

## Magnetic excitations above the critical temperature\*

S. H. Liu

Ames Laboratory—Energy Research and Development Administration and Department of Physics, Iowa State University, Ames, Iowa 50010  
(Received 24 March 1975)

We present a simple but physically motivated approximation scheme for the dynamical spin correlation function of the Heisenberg model above the critical temperature. Built upon the spin-diffusion theory, it describes the short-wavelength and high-frequency excitations as magnonlike modes which exist within clusters of highly correlated spins. The modes are damped by both the finite size and the finite lifetime of the clusters. The agreement between the theory and the neutron scattering data on a variety of systems is reasonably good.

Inelastic neutron scattering experiments have been carried out on many magnetic materials above the critical temperature.<sup>1-8</sup> The results may be summarized by the criterion first proposed by Marshall.<sup>9</sup> When the wavelength of the excitation is long compared with the spin correlation length  $\lambda$ , the mode is well described by the spin-diffusion theory.<sup>10-14</sup> When the wavelength is short compared with  $\lambda$ , the spectrum of the excitations bears remarkable resemblance to the magnon modes below the critical temperature. (The critical behavior of chromium is better described by the itinerant theory.<sup>3,4</sup>) There have been many theoretical discussions on the latter type of modes.<sup>15-23</sup> Most of these works attempt to extend the spin diffusion theory by more careful mathematical methods.

In this paper we discuss a very different approach to this problem. A preliminary account of our method was reported elsewhere.<sup>24</sup> Instead of trying to improve on the mathematical technique, we work from the physical nature of the short-wavelength modes. Although there is no long-range order above the critical temperature, the system retains various degrees of short-range order up to rather high temperatures. Thus if the wavelength of an excitation is less than the average size of the correlated cluster  $\lambda$ , or  $q\lambda > 1$ , where  $q$  is the wave vector, the mode can make a few complete oscillations within a collection of correlated spins. Then the nature of this mode should be very similar to the magnon mode in the ordered phase. The finite size of the cluster of spins gives rise to a damping mechanism of the mode. In addition, the average cluster decays away due to spin diffusion with a lifetime given by  $\tau = \lambda^2/\Lambda$ , where  $\Lambda$  is the spin-diffusion constant. Thus it is also necessary for the frequency  $\omega$  of the mode to satisfy  $\omega\tau > 1$  in order to have a well-defined mode. We will show that the shape and width of the neutron scattering response function depend on the interplay between the two conditions  $q\lambda > 1$  and  $\omega\tau > 1$ .

We will demonstrate our method by using the

simple ferromagnet as an example. The inelastic neutron scattering cross section is proportional to the dynamical spin correlation function<sup>10</sup>

$$G(\vec{q}, \omega) = \sum_j \int_{-\infty}^{\infty} \langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle e^{i\vec{q} \cdot \vec{R}_{ij} - i\omega t} dt, \quad (1)$$

where  $\vec{R}_{ij}$  is the vector distance between the spins  $\vec{S}_i$  and  $\vec{S}_j$ . In the spin-diffusion region, i. e.,  $q\lambda \ll 1$ , the two-spin correlation function has the form

$$\langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle \cong S(S+1) \rho(\vec{R}_{ij}, t), \quad (2)$$

where  $\rho(\vec{R}, t)$  is the normalized correlation function satisfying  $\rho(0, 0) = 1$ . The spin-diffusion theory gives the Fourier transform<sup>10,11</sup> of  $\rho(\vec{R}, t)$ ,

$$\rho(\vec{q}, \omega) = \frac{C}{q^2 + \kappa^2} \frac{2\Lambda q^2}{\Lambda^2 q^4 + \omega^2}, \quad (3)$$

where  $C = kT/2D$ ,  $\kappa = \lambda^{-1}$ , and  $D$  is the stiffness constant.

Now we consider the situation where  $q\lambda > 1$ . As pointed out in Ref. 24 the dominant contributions to the neutron scattering response function come from pairs of spins which are roughly one wavelength apart. When  $q\lambda > 1$ , it is highly likely that the pairs will fall within the same cluster of correlated spins. We propose to describe the dynamics of such pairs by the spin-wave theory. If two spins fall in two different clusters, we use the spin-diffusion description for the dynamics of the pair. The probability that an arbitrary spin pair is correlated is given by the spin-diffusion correlation function  $\rho(\vec{R}, t)$ . Hence our approximation scheme is as follows:

$$\langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle \cong \langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle_{\text{SW}} \rho(\vec{R}_{ij}, t) + \langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle_{\text{SD}} [1 - \rho(\vec{R}_{ij}, t)], \quad (4)$$

where the subscript SW indicates that the two-spin correlation function is to be calculated by the spin-wave theory and SD indicates that the spin-diffusion theory is employed. The first term de-

pend on the model, but the second term is simply

$$S(S+1)\rho(\vec{R}_{ij}, t)[1 - \rho(\vec{R}_{ij}, t)]. \quad (5)$$

A simple isotropic ferromagnet has the spin-wave dispersion relation  $\omega_q = Dq^2$ , where  $D$  is the stiffness constant. For each cluster of ordered spins we can define a local  $z$  axis which is in the direction of the ordered moments. Then

