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CRITICAL SCATTERING OF NEUTRONS FROM CoO (1)

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Résumé. — La distribution angulaire des neutrons diffusés par CoO immédiatement au-dessus de la température de Néel est observée et analysée dans l’approximation élastique, donnant la fonction de corrélation pour la direction des spins dans chaque sous-réseau. Les raies observées sont comparées avec celles calculées dans l’approximation du champ moléculaire.

Abstract. — The angular distribution of neutrons scattered from CoO just above the Néel temperature is observed and analyzed in the elastic approximation. It gives the correlation function for the direction of spins within each magnetic sublattice. The peaks observed are compared with those calculated from molecular field theory.

I. Introduction. — The critical magnetic scattering of neutrons from ferromagnetic substances has been the subject of several experimental and theoretical studies [1]. The phenomenon was explained by Van Hove as arising from the fluctuations of the magnetization near the Curie point. For the critical magnetic scattering of neutrons these fluctuations play the same role as do the density fluctuations in gases and liquids for the critical scattering of light and X-rays. Although no theory existed for the case of antiferromagnetic substances at the time when this work was taken up, we expected that the phenomenon would also appear in these substances near the Néel temperature. Earlier experiments by Shull et. al. [2] had also shown that residual magnetic order existed in antiferromagnetic substances above the Néel temperature. Since this work was first reported [3] theoretical treatments of the subject by de Gennes [4] and by Elliott and Marshall [5] have appeared and we will here also give a comparison with theory.

II. Theory. — In the case of ferromagnetics Van Hove has introduced the time and space dependent correlation function for spin directions \( \gamma(r, t) = < S_0(0) S_r(t) > \). The magnetic neutron scattering cross section can be expressed as the Fourier transform of this function. For temperatures close to \( T_c \) all the scattering becomes elastic and it is sufficient to use the static approximation where one neglects the time dependence. The scattering formulae may readily be extended to the case of antiferromagnetic crystals. As for the ferromagnetic case we get the formula:

\[
\frac{d\sigma}{d\Omega} = \frac{2}{3} \left( 1, \frac{\hat{e}^2}{mc^2} \right)^2 |F(k)|^2 \sum_r < S_0 S_r > e^{ikr} \tag{1}
\]

\( e \) and \( m \) are the electron charge and mass respectively, \( c \) the velocity of light, \( F(k) \) the magnetic form factor and \( k \) the scattering vector. Because of the staggered spin arrangement the correlation function is now an oscillating function whose absolute value represents the correlation for the spins within each magnetic sublattice. The general features of the scattering have been discussed in the molecular field approximation by de Gennes. He introduces the wavelength dependent susceptibility \( \chi(k) \) which describes the response to a weak external field of constant direction but of sinusoidal amplitude with wave vector \( k \). It is related to the spin correlations by

\[
\frac{\chi(k)}{\chi_1} = \frac{1}{S(S+1)} \sum_r < S_0 S_r > e^{ikr} \tag{2}
\]

\( \chi_1 \) being the paramagnetic susceptibility. What de Gennes does is to calculate \( \chi(k) \) in the molecular field approximation. Crystals with MnO structure have eight magnetic sublattices and he finds the expansion

\[
\frac{\chi_1}{\chi(k)} = 1 + \frac{\Delta - T_N}{T} s_1(k) + \frac{T_N}{T} s_4(k) \tag{3}
\]

where \( T_N \) and \( \Delta \) are the Néel and the paramagnetic Curie temperatures respectively. The functions \( s(k) \) depend upon the crystals and scattering geometry. If

\[
\Delta < 5 T_N \tag{4}
\]

the \( \chi_1/\chi(k) \) has a minimum for \( k = \tau_s \) where \( \tau_s \) is a reciprocal lattice vector corresponding to a super-lattice reflection. According to (1) and (2) this

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means that the scattering will be concentrated around $T_g$. The criterion (4) is the same as that arrived at by Anderson [6] for the stability of the same spin arrangement below $T_N$.

III. Experiment. — 1 Å neutrons were reflected from the (111) planes (4) of a single crystal of CoO. A plutonium filter was used to remove the 0.5 Å neutrons reflected into the same angle by the (222) planes. The crystal temperature could be varied by a combined dry ice cooling and an electric heating system. The temperature uncertainty due to gradients and drifting was estimated to be 0.2°.

The upper curve in figure 1 shows the temperature variation of the intensity in the (111) peak. The steep part of the curve extrapolates to zero at 19°C which agrees favourably with the tabulated values of the Néel temperature. The intensity which remains at higher temperatures has a wider angular distribution than the sharp Bragg peaks. When rotating the crystal 1.7° away from the Bragg setting we obtain the lower curve which shows the characteristic behaviour of critical scattering. This component belongs to the diffuse peak of critical scattering. For $T < T_N$ the diffuse peak is superimposed on the narrow Bragg peaks and remains in isolation for $T > T_N$. If we keep the counter fixed and rotate the crystal through the Bragg position we obtain the curves in figure 2 for different temperatures.

IV. Discussion. — The sharpening of the peaks as $T \to T_N$ displayed by figure 2 means that the region of coherence or the range of the spin correlations increases. More quantitative information can be obtained by Fourier inversion of the scattering data. Formula (1) tells that this will give the correlation function. If we assume that the crystal has a radial distribution of atoms with no directional effects in the spin correlations we get the curves on figure 3. The spin correlation is here defined by $[S(S + 1)]^{-1} < S_0 S_r >$ and should be equal to one for $r = 0$. When our value is lower it means that we have not been able to isolate the whole critical intensity from the background. It is seen from the figure that the short range correlations are practically unchanged over a temperature interval of 30° whereas the long range part changes drastically. This behaviour is reflected strongly in the curves of figure 2 since the number...
of atoms which contributes to the scattering increases as the third power of the correlation range. At \( T_s + 31^\circ \) the range is short enough that we can synthesize the peak by means of (1) using the real crystal structure. This is done in figure 4. Taking only the correlation between nearest neighbours into account gives too broad a peak, reasonable agreement is obtained by extending the calculation to the shell of 10th nearest neighbours. In the calculations the correlation was given the value 60 % between nearest neighbours and decreasing to 10 % between 5th n.n. and 4 % between 10th n.n.

If we apply the formulae (1) to (3) we can see to what extent the molecular field calculations reproduce the scattering data. This is shown in figure 5. Some of the discrepancy could be due to the fact that we have assumed that all the scattering is elastic. The values of \( \Delta \) reported [7], [8] differ, the effect of \( \Delta \) on our theoretical curves is however very small.

Elliott and Marshall [5] have carried out a Bethe-Peierls type calculation of the problem. They consider only the case of two sublattices however, and no comparison with their results is given here.

Our data give the conclusive result that correlation effects exist in antiferromagnetic substances for \( T > T_N \). The correlations are fairly well described by molecular field theory.

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[1] For references, see e.g. LOWDE (R. D.), Rev. Mod. Physics, 1958, 30, 69.