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Critical Magnetic Scattering from $KMnF_3$ *

M. J. Cooper and R. Nathans

Brookhaven National Laboratory, Upton, New York

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Abstract

Critical magnetic scattering experiments on the simple ionic antiferromagnet KMnF_3 are described. The observed intensity distributions are compared with those calculated from the Van Hove theory for quasi-elastic scattering using an experimentally determined resolution function. Excellent agreement is obtained with the Ornstein-Zernike form for the radial dependence of the correlation function in the temperature range $1.002 < T/T_c < 1.1$. The correlation range is found to increase from $12\text{\AA}$ at $T/T_c = 1.1$ to $200\text{\AA}$ at $T/T_c = 1.002$ and the inverse staggered susceptibility is found to follow very closely a $4/3$ power law dependence on $T-T_c$. The results are also compared with recent results on ferromagnetic metals.

INTRODUCTION

There has been substantial interest recently in the study of the correlation of the fluctuations involved in phenomena exhibiting second order phase transitions, for example, structural or magnetic order-disorder phenomena. Interest is centered on the form of the pair distribution function which contains information on the nature of the interaction of the positional or spin fluctuation of one particular atom on that of another atom located at some other lattice point in the matrix. The pair distribution depends in general on position and time, the time dependence reflecting the continuously changing arrangement in the correlation configurations in a given microscopic volume.

As a practical matter, the information we are seeking comes from the measurement of the angular and energy distribution of monochromatic radiation scattered from the system at temperatures close to the transition, that is in the so-called critical scattering region. The radiation may be either light from laser beams, x-rays, or neutrons, but in the case of magnetic ordering the strong interaction between the neutron magnetic moment and that of the atomic spins makes neutrons a unique tool in the investigation of these systems, and critical scattering in both ferromagnetic and antiferromagnetic systems has been observed.

The suggestion that the critical magnetic scattering is a consequence of the large scale fluctuations in the density of magnetization which occur in the neighborhood of the ordering temperature was made by Van Hove¹ who subsequently developed a theory of critical magnetic scattering based on the Ornstein-Zernike² treatment of the critical scattering of

light in gases. Until now experimental studies with neutrons have, for the most part, been concerned with ferromagnetic systems³ and taken as a whole, show a remarkably good agreement with the predictions of Van Hove, although small deviations have been observed at temperatures very close to the transition point.

Critical scattering in antiferromagnetic systems is however still relatively unexplored. Although CoO and $\alpha\text{-Fe}_2\text{O}_3$ were studied by McReynolds and Riste⁴, their data were not sufficiently accurate to make a detailed comparison with theory. For this reason a detailed study of the critical magnetic scattering from a simple ionic antiferromagnet, KMnF_3 , was undertaken and the results are presented in this paper.

THEORY OF CRITICAL MAGNETIC SCATTERING

A review of the theory of critical magnetic scattering has been given recently by de Gennes⁵, in which he expresses the cross section for diffuse scattering of neutrons from a magnetic system as

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_f}{k_i} \left(1.91 \frac{e}{mc}\right)^2 \sum_{ij} F_j^*(\vec{k}) F_i(\vec{k}) \quad (1)$$

$$\times \frac{1}{2\pi} \int dt e^{-i\omega t} \langle S_{i\perp}(0) \cdot S_{j\perp}(t) \rangle \exp[i\vec{k} \cdot (\vec{R}_i - \vec{R}_j)]$$

where $F_i(\vec{k})$ and $F_j(\vec{k})$ are the amplitude form factors of the spins at the i th and j th atoms, normalized to give $F(0) = 1$, $\vec{R}_i$ and $\vec{R}_j$ are the radius vectors, measured from some origin, to the i th and j th atoms, and $\vec{k}$ is the scattering vector. $\langle S_{i\perp}(0) \cdot S_{j\perp}(t) \rangle$ is the correlation function which in general is a function of $\vec{R} (= \vec{R}_i - \vec{R}_j)$ and t . In

