

MAGNETIC EXCITATIONS ABOVE THE CRITICAL TEMPERATURE

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TABLE OF CONTENTS

	Page
Abstract	iv
I. INTRODUCTION	1
II. REVIEW OF PREVIOUS THEORIES	12
III. SERIES EXPANSION FOR THE TIME DEPENDENT SPIN PAIR CORRELATION FUNCTION	36
IV. CALCULATION OF THE LOWEST ORDER COEFFICIENTS IN THE SERIES EXPANSION FOR THE TRANSVERSE AND LONGITUDINAL CORRELATION FUNCTIONS	48
A. Importance of Short Range Order above the Critical Temperature	48
B. Determination of $G^T(\vec{q}, \omega)$	55
C. Determination of $G^{ZZ}(\vec{q}, \omega)$	67
D. Transverse and Longitudinal Contributions to the Line Shape	70
E. Comparison with Previous Theories	72
V. DISCUSSION OF RESULTS	77
VI. CONCLUSION	83
VII. REFERENCES	86
VIII. ACKNOWLEDGMENTS	89
IX. APPENDIX A: CALCULATION OF $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ AND $\langle G_{\vec{q}}^{(3)} \rangle^{+-}$	90
X. APPENDIX B: TEMPERATURE DEPENDENCE OF THE BACKGROUND TERMS IN $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ AND $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$	94
XI. APPENDIX C: CALCULATION OF $\langle G_{\vec{q}}^{(0)} \rangle^{ZZ}$ AND $\langle G_{\vec{q}}^{(2)} \rangle^{ZZ}$	96

Magnetic excitations above the
critical temperature

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In this work, a new approach has been developed to obtain an approximate expression for the dynamical spin correlation function. This involves the expansion of the dynamical correlation function in a Taylor series in powers of the time whose coefficients are equal to progressively higher order static multiple spin correlation functions. The lowest order coefficients in this series expansion are then evaluated through the use of a decoupling scheme which explicitly takes into account positional spin correlations above the critical temperature. As a consequence of this method, when $r < q^{-1} < \lambda$ where r is the range of the exchange interaction, λ is the two-spin correlation length, and \vec{q} is the wave vector of the spin fluctuations, the dominant contribution to these coefficients is found to be caused by spin clusters involving strongly correlated spins. The degree of local order in this spin system is measured by a time-independent local order parameter¹ ρ_{ij} , the static two-spin correlation function. The resulting expressions for the zeroth, first, and second order coefficients are incorporated as leading terms in a resummation that is used to obtain an approximate

¹For a discussion of local order in one dimensional magnetic systems see F. B. McLean and M. Blume, Phys. Rev. B7, 1149 (1973).

v

expression for the Fourier transform of the dynamical spin correlation function, $G(\vec{q}, \omega)$ which is proportional to the inelastic neutron magnetic scattering cross section. When $q^{-1} < \lambda$ distinct peaks appear in the line shape of $G(\vec{q}, \omega)$ when ω is held constant and q is varied. These peaks occur at $q = \pm q_0$ where $\omega(q_0) = \omega$, which is the frequency of a spin wave of wave vector q_0 . The positions of these spin wave peaks compare favorably with the experimental data on Ni ($T_c = 631$ K) obtained by Mook, et al.² In the temperature region between 656 K ($T_c + 25$ K) - 706 K the experimental and theoretical FWHM have the same slope however outside this region pronounced discrepancies occur which can only be accounted for by a more refined theory.

²H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Letters 30, 556 (1973).

1. INTRODUCTION

In examining the low temperature thermodynamic behavior of ferromagnets Bloch^{1,2} found that the eigenfunctions corresponding to the excitations from the ground state of the exchange Hamiltonian were not characterized by states involving one spin reversal but rather by a linear combination of these states each containing a spin reversal located at a different magnetic ion site. The wave nature of these proper linear combinations was demonstrated by Slater³ in showing that the quantum mechanical problem of one spin reversal could be exactly solved. Thus physically a spin wave may be viewed as a spin reversal propagating through the crystal via exchange interactions between neighboring spins. In Bloch's calculation it was assumed that the number of spin waves excited at low temperatures was so small that all interactions between them could be neglected, and consequently the low-lying excited states in a ferromagnet could be adequately approximated by a linear superposition of these sinusoidal spin waves.

Early attempts^{4,5} at studying deviations from Bloch's theory due to spin wave interactions seemed to indicate that the region of validity for spin wave theory was closely confined to temperatures near absolute zero ($\sim 0.1T_c$). This result was obtained by Van Kranendonk⁶ who analyzed spin wave interactions within the framework of a gas of spin deviations interacting via a hard core potential. Dyson,^{7,8} on the other hand, in studying the interaction between two spin waves not only found that the linear superposition principle discussed by Bloch was not quite correct but also that Van Kranendonk had over-estimated the interaction

between spin waves. He distinguished two types of these interactions. The first is a consequence of the nonorthogonality of the spin wave states containing more than one spin wave and is called the kinematical interaction. This is a repulsive interaction whose physical significance is that no more than $2S$ units of spin can be attached to the same ion simultaneously. The other interaction arises from the fact that the exchange Hamiltonian is not diagonal in these states. This is called the dynamical interaction which causes a shift in the spin wave energy due to the presence of other spin waves. Dyson has shown that the kinematical interaction makes no contribution to the temperature expansion of the magnetization to finite order. The leading term in this expansion goes as $T^{3/2}$ and is produced by the linear spin wave theory developed by Bloch. The leading term caused by dynamical spin wave interactions is of order T^4 which implies that even the dynamical interaction is small at low T . Furthermore, at temperatures up to $0.5T_c$ where T_c is the Curie temperature it was found that the linear, noninteracting spin wave theory discussed by Bloch was quite adequate in describing the behavior of Heisenberg ferromagnets. The physical implications of Dyson's approach have been discussed by Keffer and Loudon⁹ who obtained the T^4 contribution to the magnetization by assuming that when spin waves are excited at low temperatures they are superimposed onto the instantaneous background of spin wave fluctuation already existing in the system. Dyson's results have been approximately reproduced by Oguchi¹⁰ who used the Holstein-Primakoff formalism,¹¹ in which the spin operators are written in terms of the creation and annihilation operators of the

harmonic oscillator, in order to obtain the T^4 contribution to the spontaneous magnetization.

The temperature dependence of the spin wave frequency produced by the lowest order nonlinear terms in the isotropic Heisenberg exchange interaction arising from the H-P expansion of the spin operators has been studied by M. Bloch.¹² These lowest terms have the form

$$H_{\text{ex}}^{(4)} = \frac{1}{2} \sum_{\vec{k}, \vec{k}', \vec{k}''} [J(\vec{k}) + J(\vec{k} + \vec{k}' - \vec{k}'') - 2J(\vec{k} - \vec{k}')] \cdot a_{\vec{k}}^{\dagger} a_{\vec{k}'}^{\dagger} a_{\vec{k}''} a_{\vec{k} + \vec{k}' - \vec{k}''} \quad (1.1)$$

where $a_{\vec{k}}$ and $a_{\vec{k}}^{\dagger}$ are respectively the annihilation and creation operators of a magnon with wave vector \vec{k} , and $J(\vec{k})$ is the Fourier transform of the exchange interaction. These spin wave interactions cause a change (renormalization) in the spin wave frequency which for nearest-neighbor interactions is

$$\omega_{\vec{k}}(T) = \omega_{\vec{k}} [1 - (z JS^2 N)^{-1} \sum_{\vec{k}'} \omega_{\vec{k}'} \langle n_{\vec{k}'} \rangle] \quad (1.2)$$

where z is the number of nearest neighbors, and

$$\omega_{\vec{k}} = 2Jz S[\gamma(0) - \gamma(\vec{k})] \quad (1.3)$$

is the noninteracting spin wave frequency and

$$\gamma(\vec{k}) = \frac{1}{z} \sum_{\vec{\delta}} e^{i\vec{\delta} \cdot \vec{k}} \cdot (\vec{\delta} = \text{n.n. lattice vector}) \quad (1.4)$$

Also,

$$\langle n_{\vec{k}} \rangle = \frac{1}{e^{\beta \omega_{\vec{k}}(T)} - 1} \quad (1.5)$$

which is the magnon thermal occupation number. Consequently as the temperature increases the number of excitations increases and the spin wave frequency decreases. M. Bloch has determined that solutions to the coupled equations (1.2) and (1.5) exist even up to temperatures within a few percent of Curie temperature calculated by the Bethe-Perierls-Weiss method.¹³ This nearest neighbor calculation has been extended by Horwitz and Mattis¹⁴ to the case of non-nearest neighbor interactions. From all of these studies then, it is evident that magnon-magnon interactions remain relatively weak over a broad temperature range; however, all these attempts indicate that the spin wave theory becomes meaningless near and especially above T_c in the paramagnetic phase where no long range order exists. Therefore no spin wave excitations should be observed in this temperature region. Recent experimental observations have largely substantiated these theoretical calculations, but some significant deviations have been found.

The specific experiments that will now be discussed in this connection investigate the behavior of spin waves via the inelastic scattering of neutrons. The reason for this emphasis lies with the fact that magnetic coupling energies between atoms in solids are typically on the order of a few milli-electron volts and consequently their effects are conveniently observable in thermal neutron experiments.

In their study of the spin wave excitations in iron using a triple axis neutron spectrometer, Shirane, Minkiewicz, and Nathans¹⁵ found that the spin wave modes were well resolved to temperatures very near the Curie temperature, $T_c \cong 1042^\circ\text{K}$. (See Figure 1.) Employing a constant momentum scan where the wave vector $q \sim 0.1\text{\AA}^{-1}$, they observed, however, no distinct spin wave peaks in the scattering data when the temperature was raised above T_c .

An examination of the spin fluctuations in Ni near the critical temperature by Minkiewicz, Collins, Nathans and Shirane,¹⁶ revealed that below T_c the magnon peaks were resolved, but above T_c they found that the long wavelength ($q = 0.075\text{\AA}^{-1}$) excitations could be adequately described by the spin diffusion process in which a unit of spin is transferred randomly from one point in the lattice to another with a probability given by the ordinary theory of atomic diffusion. (See Figure 2.)

Recently, Mook, Lynn and Nicklow¹⁷ have looked at the temperature dependence of short wavelength ($q > 0.240\text{\AA}^{-1}$ and $E > 12\text{ meV}$) spin waves in nickel and iron. Using a constant energy scan technique which was made necessary because of the steepness of the magnon dispersion curves for Ni and Fe at high energies, they observed well resolved excitations above T_c that had a remarkable similarity to spin wave modes below the critical temperature. (See Figure 3.) This phenomenon is not, however, limited to metals such as iron and nickel, but has also been observed in the inelastic scattering of neutrons from RbMnF_3 ,¹⁸ (Figure 4), a magnetic insulator which behaves very much like an idealized Heisenberg

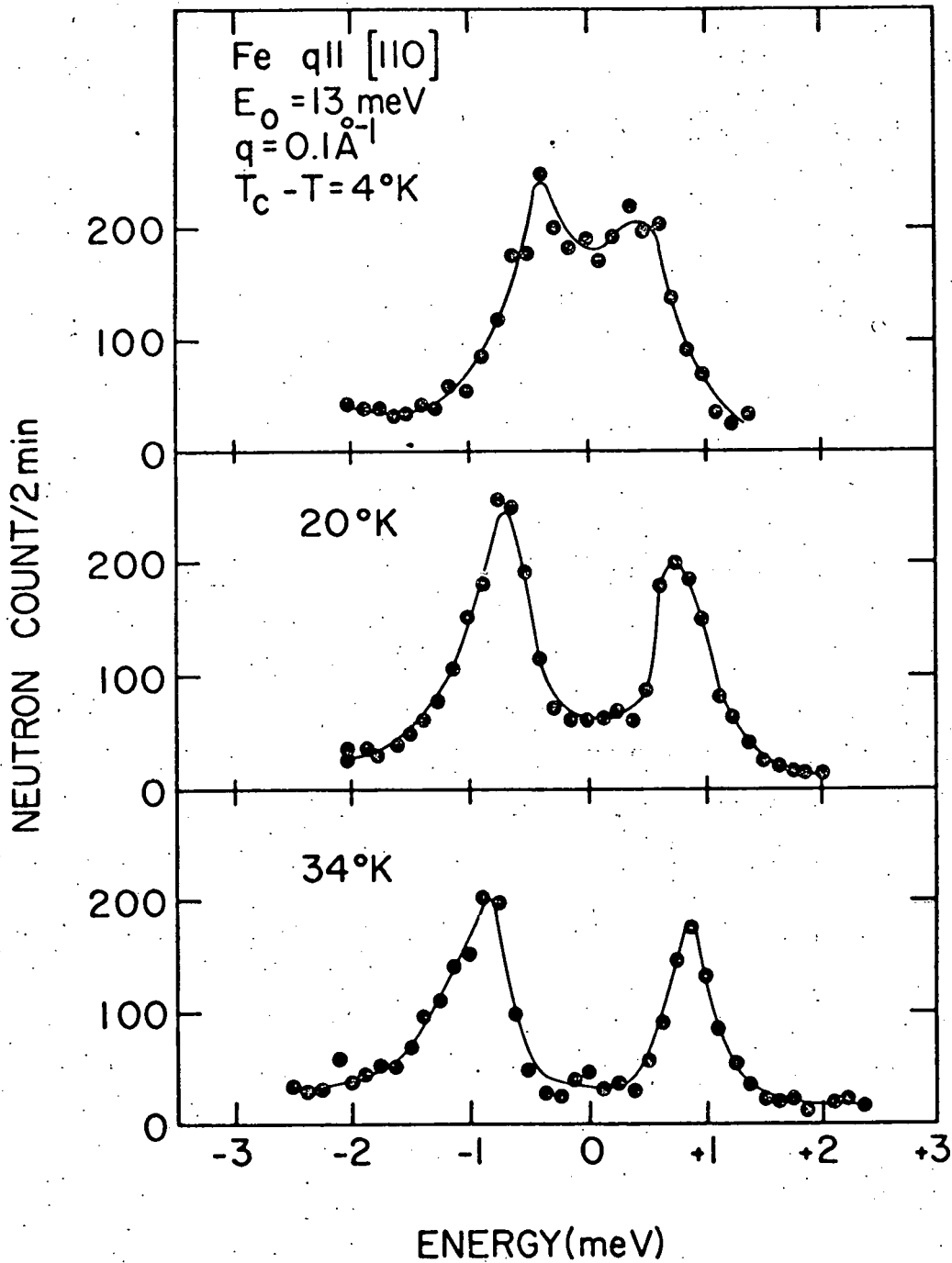


Figure 1. Inelastic neutron scattering intensity for Fe below the Curie temperature $T_c = 1042^\circ \text{K}$. Constant momentum scan.

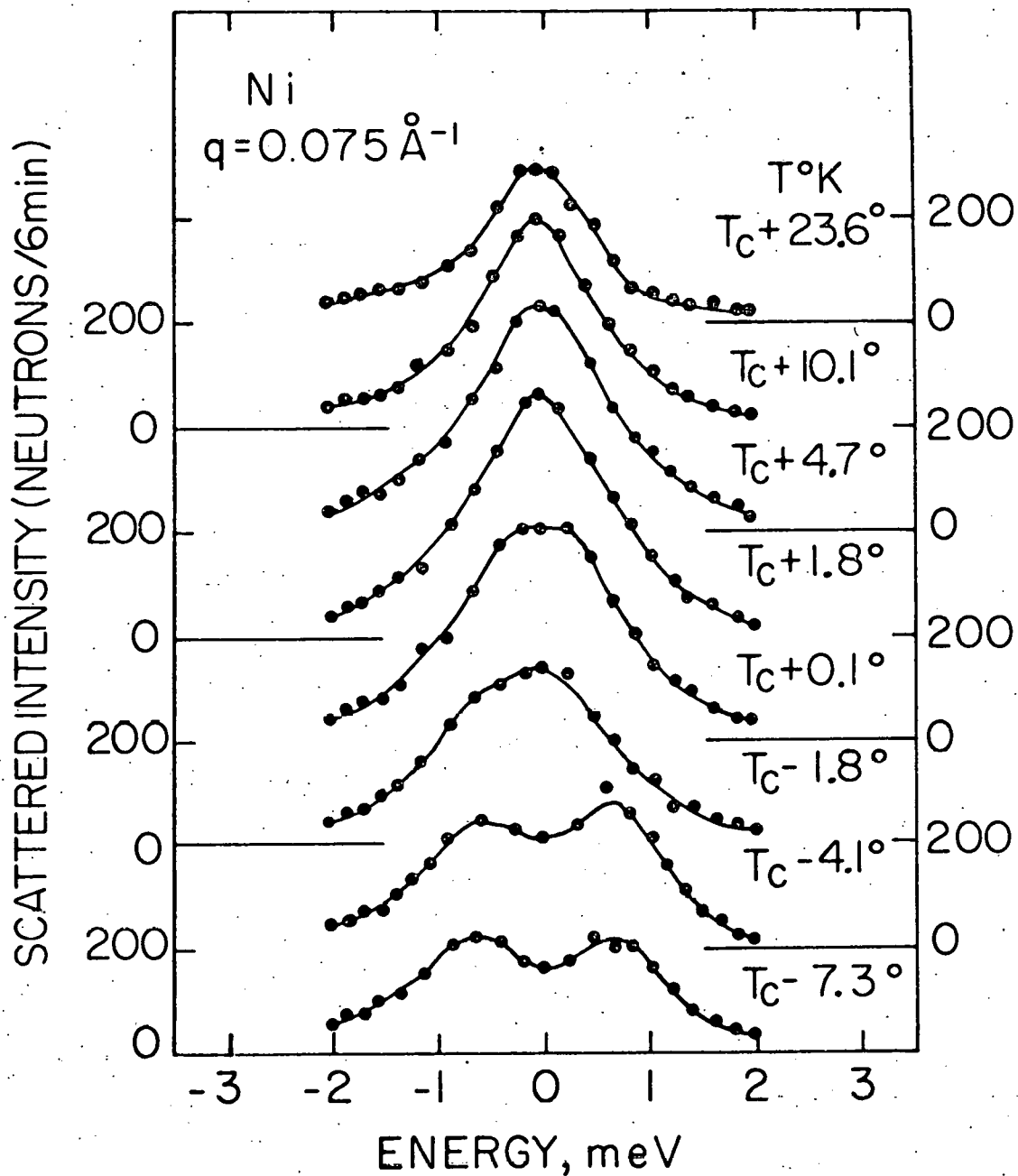


Figure 2. Constant momentum scan for Ni above and below $T_c = 631^\circ\text{K}$. \vec{q} is in the [111] direction.