$$\begin{aligned} \langle \vec{S}_i(t) \cdot \vec{S}_j(0) \rangle_{sw} &= \langle S_i^z(t) S_j^z(0) \rangle_{sw} \\ &+ \frac{1}{2} \langle S_i^+(t) S_j^-(0) \rangle_{sw} + \frac{1}{2} \langle S_i^-(t) S_j^+(0) \rangle_{sw} \end{aligned} \quad (6)$$

and

$$\langle S_i^z(t) S_j^z(0) \rangle_{sw} \cong S^2 - \frac{2S}{N} \sum_{\vec{q}'} n_{q'},$$

$$\langle S_i^+(t) S_j^-(0) \rangle_{sw} \cong \frac{2S}{N} \sum_{\vec{q}'} (n_{q'} + 1) \exp(i\vec{q}' \cdot \vec{R}_{ij} - i\omega_{q'} t), \quad (7)$$

$$\langle S_i^-(t) S_j^+(0) \rangle_{sw} \cong \frac{2S}{N} \sum_{\vec{q}'} n_{q'} \exp(i\vec{q}' \cdot \vec{R}_{ij} + i\omega_{q'} t),$$

where  $n_{q'} = (e^{\beta\omega_{q'}} - 1)^{-1}$  and where  $N$  is the total number of spins. We have ignored the two- and more-than-two-magnon terms. Putting these results into Eq. (4) and taking the Fourier transform, we obtain

$$\begin{aligned} G(\vec{q}, \omega) &= \left( S^2 - \frac{2S}{N} \sum_{\vec{q}'} n_{q'} \right) \rho(\vec{q}, \omega) + \frac{S}{N} \sum_{\vec{q}'} [n_{q'} \rho(\vec{q} - \vec{q}', \omega + \omega_{q'}) + (n_{q'} + 1) \rho(\vec{q} - \vec{q}', \omega - \omega_{q'})] \\ &+ S(S+1) \left( \rho(\vec{q}, \omega) - \frac{1}{N} \sum_{\vec{q}'} \int \frac{d\omega'}{2\pi} \rho(\vec{q}', \omega') \rho(\vec{q} - \vec{q}', \omega - \omega') \right). \end{aligned} \quad (8)$$

The first term is the central peak term because at any finite  $\vec{q}$  the function  $\rho(\vec{q}, \omega)$  peaks up at  $\omega = 0$ . In the limit of ordered spins this term becomes elastic ( $\omega = 0$ ) and describes the magnetic Bragg scattering. The next term consists of the excitation and deexcitation of single magnons, and these one-magnon lines are broadened by folding with the spin-diffusion correlation function. The last term is a background term contributed by loosely correlated pairs.

We now extend the result to antiferromagnetic systems. The spin correlation function which exhibits the critical behavior is the Fourier transform  $\rho(\vec{Q}, 0)$ , where  $\vec{Q}$  is the characteristic wavelength of the spin ordering. For a simple two-lattice antiferromagnetic system  $\vec{Q}$  is one-half of a reciprocal-lattice vector. A suitable measure of the degree of order is  $\rho(\vec{R}_{ij}, t) \cos(\vec{Q} \cdot \vec{R}_{ij})$ , which is then used in place of  $\rho(\vec{R}_{ij}, t)$  in Eq. (4). The result of the calculation is as follows:

$$\begin{aligned} G(\vec{q}, \omega) &= \left( S^2 - \frac{2S}{N} \sum_{\vec{q}'} [n_{q'} \cosh^2 \theta_{q'} + (n_{q'} + 1) \sinh^2 \theta_{q'}] \right) \frac{1}{2} [ \rho(\vec{q} + \vec{Q}, \omega) + \rho(\vec{q} - \vec{Q}, \omega) ] \\ &+ \frac{S}{2N} \sum_{\vec{q}'} \{ (n_{q'} + 1) [ e^{2\theta_{q'}} \rho(\vec{q} - \vec{q}' + \vec{Q}, \omega - \omega_{q'}) + e^{2\theta_{q'}} \rho(\vec{q} - \vec{q}' - \vec{Q}, \omega - \omega_{q'}) + 2e^{-2\theta_{q'}} \rho(\vec{q} - \vec{q}', \omega - \omega_{q'}) ] \\ &+ n_{q'} [ e^{2\theta_{q'}} \rho(\vec{q} - \vec{q}' + \vec{Q}, \omega + \omega_{q'}) + e^{2\theta_{q'}} \rho(\vec{q} - \vec{q}' - \vec{Q}, \omega + \omega_{q'}) + 2e^{-2\theta_{q'}} \rho(\vec{q} - \vec{q}', \omega + \omega_{q'}) ] \} \\ &+ (\text{background term}). \end{aligned} \quad (9)$$

In the above equation

$$\omega_q = 2S [A(\vec{q}) - B(\vec{q})]^{1/2}, \quad \tan 2\theta_q = [A(\vec{q}) - B(\vec{q})] / [A(\vec{q}) + B(\vec{q})],$$

$$A(\vec{q}) = J(\vec{Q}) - J(\vec{q}), \quad B(\vec{q}) = J(\vec{Q}) - \frac{1}{2} J(\vec{Q} + \vec{q}) - \frac{1}{2} J(\vec{Q} - \vec{q}),$$

where  $J(\vec{q})$  is the Fourier transform of the exchange interaction and both  $\vec{q}$  and  $\vec{q}'$  are measured from  $\vec{Q}$ .