order to obtain this function, which we call $\Gamma_{\vec{R}}(t)$, it is necessary to measure both the angular and energy dependence of the scattering. The complete determination of $\Gamma_{\vec{R}}(t)$ is usually an exceedingly difficult problem due to intensity and resolution limitations, since the inelasticity involved in the neutron critical scattering is small compared to the energy of the neutrons. If, however, it is established that the energy transfer to the system (that is, the range of ω values) is small compared to the incident neutron energy then we may write for the cross section integrated over the final neutron energies

$$\frac{d\sigma}{d\Omega} = \left(1.91 \frac{e^2}{mc^2} \right)^2 \sum_{ij} F_j^* F_i < S_{i1} \cdot S_{j1} > \exp(i\vec{k}_0 \cdot \vec{R}) \quad (2)$$

This is the quasi-elastic approximation and it states that by measuring only the angular dependence of the scattering we can determine the radial extension of the pair correlation function. In our work we were able to demonstrate by energy analysis of the outgoing neutrons that $\Delta E/E$ was small compared to the energy of the incident neutrons ($\approx 1000^\circ K$) thus justifying the use of the quasi-elastic approximation in the interpretation of the data.

In the temperature region close to the transition point we have according to Van Hove that the asymptotic form of $\Gamma_{\vec{R}}(0)$ may be represented by the Ornstein-Zernike form, namely

$$\Gamma_{\vec{R}}(0) = \delta_{0\vec{R}} \frac{k_B T}{4\pi A} \frac{\exp(-\kappa_1 R)}{R} \quad (3)$$

using the notation given by de Gennes; κ_1 is an inverse range parameter and A is an energy parameter, related to the amount of energy necessary for twisting the magnetization. This form of the correlation function gives a cross section

$$\frac{d\sigma}{d\Omega}(q) \propto \delta_{0\beta} \frac{T/A}{\kappa_1^2 + q^2} \quad (4)$$

q being the magnitude of the reciprocal lattice vector defining displacement from the antiferromagnetic reciprocal lattice point. As we approach the critical temperature $\kappa_1 \rightarrow 0$ and we shall expect to observe sharper and more intense peaks around the antiferromagnetic reciprocal lattice points. This is the so-called critical scattering, which, according to the quasi-elastic approximation, will have a Lorentzian radial distribution.

In ferromagnetic materials there is a simple relation between κ_1^2 and the susceptibility, $\kappa_1^2 \propto 1/\chi$, but in the case of an antiferromagnet we must consider a quantity known as the staggered susceptibility χ_s , which is the susceptibility for the wave vector defining the antiferromagnetic reciprocal lattice point, and we have the relation $\chi_s = V/\chi_1^2$.

Recent calculations for a Heisenberg ferromagnet^{6,7} predict that the susceptibility varies as $(T-T_c)^{-4/3}$. No similar calculations have been carried out for an antiferromagnetic system, and one of the incentives in our examining a simple antiferromagnetic system was to investigate what differences, if any, exist in the temperature dependence of χ_s when compared with similar measurements made on ferromagnetic systems. We may note that unlike ferromagnetic systems where the susceptibility

may be obtained from magnetization measurements the staggered susceptibility is a quantity which can only be evaluated from scattering experiments.

The Ornstein-Zernike radial dependence is only expected to be applicable over a limited temperature range near the transition temperature. At higher temperatures we have, at present, no detailed calculation predicting either the temperature dependence of the susceptibility or the radial form of the pair correlation function. The Opechowski expansion for the susceptibility can be applied at temperatures considerably above the transition point, and Saenz⁸ has used this expansion to calculate the angular dependence of the neutron cross section in terms of the exchange coupling constants of the particular material. Unfortunately the relationships are so complex that the calculation of the cross section only becomes practicable at temperatures several times the critical temperature. We have, however, made a study of the neutron scattering over an extended temperature range in the paramagnetic region to examine the manner in which the observed angular dependence of the scattering deviated from the Ornstein-Zernike form.