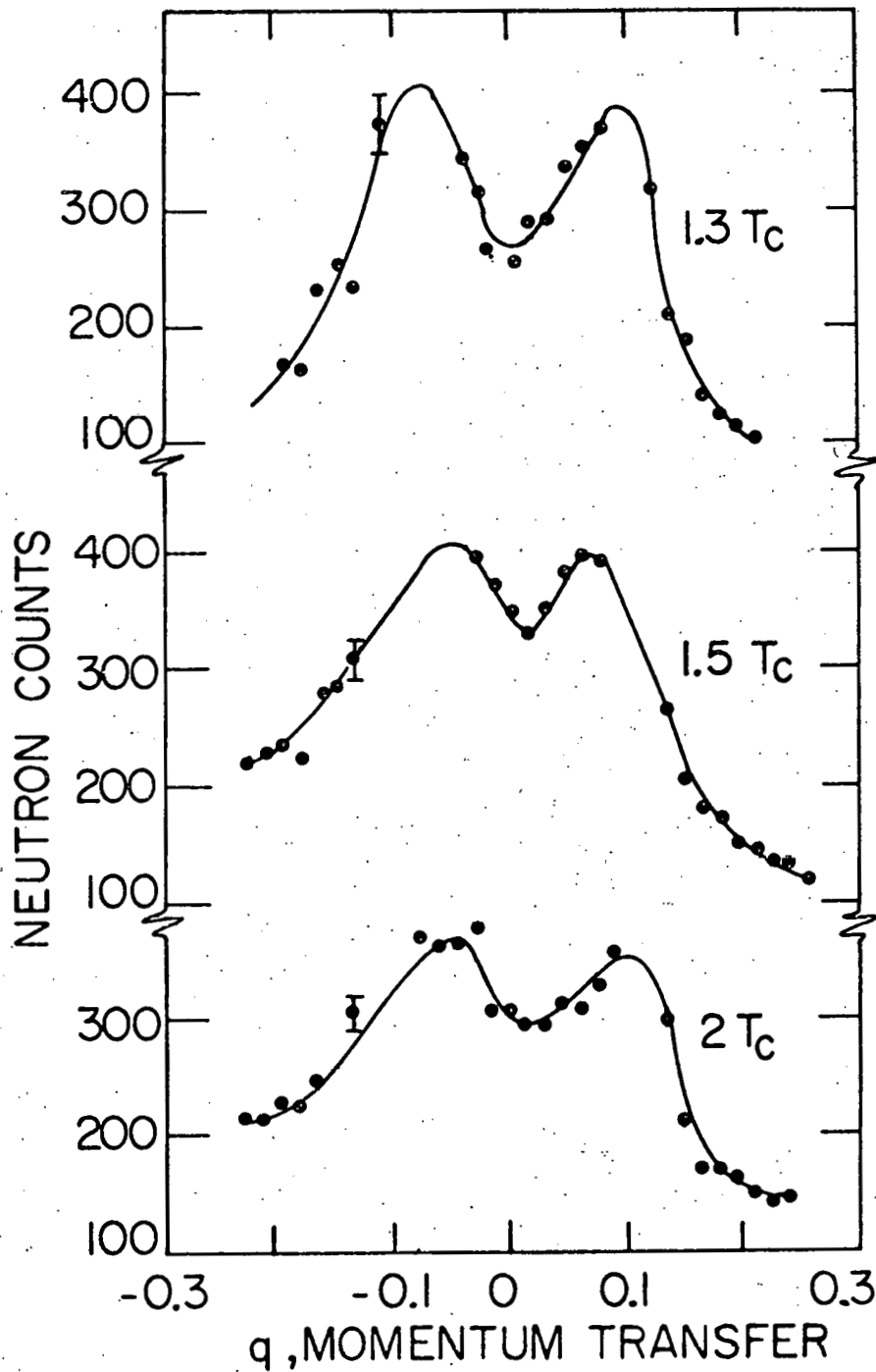


Figure 3. Constant energy scan for Ni above the Curie temperature about the (111) reciprocal lattice point when the energy transfer $\Delta E = 12.4$ meV. q is in zone boundary units of $2\sqrt{3}/a$.

magnet.¹⁹ It is this observation which has motivated us to examine the magnetic excitations of the isotropic Heisenberg model in the paramagnetic phase.

Although we will be primarily interested in the behavior of spin fluctuations in three dimensional magnets such as metals like Ni and Fe and insulators like RbMnF_3 , much work has recently been done in studying the spin dynamics of one dimensional magnets. The experimental investigations have centered on the linear-chain antiferromagnet $(\text{CD}_3)_4\text{NMnCl}_3$ (TMMC).^{20,21} The reason for the interest in 1-d magnetic structures is that the persistence of quasi-spin wave excitations in the paramagnetic regime is more pronounced in the case of systems of lower dimensionality. Furthermore, in 1-d systems there is no finite ordering temperature, i.e., long range order exists only at $T = 0$.²² However, experiments on TMMC have demonstrated that there is a highly developed short range order at temperatures near absolute zero. Tomita and Mashiyama²³ have examined the characteristics of magnon modes in a linear chain of classical spins using the Heisenberg model, and they found fairly good agreement between their calculated inelastic neutron scattering cross section and the experimental data of TMMC.

From all these experiments, then, it is quite evident that spin wave theory has a much broader region of applicability than was predicted by the early theories. More recently there have been several attempts to account for the experimentally observed behavior of spin fluctuations in three dimensional systems at high temperatures and near T_c . In the

next section the most salient features of these theories will be discussed along with an examination of their shortcomings.

II. REVIEW OF PREVIOUS THEORIES

The various theories that have been developed which quantitatively describe spin fluctuations in the paramagnetic phase and their corresponding spectral density fall into two general categories: those which deal directly with the multiple spin time dependent correlation function and attempt to construct a decoupling scheme for this function such as

$$\begin{aligned} \langle s_{\vec{q}_1}^-(t) s_{\vec{q}_2}^+(t), s_{\vec{q}_3}^-(0) s_{\vec{q}_4}^+(0) \rangle \simeq & \delta(\vec{q}_1 + \vec{q}_4) \delta(\vec{q}_2 + \vec{q}_3) \beta^{-1} \\ & \times \langle s_{\vec{q}_1}^-(t), s_{-\vec{q}_1}^+(0) \rangle \langle s_{\vec{q}_2}^-(t) s_{-\vec{q}_2}^+(0) \rangle \end{aligned} \quad (II.1)$$

and those which characterize the spectral shape of the associated excitations in terms of its lowest order moments, namely the zeroth, second, and fourth moments, and relates these moments to higher order static correlation functions.

One of the earliest discussions of this problem was given by Marshall²⁴ in which he described very qualitatively the temperature dependence of spin waves. In his work it was noted that just above T_c when the momentum transfer of the scattered neutrons is large, short wavelength spin fluctuations are excited which behave very much like magnon modes at low temperatures. These excitations were called remnant spin waves. Within this framework some of the experimental observations

can be readily understood: when neutrons are scattered at large wave vectors just above T_c where $q > \chi$, the inverse correlation length, then remnant spin waves should be observed. However, in the case of scattering at smaller wave vectors where $q < \chi$ the excitations span many clusters of spins that are correlated in different directions and consequently must behave very differently from spin waves. Although this picture satisfies the experimental observations in a qualitative way, the previous theoretical attempts to account for the observed spectral shape near the critical point run into difficulties which will be evident in the following discussion.

The first extensive theoretical investigation of spin fluctuations above the critical temperature was done by Resibois and De Leener²⁵⁻²⁸ who concentrated on an approximate evaluation of the spin autocorrelation function

$$I_{ab}^{\alpha\beta}(t) = \langle S_a^\alpha(t) S_b^\beta(0) \rangle, \quad (11.2)$$

which plays a central role in neutron inelastic scattering measurements. In this expression $S_a^\alpha(t)$ is the Heisenberg representation of the α^{th} component of the spin operator located at the lattice site a , and $\langle \dots \rangle$ is an average over an equilibrium canonical ensemble. Using a localized spin model we can write the autocorrelation function in the following manner

$$I_{ab}^{\alpha\beta}(t) = \sum_{m,m'} \langle m | S_a^\alpha | m' \rangle \langle m' | \rho^\beta(t|b) | m \rangle, \quad (11.3)$$

where

$$\rho^\beta(t|b) = e^{-iHt} S_b^\beta \rho^{\text{eq}} e^{iHt}, \quad (11.4)$$

with

$$\rho^{\text{eq}} = \frac{e^{-\beta H}}{\text{Tr}(e^{-\beta H})}. \quad (11.5)$$

Here H is the isotropic Heisenberg exchange Hamiltonian.

As a starting point in their analysis of the autocorrelation function a formal series solution was obtained for the equation of motion for the matrix element of $\rho^\beta(t|b)$ where

$$i \frac{\partial}{\partial t} \{ \langle m | \rho^\beta(t|b) | m' \rangle \} = \lambda \langle m | [H, \rho^\beta(t|b)] | m' \rangle. \quad (11.6)$$

The separate terms in the series expansion were examined via a diagrammatical technique and since they were interested in the long-time behavior of $I_{ab}^{\alpha\beta}(t)$ an infinite series of diagrams had to be considered. A resummation procedure was subsequently developed which was based physically on the idea that two interacting spins are not isolated but interact with the "bath" composed of the remaining spins. Using this approximation and taking the Weiss limit ($z \rightarrow \infty$) where z is the number of neighbors in the range of the exchange interaction, Resibois and De Leener were able to obtain approximate expressions for the direct and indirect autocorrelation functions which were valid for both short and long times.

In extending their theory to finite temperatures in the neighborhood of the critical temperature the following non-Markoffian kinetic equation for the autocorrelation function $\Gamma_{ab}^{\alpha\beta}(t)$ was derived:

$$\frac{\partial}{\partial t} [\tilde{\Gamma}_q(t)] = \int_0^t \tilde{G}_q(t-t') \tilde{\Gamma}_q(t') dt', \quad (11.7)$$

where $\tilde{\Gamma}_q(t)$ is the space Fourier transform of $\Gamma_{ab}^{\alpha\beta}(t)$ and $\tilde{G}_q(t|\tilde{\Gamma}_q)$ is the Fourier transform of an infinite series of terms whose form is determined by a diagrammatical analysis which was discussed in detail. Only the lowest order "bubble" diagrams were kept in order to obtain an approximate expression for $\tilde{\Gamma}_q(t)$.

Near the critical point the shape of the frequency spectrum near T_c , which is directly connected with the observed neutron scattering cross section, is seen in Figure 5, where γ is the spectral shape function

$$\gamma(\Omega) = 1/[\Omega^2 + (1 - \epsilon\Omega^2)^2], \quad (11.8)$$

and Ω is a dimensionless frequency

$$\Omega = \omega(\alpha q^{\frac{5}{2}})^{-1}, \quad (11.9)$$

with ϵ and α as fitting parameters.

One of the major short comings of this theory is that the whole approach is only valid in the Weiss limit where there are an infinite number of neighbors; however in a realistic situation $z = 6$ or 8 ,

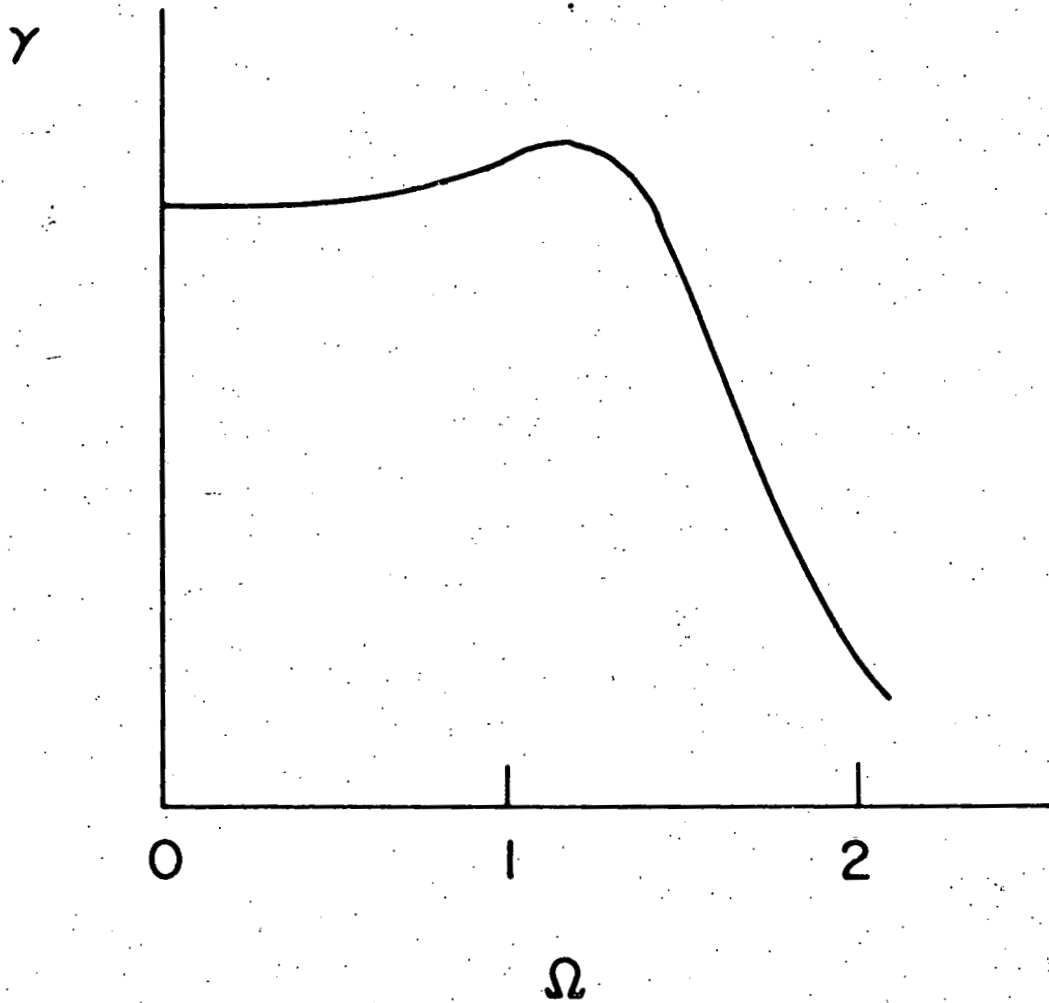


Figure 5. Dependence of the De Leener and Resibois spectral function on the dimensionless frequency, Ω .

and as we shall see the effects of short range order on the observed neutron cross section become dominant when the number of neighbors within the range of the exchange interaction is finite and small.