A special case of the helical antiferromagnet is the linear-chain model with nearest-neighbor interaction.<sup>25</sup> It is customary to express the wave vector in units of inverse lattice parameter. Then  $Q = \pi$ ,  $J(q) = 2J \cos q$ ,  $\tan 2\theta_q = \cos q$ , etc. Thus,

$$\begin{aligned} G(q, \omega) &= \left( S^2 - \frac{2S}{N} \sum_{q'} [n_{q'} (\csc q' + 1) + (n_{q'} + 1) (\csc q' - 1)] \right) \rho(q - \pi, \omega) + \frac{S}{N} \sum_{q'} \{ (n_{q'} + 1) [A_q^{(+)} \rho(q - q' + \pi, \omega - \omega_{q'}) \\ &+ A_q^{(-)} \rho(q - q', \omega - \omega_{q'})] + n_{q'} [A_q^{(+)} \rho(q - q' + \pi, \omega + \omega_{q'}) + A_q^{(-)} \rho(q - q', \omega + \omega_{q'})] \} + (\text{background term}), \end{aligned} \quad (10)$$

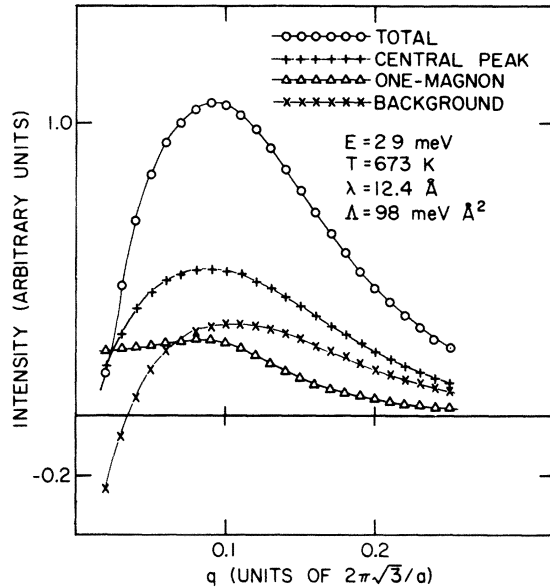


FIG. 1. Calculated line shape for a constant-energy scan on nickel above the Curie temperature (631 K).

where  $A_q^{(\pm)} = c \sin q \pm \cot q$  and the sum on  $q'$  is carried out in the magnetic Brillouin zone  $-\frac{1}{2}\pi < q \leq \frac{1}{2}\pi$ . There is a well-known infrared divergence in the simple spin-wave theory of linear-chain models. The divergence appears in all of the sums on  $q'$  at  $q' = 0$ . However, it is very easy to show that in Eq. (10) the divergent part of the central peak term cancels exactly the corresponding divergent part of the one-magnon term. Consequently, one can remove both divergences by restricting the sum on  $q'$  by  $\lambda^{-1} \leq |q| \leq \frac{1}{2}\pi$ .

At this point we would like to point out an uncertainty in the theoretical results Eqs. (8)–(10). The peaked structure of  $\rho(\vec{q}, \omega)$  implies that the major contribution to the one-magnon terms come from the region  $\vec{q}' \cong \vec{q}$  and  $\omega' \cong \omega$ . For  $|\vec{q} - \vec{q}'| < \lambda^{-1}$  we can justifiably use the form of the spin-diffusion correlation function in Eq. (3). However, in the three-dimension model the sum on  $\vec{q}'$  converges slowly, so that it may be necessary to include  $\vec{q}'$  vectors outside this range. The situation is worse for the central peak and background terms because the region of interest is outside the range of validity of Eq. (3). This uncertainty arises from the fact that we take the spin-diffusion theory as given and build a theory of short-lived magnons on it. We make no attempt to extend the spin-diffusion theory to shorter wavelengths, because in doing so we would encounter all of the difficulties mentioned in the previous theoretical works.<sup>15–23</sup> We will show that in spite of this uncertainty the theory still allows a great deal of understanding of the neutron scattering results.

It is easy to verify that the theory fails in the

infinite-temperature limit.<sup>16,21</sup> This is not a serious drawback, because our main interest is to understand the neutron data over a finite temperature range above the critical point.