CRYSTALLOGRAPHY OF KMnF_3

The structure of KMnF_3 is of the perovskite type (cubic) and becomes magnetically ordered below 88°K .⁹ The ordered magnetic structure is of the G-type, that is the manganese atoms form a simple cubic arrangement with all nearest neighbors coupled antiferromagnetically. The side of the cubic magnetic unit cell is therefore twice that of the

structural unit cell, so that the magnetic ordering gives rise to additional neutron reflections having half-integral values of h, k and l . The lowest angle magnetic reflection is therefore $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ and for the most part the scattering studied was that associated with the $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ reciprocal lattice point. However, scans were also made through $\frac{3}{2} \frac{1}{2} \frac{1}{2}$ for selected temperatures as a check on various factors which may alter the measured intensity such as extinction, multiple scattering and distortion scattering.

Crystallographic studies, using x-ray techniques, have been reported by Beckman and Knox¹⁰ who describe the distortions of the perovskite structure which are observed at low temperatures. At room temperature the structure is undistorted and the lattice constant is 4.19Å. On cooling distortions first occur at 184°K when the fluorine atoms move off their special positions and the over-all symmetry becomes orthorhombic. A further structural change is reported to occur at the Néel temperature, below which the pseudo-cell is again orthorhombic, but with different cell dimensions. Hysteresis is also reported in the temperature region immediately above the Néel temperature, when on heating from below this temperature the pseudo-cell becomes monoclinic.

The distortions are such that for neutron scattering small nuclear peaks appear at the positions of the magnetic reflections and are thus superimposed on the magnetic critical scattering around these positions. However, the sharpness of these nuclear peaks is such that they can be separated easily from the diffuse magnetic scattering down to within a small fraction of the critical temperature.

The intensity of the distortion peaks was found to be small and was observed to vary slowly with temperature. Hysteresis of the type described above was not observed in the temperature dependence of the intensity near the critical temperature. Rather the intensity of the distortion peak was found not to be exactly reproducible on cycling through the critical temperature indicating an extended range of possible distortion.

COLLECTION OF DATA

The diffractometer used was a conventional two-crystal instrument of the type described by the authors elsewhere¹¹ in a paper discussing the resolution function and its application to the measurement of diffuse scattering. Neutrons from the source were incident on a copper monochromator crystal set to reflect neutrons of wavelength $1.05\text{\AA}$ and a filter was used to reject the $\lambda/2$ component. The sample was a single crystal of KMnF_3 , roughly rectangular in shape and measuring approximately $10\text{mm} \times 6\text{mm} \times 6\text{mm}$, which was loaned to us by Dr. Guggenheim of the Bell Telephone Laboratories. The mosaic spread of this crystal was determined to be about $7'$ full width at half height.

One of the most stringent requirements in this type of measurement is maintenance of uniformity and stability of the sample temperature. In these experiments the sample was mounted in^a temperature-controlled dewar assembly. The crystal is mounted on an aluminum block which forms the bottom section of a container attached to a temperature-controlled copper block. Two radiation shields surround the sample. The

temperature of the copper block is measured using a platinum resistor embedded within it and is controlled by means of a heater and a coolant vent. By alternating the operation of these the temperature of the copper block is maintained automatically within close limits of a pre-selected temperature. The coolant used in this case was liquid nitrogen, and the temperature excursions were estimated to be less than $\pm 0.05^{\circ}\text{K}$ from the average temperature.

Because the sample is a thermal insulator there is a time lag between any change in the temperature of the copper block and that of different parts of the sample. Since this lag is much longer than the cycling period of the controller the temperature of the sample remains constant within much smaller limits than that of the copper block. These temperature effects were investigated by a careful study of the temperature and time dependence of the $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ magnetic scattering peak close to the Néel temperature, just below which the intensity is extremely temperature dependent. The temperature dependence of the scattering peak is shown in Fig. 1. It can be seen that there is no hysteresis and that the disappearance of long range order scattering is very sharp. This indicates that the sample had reached equilibrium at the pre-selected temperature and that there were no appreciable temperature gradients within the sample. Furthermore, it provided us with a precise value of the Néel temperature, viz. $88.06 \pm 0.02^{\circ}\text{K}$, ignoring any calibration error since we are only concerned with accurate values of $T - T_c$.