Another approach to the analysis of time dependent spin correlations at high temperatures was presented by Blume and Hubbard.²⁹ The basic physical idea behind their mathematical calculation is that each spin interacts with a randomly varying effective field which is produced by the neighboring fluctuating spins. The correlation functions $C_q = \langle S_{-q}^z S_q^z(t) \rangle$ are evaluated indirectly through the use of the relaxation function $\{S_{-q}^z, S_q^z(t)\}$ where by definition:

$$\{A, B(t)\} = \int_0^\beta \langle e^{\lambda H} A e^{-\lambda H} B(t) \rangle d\lambda . \quad (11.10)$$

This function describes the relaxation of the variable B for $t > 0$ when a perturbation which is proportional to A is switched on adiabatically between $t = -\infty$ and $t = 0$ and then is suddenly turned off at $t = 0$. Then when $T \rightarrow \infty$

$$\frac{\langle S_{-q}^z S_q^z(t) \rangle}{\langle S_{-q}^z S_q^z \rangle} = \frac{\{S_{-q}^z, S_q^z(t)\}}{\{S_{-q}^z, S_q^z\}} = F_q(t) \quad (11.11)$$

where $\langle . . . \rangle$ denotes an average over an equilibrium canonical ensemble, and thus

$$C_q(t) = \frac{1}{3} N^{-1} S(S+1) F_q(t) \quad (11.12)$$

Kubo's formalism³⁰ is then used to show that

$$F_q(t) = \frac{\langle \delta S_q^Z(t) \rangle}{\langle \delta S_q^Z \rangle} \quad (11.13)$$

where $\delta S_q^Z(t)$ represents the change in $S_q^Z(t)$ due to the perturbation S_{-q}^Z . Now in order to find $F_q(t)$ a formal time-order solution is obtained for the equation of motion for $\delta S_q^Z(t)$:

$$\delta \dot{S}_{q'}^Z = \sum_{q''} J(\vec{q}', \vec{q}'') S_{\vec{q}' - \vec{q}''}^+ \delta S_{\vec{q}''}^- \quad (11.14)$$

where

$$J(\vec{q}', \vec{q}'') = J(\vec{q}') - J(\vec{q}''). \quad (11.15)$$

The random effective magnetic field is then associated with the term $J(\vec{q}', \vec{q}'') S_{\vec{q}' - \vec{q}''}^+$. The solution of $F_q(t)$ consequently involves the evaluation of time dependent multiple spin correlation functions. The principal approximation made is to neglect the detailed correlations between three or more spins. This approximation is not too bad at high temperatures where it leads to errors of $O(1/z)$ where z is the number of magnetic ions lying within the range of the exchange interaction. The result of this method, valid for general times in the high temperature limit ($T \rightarrow \infty$), is seen in Figure 6. It is evident from these plots that there is no indication of propagating modes-- only one that is

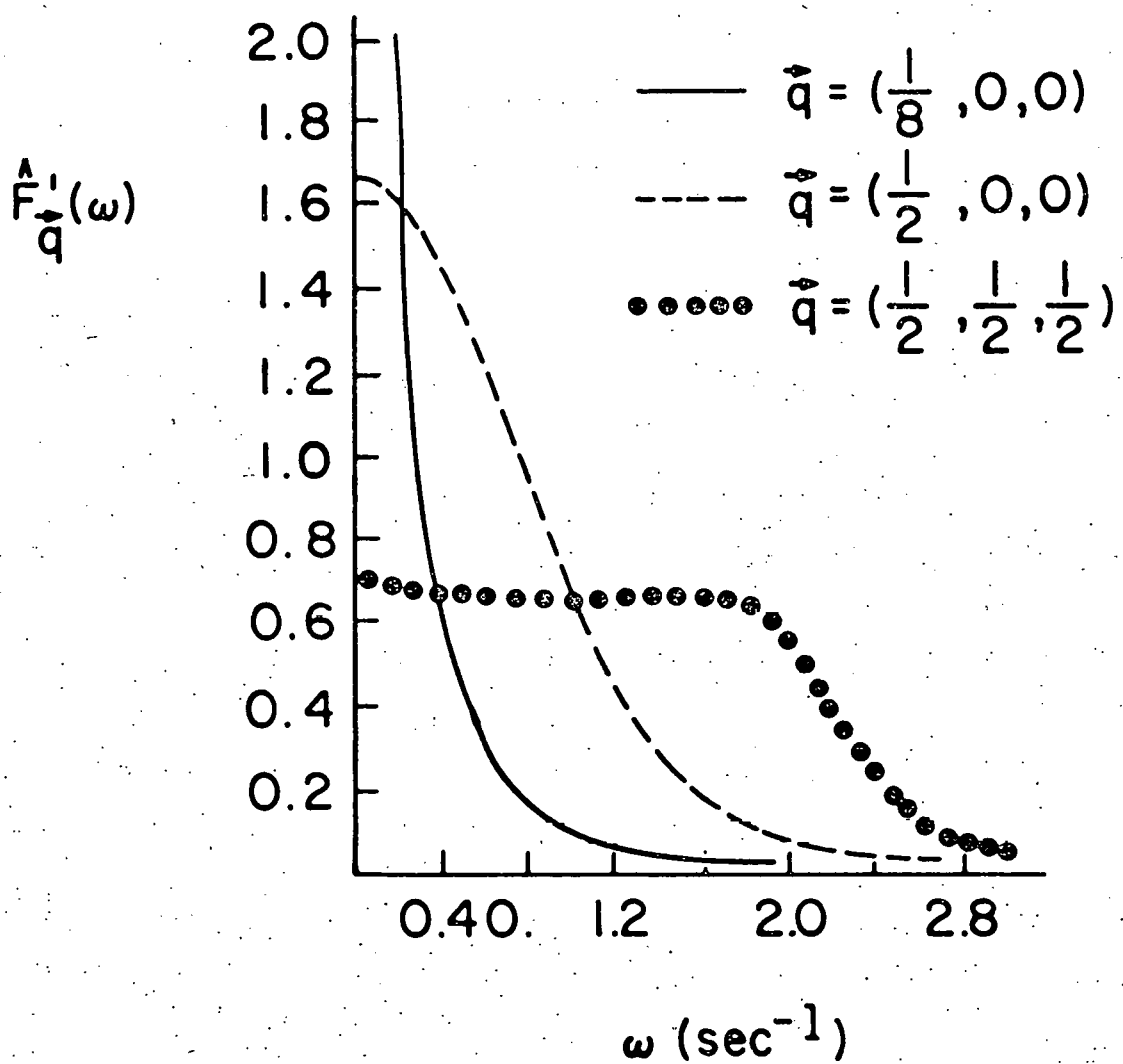


Figure 6 Fourier transform of the relaxation shape function for a simple cubic lattice obtained by Blume and Hubbard for various wave vectors, \vec{q} , where \vec{q} is in units of $2\pi/a$.

strongly overdamped when $q = (1/2, 1/2, 1/2)$. The reason for this behavior can be readily understood: the principal approximation which is discussed above can only be applied effectively at very high temperatures where there is much random spin motion. However, it cannot be extended down to near the critical temperature where significant short range order contributions to the frequency spectrum of the neutron scattering cross section are observed experimentally.^{17,18} Here the decoupling of the multiple spin correlations must be treated differently. This will be discussed in detail in Chapter IV.

The phenomenological theories based on the calculation of moments of the spectral density or related functions, e.g. the spectral shape function are of particular interest since we will be using a moment type method in studying the existence of spin wave excitations above the critical temperature. This particular approach will be discussed completely in the next section, however before this is done some previous theories in this area will be examined.

The earliest calculation of this kind was carried out by De Gennes³¹ who in studying the inelastic magnetic scattering of neutrons calculated the second and fourth moments of

$$\rho_{\chi}(\omega) = \sum_{\vec{R}} \int dt e^{i(\chi \cdot \vec{R} - \omega t)} u_{\vec{R}}(t) \quad (11.16)$$

where

$$u_{\vec{R}}(t) = \langle \vec{S}_0(0) \cdot \vec{S}_{\vec{R}}(t) \rangle / [S(S+1)] \quad (11.17)$$

which is closely related to the spin autocorrelation function $\Gamma_{ab}^{\alpha\beta}(t)$ used by Resibois and De Leener. Actually $\sum_{\alpha} \Gamma_{ab}^{\alpha\alpha}(t)$ is equivalent to $U_{\vec{R}}(t)$ to within a factor $S(S+1)$.

The moments of $\rho_{\kappa}(\omega)$ are

$$\langle \omega^{2n} \rangle_{\kappa} = \frac{\int \rho_{\kappa}(\omega) \omega^{2n} d\omega}{\int \rho_{\kappa}(\omega) d\omega} \quad (11.18)$$

The expressions for $\langle \omega^2 \rangle_{\kappa}$ and $\langle \omega^4 \rangle_{\kappa}$ are valid only in the high temperature limit and for polycrystals. The wave vector dependence of the second and fourth moments indicated that for small wave vectors ($\kappa \rightarrow 0$) the spectral density $\rho_{\kappa}(\omega)$ can be approximated by a truncated Lorentzian. At larger wave vectors where $\kappa \gtrsim \pi/b$ where b is the nearest neighbor distance the shape of $\rho_{\kappa}(\omega)$ is more closely given by a Gaussian function.

A preliminary attempt to determine whether or not propagating modes exist in a Heisenberg paramagnet was carried out by Bennett,³² who calculated the lowest order moments of an effective frequency spectrum assumed to have no delta-function of frequency terms, $\delta(\omega)$, and to have a smooth monotonic behavior. It is noted that the validity of this approach depends on the validity of the diffusion equation for the magnetization in the hydrodynamic regime³³ where $q \ll \kappa$, the long wavelength region. Then the multiple spin time dependent correlation functions appearing in the calculation are decoupled in the following way:

$$\langle s_i^z s_j^+ s_k^z s_\ell^- \rangle \approx \langle s_i^z s_k^z \rangle \langle s_j^+ s_\ell^- \rangle, \quad (11.19)$$

and

$$\langle s_i^+ s_j^- s_k^+ s_\ell^- \rangle \approx \langle s_i^+ s_j^- \rangle \langle s_k^+ s_\ell^- \rangle + \langle s_i^+ s_\ell^- \rangle \langle s_k^+ s_j^- \rangle. \quad (11.20)$$

When this approximation is used to simplify the expressions for the second and fourth moments Bennett finds that the criterion for the existence of propagating modes when $q > \xi^{-1}$ where ξ is the spin correlation length is

$$R(\vec{q}) = \langle \omega_q^2 \rangle^2 / \langle (\omega_q^2 - \langle \omega_q^2 \rangle)^2 \rangle_q \gg 1. \quad (11.21)$$

When this inequality is satisfied high frequency modes exist; however when $q < \xi^{-1}$,

$$R(\vec{q}) < 1 \quad (11.22)$$

and diffuse modes dominate. The fact that Bennett's calculations are carried out in the hydrodynamic regime is one of the most prominent shortcomings of his theory. The reason for this is found in Halperin and Hohenberg's discussion³³ of the critical and hydrodynamic regions where the latter is defined to occur when $q \ll \kappa$. However, as will be shown below, the propagating modes observed by Mook et al.¹⁷ in Ni above

the critical temperature occur when $q > \kappa$, the inverse correlation length. Consequently, the decoupling scheme used by Bennett is in question. It is apparent from his work that no attempt was made to consider the possible contribution of short range order to the shape of the frequency spectrum above T_c .

Of greater interest is the work done by Tomita and Tomita³⁴ in which theoretical line shapes are obtained for the relaxation shape function. These shapes are then compared with the neutron inelastic magnetic scattering from RbMnF_3 .¹⁸ The second and fourth moments of the relaxation function are calculated assuming nearest-neighbor interactions only. The general forms of these moments are

$$\langle \omega^2 \rangle_{\vec{k}} = \frac{[1 - \lambda(\vec{k})]}{\hbar^2 \chi^{zz}(\vec{k})} \frac{1}{N} \sum_{\vec{q}} J(\vec{q}) \langle s^+(\vec{-q}) s^-(\vec{q}) \rangle, \quad (11.23)$$

where

$$\lambda(\vec{k}) = \frac{1}{z} \sum_{\vec{a}} e^{i\vec{k} \cdot \vec{a}}, \quad (11.24)$$

and

$$J(\vec{q}) = J \sum_{\vec{a}} e^{i\vec{q} \cdot \vec{a}}, \quad (11.25)$$

In which z is the number of nearest-neighbors, \vec{a} is the nearest-neighbor position vector, J is the nearest-neighbor exchange interaction, and

$\chi^{zz}(\vec{k})$ is the longitudinal wave vector dependent susceptibility.

at small k

$$\langle \omega^4 \rangle_{\vec{k}} = \frac{k^2}{\hbar^4 \chi^{zz}(\vec{k})} \times \{\text{Set of 4-spin correlation functions}\}. \quad (11.26)$$

Then the high temperature ($T \rightarrow \infty$), finite temperature ($T > T_c$), and low temperature ($T \rightarrow 0$) behavior of these moments is discussed. At finite temperatures the random phase approximation (R.P.A.) form of the two-spin correlation function,

$$\langle s^z(-\vec{q}) s^z(\vec{q}) \rangle = \frac{Nk_B T}{\chi_0^{-1} + [J(0) - J(\vec{q})]}, \quad (11.27)$$

where χ is the paramagnetic susceptibility, is used in finding explicit expressions for $\langle \omega^2 \rangle_{\vec{q}}$ and $\langle \omega^4 \rangle_{\vec{q}}$. Also for $T > T_c$ the calculation of the fourth moment is made tractable by expressing the four-spin correlation function in terms of a product of two spin correlation functions. Bennett has also employed this same type of approximation which is valid in the high temperature limit where the number of neighbors z is large since the error introduced is of the order of $1/z$. In comparing the temperature dependence of the second moment to that of the fourth moment, Tomita and Tomita find that the fourth moment is more sensitive to fluctuations in the spin system as the temperature changes than the

second moment due to the higher order multi-spin correlations contained in $\langle \omega^4 \rangle_{\vec{q}}$.

In order to obtain their spectral line shapes, Mori's well-known continued fraction expansion³⁵ is used in evaluating the canonical correlation function

$$\psi(\vec{k}, \omega) = \int_0^{\infty} dt e^{-i\omega t} \frac{R(\vec{k}, t)}{R(\vec{k}, 0)} \quad (11.28)$$

where $R(\vec{k}, t)$ is the relaxation function. This continued fraction expansion³⁶ may be written as

$$\psi(\vec{k}, \omega) = [i\omega + a_1^2(\vec{k}) f(\omega; \{b^2(\vec{k})\})]^{-1} \quad (11.29)$$

where

$$f(\omega; \{b^2(\vec{k})\}) = [i\omega(a_2^2(\vec{k})/i\omega + \dots)]^{-1}, \quad (11.30)$$

and

$$a_1^2(\vec{k}) = \langle \omega^2 \rangle_{\vec{k}} \equiv a^2(\vec{k}) \quad (11.31)$$

$$a_2^2(\vec{k}) = \frac{\{\langle \omega^4 \rangle_{\vec{k}} - \langle \omega^2 \rangle_{\vec{k}}^2\}}{\langle \omega^2 \rangle_{\vec{k}}} \equiv b^2(\vec{k}) \quad (11.32)$$

[Note that $R(\vec{k}, t)$ is equivalent to Blume and Hubbard's $C_{\vec{k}}(t)$ and that $\psi(\vec{k}, \omega)$ is just the Laplace transform of their normalized spectral shape

function $F_{\vec{k}_2}(t)$.] Here $f(\omega; \{b^2(\vec{k})\})$ is the torque correlation function in which $\{b^2(\vec{k})\}$ corresponds to the set $\{a_2^2(\vec{k}), a_3^2(\vec{k}), \dots\}$. Then it is assumed that the torque correlation spectrum can be characterized by only $a_2^2(\vec{k})$ and further that the shape of this spectrum can be simply described by a Gaussian function. The total frequency spectrum obtained can be seen in Figure 7. Although there are peaks in the theoretical line shape calculated by Tomita and Tomita, serious discrepancies exist between their shape function and the observed inelastic neutron scattering data for RbMnF_3 .¹⁸ (See Figure 8.) Two problems arise with their approach: first, the second and fourth moments are evaluated through the use of a decoupling scheme that is valid only at high temperatures. Another difficulty concerns the particular truncation procedure used which introduces mathematically a two peaked function into the theory. It would be more meaningful if the derivation of these peaks could be based on the nature of the physical processes which govern the behavior of the spin system above the critical point such as the existence of short range order.