In our numerical calculation for the simple ferromagnet we put in the spin stiffness constant, correlation length, and spin-diffusion constant of nickel.<sup>1,7</sup> There are reasons why one should not apply the Heisenberg model to nickel; for example, the rapid broadening of the magnon line around 80 meV can be understood only on the basis of the band model.<sup>26</sup> However, aside from this fact our theory is not sensitive to the model. Away from the critical region we assume

$$\kappa = \kappa_0(T/T_c - 1)^{1/2}, \quad \Lambda = \Lambda_0(T/T_c - 1)^{1/2},$$

where  $T_c = 631$  K,  $\kappa_0 = 0.312 \text{ \AA}^{-1}$ ,  $\Lambda_0 = 380 \text{ meV \AA}^2$ , and  $D = 280 \text{ meV \AA}^2$ . We also take  $S = \frac{1}{2}$  for convenience. A typical line shape for a constant energy scan is shown in Fig. 1, where we show the total line shape and the contributions from the central peak, one-magnon, and background terms. The three components are equally important in determining the shape and width of the response function. In a constant energy scan the central peak splits into two peaks because  $\rho(\vec{q}, \omega)$  is zero at  $\vec{q} = 0$  and  $\omega \neq 0$ . We used the same input data to simulate a constant  $q$  scan, and the line shape showed only one broad peak centered at  $\omega = 0$ . Therefore we do not regard the lines observed in Refs. 7 and 8 as convincing indications of propagating modes above the critical temperature.

In Fig. 2 we compare the theoretical line shape with the experimental data at two temperatures.<sup>7</sup> The data includes an instrumental resolution whose size is given in Ref. 8 as  $0.05 \text{ \AA}^{-1}$  or  $0.02$  reduced units. This is rather small compared with the total width of the peak and is comparable to the discrepancy between the theory and the data. We also assumed that all of the experimental background came from scattering by disordered spins. Since this background was fully accounted for in our calculation, we made no adjustment of the experimental background. One can see from Fig. 2 that the comparison between theory and experiment is reasonably good. On the same graph we also show the predicted line shape at 1000 K. The calculated line is somewhat narrow compared with the data.<sup>8</sup> This may be due to the simplified assumptions we made for the temperature dependence of the correlation length and the spin diffusion constant.

There appears to be very little change in intensity as well as in linewidth with increasing temperature. This is in qualitative agreement with the observations.<sup>8</sup> To understand this phenomenon we must consider many factors. Since  $\omega = Dq^2$  for a simple ferromagnet, we have

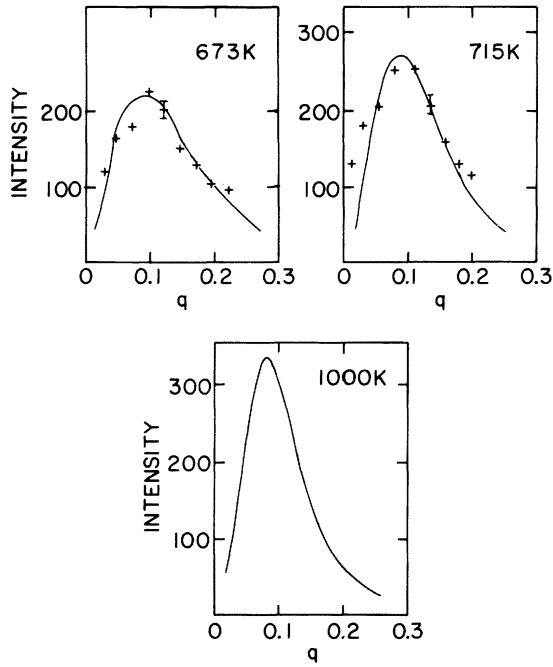


FIG. 2. Comparison between theoretical line shape and experimental data for nickel above the Curie temperature. The wave vector is in units of  $2\pi\sqrt{3}/a$ , and the intensity is in units of neutron counts per number of monitor counts, as in Ref. 7.

$$\omega\tau = (D/\Lambda)(q\lambda)^2.$$

The ratio  $D/\Lambda$  is of order unity; thus the conditions  $q\lambda > 1$  and  $\omega\tau > 1$  more or less go together. When the temperature increases, the correlation length decreases. This has the tendency of broadening the line. On the other hand, the spin diffusion also becomes faster, with the effect of cutting out the influence of the small clusters and leaving only the large clusters to respond to the excitation. This explains why the line does not actually broaden. Although the number of large clusters decreases with increasing temperature, the boson distribution function increases so that there is no net loss of intensity. The neutron scattering data on iron have the same qualitative features.<sup>8</sup>

In all of these calculations we used the simple form of the spin diffusion correlation function. Because of the uncertainty in the form of this function for large  $\vec{q}$ , we performed a series of tests by assuming different forms for the correlation function. One possible form is the Gaussian function

$$\rho(\vec{q}, \omega) = (c/\kappa^2) e^{-q^2/\kappa^2} 2\Lambda q^2 / (\Lambda^2 q^4 + \omega^2); \quad (11)$$

another form is the cutoff Lorentzian where we use the form in Eq. (3) for  $q < q_c$  and  $\rho(\vec{q}, \omega) = 0$  for  $q > q_c$ .  $q_c$  being a parameter. The results show that the line shapes change very slightly when the

Gaussian form is used, and hardly at all when the cutoff Lorentzian form is used with  $q_c \sim 0.1 \text{ \AA}^{-1}$ . This choice of  $q_c$  corresponds to the inverse of the range of spin interaction in nickel. Physically both the Gaussian and the cutoff Lorentzian forms of the correlation function suppress the number of those clusters that are much smaller than the average size  $\lambda$ . Thus it appears that these smaller clusters decay away sufficiently rapidly so that the detailed shape of  $\rho(\vec{q}, \omega)$  is not very important.

