i J_i^2\) (where \(Z_i\) is the number of \(i\) the nearest neighbours and \(J_i\) is the \(i\) th exchange integral), and that \(<E^2>\) is independent of \(Q\).

Figure 3 shows a semi-logarithmic plot of \(R(Q, \omega)\) obtained from our experimental results at 300 K, against \(E^2\). \(R(Q, \omega)\) is seen to be a Gaussian. A value of \(-0.301 \pm 0.030\) meV is obtained for \(J_i\) from these results if one assumes only nearest neighbour interactions, and this is in good agreement with values quoted elsewhere.

In the «short range order region» of temperature, Villain [6] has predicted that no collective excitations can exist in a 3-dimensional Heisenberg (isotropic) magnetic system for \(T > T_\text{C}\). Bennett [7], on the other hand, has shown that propagating modes can exist in such systems if the values of the momentum transfer, \(Q\), is greater than a certain momentum transfer, \(Q_\text{c}\), while for \(Q < Q_\text{c}\) diffusive modes dominate. Another interesting model is that of the modified Gaussian approximation used by Madhav Rao et al. [8]. This model, which has been successfully used to explain coherent neutron scattering from liquids, gives

\[
R(Q, \omega) \propto \frac{S(Q)^{3/2}}{<E^2>^{1/2}} \times \exp\left[-\frac{E^2 S(Q)}{2 <E^2>}\right]
\]

where \(<E^2>\) is the second moment of the energy distribution at large values of \(Q\) (where \(S(Q) \approx 1\)). For an energy distribution at constant \(\theta\) (as is the case in our experiments), this model predicts a peak of \(R(Q, \omega)\) at a finite value of the energy gain (say \(E_3\)). \(E_3\) is largest for small values of \(Q_0\) and it tends to zero as \(Q_0\) increases.

\(R(Q, \omega)\) obtained from our results is shown in figure 4, plotted as a function of the energy gain, \(E\).
height increases with $Q_0$. At 135 $^\circ$K, $R(Q, \omega)$ for $Q_0 = 1.30$ Å$^{-1}$ seems to be a sum of (1) a quasi-elastic peak of width less than that of the corresponding one at $Q_0 = 0.65$ Å$^{-1}$, and (2) an inelastic hump at about 5 meV. The corresponding energy spectrum for 170 $^\circ$K has a rather surprising structure, but it also seems to have an inelastic hump at 5 meV. Both the distributions at $Q_0 = 1.84$ Å$^{-1}$ show a distinct inelastic peak with the peak for 135 $^\circ$K corresponding to a larger energy shift than that at 170 $^\circ$K.

The curves calculated using eq. (1), with $<E^2> = 12.7$ meV$^2$, are shown by dot-dashed lines in figure 4. The fits are seen to be far from satisfactory. If $<E^2>$ is left as an adjustable parameter, it is seen that the value giving the best fit, increases considerably with $Q_0$, which is in disagreement with the model. Villain's predictions also fail in accounting for our results since our results indicate the presence of collective excitations. Bennet's theory seems to account for the behaviour of $R(Q, \omega)$ in a qualitative way, since $R(Q, \omega)$ points to the existence of diffusive modes for low values of $Q_0$ and at high values of $Q_0$ there seems to be a clear evidence of the existence of collective excitations. The value of $Q_0$ is however difficult to locate since the experiments have not been performed at constant $Q$ values.

Our results show that collective excitations are present even when only short range order exists in a magnetic system. However, a clearer picture of the spin dynamics in such systems can only be obtained by performing the measurements with a single crystal, at constant $Q$ values.

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