Recently Lovesey and Meserve³⁷ have employed Mori's continued fraction expansion of the relaxation spectral shape function in analyzing the frequency spectrum of the inelastic neutron scattering cross section. Here again a truncation procedure is developed in order to approximate the relaxation function, however the expansion is carried to one order farther than Tomita and Tomita's work so that a three pole approximation is constructed. The second and fourth moments involved in this calculation are evaluated at infinite temperature³⁸ and at finite temperatures

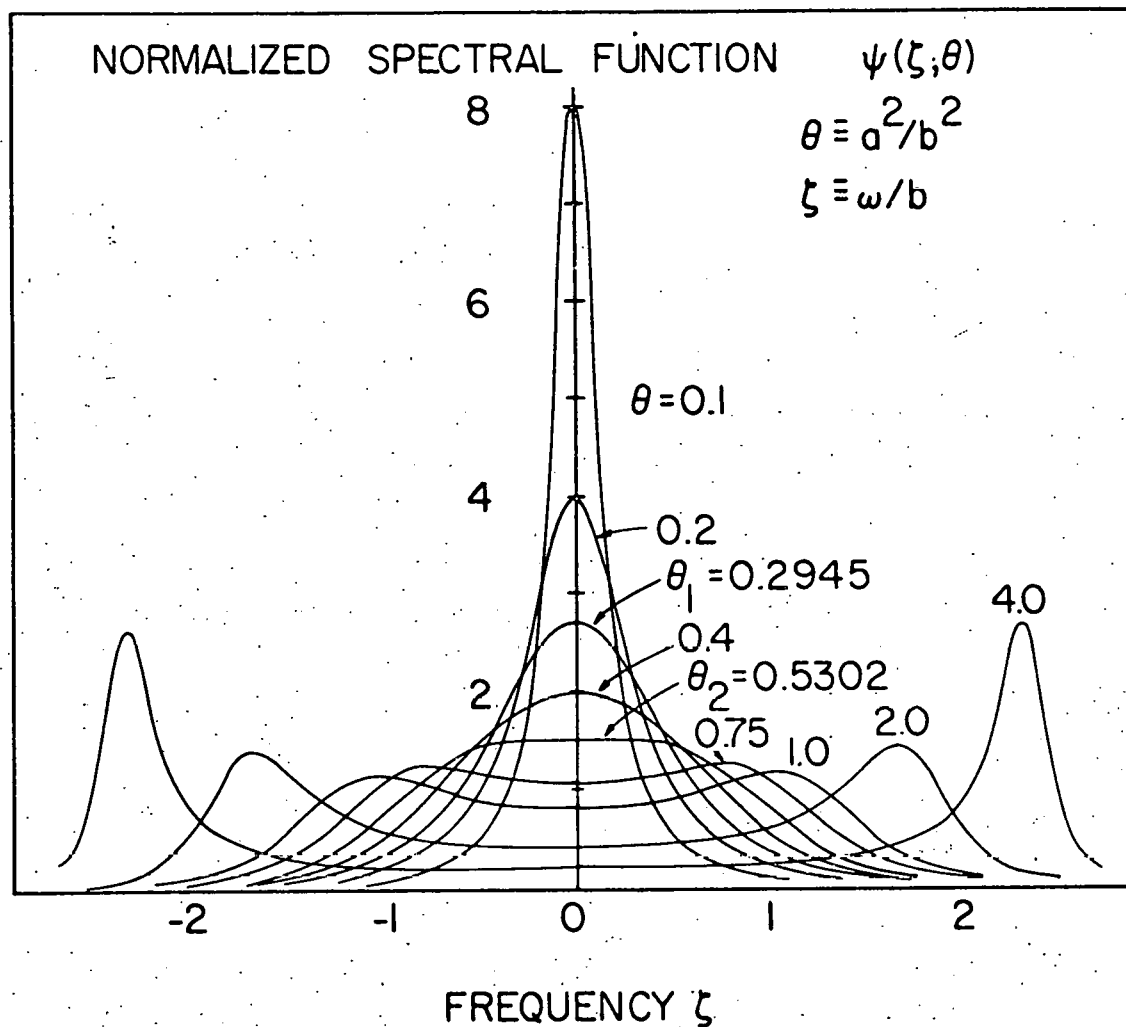


Figure 7. Spectral function obtained by Tomita and Tomita. At θ_1 the poles of ψ become complex and when $\theta > \theta_2$ peaks may be discerned.

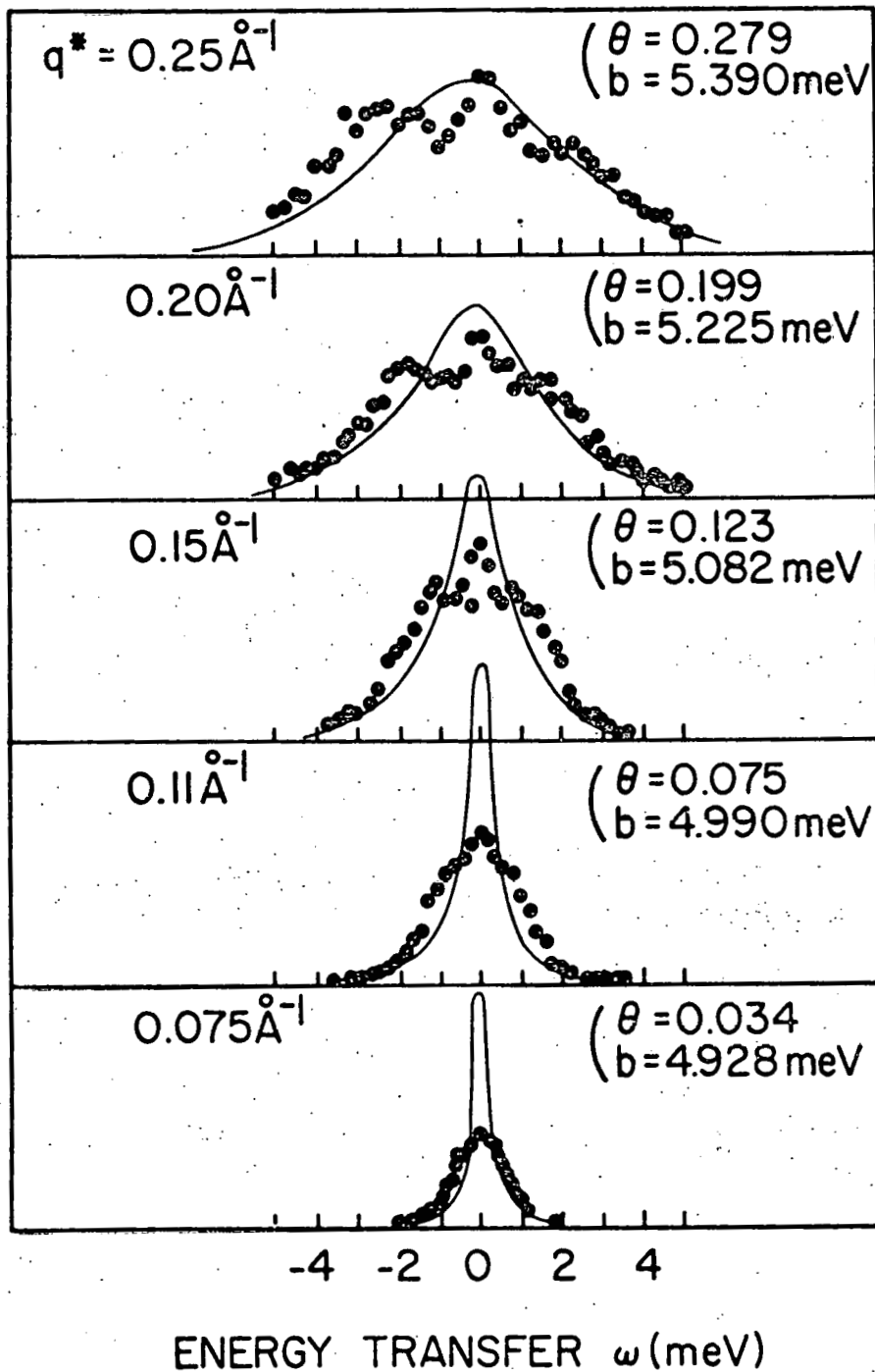


Figure 8. Comparison between the theoretical lines obtained by Tomita and Tomita and inelastic neutron scattering data for RbMnF₃. q^* is measured along the [111] direction.

$T > T_c$ where the four spin correlation functions contained in the expression for the fourth moment are approximated in terms of two-spin correlation functions in a manner quite similar to the decoupling scheme used by Tomita and Tomita. The agreement between this method and the experimental data¹⁸ obtained for RbMnF_3 is fairly good. However the same objections that were raised there also apply in this discussion: the truncation procedure automatically determines a function which has the desired number of poles in it. There is no attempt to determine from the physics of the system why the particular structure in the spectrum is observed, i.e. the possible effects of short range order near T_c are again not explicitly considered. Furthermore, a nonphysical peak in the theoretical line shape occurs at $T = \infty$ when $\vec{k} > (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

One of the most detailed examinations of the moment expansion method was presented by Reiter³⁹ in his study of spin fluctuations in Heisenberg paramagnets. Reiter obtains an infinite series expansion of the relaxation function in terms of the moments of the spectral density. The relaxation of the q^{th} component of the magnetization in the absence of any externally applied fields is given by

$$\bar{M}(\vec{q}, t) = (S^2(\vec{q}, t), S^z(-\vec{q}, 0)) = \langle S^z(-\vec{q}, 0) | S^z(\vec{q}, t) \rangle \quad (11.33)$$

where

$$(A, B) \equiv \{A, B\}, \quad (11.34)$$

which is defined in Eq. (11.10), and where

$$\langle A|B \rangle = \{A^+, B\}. \quad (11.35)$$

Thus $\Sigma(\vec{q}, t)$ is the unnormalized form of the shape function $F_q(t)$ which is used by Blume and Hubbard. [See Eq. (11.11)].

In terms of its moments

$$\Sigma(\vec{q}, t) = \sum_{n=0}^{\infty} \frac{(-it)^n}{n!} \chi(\vec{q}, 0) \langle \omega^n \rangle_q. \quad (11.36)$$

Here $\chi(\vec{q}, 0)$ is the isothermal response of the magnetization to a magnetic field of strength $(g\mu_B)^{-1}$ and wave vector \vec{q} , where

$$\langle \omega^n \rangle_q = \frac{\langle S^z(\vec{q}) | L^n | S^z(\vec{q}) \rangle}{\chi(\vec{q}, 0)}, \quad (11.37)$$

in which L is the Liouville operator of the system:

$$L0 = \frac{1}{\hbar} [0, H]. \quad (11.38)$$

Diagrammatic expansions for these moments are developed for infinite temperature and for finite temperatures above T_c . When $T > T_c$ the second moment $\langle \omega^2 \rangle_q$ and its fluctuation $\langle \Omega_q^4 \rangle = \langle (\omega^2 - \langle \omega^2 \rangle_q)^2 \rangle_q$ become

$$\langle \omega^2 \rangle_{\vec{q}} = \frac{1}{3} S(S+1) N^{-1} \sum_{\vec{q}'} [V(\vec{q}') - V(\vec{q} - \vec{q}')]^2 \rho(\vec{q}') \rho(\vec{q} - \vec{q}') \rho(\vec{q})^{-1}, \quad (11.39)$$

and

$$\begin{aligned} \langle \Omega^4 \rangle_{\vec{q}} = & \frac{2}{3} [S(S+1)]^2 N^{-2} \sum_{\vec{q}'} \sum_{\vec{q}''} [V(\vec{q} - \vec{q}')][V(\vec{q}'') - V(\vec{q} - \vec{q}' - \vec{q}'')] \\ & [V(\vec{q}' + \vec{q}'') - V(\vec{q} - \vec{q}' - \vec{q}'')][V(\vec{q}'') - V(\vec{q}')] \rho(\vec{q}) \rho(\vec{q}'') \\ & \rho(\vec{q} - \vec{q}' - \vec{q}'') \rho(\vec{q})^{-1}, \quad (11.40) \end{aligned}$$

where $\rho(\vec{q})$ is the equilibrium spin pair correlation function

$\rho(\vec{q}) \propto \langle S_{\vec{q}}^z S_{-\vec{q}}^z \rangle$. When the moment expansion is carried to finite (lowest) order the expression obtained for the relaxation function is valid for short times only.

Now in order to account for the details observed in the spectral density associated with the neutron inelastic scattering cross section, Reiter⁴⁰ obtains a set of kinetic equations for the two spin correlation functions $\langle S^-(\vec{q}_1) S^+(\vec{q}_2) \rangle$ and $\langle S^z(\vec{q}_1) S^z(\vec{q}_2) \rangle$. These kinetic equations are then used to determine $\Sigma(\vec{q}, t)$ and $\Gamma(\vec{q}_1, \vec{q}_2, t)$ which describes the decay of a fluctuation of wave vector $\vec{q} = \vec{q}_1 + \vec{q}_2$ into two fluctuations of wave vectors \vec{q}_1 and \vec{q}_2 . The expressions for $\langle \omega^2 \rangle_{\vec{q}}$ and $\langle \Omega^4 \rangle_{\vec{q}}$ are then obtained from the leading terms of series expansion in powers of $(\frac{1}{2})$ for $\Gamma(\vec{q}_1, \vec{q}_2, z)$, the Laplace transform of $\Gamma(\vec{q}_1, \vec{q}_2, t)$, and they agree completely with those derived from the diagrammatic expansion of the moments. However the physical implications of the approximations used in obtaining

the kinetic equations are much more apparent than those used in the diagram approach. In particular the coupled equations of motions for $s_z(\vec{q}_1)$, $s^+(\vec{q}_1)s^-(\vec{q}_2)$, $s^z(\vec{q}_1)s^z(\vec{q}_2)$ and $s^-(\vec{q}_1)s^z(\vec{q}_2)s^+(\vec{q}_3)$ are closed by decoupling the four-spin correlation functions in the equation of motion for $\langle s^-(\vec{q}_1)s^z(\vec{q}_2)s^+(\vec{q}_3) \rangle$ as follows:

$$\begin{aligned} \langle s^-(\vec{q}_1)s^z(\vec{q}_2)s^+(\vec{q}_3)s^+(\vec{q}_4) \rangle \cong \\ \langle s^z(\vec{q}_2)s^z(\vec{q}_3) \rangle \langle s^-(\vec{q}_1)s^+(\vec{q}_4) \rangle, \end{aligned} \quad (11.41)$$

and

$$\begin{aligned} \langle s^-(\vec{q}_1)s^-(\vec{q}_2)s^+(\vec{q}_3)s^+(\vec{q}_4) \rangle \cong \\ \langle s^-(\vec{q}_1)s^+(\vec{q}_3) \rangle \langle s^-(\vec{q}_2)s^+(\vec{q}_4) \rangle + \\ \langle s^-(\vec{q}_1)s^+(\vec{q}_4) \rangle \langle s^-(\vec{q}_2)s^+(\vec{q}_3) \rangle. \end{aligned} \quad (11.42)$$

As noted by Reiter this decoupling method neglects those correlations which are nonvanishing when all the spins in $\langle \dots \rangle$ are "close" to one another. This is valid when the range of the exchange interaction is much larger than the range of the four-spin cumulant correlation function. The error introduced is $O(1/c)$, where c is the number of spins within the range of the exchange interaction. It is precisely these terms which Reiter neglects which will be found to become important near T_c .

The high temperature approximation for $\Gamma(\vec{q}_1, \vec{q}_2, z)$ which is used to obtain the overall shape of the spectral density is

$$\Gamma(q_1, q_2, z) \approx \{N^{-\frac{1}{2}} \frac{2}{3} S(S+1) \frac{[\rho(\vec{q}_2) - \rho(\vec{q}_1)]}{\rho(\vec{q}_1 + \vec{q}_2)}\} / \nu(z) \quad (11.43)$$

where $\nu(z)$ is determined in such a manner that the correct forms of $\langle \omega^2 \rangle_q$ and $\langle \Omega^4 \rangle_q$ are maintained. This constant relaxation time approximation is then used to obtain theoretical line shapes which are compared with the neutron inelastic scattering data for RbMnF_3 .¹⁸ It is evident that Reiter's approach does describe the shape of the frequency spectrum at high temperatures very adequately, however at low temperatures near T_c there are discrepancies especially near $\omega = 0$.

The shortcomings that are inherent in the theoretical work dealing with spin fluctuations in three dimensional Heisenberg ferromagnets are not however present in the studies of one dimensional (1d) Heisenberg systems. Fisher⁴¹ has obtained exact solutions for both the partition function and the spin pair correlation function for a one dimensional Heisenberg ferromagnet and antiferromagnet in the case where $S \rightarrow \infty$. In this limit the classical spin Hamiltonian is obtained i.e., the spin operators in the Hamiltonian commute. Fisher's results have been extended by Tomita and Mashiyama who have rigorously calculated exact expressions for the second, fourth and sixth moments of the time-dependent spin correlation functions for a Heisenberg 1d chain of classical spins with nearest-neighbor interactions only. Mori's continued fraction

expansion was then used to obtain the spectral shape of the inelastic scattering cross section. Their calculated result compares quite favorably to the inelastic neutron scattering data of THMC obtained by Hutchings et al.²⁰ who determined that this system is adequately described by the classical Heisenberg's 1d antiferromagnet with nearest neighbor interactions when $T \gtrsim 1.1 \text{ } ^\circ\text{K}$. In both the theory and experiment, well-defined spin wave modes were observed in the paramagnetic region. These short wavelength spin wave excitations occur because of the existence of a highly-developed short range order just above the critical point which in the case of a 1d ferromagnet is $T_c = 0^\circ\text{K}$.

Recently McLean and Blume⁴² have studied the spin dynamics at finite temperature of 1d Heisenberg magnetic chains involving spins of arbitrary size. They examined both the long wavelength and the short wavelength spin fluctuation in this system using Blume and Hubbard's technique which has been discussed above. The existence of short range order was explicitly allowed for through the introduction of a local order parameter Δ where Δ is taken to be the square root of the near-neighbor correlation. When this parameter was included in their calculation it was found that the short wavelength excitations, $q \gg 1/\ell_c$, where ℓ_c is the correlation length, are spin-wave-like and that the long wavelength fluctuations, $q \lesssim 1/\ell_c$ are diffusive in nature and are associated with variations in the local order parameter. Again the comparison between the experimental data of THMC determined by Hutchings et al.²⁰ and their theoretical scattering function is quite good.

The major point of this whole discussion is that none of the existing theories adequately describes the structure observed in the inelastic neutron scattering data near T_c at large wave vectors except in one dimension. In all the 3d theories discussed above one of the major shortcomings was the way in which the multiple spin correlation functions were decoupled when $T > T_c$. The region of validity of this decoupling scheme and the motivation for its use will be examined shortly in connection with a procedure we have developed in which the positional correlation of the spins is considered in detail. From this calculation the importance of short range order and its effect on the spectral density when $T \gtrsim T_c$ will become quite apparent.