We then applied the theory to a simple antiferromagnet,  $\text{RbMnF}_3$ . The magnetic lattice of this material has a simple cubic structure with lattice spacing  $a = 4.186 \text{ \AA}$ .<sup>27</sup> The important exchange interaction is between nearest neighbors with  $J = 1.99 \text{ K}$ .<sup>28</sup> The spin for Mn ions is  $S = \frac{5}{2}$ , and the Néel temperature is  $T_N = 83 \text{ K}$ .

Tucciarone *et al.* followed up their early work<sup>14</sup> by a detailed study of the inelastic neutron scattering line shape at the critical temperature.<sup>29</sup> The authors analyzed the data in terms of the dynamic scaling theory<sup>13</sup> and found very good agreement. At the critical temperature the spin-diffusion correlation function is given by

$$\rho(\vec{Q} + \vec{q}, \omega) \cong (A/q^{2-\eta}) \Gamma(q) / [\Gamma^2(q) + \omega^2], \quad (12)$$

where  $\vec{q}$  is a small deviation from  $\vec{Q}$ ,  $\eta = 0.05$ ,  $\Gamma(q) = Cq^{3/2}$ , and  $A$  is a normalization constant. The spin correlation length is infinitely large.

It is difficult to apply the theory at the critical temperature for two reasons: (i) the nonintegral exponents in expression for  $\rho(\vec{q}, \omega)$  makes the integration of  $\vec{q}'$  in Eq. (9) impossible to work out; and (ii) the expression in Eq. (12) only holds for small  $\vec{q}$ , and the correlation function for large  $\vec{q}$  is unknown. Nevertheless, some progress can be made by observing that the spin-diffusion correlation function peaks sharply at  $\vec{q}' = \vec{q}$  and that at the critical temperature the major damping mechanism for the magnons is the magnon-magnon interaction. Thus we may ignore the Fourier component of the correlation function for wave vectors  $q - q' \pm Q$  completely and approximate

$$\rho(\vec{q} - \vec{q}', \omega - \omega_{q'}) \cong \delta(\vec{q} - \vec{q}') 2\Gamma'(q) / [(\omega - \omega_q)^2 + \Gamma'^2(q)], \quad (13)$$

where  $\Gamma'(q) = Dq^2$  as given by the hydrodynamic theory.<sup>30</sup> For small  $q$  the factor  $e^{-2\theta q} \cong 2\sqrt{3} (aq)^{-1}$  and the magnon frequency  $\omega_q = cq$ , where  $c = 2\sqrt{3} J/a$ . These enabled us to find

$$G(\vec{q}, \omega) \cong \frac{A}{q^{2-\eta}} \frac{\Gamma(q)}{\Gamma^2(q) + \omega^2} + \frac{2\sqrt{3}S}{aq} \times \left( \frac{2(n_q + 1)\Gamma'(q)}{(\omega - \omega_q)^2 + \Gamma'^2(q)} + \frac{2n_q\Gamma'(q)}{(\omega + \omega_q)^2 + \Gamma'^2(q)} \right). \quad (14)$$

The background term was found to be negligible.

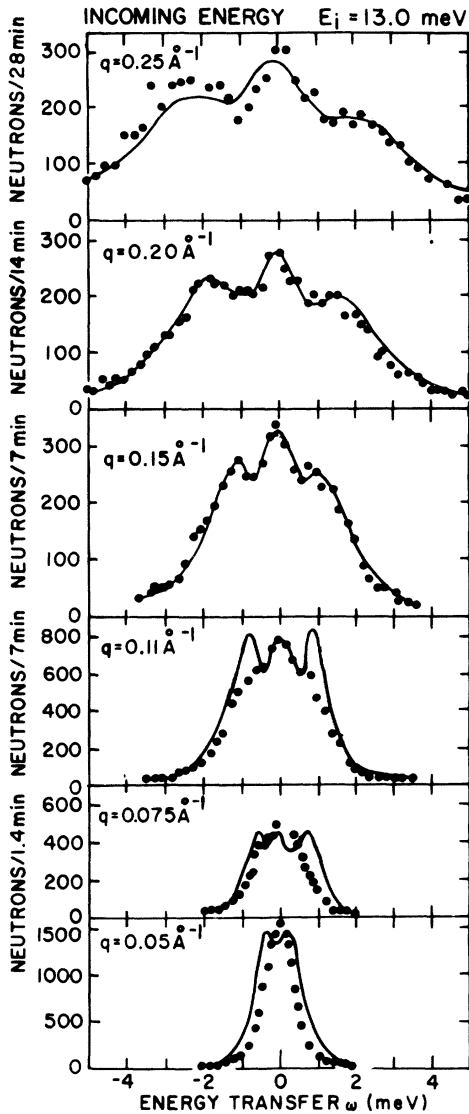


FIG. 3. Comparison between theoretical and experimental line shapes for constant- $q$  scans on  $\text{RbMnF}_3$  at the critical temperature (83 K).