Because of the wavelength spread of the neutron beam and the finite apertures of the collimators, neutrons detected at a given counter and

crystal setting will be spread over a range of wavelength and angle. This means that we are averaging the intensity distribution over some volume in reciprocal space. The probability distribution in reciprocal space is the resolution function for the given conditions and the form of this function has been described by the authors elsewhere.¹¹ In order to take full advantage of the properties of the resolution function a crystal (φ) scan was chosen for the data collection. This also has the added advantage that the value of the magnetic form factor and the isotropic background do not vary during the scan. One or two θ - 2θ scans were also taken but due to these varying terms and the poorer resolution they were not analyzed in very great detail. The crystal was mounted with a $\{110\}$ axis vertical and φ scans were taken in the quadrant between $[001]$ and $[110]$ directions.

EXPERIMENTAL RESULTS

In Fig. 2 we show a number of 90° φ scans through $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ at various temperatures in the range T_c to $T_c + 200^\circ K$. These scans show the expected increase of the diffuse peak as the temperature of the sample is lowered towards the Néel temperature. No change in the diffuse peak was observed to be associated with the structural transition at $184^\circ K$. Even at room temperature, which is more than three times the Néel temperature, there is an appreciable peak and the diffuse scattering clearly extends over the entire Brillouin zone. A network of scans in reciprocal space at room temperature did not indicate any asphericity in the distribution other than that due to zone overlap. At lower temperatures, this overlap becomes less important with respect to the peak but it is

nevertheless present and the background was therefore always estimated from the scattering observed near the [001] direction. No significant effects of multiple scattering were observed.

Because of the low intensity observed at the higher temperatures a loose collimation was used initially and data were collected at temperatures from 288°K down to 88.3°K. However, the diffuse peak becomes so sharp near the Néel point that the resolution correction becomes exceedingly large. 10 degree scans were therefore repeated for temperatures from 97° to 88.7°K and from 88.7° to 88.2°K with increasingly tighter collimation, as permitted by the increased intensity in the peak. In each case the resolution function was determined experimentally by scanning the antiferromagnetic $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ peak at 84°K.

Data were also collected for φ scans at several temperatures below the Néel point in order to investigate the symmetry of the critical scattering about the transition point. By comparing the critical scattering at temperature increments equally above and below the Néel point it was observed that the angular dependence of the scattering about the $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ position was almost identical for $\Delta T = T - T_c = T_c - T$.

If we now return to the data presented in Fig. 1, we can subtract off the critical scattering below the Néel temperature giving the broken curve as the peak height of the antiferromagnetic reflection. The resultant curve indicates a power law temperature variation of the magnetization close to the Néel temperature with an exponent of about 0.37. This is of course based on the peak height and not the integrated intensity. Data in the region $1^\circ < T_c - T < 10^\circ\text{K}$ gave an exponent close

to the expected 0.33.

As noted previously, experiments were also carried out to investigate the magnitude of any inelasticity in the critical scattering. For these the detector of the diffractometer was replaced by an analyzing spectrometer. The analyzing crystal was then set to select the optimum wavelength neutrons from the sample and a sample φ scan was made at 88.7°K as before, keeping the analyzer fixed, thus adopting the "elastic" diffraction method discussed by Caglioti.¹² The excellent agreement between this "elastic" run and runs taken without an energy analyzing crystal can be taken as an indication of the quasi-elastic nature of the scattering in this temperature region. As a further check the energy distribution of the scattered neutrons was investigated at 88.7°K for two appreciably different values of q , the inelasticity being a function of q^2 . The resultant scaled intensities as a function of analyzer angle are shown in Fig. 3. Although the ratio of q^2 values is almost 10, no significant broadening of the curve is observed. A similar curve for diffuse scattering from a nickel powder sample is shown by the solid curve for reference. Any inelasticity present is clearly much less than the energy resolution of the instrument, estimated to be $< \approx 3\%$, and the data were therefore analyzed in terms of the quasi-elastic approximation.