III. SERIES EXPANSION FOR THE TIME DEPENDENT SPIN PAIR CORRELATION FUNCTION

Since inelastic neutron scattering has been extensively used to examine the magnetic excitations above the critical temperature a method will be developed to obtain theoretical line shapes for the frequency and wave vector spectrum of the neutron inelastic scattering cross section. Of fundamental importance in this discussion is the fact that the magnetic scattering cross section is related to the time dependent spin pair correlation function.⁴³ Consequently if an expression for this function can be derived it will be possible to generate line shapes which can be compared with experiment.

The relation between the neutron magnetic scattering cross section and the spin pair correlation function has been shown by Van Hove:⁴³

$$\frac{d^2\sigma}{d\Omega' d\omega'} = \frac{k'}{k} \frac{\gamma e^2}{mc^2} \frac{|f(\vec{q})|^2}{\hbar} \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{q}_\alpha \hat{q}_\beta) G^{\alpha\beta}(\vec{q}, \omega) \quad (III.1)$$

where

$$G^{\alpha\beta}(\vec{q}, \omega) = \frac{1}{2\pi} \sum_{\vec{r}} \int_{-\infty}^{\infty} dt e^{i(\vec{q}\cdot\vec{r}-\omega t)} \langle S_0^\alpha(0) S_{\vec{r}}^\beta(t) \rangle, \quad (III.2)$$

and $\hbar\vec{q} = \hbar(\vec{k} - \vec{k}')$ is the momentum transfer vector in which \vec{k} and \vec{k}' are respectively the incident and scattered wave vectors, $f(\vec{q})$ is the magnetic form factor, and $G^{\alpha\beta}(\vec{q}, \omega)$ is the space and time Fourier transform of the spin pair correlation function. Consequently it is evident from Eq. (III.1) that the scattering is a measure of the spin correlations in the system and furthermore that all information concerning the

frequency distribution of the neutron scattering cross section is contained in $G^{\alpha\beta}(\vec{q}, \omega)$.

In order to simplify the following calculation a system will be considered in which the spins are localized at the ionic sites of the lattice, and so

$$\vec{S}(\vec{r}) = \sum_i \vec{S}_i \delta(\vec{r} - \vec{R}_i). \quad (III.3)$$

Then

$$\vec{S}_{\vec{q}} = \frac{1}{\sqrt{N}} \sum_i \vec{S}_i e^{i\vec{q} \cdot \vec{R}_i}, \quad (III.4)$$

where the sum on i is carried over all N lattice vectors \vec{R}_i . For this localized spin system, $G^{\alpha\beta}(\vec{q}, \omega)$ can be written in the following way:

$$G^{\alpha\beta}(\vec{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\beta}(t) \rangle e^{-i\omega t} dt. \quad (III.5)$$

When this expression is integrated over all frequencies we obtain

$$\begin{aligned} \int_{-\infty}^{\infty} G^{\alpha\beta}(\vec{q}, \omega) d\omega &= \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\beta}(t) \rangle e^{-i\omega t} dt \\ &= \int_{-\infty}^{\infty} \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\beta}(t) \rangle \delta(t) dt \\ &= \langle S_{\vec{q}}^{\alpha} S_{-\vec{q}}^{\beta} \rangle, \end{aligned} \quad (III.6)$$

which is just the equilibrium spin pair correlation function. For a system with isotropic interactions the neutron scattering cross section, Eq. (III.1), reduces to

$$\frac{d^2\sigma}{d\Omega' dE'} \propto \sum_{\alpha} [1 + (\hat{q}_{\alpha})^2] G^{\alpha\alpha}(\vec{q}, \omega), \quad (III.7)$$

where $\alpha = x, y, z$.

In the paramagnetic phase

$$\langle S_{\vec{q}}^x(0) S_{-\vec{q}}^x(t) \rangle = \langle S_{\vec{q}}^y(0) S_{-\vec{q}}^y(t) \rangle = \langle S_{\vec{q}}^z(0) S_{-\vec{q}}^z(t) \rangle \quad (III.8)$$

since there is no overall preferred direction of spin orientation.

This implies that

$$G^{xx}(\vec{q}, \omega) = G^{yy}(\vec{q}, \omega) = G^{zz}(\vec{q}, \omega) \equiv G(\vec{q}, \omega). \quad (III.9)$$

Consequently, above T_c

$$\frac{d^2\sigma}{d\Omega' dE'} \propto G(\vec{q}, \omega). \quad (III.10)$$

Therefore, since the neutron scattering cross section is directly proportional to $G(\vec{q}, \omega)$ in this temperature region, a comparison between $G(\vec{q}, \omega)$ and the neutron cross section determined from the measured scattering intensity can be readily accomplished. However, before we consider the properties of this function in more detail, its relation to the spin relaxation function and spectral density function which have been used in prior theoretical work (see Chapter II) will be examined.

In studying the behavior of spontaneous spin fluctuations, Marshall and Lowde⁴⁴ have examined the properties of the spin relaxation function

$$R^{\alpha\beta}(\vec{q}, t) = \{S_{-\vec{q}}^{\alpha}(0), S_{\vec{q}}^{\beta}(t)\}, \quad (III.11)$$

which describes how a spin system relaxes after a disturbance is removed.

The Fourier transform of $R^{\alpha\beta}(\vec{q}, t)$ is

$$r^{\alpha\beta}(\vec{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} R^{\alpha\beta}(\vec{q}, t) dt. \quad (III.12)$$

When $T > T_c$, then by definition

$$\{S_{-\vec{q}}^{\alpha}(0), S_{\vec{q}}^{\beta}(t)\} = \int_0^{\beta} d\lambda \langle e^{\lambda H} S_{-\vec{q}}^{\alpha}(0) e^{-\lambda H} S_{\vec{q}}^{\beta}(t) \rangle,$$

and

$$r^{\alpha\beta}(\vec{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{-i\omega t} \int_0^{\beta} d\lambda \langle e^{\lambda H} S_{-\vec{q}}^{\alpha}(0) e^{-\lambda H} e^{iHt} S_{\vec{q}}^{\beta}(0) e^{-iHt} \rangle. \quad (III.13)$$

Therefore,

$$\begin{aligned} r^{\alpha\beta}(\vec{q}, \omega) &= \frac{1}{2} \sum_{ij} \int_{-\infty}^{\infty} dt e^{-i[\omega - (E_j - E_i)]t} \\ &\quad \times \int_0^{\beta} d\lambda Z^{-1} e^{-\beta E_i - \lambda(E_j - E_i)} \\ &\quad \times \langle i | S_{-\vec{q}}^{\alpha}(0) | j \rangle \langle j | S_{\vec{q}}^{\beta}(0) | i \rangle, \end{aligned} \quad (III.14)$$

where

$$S_{\vec{q}}^{\alpha}(t) = e^{iHt} S_{\vec{q}}^{\alpha}(0) e^{-iHt}, \quad (III.15)$$

$|i\rangle$ and $|j\rangle$ are eigenstates of the spin Hamiltonian H with energies E_i and E_j respectively, and Z is the partition function. Then

$$\begin{aligned}
 r^{\alpha\beta}(\vec{q}, \omega) &= \sum_{ij} \delta[\omega - (E_j - E_i)] Z^{-1} e^{-\beta E_i} \\
 &\quad \langle i | S_{-\vec{q}}^{\alpha}(0) | j \rangle \langle j | S_{\vec{q}}^{\beta}(0) | i \rangle \int_0^{\beta} e^{-\lambda \omega} d\lambda, \\
 &= \frac{(1 - e^{-\beta \omega})}{\omega} \frac{1}{2\pi N} \int_{-\infty}^{\infty} e^{-i\omega t} Z^{-1} \\
 &\quad \times \sum_{ij} e^{-\beta E_i} \langle i | S_{-\vec{q}}^{\alpha}(0) | j \rangle \langle j | e^{iHt} S_{\vec{q}}^{\beta}(0) e^{-iHt} | i \rangle, \\
 &= N \frac{(1 - e^{-\beta \omega})}{\omega} \int_{-\infty}^{\infty} e^{-i\omega t} \langle S_{-\vec{q}}^{\alpha}(0) S_{\vec{q}}^{\beta}(t) \rangle, \quad (III.16)
 \end{aligned}$$

so

$$G^{\alpha\beta}(\vec{q}, \omega) = \frac{N^{-1} \omega}{(1 - e^{-\beta \omega})} r^{\alpha\beta}(\vec{q}, \omega). \quad (III.17)$$

In the above calculation $\hbar = 1$ and the factor $\omega/(1 - e^{-\beta \omega})$ takes into account the detailed balance between the magnon absorption and emission processes that occur when neutrons are scattered inelastically by the spin system.

The relation between $G^{\alpha\beta}(\vec{q}, \omega)$ and the dynamical susceptibility can be derived in the following way. In a straight forward proof it can be shown

$$\frac{\partial}{\partial t} \{A, B(t)\} = i \langle [A, B(t)] \rangle. \quad (III.18)$$

Consequently,

$$\begin{aligned} \frac{\partial}{\partial t} R^{\alpha\beta}(\vec{q}, t) &= - \langle [S_{-\vec{q}}^{\alpha}(0), S_{\vec{q}}^{\beta}(t)] \rangle \\ &= - \frac{N}{g^2 \mu^2} \chi^{\alpha\beta}(\vec{q}, t), \end{aligned} \quad (III.19)$$

where $\chi^{\alpha\beta}(\vec{q}, t)$ is the dynamical response function which determines the response of the \vec{q}^{th} wavevector of the magnetization to an applied field $\vec{H}(\vec{q}, t)$:

$$M^{\alpha}(\vec{q}, t) = \int dt' \chi^{\alpha\beta}(\vec{q}, t - t') H^{\beta}(\vec{q}, t') \quad (III.20)$$

in the case where the system is translationally invariant and stationary. Since the principle of causality imposes the following restriction on the response function:

$$\chi^{\alpha\beta}(\vec{q}, t) = 0, \quad (III.21)$$

when $t < 0$, then we can define the generalized susceptibility to be³⁰

$$\chi^{\alpha\beta}(\vec{q}, \omega) = \int_0^{\infty} e^{-i\omega t} \chi^{\alpha\beta}(\vec{q}, t) e^{-\epsilon t} dt \quad (III.22)$$

where $\epsilon \rightarrow 0^+$. The presence of $e^{-\epsilon t}$ insures that the response is adiabatic.

Using this definition and Eq. (III.18) we obtain

$$\chi^{\alpha\beta}(\vec{q}, \omega) = \frac{-g^2 \mu^2}{N} \int_0^{\infty} e^{-i\omega t} \frac{\partial}{\partial t} R^{\alpha\beta}(\vec{q}, t) dt, \quad (III.23)$$

which when integrated by parts becomes

$$\chi^{\alpha\beta}(\vec{q}, \omega) = \frac{g\mu^2}{N} R^{\alpha\beta}(\vec{q}, 0) - \frac{g\mu^2}{N} i\omega \int_0^{\infty} e^{-i\omega t} R^{\alpha\beta}(\vec{q}, t) dt. \quad (III.24)$$

Since the canonical correlation function has the property

$$\{A, B\} = \{B, A\}, \quad (III.25)$$

then

$$R^{\alpha\beta}(\vec{q}, t) = R^{\beta\alpha}(-\vec{q}, -t) = R^{\alpha\beta}(-\vec{q}, t)^* \quad (III.26)$$

This result along with Eq. (III.24) implies that

$$\chi^{\alpha\beta}(\vec{q}, \omega) = \chi^{\alpha\beta}(-\vec{q}, -\omega)^*. \quad (III.27)$$

Consequently,

$$\begin{aligned} \chi^{\alpha\beta}(\vec{q}, \omega) - \chi^{\beta\alpha}(\vec{q}, \omega)^* &= -\frac{g\mu^2}{N} i\omega \int_{-\infty}^{\infty} e^{-i\omega t} R(\vec{q}, t) dt \\ &= \frac{-g\mu^2}{N} 2\pi i\omega r^{\alpha\beta}(\vec{q}, \omega). \end{aligned} \quad (III.28)$$

Now when $\alpha = \beta$, the above equation becomes

$$\frac{\chi^{\alpha\alpha'}(\vec{q}, \omega)}{\omega} = \frac{g\mu^2}{N} 2 \operatorname{Re}[r^{\alpha\alpha}(\vec{q}, \omega)], \quad (III.29)$$

where $\chi^{\alpha\alpha'}$ is the imaginary component of the generalized susceptibility which in this case governs the dissipation of energy caused by the excitation of spin fluctuations in the system. Then using Eq. (III.17)

we obtain

$$\frac{\chi^{\alpha\alpha'}(\vec{q}, \omega)}{\omega} = \frac{(1 - e^{-\beta\omega})}{\beta\omega} G^{\alpha\alpha'}(\vec{q}, \omega), \quad (III.30)$$

which is simply a statement of the fluctuation-dissipation theorem.

Here the term on the left hand side is usually referred to as the spectral density. Thus because of the above interrelationships between $G^{\alpha\beta}(\vec{q}, \omega)$, the relaxation function, the spectral density, and the generalized susceptibility, it is desirable to obtain an explicit expression for $G^{\alpha\beta}(\vec{q}, \omega)$.

Below the critical temperature the spin system exhibits long range order, and as $T \rightarrow 0^\circ\text{K}$ the spins take on their maximum allowable quantum number along the axis of quantization, namely S . Consequently, at temperatures near absolute zero spin wave theory may be used to evaluate the spin pair correlation functions, and thus in this temperature region we find that⁴⁴

$$G^{xx}(\vec{q}, \omega) \propto \frac{1}{2} \{ \delta[\omega - \omega(\vec{q})] + S[\omega + \omega(\vec{q})] \}, \quad (III.31)$$

where $\hbar\omega(\vec{q})$ is the energy of a noninteracting spin wave mode of wave vector \vec{q} :

$$\hbar\omega(\vec{q}) = 2\hbar S[J(0) - J(\vec{q})]. \quad (III.32)$$

Here $J(\vec{q})$ is the Fourier transform of the exchange interaction. So at low temperatures $G^{xx}(\vec{q}, \omega)$ is expressed in terms of delta function singularities which occur at frequencies corresponding to the absorption

or emission of a spin wave mode excited by the inelastic scattering of neutrons. As the temperature increases, spin wave interactions become important and act to give a finite line width to the peaks in $G^{xx}(\vec{q}, \omega)$. However at temperatures near the critical temperature an exact form for $G^{\alpha\alpha}(\vec{q}, \omega)$ is impossible to obtain.

In order to clearly ascertain what difficulties are involved in the calculation $G^{\alpha\alpha}(\vec{q}, \omega)$ near T_c ($T \gtrsim T_c$), we will now construct a series expansion for the dynamical spin correlation function

$$g^{\alpha\alpha}(\vec{q}, t) = \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\alpha}(t) \rangle \quad (III.33)$$

and use the result to obtain $G^{\alpha\alpha}(\vec{q}, \omega)$, since

$$G^{\alpha\alpha}(\vec{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} g^{\alpha\alpha}(\vec{q}, t) e^{-i\omega t} dt. \quad (III.34)$$

We begin by expanding $g^{\alpha\alpha}(\vec{q}, t)$ into a Taylor series in powers of t .

$$g^{\alpha\alpha}(\vec{q}, t) = \sum_n \frac{(-it)^n}{n!} \langle G_{\vec{q}}^{(n)} \rangle^{\alpha\alpha} \quad (III.35)$$

where

$$\langle G_{\vec{q}}^{(n)} \rangle^{\alpha\alpha} = \left[\left(\frac{1}{i} \frac{\partial}{\partial t} \right)^n \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\alpha}(t) \rangle \right]_{t=0}. \quad (III.36)$$

As a consequence of the fact that $S_{-\vec{q}}^{\alpha}(t)$ is a Heisenberg operator, i.e.

$$S_{-\vec{q}}^{\alpha}(t) = e^{iHt} S_{-\vec{q}}^{\alpha}(0) e^{-iHt}, \quad (III.37)$$

when H is time independent, and

$$\dot{s}_{-\vec{q}}^{\alpha}(t) = i[H, s_{-\vec{q}}^{\alpha}(t)], \quad (III.38)$$

then the coefficients in the above series expansion can be expressed in terms of multiple commutators of the Heisenberg spin Hamiltonian

$$H = - \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j = - \sum_{\vec{q}} J(\vec{q}) \vec{S}_{\vec{q}} \cdot \vec{S}_{-\vec{q}}. \quad (III.39)$$

So through the use of Eq. (III.38) and cyclic property of the trace it can be readily shown that

$$\langle G_{\vec{q}}^{(0)} \rangle^{\alpha\alpha} = \langle s_{\vec{q}}^{\alpha} s_{-\vec{q}}^{\alpha} \rangle, \quad (III.40)$$

$$\langle G_{\vec{q}}^{(1)} \rangle^{\alpha\alpha} = \langle s_{\vec{q}}^{\alpha} [H, s_{-\vec{q}}^{\alpha}] \rangle, \quad (III.41)$$

$$\langle G_{\vec{q}}^{(2)} \rangle^{\alpha\alpha} = \langle [s_{\vec{q}}^{\alpha}, H] [H, s_{-\vec{q}}^{\alpha}] \rangle, \quad (III.42)$$

etc. Since each commutation with H brings in another pair of spins, it is evident that the coefficients $\langle G_{\vec{q}}^{(n)} \rangle^{\alpha\alpha}$ in the series expansion of the time-dependent spin correlation function are equal to progressively higher order static multi-spin correlation functions.