There are four parameters,  $A$ ,  $C$ ,  $D$ , and the scale factor, in the theory. We calculated the theoretical line shape for  $q=0.25 \text{ \AA}^{-1}$ , folded with the resolution function given in Ref. 29, and adjusted the parameters for a best fit. The magnon velocity was not adjusted. We then used the same set of parameters to calculate the line shapes for  $q=0.2, 0.15, 0.11, 0.075$ , and  $0.05 \text{ \AA}^{-1}$ . The results are shown in Fig. 3 together with the data from Ref. 29. One can see that for large values of  $q$  the theory reproduces the data very well, but for small- $q$  values the magnon peaks stand out too sharply. This discrepancy arises because our theory does not satisfy the dynamic scaling law,<sup>15</sup> which says that both  $\omega_q$  and  $\Gamma'(q)$  should be proportional to

$q^{3/2}$ . Compared with our theory with  $\omega_q \propto q$  and  $\Gamma'(q) \propto q^2$ , the magnon frequency in our theory is too high and the damping too low for small values of  $q$ . This results in three distinct peaks in the theoretical curve even for the smallest  $q$ . Therefore the failure of our theory in the small- $q$  region comes about because the critical fluctuation alters the nature of the modes in a fundamental manner.<sup>31-33</sup> It is beyond the scope of our theory to elucidate this effect.

We have also tested the theory on the one-dimensional antiferromagnetic system TMMC [ $(\text{CD}_3)_4\text{NMnCl}_3$ ]. In this material the ordering temperature is suppressed by the dimensionality so that there is no evidence of phase transition down to 1.5 K.<sup>5</sup> All of the inelastic neutron scattering measurements were done above the critical temperature, which is 0 K for a linear chain. The static correlation function for a classical Heisenberg chain was given by Fisher.<sup>25</sup> It appears that the spin value of  $\frac{5}{2}$  for the Mn ions is large enough for the classical theory to work very well.<sup>5</sup> On the other hand, the critical dynamics of the system has not been adequately explored. Therefore we chose to approximate

$$\rho(q, \omega) = 2\pi\rho(q)\delta(\omega),$$

with

$$\rho(q) = (1 - u^2)/(1 + u^2 + 2u \cos q),$$

where  $u = \coth(2J/kT) - kT/2J$  and  $J = 7.7 \text{ K}$ . This approximation has two effects, i.e., it underestimates the linewidth by ignoring the decay rate of the correlated clusters and it prevents us from drawing any conclusion about the central peak. Numerical calculation shows that the first effect is probably not important because the shapes of the one-magnon lines are well accounted for over a wide range of temperature and wave vector. The second effect is beyond our concern at this moment because there exists no data in the central peak region.

In Figs. 4 and 5 we compare the calculated line shape with the data. To make a realistic comparison we folded the calculated curves with a resolution function having a full width of 0.3 meV. Because we concentrated on comparing the line shape, we used an adjustable intensity scale factor. The variation of the intensity scale factor is less than a factor of 2 for two values of  $q$  and five values of the temperature. The theoretical line shapes fit the data exceedingly well.

The temperature dependence of the lines in Figs. 4 and 5 is strikingly different from that of the lines in Fig. 2. The response function for TMMC broadens and the peak value decreases steadily with increasing temperature. As mentioned before the line broadening is due to the effect of the small

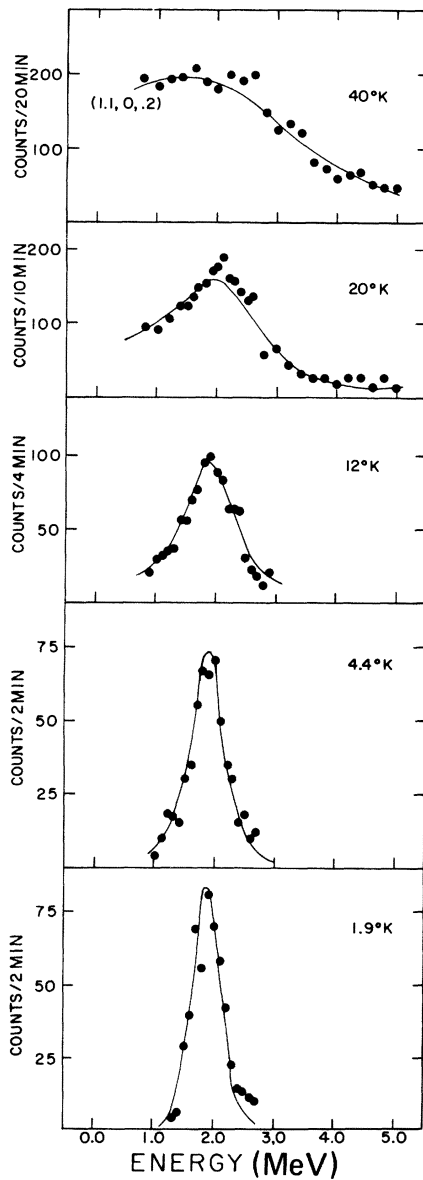


FIG. 4. Comparison between theoretical and experimental line shape for constant- $q$  scans on TMMC. The wave vector is  $0, 1(2\pi/a)$  measured from the magnetic reciprocal-lattice point  $Q = \pi/a$ .