ANALYSIS OF RESULTS

a. Near the Néel Temperature

We shall consider first the data collected with the tighter collimation systems in the temperature range 88.2° to 97°K, i.e. $1.002 < T/T_c < 1.1$,

over which the Ornstein-Zernike theory is expected to apply.

For the Ornstein-Zernike relation the cross section is inversely proportional to $\kappa_1^2 + q^2$ and it has therefore become customary to consider plots of the inverse intensity against q^2 , which should therefore be linear and provide the values of κ_1^2 on extrapolation. However, it should be pointed out that this type of plot may in fact be misleading since the achievement of a linear plot is not sufficient evidence that an adequate resolution correction has been made. This is particularly so because the effect of resolution is to elevate the curve in such a way that a considerable position may remain almost linear and the bending down which occurs at small values of $\kappa_1^2 + q^2$ is very similar to that predicted by such theories as that of Fisher.¹³ Hence both the estimation of κ_1^2 and observed deviations from the Ornstein-Zernike theory will depend on the reliability of the resolution function used.

For a given cross section $\sigma(\bar{q})$, mosaic spread $M(\bar{q}_m)$ and resolution function $R(\bar{q}, \varphi, \psi, 2\theta)$, where $\bar{q}_m$ is the vector displacement in reciprocal space from the position for optimum reflectivity of the sample, φ and ψ are the crystal angles and 2θ the counter angle, the observed intensity will be

$$I(\varphi, \psi, 2\theta) = \int \left\{ \int \sigma(\bar{q} + \bar{q}_m) M(\bar{q}_m) d\bar{q}_m \right\} R(\bar{q}, \varphi, \psi, 2\theta) d\bar{q} \quad (5)$$

The resolution function determined with the antiferromagnetic peak is

$$R'(\bar{q}, \varphi, \psi, 2\theta) = \int M(\bar{q}_m) R(\bar{q} + \bar{q}_m, \varphi, \psi, 2\theta) d\bar{q}_m \quad (6)$$

However, if the mosaic spread is small, as in this case, we can approximate equation (5) by

$$I(\varphi, \psi, 2\theta) = \int \sigma(\bar{q}) R'(\bar{q}, \varphi, \psi, 2\theta) d\bar{q} \quad (7)$$

By inverting this relationship it should then be possible to determine $\sigma(\bar{q})$ from a knowledge of I throughout a complete Brillouin zone. However, in practice, this is not known sufficiently accurately for the whole zone so that data collection was restricted to a single φ scan and the observed intensity was compared directly with the theoretical intensity calculated from equation (7) for an Ornstein-Zernike cross section, as given in equation (4).

The cross section contains two unknown parameters, κ_1 and A . By calculating the theoretical intensity distributions for various values of κ_1 we were able to determine the absolute values of κ_1 and the relative values of A to give the best fit to the experimental distributions. As expected A was found to vary slowly with increasing temperature. The staggered susceptibility is inversely proportional to $\kappa_1^2 A/A_c$ and we plot this quantity in Fig. 4 as a function of $T-T_c$ on a log-log plot. The straight line represents a slope of 4/3 showing the excellent agreement with a 4/3 power law over this temperature range. Plots of $(T-T_c)^{1/3}/\chi$ vs. T gave the value of the exponent as -1.33 ± 0.05 and Fig. 5 shows a linear plot of $\kappa_1^2 A/A_c$ against $(T-T_c)^{1.33}$. These figures also illustrate the excellent agreement between data taken with the two sets of collimators,

giving added confidence in the resolution functions used.