The problem of calculating the time dependent spin correlation function now becomes clear when Eq. (III.35) is examined: at temperatures near T_c critical spin fluctuations become very important and consequently an expression for $G^{\alpha\alpha}(\vec{q}, \omega)$ must be obtained which is valid not only for

short times but also for long time intervals. In order to do this we must sum over an infinite number of terms in the moment expansion. This is clearly an impossible task especially since the problem is complicated further by the fact that no small parameter can be found which would allow for an obvious resummation procedure involving a subset of terms whose contribution to the series is dominant. We must use our intuition concerning the significance of the various physical processes in order to determine the dominant contribution to the above series expansion for the time dependent spin correlation function.

In carrying out this procedure (Chapter IV) we will find that we need to have a detailed knowledge of the equilibrium spin pair correlation function such as $\langle S_{\vec{q}}^{\alpha} S_{-\vec{q}}^{\alpha} \rangle$ which has been discussed at length by Kadanoff et al.⁴⁵ and Halperin and Hohenberg³³ who showed that at small wave vectors above T_c

$$\langle S_{\vec{q}}^{\alpha} S_{-\vec{q}}^{\alpha} \rangle \propto \frac{1}{q^2 + 1/\lambda^2} \quad (\text{III.43})$$

where λ is the correlation length whose physical meaning is that spins lying within this distance do not behave independently. Thus coherent spin fluctuations may be excited within this region.

From scaling theory,⁴⁵ λ exhibits the following temperature dependence

$$\lambda \propto (T - T_c)^{-\nu} \quad (\text{III.44})$$

In the random phase approximation $\nu = \frac{1}{2}$. However, in more exact theories $\nu \approx \frac{2}{3}$, within the critical region where $k\lambda \gg 1$ and where both k^{-1} and λ are macroscopic lengths. However, since we will be examining the behavior of spin fluctuations outside the critical region where $k\lambda \gtrsim 1$, $\nu = \frac{1}{2}$ can be used.

The experimental determination of the correlation length follows directly from the definition of the neutron scattering cross section where, from Eqs. (III.1), (III.2) and (III.5),

$$\frac{d^2\sigma}{d\Omega' d\omega'} \propto \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\alpha}(t) \rangle dt \quad (III.45)$$

then integrating over all energies of the scattered neutrons we obtain

$$\int \frac{d^2\sigma}{d\Omega' d\omega'} d\omega \propto \frac{1}{2\pi} \int d\omega \int dt e^{-i\omega t} \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\alpha}(t) \rangle \propto \langle S_{\vec{q}}^{\alpha} S_{-\vec{q}}^{\alpha} \rangle. \quad (III.46)$$

Consequently, for small q

$$\int \frac{d^2\sigma}{d\Omega' d\omega'} d\omega \propto \frac{1}{q^2 + (1/\lambda)^2}. \quad (III.47)$$

Thus λ may be obtained by fitting the expression on the left hand side of Eq. (III.47) to the experimentally determined total integrated scattering intensity.

IV. CALCULATION OF THE LOWEST ORDER COEFFICIENTS IN THE SERIES
EXPANSION FOR THE TRANSVERSE AND LONGITUDINAL
CORRELATION FUNCTIONS

A. Importance of Short Range Order above the Critical Temperature

For the present study the isotropic Heisenberg model will be examined:

$$H = - \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j = - \sum_{\vec{q}} J(\vec{q}) \vec{S}_{\vec{q}} \cdot \vec{S}_{-\vec{q}}, \quad (\text{IV.1})$$

where

$$J(\vec{q}) = \sum_j J_{ij} e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.2})$$

and

$$\vec{S}_{\vec{q}} = \frac{1}{\sqrt{N}} \sum_j \vec{S}_j e^{-i\vec{q} \cdot \vec{R}_j}. \quad (\text{IV.3})$$

Here J_{ij} gives the exchange interaction between the i^{th} and j^{th} spins $J_{ij} = J(\vec{R}_i - \vec{R}_j)$. In practice these exchange parameters are considered to be phenomenological constants whose values are determined by fitting this model to the experimental data.

The motivation for our consideration of this particular system comes from the fact that it is inherently much simpler to treat than the itinerant electron theory of ferromagnetism and from the fact that one observes magnon-like excitations above the critical temperature in

neutron scattering experiments not only in metals such as nickel¹⁷ and iron⁴⁶ but also in magnetic insulators like RbMnF_3 .¹⁸

From Eq. (III.10) it is evident that in order to determine the shape of the inelastic neutron scattering cross section due to spin fluctuations above the critical point the function $G^{\alpha\alpha}(\vec{q}, \omega)$ must be calculated. This calculation will now be accomplished through the use of the series expansion of the dynamical spin correlation function in powers of t , Eq. (III.35). The usefulness of this approach lies in the fact that the coefficients, $\langle G_{\vec{q}}^{(n)\alpha\alpha} \rangle$, in this power series can be related to static multiple spin correlation functions. We can readily evaluate the lowest order coefficients by employing a physical approximation which is developed in this section. Because of the resulting simple expressions for the coefficients of the zeroth through second power of t , we will incorporate them as leading terms in a resummation which is used to determine an approximate expression for the dynamical spin correlation function, Eq. (III.33), from which $G^{\alpha\alpha}(\vec{q}, \omega)$ can be found by Fourier transformation.

We now proceed with a calculation of $G^{\alpha\alpha}(\vec{q}, \omega)$ above the critical temperature. In Chapter III we define

$$G^{\alpha\alpha}(\vec{q}, \omega) = \int_{-\infty}^{\infty} g^{\alpha\alpha}(\vec{q}, t) e^{-i\omega t} dt, \quad (\text{IV.4})$$

where $\alpha = x, y, z$ and

$$g^{\alpha\alpha}(\vec{q}, t) = \langle S_{\vec{q}}^{\alpha}(0) S_{-\vec{q}}^{\alpha}(t) \rangle. \quad (\text{IV.5})$$

Then using the expansion of $g^{\alpha\alpha}(\vec{q}, t)$ in powers of t , we obtain

$$G^{\alpha\alpha}(\vec{q}, \omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} [\langle G_{\vec{q}}^{(0)} \rangle^{\alpha\alpha} - i \langle G_{\vec{q}}^{(1)} \rangle^{\alpha\alpha} t - \frac{1}{2!} \langle G_{\vec{q}}^{(2)} \rangle^{\alpha\alpha} t^2 + \frac{i}{3!} \langle G_{\vec{q}}^{(3)} \rangle^{\alpha\alpha} t^3 + \dots]. \quad (\text{IV.6})$$

The multiple commutator expressions for the lowest order coefficients in the above series are given by Eqs. (III.40) - (III.42) and

$$\langle G_{\vec{q}}^{(3)} \rangle^{\alpha\alpha} = \langle S_{\vec{q}}^{\alpha}, H [H, [H, S_{-\vec{q}}^{\alpha}]] \rangle, \quad (\text{IV.7})$$

where H is the Heisenberg exchange Hamiltonian. It will be very apparent from the complete calculation of $\langle G_{\vec{q}}^{(3)} \rangle^{\alpha\alpha}$ (Appendix A) that an explicit derivation of any of the higher order coefficients would be extremely tedious, and consequently we will carry out detailed calculations for only $\langle G_{\vec{q}}^{(0)} \rangle$ through $\langle G_{\vec{q}}^{(3)} \rangle$.

Our calculation of these coefficients can be simplified by making the following observation. When $T \gtrsim T_c$, and the neutron scattering wave vector satisfies the inequality $q > \lambda^{-1}$ where λ is the spin pair correlation length discussed in Chapter III, then from the theory of Fourier transformations the dominant contribution to the spatial sum in the equation for the Fourier transform of the dynamical spin correlation function,

$$g^{\alpha\alpha}(\vec{q}, t) = \langle s_{\vec{q}}^{\alpha}(0) s_{-\vec{q}}^{\alpha}(t) \rangle = \sum_j \langle s_i^{\alpha}(0) s_j^{\alpha}(t) \rangle e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.8})$$

where

$$\vec{R}_{ij} = \vec{R}_i - \vec{R}_j, \quad (\text{IV.9})$$

comes from those terms in which

$$|\vec{R}_{ij}| < \frac{1}{q} < \lambda, \quad (\text{IV.10})$$

so that the spins at sites i and j lie inside a distance λ from each other. (It should be noted that our interest in this wave vector region is generated by the fact that the neutron scattering experiments on Ni done by Mook et al.¹⁷ are carried out in this region.) Proceeding further, if we assume the range of the exchange interaction r to be finite and

$$r < \frac{1}{q} < \lambda, \quad (\text{IV.11})$$

then to a good approximation the dominant contribution to the lowest

order coefficients in the series expansion of the spin correlation function,

$$g^{\alpha\alpha}(\vec{q}, t) = \sum_j \left(\langle G_{ij}^{(0)} \rangle^{\alpha\alpha} - i \langle G_{ij}^{(1)} \rangle^{\alpha\alpha} t - \frac{1}{2} \langle G_{ij}^{(2)} \rangle^{\alpha\alpha} t^2 + \dots \right) e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.12})$$

where

$$\langle G_{ij}^{(n)} \rangle^{\alpha\alpha} = \left[\left(\frac{1}{i} \frac{\partial}{\partial t} \right)^n \langle S_i^\alpha(0) S_j^\alpha(t) \rangle \right]_{t=0} \quad (\text{IV.13})$$

comes from spins which lie within regions whose sizes are measured by λ . The reason for this is that each succeeding coefficient involves a higher order time derivative of the dynamical correlation function which introduces a multiple commutator of the Heisenberg Hamiltonian H with the spin operator. Each commutation of H with S_j^α brings in the appropriate components of a set of spins at sites l which are coupled to S_j^α via the exchange interaction J_{jl} . Consequently, when $r < q^{-1} < \lambda$, all of the spins at sites l lie within a correlated spin cluster. Thus when the above inequality, Eq. (IV.11) holds, the major contribution to the dynamical spin correlation function $g^{\alpha\alpha}(\vec{q}, t)$ above the critical temperature is due to clusters of correlated spins. Hence it is evident that positional spin correlations are very important near the critical temperature and must be explicitly accounted for. In order to do this we must introduce a local order parameter which is a measure of the

degree of spin order within a correlated cluster. McLean and Blume⁴² first introduced the concept of a local order parameter so that they could account for the existence of short range order in one dimensional Heisenberg magnets. This parameter was oriented in the direction of the local magnetization. The underlying assumption of their theory is that the spatial and temporal variations of the parameter occur slowly near $T = 0$ so that in the first approximation the local order parameter is constant with respect to the propagation of short wavelength ($q \gg \lambda^{-1}$) spin waves.

This definition will be followed rather closely here. However, in order to simplify our calculations, we will construct a time independent local order parameter. Since the normalized correlation function $\langle \vec{S}_i \cdot \vec{S}_j \rangle / S(S+1)$ gives the probability that spins on sites i and j are correlated with each other and thus is a measure of the spin alignment, we will take this to be our local order parameter. This parameter also has a direction which is that of the local magnetization associated with the spin cluster. We define this to be the direction of the local z axis.

Consequently, in discussing the contributions to the lowest order coefficients in the power series expansion of the dynamical spin correlation function, we explicitly account for the existence of short range order above the critical temperature by approximating the system as a collection of correlated spin clusters whose orientations are distributed randomly and whose degree of spin alignment is given by the local order parameter.

When the system is approximated in this way, the time dependent spin pair correlation function $\langle S_i^\alpha(0) S_j^\alpha(t) \rangle$ can be written in the following way:

$$\langle S_i^\alpha(0) S_j^\alpha(t) \rangle = \frac{1}{3} [\langle S_i^Z(0) S_j^Z(t) \rangle + \frac{1}{2} (\langle S_i^+(0) S_j^-(t) \rangle + \langle S_i^-(0) S_j^+(t) \rangle)], \quad (IV.14)$$

where

$$S_j^\pm = S_j^X \pm i S_j^Y. \quad (IV.15)$$

Here these spin operators are defined with respect to the local z axis of the spin cluster which is the axis of quantization. The factor of $\frac{1}{3}$ takes into account the fact that the local z axis is randomly oriented in the spin system under consideration.

As a consequence of Eq. (IV.4), we can write

$$G^{\alpha\alpha}(\vec{q}, \omega) = \frac{1}{3} G^{ZZ}(\vec{q}, \omega) + \frac{1}{6} [G^{+-}(\vec{q}, \omega) + G^{-+}(\vec{q}, \omega)], \quad (IV.16)$$

where we can define

$$G^T(\vec{q}, \omega) = \frac{1}{2} [G^{+-}(\vec{q}, \omega) + G^{-+}(\vec{q}, \omega)]. \quad (IV.17)$$

Then Eq. (IV.17) has the same form as that derived by McLean and Blume.⁴²

It should be noted however that it is valid only for $T > T_c$ when the system is approximated in the above discussed manner. Thus one of the most significant features of the inclusion of local order above the critical point is that instead of a single correlation function both

the transverse and longitudinal correlations with respect to the axis of quantization, the local z axis, must be considered.

In Section B we will calculate the first and second order coefficients of the powers of t in the series expansion for the transverse dynamical spin correlation function $g^T(\vec{q}, t)$ where

$$g^T(\vec{q}, t) = \frac{1}{2}[g^{+-}(\vec{q}, t) + g^{-+}(\vec{q}, t)]. \quad (\text{IV.18})$$

An approximate expression for $g^T(\vec{q}, t)$ will then be obtained by a resummation and $G^T(\vec{q}, \omega)$ obtained by Fourier transformation. In Section C an equation for $G^{ZZ}(\vec{q}, \omega)$ will be determined.

B. Determination of $G^T(\vec{q}, \omega)$

Since $G^{+-}(\vec{q}, \omega)$ and $G^{-+}(\vec{q}, \omega)$ may be calculated in a parallel manner, we will concentrate on the evaluation of $G^{+-}(\vec{q}, \omega)$. From this discussion the expression for $G^{-+}(\vec{q}, \omega)$ will follow directly.

As discussed in Chapter III, Eq. (III.35), the dynamical spin correlation function can be expanded in a Taylor series in powers of t .

Thus

$$g^+(\vec{q}, t) = \sum_{n=0}^{\infty} \frac{(-it)^n}{n!} \langle G_{\vec{q}}^{(n)} \rangle^{+-}, \quad (\text{IV.19})$$

and so

$$G^{+-}(\vec{q}, \omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} [\langle G_{\vec{q}}^{(0)} \rangle^{+-} - i \langle G_{\vec{q}}^{(1)} \rangle t - \frac{1}{2!} \langle G_{\vec{q}}^{(2)} \rangle t^2 - \dots]. \quad (\text{IV.20})$$

From Eqs. (III.40 and III.42) we have the result that

$$\langle G_{\vec{q}}^{(0)} \rangle^{+-} = \langle S_{\vec{q}}^+ S_{-\vec{q}}^- \rangle, \quad (\text{IV.21})$$

$$\langle G_{\vec{q}}^{(1)} \rangle^{+-} = \langle S_{\vec{q}}^+ [H, S_{-\vec{q}}^-] \rangle \quad (\text{IV.22})$$

and

$$\langle G_{\vec{q}}^{(2)} \rangle^{+-} = \langle [S_{\vec{q}}^+, H] [H, S_{-\vec{q}}^-] \rangle. \quad (\text{IV.23})$$

Now in order to determine exactly how the presence of positional correlations of the spins above T_c affects the calculation of the lowest order coefficients, the spatial Fourier transform of the first order coefficient will be examined:

$$\langle G_{\vec{q}}^{(1)} \rangle^{+-} = \sum_j \langle G_{ij}^{(1)} \rangle^{+-} e^{i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.24})$$

where

$$\langle G_{ij}^{(1)} \rangle^{+-} = \langle S_i^+ [H, S_j^-] \rangle. \quad (\text{IV.25})$$

Here Eqs. (IV.1-IV.3) have been used in the derivation of these expressions.