clusters which do not decay away very fast. In the meantime the temperature is low compared with the magnon energies; thus there is a less dramatic increase of the magnon population with increasing temperature, certainly not enough to compensate for the decrease due to the reduced number of large clusters. Hence the difference between the dispersion relations of the simple ferromagnet and the linear antiferromagnet can account for the difference in the temperature dependence of their neutron scattering lines.

The neutron scattering lines of the magnetic ex-

citons in cubic Pr and  $\text{Pr}_3\text{Tl}$  show a temperature dependence similar to the magnetic excitations in TMMC.<sup>6</sup> The dispersion relation of the magnetic excitons has weak dependence on  $q$ . Therefore we expect the  $\omega\tau$  effect to be weak and the  $q\lambda$  effect to dominate. The observed magnetic excitons above the critical temperature in the range  $q \gtrsim 0.4 \text{ \AA}^{-1}$  is very likely a result of the short-range order. The question of soft modes in these materials can be settled only by performing the experiment at much smaller values of  $q$ .

Finally, we may draw from our numerical experience some general conclusions about the nature of the magnetic excitations above the critical temperature. For an isotropic ferromagnet with  $\omega_q = Dq^2$ , the spin-diffusion mode and the magnon mode have nearly the same energy. The result is a strong damping of the magnon mode, so that the most likely line shape for a constant- $q$  scan is a

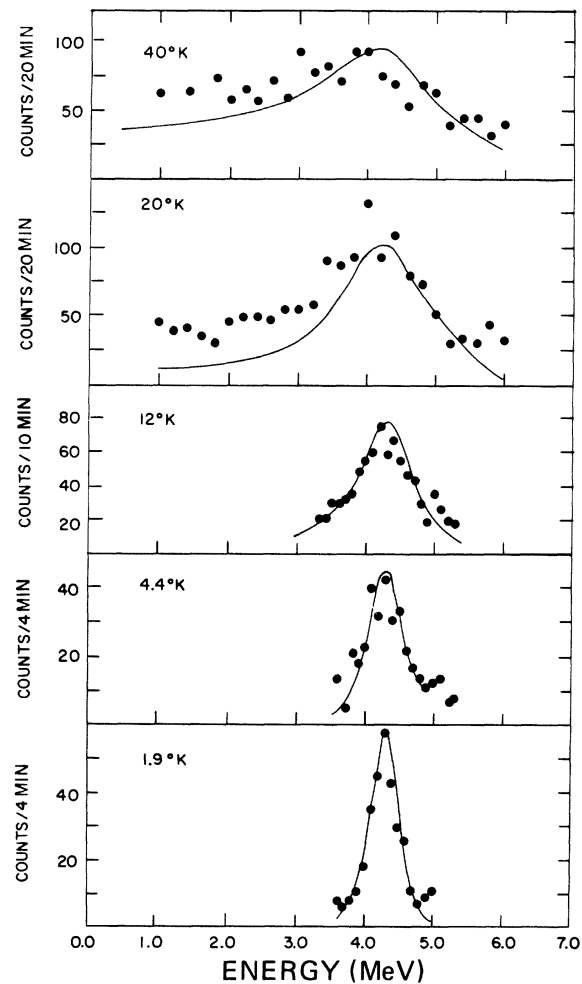


FIG. 5. Comparison between theoretical and experimental line shape for constant- $q$  scans on TMMC. The wave vector is  $0, 25(2\pi/a)$  measured from the magnetic reciprocal-lattice point  $Q = \pi/a$ .

broad diffusion peak at  $\omega = 0$ . For a constant- $\omega$  scan the diffusion peak splits into two. Recent data on EuO given by Passel *et al.* is in agreement of this conclusion.<sup>34</sup> For an isotropic antiferromagnet with  $\omega_q = cq$ , the magnon energy stands above the energy of the spin diffusion mode so that

a constant- $q$  scan with sufficient resolution may reveal a three-peak structure. If the anisotropy of a ferromagnetic or antiferromagnetic system is high, so that  $\omega$  is nearly independent of  $q$ , the likelihood of seeing a three-peak structure is even more favorable.<sup>35</sup>

\*Prepared for the Energy Research and Development Administration under contract No. W-7405-eng-82.