Figure 6 shows a typical intensity plot at 88.7°K . The top curve is the theoretical Lorentzian curve uncorrected for resolution and the lower curves are the theoretical curves for the experimental resolutions. The circles represent the smoothed experimental data. No significant deviations from the Ornstein-Zernike form were observed in this data which extend over the range $0 < q < 0.1 \text{\AA}^{-1}$.

b. Intermediate Temperature Range

The data taken with the looser collimation were analyzed as before and satisfactory agreement with the more accurate data was achieved over the temperature range up to 97°K and agreement with the Ornstein-Zernike form was observed out to about $q = 0.25 \text{\AA}^{-1}$. Above this temperature fairly good agreement with the calculated Ornstein-Zernike intensity distributions could be obtained up to a temperature of about $2T_c$. However, the scaling factor ($\propto T/A$) was found to remain remarkably constant and the inverse of the staggered susceptibility was found to deviate from the $4/3$ powerlaw, increasing too rapidly. It seems probable, therefore, that the main deviation from theory appears in the scaling factor which would be expected to increase appreciably over this temperature range, and that the observed intensities are therefore smaller than the theoretical. At $2T_c$ the correlation range has dropped to about the nearest neighbor distance.

Near room temperature ($>3T_c$) deviation from the Ornstein-Zernike theory is even more pronounced. However, an approximate calculation of the expression given by Sáenz⁸ using $J_1 = 3.1^{\circ}\text{K}$ and $J_2 = 0.3^{\circ}\text{K}$, was found

to give reasonable agreement for this temperature.

DISCUSSION

The observed magnetic scattering from KMnF_3 is found to agree remarkably well with that calculated from the Van Hove theory for quasi-elastic scattering, using an experimentally determined resolution function, in the temperature range $1.002 < T/T_c < 1.1$. The temperature dependence of the staggered susceptibility is found to follow very closely a $4/3$ power law, even down to temperatures very close to the Néel point. However, at these temperatures the peak becomes so sharp that interpretation of the data depends quite critically on the reliability of the resolution function and the relative uncertainty in the value of the staggered susceptibility becomes much larger. Although there is no evidence of deviations from the $4/3$ power law or the Ornstein-Zernike form near the Néel temperature it is clear that a satisfactory investigation of possible small deviations will require the use of very narrow angular collimation and this in turn will necessitate the use of higher neutron fluxes to provide a sufficient scattered intensity. Deviations from the Ornstein-Zernike form are observed, as expected, at higher temperatures.

In order to compare the present results for KMnF_3 with recent results for iron we have included in Fig. 4 the line of slope $4/3$ derived from the data given by Passell et al³ for corresponding values of T/T_c for iron. It can be seen that at a given T/T_c the correlation range in KMnF_3 is about three times that in iron. A further difference occurs in the angular range over which the Ornstein-Zernike form applies.

In iron there is evidence for a q^4 term becoming important at about $q = 0.08\text{\AA}^{-1}$ whereas in KMnF_3 the Ornstein-Zernike form is found to apply over a range of q of at least three times this value. This is consistent with the interpretation that in KMnF_3 the interaction is essentially a nearest neighbor coupling in contrast to the longer range interaction which occurs in the ferromagnetic metals.³

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FIGURE CAPTIONS

- Fig. 1. Observed temperature dependence of the $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ peak intensity close to the Néel temperature. The broken curve represents the antiferromagnetic Bragg peak intensity determined by assuming the critical scattering to be symmetrical about the Néel temperature.
- Fig. 2. φ scans through $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ at various temperatures in the range T_c to $T_c + 200^\circ\text{K}$.
- Fig. 3. Observed intensity as a function of analyzer misset for two different q values at 88.7°K . The solid curves represent a similar function obtained for a nickel powder sample.
- Fig. 4. Log-log plot of $k_1^2 A/A_c$ as a function of $T-T_c$. The straight lines have slopes of $4/3$, the upper line corresponding to the data of Passel et al. for iron for equivalent T/T_c values.
- Fig. 5. Linear plot of $k_1^2 A/A_c$ as a function of $(T-T_c)^{4/3}$.
- Fig. 6. Observed intensity distributions at 88.7°K for two experimental collimations. The upper curve represents the theoretical distribution and the lower curves the same distribution after correction for instrumental resolution.

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

FIGURE 6