Using Eq. (IV.1) and the following commutator relations

$$[S_i^z, S_j^+] = \delta_{ij} S_i^+, \quad (\text{IV.26})$$

$$[S_i^z, S_j^-] = \delta_{ij} S_i^-, \quad (\text{IV.27})$$

and

$$[s_i^+, s_j^-] = 2\delta_{ij} s_i^z, \quad (IV.28)$$

we can expand Eq. (IV.25) and get

$$\langle G_{ij}^{(1)} \rangle^{+-} = -2 \sum_{j\ell} J_{j\ell} \{ \langle s_i^+ s_\ell^- s_j^z \rangle - \langle s_i^+ s_j^- s_\ell^z \rangle \}. \quad (IV.29)$$

Thus

$$\begin{aligned} \langle G_{\vec{q}}^{(1)} \rangle^{+-} &= -2 \sum_{j\ell} J_{j\ell} [\langle s_i^+ s_\ell^- s_j^z \rangle \\ &\quad - \langle s_i^+ s_j^- s_\ell^z \rangle] e^{-i\vec{q} \cdot \vec{R}_{ij}}. \end{aligned} \quad (IV.30)$$

Then, as discussed in Section A, when the wave vector \vec{q} satisfies the inequality $r < q^{-1} < \lambda$, the dominant contribution to $\langle G_{\vec{q}}^{(1)} \rangle$ comes from those terms in the sums over j and ℓ in Eq. (IV.28) in which all the spins at sites i , ℓ and j lie within a strongly correlated spin cluster where the degree of spin order in these clusters is determined by the local order parameter. In order to quantitatively determine this contribution, we define P to be the probability that the spins at sites i , j , and ℓ all lie within a cell whose size is measured by λ . Consequently, $(1 - P)$ is just the probability that the spins on sites i , j , and ℓ are not all contained within this cell. Thus we can write $\langle G_{\vec{q}}^{(1)} \rangle^{+-}$ in the following way:

$$\langle G_{\vec{q}}^{(1)} \rangle^{+-} \cong \sum_j (A_{ij}^{+-}(1) + B_{ij}^{+-}(1)) e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (IV.31)$$

where

$$A_{ij}^{+-}(1) = \langle S_i^+ [H, S_j^-] \rangle_{sw} P, \quad (IV.32)$$

$$B_{ij}^{+-}(1) = \langle S_i^+ [H, S_j^-] \rangle (1 - P) \quad (IV.33)$$

in which the significance of $\langle \dots \rangle_{sw}$ is described directly below.

An expression for P may be determined in the following way. Since we have assumed that the range of the exchange interaction is much less than the two spin correlation length λ , then if the spins at sites i and j are within a distance λ of each other, all the spins at sites i , j , and l will lie within this region. So the probability P may simply be equated to the normalized two-spin correlation function

$$P \equiv \rho_{ij} = \frac{\langle \vec{s}_i \cdot \vec{s}_j \rangle}{S(S+1)}. \quad (IV.34)$$

When the spins on sites i and j are aligned, $\rho_{ij} = 1$; and when they are statistically independent, $\rho_{ij} = 0$. Thus P is the magnitude of the local order parameter in the spatial region near the pair of spins i and j .

In evaluating $A_{ij}^{+-}(1)$, we note that all the spins contained in the multi-spin correlation functions comprising $A_{ij}^{+-}(1)$ fall within correlated spin clusters. Therefore, these correlation functions will be approximated through the use of spin wave theory for an infinite crystal, which is symbolized by $\langle \dots \rangle_{sw}$. This is done by letting

$$S_i^+ \approx \sqrt{2S} a_i, \quad (IV.35)$$

$$S_i^- \simeq \sqrt{2S} a_i, \quad (\text{IV.36})$$

and

$$S_i^z = -S + a_i^\dagger a_i, \quad (\text{IV.37})$$

where

$$a_i = \frac{1}{\sqrt{N}} \sum_{\vec{k}} a_{\vec{k}} e^{i\vec{k} \cdot \vec{R}_i}, \quad (\text{IV.38})$$

$$a_i^\dagger = \frac{1}{\sqrt{N}} \sum_{\vec{k}} a_{\vec{k}}^\dagger e^{-i\vec{k} \cdot \vec{R}_i}, \quad (\text{IV.39})$$

in which $a_{\vec{k}}^\dagger$ and $a_{\vec{k}}$ are the magnon creation and annihilation operators respectively. The above expressions are just the Holstein-Primakoff spin operator expansions¹¹ in which only the lowest order terms retained. Then

$$\langle S_i^+ S_\ell^- S_j^z \rangle \simeq S (2S) \langle a_i a_\ell^\dagger \rangle, \quad (\text{IV.40})$$

and $A_{ij}^{+-}(1)$ has the form

$$A_{ij}^{+-}(1) = -2S \sum_{\ell} J_{j\ell} \{2S[\langle a_i a_\ell^\dagger \rangle - \langle a_i a_j^\dagger \rangle]\} \rho_{ij}, \quad (\text{IV.41})$$

where

$$\langle a_i a_\ell^\dagger \rangle = \frac{1}{N} \sum_{\vec{k}} (1 + \langle n_{\vec{k}} \rangle) e^{i\vec{k} \cdot \vec{R}_{i\ell}}, \quad (\text{IV.42})$$

since $[a_{\vec{k}}, a_{\vec{k}'}^\dagger] = \delta_{\vec{k}, \vec{k}'}$ and $n_{\vec{k}} = a_{\vec{k}}^\dagger a_{\vec{k}}$ which is the magnon number operator.

It should be noted that by using this approximation we assume that the spins within the spin cluster tend to be aligned along the local z axis of the cluster.

We will now evaluate the multiple spin correlation functions in $B_{ij}^{+-}(1)$. Since the spins in this term lie outside the spin clusters the multiple spin correlation functions may be approximated through the use of the cumulant expansion method employed by Reiter;⁴⁰

$$\langle S_i^+ S_\ell^- S_j^z \rangle \simeq \langle S_i^+ S_\ell^- \rangle \langle S_j^z \rangle, \quad (\text{IV.43})$$

where $\langle \dots \rangle$ is an average over an equilibrium canonical ensemble.

However, $\langle S_j^z \rangle = 0$ when $T > T_c$, since there is no preferred direction for spins lying outside the spin clusters. Consequently,

$$B_{ij}^{+-}(1) = 0. \quad (\text{IV.44})$$

The above argument can be extended to include all odd ordered coefficients so that

$$B_{ij}^{+-}(2n+1) = 0; \quad n=0, 1, 2, \dots \quad (\text{IV.45})$$

This is a result of the fact that the multi-spin correlation functions comprising $B_{ij}^{+-}(2n+1)$ must be rotationally invariant because the collection of spins lying outside the spin clusters have this symmetry property. But the correlation functions in $B_{ij}^{+-}(2n+1)$ contain odd numbers of spins, and thus the requirement of rotational invariance forces these equilibrium ensemble averages to be identically equal to zero.

As a consequence of this result $\langle G_{\vec{q}}^{(1)} \rangle^{+-}$ is determined solely by the spin cluster contribution, $A_{ij}^{+-}(1)$. We can obtain an explicit expression for the first order coefficient by taking the Fourier transform of $A_{ij}^{+-}(1)$. Since

$$\rho_{ij} = \frac{1}{N} \sum_{\vec{q}} \rho(\vec{q}) e^{i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.46})$$

where

$$\langle S_i^z S_j^z \rangle = \frac{1}{2} \langle S_i^+ S_j^- \rangle = \frac{1}{3} S(S+1) \rho_{ij}, \quad (\text{IV.47})$$

then for $T \gtrsim T_c$ and $r < q^{-1} < \lambda$

$$\langle G_{\vec{q}}^{(1)} \rangle^{+-} = \frac{2S}{N} \sum_{\vec{q}'} \omega(\vec{q}') [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'), \quad (\text{IV.48})$$

with

$$\langle n_{\vec{q}'} \rangle = \frac{1}{e^{\beta\omega(\vec{q}')} - 1}, \quad (\text{IV.49})$$

and

$$\omega(\vec{q}') = 2S[J(0) - J(\vec{q}')]. \quad (\text{IV.50})$$

The calculation of the second order coefficient $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$ is done in an analogous manner. Since

$$\langle G_{\vec{q}}^{(2)} \rangle^{+-} = \sum_j \langle G_{ij}^{(2)} \rangle^{+-} e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.51})$$

then when $r < q^{-1} < \lambda$ the dominant contribution comes from those terms

in the sum on j in which all the spins lie within a correlated spin cluster. A quantitative determination of this contribution can be made by approximating $\langle G_{ij}^{(2)} \rangle^{+-}$ as

$$\langle G_{ij}^{(2)} \rangle^{+-} = A_{ij}^{+-}(2) + B_{ij}^{+-}(2), \quad (\text{IV.52})$$

where

$$A_{ij}^{+-}(2) = \langle G_{ij}^{(2)} \rangle_{sw}^{+-} \rho_{ij}, \quad (\text{IV.53})$$

and

$$B_{ij}^{+-}(2) = \langle G_{ij}^{(2)} \rangle^{+-} (1 - \rho_{ij}). \quad (\text{IV.54})$$

Spin wave theory is used in evaluating $A_{ij}^{+-}(2)$ and the result is

$$\begin{aligned} A_{ij}^{+-}(2) = 4S^2 \sum_{mn} J_{in} J_{jm} \{ (2S) [\langle a_n a_m^\dagger \rangle \\ - \langle a_i a_m^\dagger \rangle - \langle a_n a_j^\dagger \rangle + \langle a_i a_j^\dagger \rangle] \} \rho_{ij}, \end{aligned} \quad (\text{IV.55})$$

where as long as $r \ll \lambda$, ρ_{ij} is the probability that the spins on sites i, j, n, m all lie within a correlated spin cluster.

Again since the spins in $B_{ij}^{+-}(2)$ lie outside the spin clusters, Reiter's decoupling scheme can be used to approximate the four-spin correlation functions in terms of products of equilibrium spin pair correlation functions:

$$\langle s_i^+ s_m^z s_j^- \rangle \simeq \langle s_i^+ s_j^- \rangle \langle s_m^z s_n^z \rangle, \quad (\text{IV.56})$$

which implies that the spin pair at sites i and j is statistically independent of the spin pair at m and n . Thus

$$\begin{aligned}
 B_{ij}^{+-}(2) &= 4 \sum_{mn} J_{in} J_{jm} [\langle S_n^+ S_m^- \rangle \langle S_i^z S_j^z \rangle \\
 &\quad - \langle S_i^+ S_m^- \rangle \langle S_n^z S_j^z \rangle - \langle S_n^+ S_j^- \rangle \langle S_i^z S_m^z \rangle \\
 &\quad + \langle S_i^+ S_j^- \rangle \langle S_n^z S_m^z \rangle] (1 - \rho_{ij}). \tag{IV.57}
 \end{aligned}$$

Upon taking the Fourier transform of $A_{ij}^{+-}(2)$ and $B_{ij}^{+-}(2)$ we obtain

$$\begin{aligned}
 \langle G_{\vec{q}}^{(2)} \rangle^{+-} &= \frac{2S}{N} \sum_{\vec{q}'} [\omega(\vec{q}')]^2 [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}') \\
 &\quad + \frac{8}{N} \left[\frac{1}{3} S(S+1) \right]^2 \left\{ \sum_{\vec{q}'} [J(\vec{q}') - J(\vec{q} - \vec{q}')]^2 \rho(\vec{q}') \rho(\vec{q} + \vec{q}') \right. \\
 &\quad \left. - \sum_{\vec{q}'\vec{q}''} [J(\vec{q}') - J(\vec{q}' + \vec{q}'')]^2 \rho(\vec{q}') \rho(\vec{q}' + \vec{q}'') \rho(\vec{q} - \vec{q}'') \right\}. \tag{IV.58}
 \end{aligned}$$

The first term in the above equation is the Fourier transform of $A_{ij}^{+-}(2)$, and thus it gives the spin cluster contribution to $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$, i.e. it is due to the local order above the critical point. Both the second and third terms on the right hand side of Eq. (IV.58) come from $B_{ij}^{+-}(2)$. It should be noted that the second term is quite similar to the expression Reiter³⁹ has derived for the second moment of the spectral density except for the factor $\rho(\vec{q})$ which appears in the denominator of his equation. The third term plays a very important role at temperatures near T_c because as $T \rightarrow T_c$, $\rho(\vec{q})$ becomes strongly peaked at $\vec{q} = 0$ for

a ferromagnet. The width of this peak is measured by λ^{-1} . Consequently since we are considering only those wave vectors which satisfy $q > \lambda^{-1}$ the third term tends to cancel the second. This behavior will be discussed in detail through the use of a specific model in Appendix B. Therefore, when $T \gtrsim T_c$ the short range order contribution dominates near the critical temperature, and

$$\langle G_{\vec{q}}^{(2)} \rangle^{+-} \approx \frac{2S}{N} \sum_{\vec{q}'} [\omega(\vec{q}')]^2 [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.59})$$

In like manner it can be readily shown (Appendix A) that the dominant contribution to $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ is also due to correlated spin clusters:

$$\langle G_{\vec{q}}^{(0)} \rangle^{+-} = \frac{2S}{N} \sum_{\vec{q}'} [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.60})$$

Now by examining the results we obtained for the first three coefficients, Eqs. (IV.60), (IV.48) and (IV.59), in the series expansion of $g^{+-}(\vec{q}, t)$ we observe that they have a very simple form which leads us to postulate that when $T \gtrsim T_c$ and $r < q^{-1} < \lambda$,

$$\langle G_{\vec{q}}^{(n)} \rangle^{+-} \approx \frac{2S}{N} \sum_{\vec{q}'} [\omega(\vec{q}')]^n [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.61})$$

for $n > 2$. However, we are not able to give a general proof for this expression because as the order of the coefficient increases more and more spins are contained within a cluster of size λ , and therefore the criterion, Eq. (IV.11), which determines the validity of our decoupling procedure will break down eventually, but the discrepancies that do

cor should come in the higher order coefficients which affect only the behavior of the wings of the line shape. Consequently, a good result for the central line shape may still be expected. When Eq. (IV.61) is substituted into Eq. (IV.20), then

$$G^{+-}(\vec{q}, \omega) = \frac{2S}{N} \sum_{\vec{q}'} \left\{ \frac{1}{2} \int_{-\infty}^{\infty} dt e^{-i\omega t} \left[\sum_m \frac{(it)^m}{m!} (\omega(\vec{q}'))^m \right] \right\} \\ \times [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.62})$$

Upon completing the integration we obtain

$$G^{+-}(\vec{q}, \omega) = \frac{2S}{N} \sum_{\vec{q}'} \delta[\omega - \omega(\vec{q}')] [\langle n_{\vec{q}'} \rangle + 1] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.63})$$

In a similar fashion we can determine $G^{-+}(\vec{q}, \omega)$. The only difference is that the factor $\langle n_{\vec{q}'} \rangle + 1$ in Eq. (IV.63) is replaced by $\langle n_{\vec{q}'} \rangle$, and $\delta(\omega - \omega(\vec{q}')) \rightarrow \delta(\omega + \omega(\vec{q}'))$. Thus from Eq. (IV.17), $G^T(\vec{q}, \omega)$, the time Fourier transform of the transverse dynamical spin correlation function, becomes

$$G^T(\vec{q}, \omega) = \frac{S}{N} \sum_{\vec{q}'} [\langle n_{\vec{q}'} \rangle \delta(\omega - \omega(\vec{q}')) \\ + \langle n_{\vec{q}'} \rangle \delta(\omega + \omega(\vec{q}'))] \rho(\vec{q} - \vec{q}'). \quad (\text{IV.64})$$

It should be noted that the above expression contains only the one-magnon contribution to the transverse correlation function. In order to determine the first order nonlinear contribution to $G^T(\vec{q}, \omega)$, we must include the next order term in the Holstein-Primakoff spin operator

expansions for S_i^+ and S_i^- , so that

$$S_i^+ \approx \sqrt{\frac{2S}{N}} \sum_{\vec{k}} e^{i\vec{k} \cdot \vec{R}_i} a_{\vec{k}} - \frac{1}{\sqrt{8NS}} \sum_{\vec{k}, \vec{k}', \vec{k}''} e^{-i(\vec{k} - \vec{k}' - \vec{k}'') \cdot \vec{R}_i} a_{\vec{k}}^\dagger a_{\vec{k}'} a_{\vec{k}''}, \quad (IV.65)$$

and

$$S_i^- \approx \sqrt{\frac{2S}{N}} \sum_{\vec{k}} e^{-i\vec{k} \cdot \vec{R}_i} a_{\vec{k}}^\dagger - \frac{1}{\sqrt{8NS}} \sum_{\vec{k}, \vec{k}', \vec{k}''} e^{-i(\vec{k} + \vec{k}' - \vec{k}'') \cdot \vec{R}_i} a_{\vec{k}}^\dagger a_{\vec{k}'}^\dagger a_{\vec{k}''}. \quad (IV.66)$$

Then using the same procedure as outlined above we can obtain not only the one magnon contribution but also the lowest order nonlinear contribution to $G^T(\vec{q}, \omega)$:

$$G^T(\vec{q}, \omega) = \frac{1}{N} (S - \frac{1}{N} \sum_{\vec{q}''} \langle n_{\vec{q}''} \rangle) \sum_{\vec{q}'} \{ (\langle n_{\vec{q}'} \rangle + 1) \delta[\omega - \omega(\vec{q}')] + \langle n_{\vec{q}'} \rangle \delta[\omega + \omega(\vec{q}')] \} \rho(\vec{q} - \vec{q}'). \quad (IV.67)$$

The characteristics of the line shape generated by $G^T(\vec{q}, \omega)$ will be discussed in Section D.

C. Determination of $G^{ZZ}(\vec{q}, \omega)$

As in the above discussion for $G^T(\vec{q}, \omega)$, an approximate expression for $G^{ZZ}(\vec{q}, \omega)$ will be obtained by another Taylor series expansion in powers of t . The lowest order coefficients in this series will be calculated and used to infer an approximate series expression for the longitudinal spin correlation function. Upon Fourier transformation of this result $G^{ZZ}(\vec{q}, \omega)$ is obtained.