- <sup>1</sup>V. J. Minkiewicz, M. F. Collins, R. Nathans, and G. Shirane, *Phys. Rev.* **182**, 624 (1968); M. F. Collins, V. J. Minkiewicz, R. Nathans, L. Passell, and G. Shirane, *ibid.* **179**, 417 (1969).
- <sup>2</sup>R. Nathans, R. Menzinger, and S. J. Pickart, *J. Appl. Phys.* **39**, 1237 (1968).
- <sup>3</sup>S. K. Sinha, S. H. Liu, L. D. Muhlestein, and N. Wakabayashi, *Phys. Rev. Lett.* **23**, 311 (1960).
- <sup>4</sup>J. Als-Nielsen, J. D. Axe, and G. Shirane, *J. Appl. Phys.* **42**, 1666 (1971).
- <sup>5</sup>M. T. Hutchings, G. Shirane, R. J. Birgeneau, and S. L. Holt, *Phys. Rev. B* **5**, 1999 (1972).
- <sup>6</sup>R. J. Birgeneau, J. Als-Nielsen, and E. Bucher, *Phys. Rev. Lett.* **27**, 1530 (1971).
- <sup>7</sup>H. A. Mook, J. W. Lynn, and R. M. Nicklow, *Phys. Rev. Lett.* **30**, 556 (1973).
- <sup>8</sup>H. A. Mook, J. W. Lynn, and R. M. Nicklow, *AIP Conf. Proc.* **18**, 781 (1974); J. W. Lynn, *Phys. Rev. B* **11**, 2624 (1975).
- <sup>9</sup>W. C. Marshall, in *Conference on Critical Phenomena*, Natl. Bur. Stand. Miscellaneous Publication No. 273 (U. S. GPO, Washington, D. C., 1965), p. 135.
- <sup>10</sup>L. van Hove, *Phys. Rev.* **95**, 249 (1954).
- <sup>11</sup>P. de Gennes, *J. Phys. Chem. Solids* **4**, 223 (1958).
- <sup>12</sup>H. Mori and K. Kawasaki, *Prog. Theor. Phys.* **27**, 529 (1962).
- <sup>13</sup>B. I. Halperin and P. C. Hohenberg, *Phys. Rev. Lett.* **19**, 700 (1967); *Phys. Rev.* **177**, 952 (1969).
- <sup>14</sup>A. Tucciarone, J. M. Hastings, and L. M. Corliss, *Phys. Rev. Lett.* **26**, 257 (1971).
- <sup>15</sup>P. Resibois and M. DeLeneer, *Phys. Rev.* **178**, 819 (1969).
- <sup>16</sup>M. Blume and J. Hubbard, *Phys. Rev. B* **1**, 3815 (1970).
- <sup>17</sup>F. B. McLean and M. Blume, *Phys. Rev. B* **7**, 1149 (1973).
- <sup>18</sup>F. Carboni and P. M. Richards, *Phys. Rev.* **177**, 889 (1969).
- <sup>19</sup>H. Tomita and H. Mashiyama, *Prog. Theor. Phys.* **48**, 1133 (1972).
- <sup>20</sup>S. W. Lovesey and R. A. Meserve, *J. Phys. C* **6**, 79 (1973).
- <sup>21</sup>G. F. Reiter, *Phys. Rev. B* **5**, 222 (1971); **7**, 3325 (1973).
- <sup>22</sup>C. W. Myles and P. A. Fedders, *Phys. Rev. B* **9**, 4872 (1974).
- <sup>23</sup>S. A. Scales and H. A. Gersch, *Phys. Rev. Lett.* **28**, 917 (1972).
- <sup>24</sup>S. H. Liu and P. A. Swanson, *AIP Conf. Proc.* **24**, 163 (1975).
- <sup>25</sup>M. E. Fisher, *Am. J. Phys.* **32**, 343 (1964).
- <sup>26</sup>J. B. Sokoloff, *Phys. Rev. Lett.* **31**, 1417 (1973).
- <sup>27</sup>O. Beckman and K. Knox, *Phys. Rev.* **121**, 376 (1961).
- <sup>28</sup>D. T. Teaney, M. J. Freiser, and R. W. H. Stevenson, *Phys. Rev. Lett.* **9**, 212 (1962).
- <sup>29</sup>A. Tucciarone, H. Y. Lau, L. M. Corliss, A. Delapalme, and J. M. Hastings, *Phys. Rev. B* **4**, 3206 (1971).
- <sup>30</sup>B. I. Halperin and P. C. Hohenberg, *Phys. Rev.* **188**, 898 (1969).
- <sup>31</sup>K. Kawasaki, *Phys. Rev.* **150**, 291 (1966); *Ann. Phys. (N. Y.)* **61**, 1 (1970).
- <sup>32</sup>K. Kawasaki and M. Tanaka, *Proc. Phys. Soc. Lond.* **90**, 791 (1967).
- <sup>33</sup>L. P. Kadanoff and J. Swift, *Phys. Rev.* **166**, 89 (1968); J. Swift and L. P. Kadanoff, *Ann. Phys. (N. Y.)* **50**, 312 (1968).
- <sup>34</sup>L. Passell (private communication).
- <sup>35</sup>V. J. Minkiewicz, K. Gesi, and E. Hirahara, *J. Appl. Phys.* **42**, 1374 (1971).