The longitudinal spin correlation function is given by

$$g^{ZZ}(\vec{q}, t) = \langle S_{\vec{q}}^Z(0) S_{-\vec{q}}^Z(t) \rangle, \quad (\text{IV.68})$$

whose Taylor series in powers of t is

$$g^{ZZ}(\vec{q}, t) = \sum_n \frac{(-it)^n}{n!} \langle G_{\vec{q}}^{(n)} \rangle^{ZZ}, \quad (\text{IV.69})$$

where

$$\langle G_{\vec{q}}^{(n)} \rangle^{ZZ} = \left[\left(\frac{1}{i} \frac{\partial}{\partial t} \right)^n \langle S_{\vec{q}}^Z(0) S_{-\vec{q}}^Z(t) \rangle \right]_{t=0}. \quad (\text{IV.70})$$

The first three coefficients in this series are

$$\langle G_{\vec{q}}^{(0)} \rangle^{ZZ} = \langle S_{\vec{q}}^Z S_{-\vec{q}}^Z \rangle, \quad (\text{IV.71})$$

$$\langle G_{\vec{q}}^{(1)} \rangle^{ZZ} = \langle S_{\vec{q}}^Z [H, S_{-\vec{q}}^Z] \rangle, \quad (\text{IV.72})$$

$$\langle G_{\vec{q}}^{(2)} \rangle^{zz} = \langle [S_{\vec{q}}^z, H] [H, S_{-\vec{q}}^z] \rangle. \quad (\text{IV.73})$$

The calculation of these coefficients closely parallels the calculation carried out for $g^T(\vec{q}, t)$. Consequently, we will only discuss the most salient parts. For example, the spatial Fourier transform of $\langle G_{\vec{q}}^{(1)} \rangle^{zz}$ is given by

$$\langle G_{\vec{q}}^{(1)} \rangle^{zz} = \sum_j (G_{ij}^{(1)})^{zz} e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{IV.74})$$

where

$$\begin{aligned} (G_{ij}^{(1)})^{zz} &= \langle S_i^z [H, S_j^z] \rangle \\ &= - \sum_m J_{jm} (\langle S_i^z S_j^+ S_m^- \rangle - \langle S_i^z S_m^+ S_j^- \rangle) \end{aligned} \quad (\text{IV.75})$$

which has been derived through the use of Eqs. (IV.26) - (IV.28).

From our discussion in Sections A and B, the dominant contribution to $\langle G_{\vec{q}}^{(1)} \rangle^{zz}$ comes from clusters of strongly correlated spins. We can thus make the same approximations in order to determine the three-spin static correlation functions. The result we obtain is

$$\begin{aligned}
\langle G_{\vec{q}}^{(1)} \rangle^{zz} &= \frac{1}{2N^2} \sum_{\vec{q}' \vec{q}''} \{ [\omega(\vec{q}'') - \omega(\vec{q}')] \langle n_{\vec{q}'} \rangle (\langle n_{\vec{q}''} \rangle + 1) \\
&\quad + [\omega(\vec{q}') - \omega(\vec{q}'')] \langle n_{\vec{q}''} \rangle (\langle n_{\vec{q}'} \rangle + 1) \} \\
&\quad \times \rho(\vec{q} - \vec{q}' + \vec{q}''). \tag{IV.76}
\end{aligned}$$

In a similar manner the dominant contribution to $\langle G_{\vec{q}}^{(0)} \rangle^{zz}$ and $\langle G_{\vec{q}}^{(2)} \rangle^{zz}$ can be found. The results for these coefficients are obtained in

Appendix C. Then by taking

$$\begin{aligned}
\langle G_{\vec{q}}^{(m)} \rangle^{zz} &= \frac{1}{2M^2} \sum_{\vec{q}' \vec{q}''} \{ [\omega(\vec{q}') - \omega(\vec{q}'')]^m (1 + \langle n_{\vec{q}'} \rangle) \langle n_{\vec{q}''} \rangle \\
&\quad + [\omega(\vec{q}'') - \omega(\vec{q}')]^m (1 + \langle n_{\vec{q}''} \rangle) \langle n_{\vec{q}'} \rangle \} \rho(\vec{q} + \vec{q}' - \vec{q}'') \tag{IV.77}
\end{aligned}$$

to be the general form for the coefficients of the powers of t in the series expansion for the longitudinal dynamical spin correlation function, we finally obtain, upon Fourier transformation, the following approximate expression for $G^{zz}(\vec{q}, \omega)$:

$$\begin{aligned}
G^{zz}(\vec{q}, \omega) = & \left[S - \frac{1}{N} \sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle \right]^2 \rho(\vec{q}) \delta(\omega) \\
& + \frac{1}{2N^2} \sum_{\vec{q}', \vec{q}''} \{ \delta(\omega + \omega(\vec{q}') - \omega(\vec{q}'')) (1 + \langle n_{\vec{q}'} \rangle) \langle n_{\vec{q}''} \rangle \\
& + \delta(\omega + \omega(\vec{q}') - \omega(\vec{q}'')) (1 + \langle n_{\vec{q}''} \rangle) \langle n_{\vec{q}'} \rangle \} \\
& \times \rho(\vec{q} - \vec{q}' + \vec{q}''). \tag{IV.78}
\end{aligned}$$

where the first term in the above expression is due to the Holstein-Primakoff spin operator expansion for $S_{\vec{q}}^z$. The physical implications of this result will be discussed in the next section.

D. Transverse and Longitudinal Contributions to the Line Shape

Using the equations that have been derived for $G^T(\vec{q}, \omega)$ and $G^{zz}(\vec{q}, \omega)$ we will determine the frequency distribution and wave vector distribution of the neutron magnetic scattering cross section.

The relevant function which contains all the information concerning these distributions is $G(\vec{q}, \omega)$ which, as can be seen from Eq. (IV.16) is equal to the sum of $G^T(\vec{q}, \omega)$ and $G^{zz}(\vec{q}, \omega)$ in our model. Thus in order to gain a quantitative understanding of the line shape characteristics and their physical significance we will consider these functions separately.

In Eq. (IV.67) for $G^T(\vec{q}, \omega)$, we first vary ω keeping \vec{q} fixed which corresponds to a constant momentum scan. When this is done it is

evident that two peaks will be present in the line shape. The broadening of these peaks is determined not only by the Bose-Einstein distribution function $\langle n_{\vec{q}} \rangle$ but also by $\rho(\vec{q} - \vec{q}')$. The contribution of $\rho(\vec{q} - \vec{q}')$ to this broadening is particularly interesting because we have chosen ρ_{ij} as our local order parameter and so $\rho(\vec{q} - \vec{q}')$ is a measure of the spatial fluctuations of the correlated spin clusters. Thus these fluctuations of the local order regions contribute directly to the line broadening. Since the width of $\rho(\vec{q} - \vec{q}')$ is on the order of $1/\lambda$, when $1/\lambda \ll q$, ρ becomes strongly peaked about $\vec{q}' = \vec{q}$. Consequently, distinct peaks will be obtained at $\omega = \pm \omega(\vec{q})$ which correspond to the emission and absorption of magnons. When \vec{q} is varied while ω is fixed two peaks are found at $|\vec{q}| = \pm |\vec{q}_0|$ (when $1/\lambda \ll q$), where $\omega(\vec{q}_0) = \omega$, which are also broadened by $\langle n_{\vec{q}} \rangle$ and $\rho(\vec{q} - \vec{q}')$. The factor multiplying the sums over \vec{q}' in $G^T(\vec{q}, \omega)$,

$$S = \sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle, \quad (\text{IV.79})$$

also has physical significance because from elementary spin wave theory we know that

$$\sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle \propto \Delta M = M_0 - M(T) \quad (\text{IV.80})$$

where M_0 is the magnetization for the completely ordered state which in our model would occur at $T = T_c$. Therefore

$$S = \sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle \propto M(T), \quad (\text{IV.81})$$

and so the size of $G^T(\vec{q}, \omega)$ is modified by a term which is proportional to the temperature dependent magnetization.

The characteristics of the line shape contribution from $G^{ZZ}(\vec{q}, \omega)$ can be discerned directly from Eq. (IV.78). The first term in this expression is a central peak which is a delta function of the frequency at $\omega = 0$, in our approach. In the limit as $T \rightarrow T_c$, $\rho(\vec{q}) \rightarrow \delta(\vec{q})$ and the first term gives the elastic Bragg scattering contribution to the cross section. Consequently, when $T > T_c$ it is apparent that our central peak is the remnant of the elastic Bragg peak which is diffused spatially due to the fluctuations in the local order parameter as measured by $\rho(\vec{q})$. The model that we have constructed gives a central peak that has a zero frequency width. This implies that the decay time of the local order parameter is infinite,⁴² which is to be expected because our local order parameter is time independent. It is also evident upon examination of the expression for $G^{ZZ}(\vec{q}, \omega)$ that there are two-magnon terms contained in it which are also broadened by convolution with $\rho(\vec{q})$.

E. Comparison with Previous Theories

Although we have not actually determined the moments of the spectral shape function in Sections B and C, we can still compare our calculation with moment expansion theories since $G^{\alpha\alpha}(\vec{q}, \omega)$ is closely related to the shape function, $R^{\alpha\alpha}(\vec{q}, \omega)$:

$$R^{\alpha\alpha}(\vec{q}, \omega) = \frac{G^{\alpha\alpha}(\vec{q}, \omega)}{\int_{-\infty}^{\infty} G^{\alpha\alpha}(\vec{q}, \omega) d\omega} = \frac{G^{\alpha\alpha}(\vec{q}, \omega)}{\langle S_{\vec{q}}^{\alpha} S_{-\vec{q}}^{\alpha} \rangle}.$$

Thus we can relate the lowest order coefficients of the powers of t in the Taylor series expansion for the dynamical spin correlation function to the moments of $R^{\alpha\alpha}(\vec{q}, \omega)$. From Eqs. (III.36) and (III.40) - (III.42), we find that the second moment is given by

$$\langle \omega_{\vec{q}}^2 \rangle^{\alpha\alpha} = \frac{\langle G_{\vec{q}}^{(2)} \rangle^{\alpha\alpha}}{\langle G_{\vec{q}}^{(0)} \rangle^{\alpha\alpha}}, \quad (\text{IV.83})$$

and that the fourth moment is simply

$$\langle \omega_{\vec{q}}^4 \rangle^{\alpha\alpha} = \frac{\langle G_{\vec{q}}^{(4)} \rangle^{\alpha\alpha}}{\langle G_{\vec{q}}^{(0)} \rangle^{\alpha\alpha}}. \quad (\text{IV.84})$$

In Reiter's derivation of the second and fourth moments of the spectral density above the critical temperature,⁴⁰ the fluctuation of the second moment, defined by,

$$\langle \Omega_{\vec{q}}^4 \rangle^{\alpha\alpha} = \langle \omega_{\vec{q}}^4 \rangle^{\alpha\alpha} - (\langle \omega_{\vec{q}}^2 \rangle^{\alpha\alpha})^2, \quad (\text{IV.85})$$

satisfies the following relation

$$\frac{\langle \omega_{\vec{q}}^4 \rangle^{\alpha\alpha}}{(\langle \omega_{\vec{q}}^2 \rangle^{\alpha\alpha})^2} \approx \frac{3}{2}, \quad (\text{IV.86})$$

when $q \gg \lambda^{-1}$. However, in our calculation through the use of the results from Sections B and C along with Eqs. (IV.83) and (IV.84) we obtain

$$\langle \omega_{\vec{q}}^2 \rangle^{\alpha\alpha} \approx [\omega(\vec{q})]^2, \quad (\text{IV.87})$$

and

$$\langle \omega_{\vec{q}}^4 \rangle^{\alpha\alpha} \approx [\omega(\vec{q})]^4, \quad (\text{IV.88})$$

when $r < q \ll \lambda^{-1}$ and $T \gtrsim T_c$. Here $\omega(\vec{q})$ is the spin wave frequency defined by Eq. (IV.50). Consequently,

$$\langle \omega_{\vec{q}}^4 \rangle^{\alpha\alpha} \approx 0. \quad (\text{IV.89})$$

Then from Marshall and Lowde's discussion of moment theory we see that in our calculation when the temperature approaches T_c and $r < q^{-1} < \lambda$ the spectral shape function will have well-resolved peaks corresponding to the excitation and absorption of spin waves. In Reiter's calculation, however, no spin wave peaks are found in the theoretical line shape when $q > \lambda^{-1}$ and $T > T_c$. His result is contradicted by the experimental observation of quasi-spin wave peaks in the neutron scattering cross

section of Ni¹⁷ and RbMnF₃.¹⁸ The most important reason for this difficulty in Reiter's approach can be traced to the method he used to decouple the multiple-spin correlation functions.

As was stated in Section B, Reiter decoupled the four-spin correlation functions in the following way:

$$\langle S_n^+ S_\ell^Z S_m^- S_j^Z \rangle \simeq \langle S_n^+ S_m^- \rangle \langle S_\ell^Z S_j^Z \rangle, \quad (\text{IV.90})$$

of spins. This particular decoupling scheme, that all spin clusters are statistically independent from every other pair, is valid at high temperatures, where there is much spin disorder. In this temperature region the spin correlation length λ , which is a measure of the size of a correlated spin cluster, is quite small. Therefore, when $q \ll \lambda^{-1}$ where q is the spin fluctuation wave vector, these fluctuations span many spin clusters oriented randomly and thus they must behave quite differently from spin waves.

Reiter's decoupling scheme is quite analogous to one which was developed in order to treat phonon-phonon interactions and which has been quite successful in describing the phenomenon of acoustic attenuation. In studying the attenuation of long wavelength sound waves in Helium II, Kawasaki⁴⁶⁻⁴⁸ found that when $q \ll \lambda^{-1}$, where q is the wave vector of the sound wave, and λ^{-1} is the inverse correlation length of the particle fluctuations, the correlation functions containing multiple phonon creation and annihilation operators found in the expression for the attenuation constant can be approximated by products of pair correlation functions.

Although this particular decoupling procedure was found to be quite useful in the acoustic attenuation problem, its application to the problem of spin fluctuations leads to difficulties because it does not adequately account for the positional spin correlations that are important when $T \gtrsim T_c$ and $q^{-1} \ll \lambda$ which as we have seen must be taken into consideration in the determination of the dynamical spin correlation function.

V. DISCUSSION OF RESULTS

We now proceed with the evaluation of the expressions we obtained for the Fourier transforms of the transverse and longitudinal dynamical spin correlation functions. In order to accomplish this the sums over the wave vectors \vec{q}' and \vec{q}'' in Eqs. (IV.67) and (IV.78) must be carried out. Since these wave vectors can take on a continuous set of values the sums can be converted to integrals which can be simplified through the use of the following approximations: the spin wave dispersion curve is approximated by a parabola so that

$$J(0) - J(\vec{q}) = \alpha q^2, \quad (V.1)$$

and

$$\omega_q = 2S\alpha q^2 \equiv Dq^2, \quad (V.2)$$

where α and D are fitting parameters and D is often referred to as the spin wave stiffness.

As in other theories dealing with spin-spin interactions above T_c , our theory requires that we have an explicit expression for the static spin pair correlation function. Consequently we have used a Gaussian function of the form $\rho(\vec{q}) \propto e^{-\lambda^2 q^2}$ where $\lambda = (2D/k_B(T - T_c))^{1/2}$.

These approximations allow us to evaluate the integral expression for $G^T(\vec{q}, \omega)$ analytically, however $G^{zz}(\vec{q}, \omega)$ must be evaluated through the use of numerical integration techniques. Before we can determine

the numerical values for these functions, the values of λ , the two-spin correlation length, and D , the spin wave stiffness, must be obtained from experimental data. The spin wave stiffness constant we use is $D = 280 \text{ meV} \cdot \text{\AA}^2$. In the case of Ni, neutron inelastic scattering experiments have indicated that at $T_c + 50 \text{ K}$ ($T_c \cong 631 \text{ K}$), $\lambda^{-1} = 0.11 \text{\AA}^{-1}$. The value at any other temperature can then be calculated with the help of the temperature scaling law for the inverse correlation length, Eq. (III.44).

(As an interesting aside it should be noted that this temperature dependence yields $\lambda^{-1} = 0.15 \text{\AA}^{-1}$ at 711 K ($T_c + 80 \text{ K}$). This result indicates that the high temperature spin wave dispersion curve reported by Mook, Lynn, and Nicklow¹⁷ lies in the short wavelength region $q > \lambda^{-1}$).

The function $G(\vec{q}, \omega)$ can now be evaluated numerically. In Figure 9 a typical set of line shapes is shown for the case where ω is held constant and q is varied. (A constant energy scan.) The most salient characteristic of these line shapes is that they contain distinct peaks which are located at $q = \pm q_0$, where $\hbar\omega(q_0) \cong \hbar\omega$ is the energy of a spin wave whose wave vector is q_0 .

Although these line shapes have the same qualitative appearance as those obtained experimentally in the inelastic scattering of neutrons from Ni, a more detailed comparison reveals definite discrepancies between our simple theory and the Ni data. In Figure 10 we compare our numerical results for the full-width-at-half-maximum (FWHM) for the peak in $G(\vec{q}, \omega)$ at various temperatures with the experimental curve for Ni for an energy transfer $\Delta E = \hbar\omega = 12.4 \text{ meV}$, where the instrumental resolution

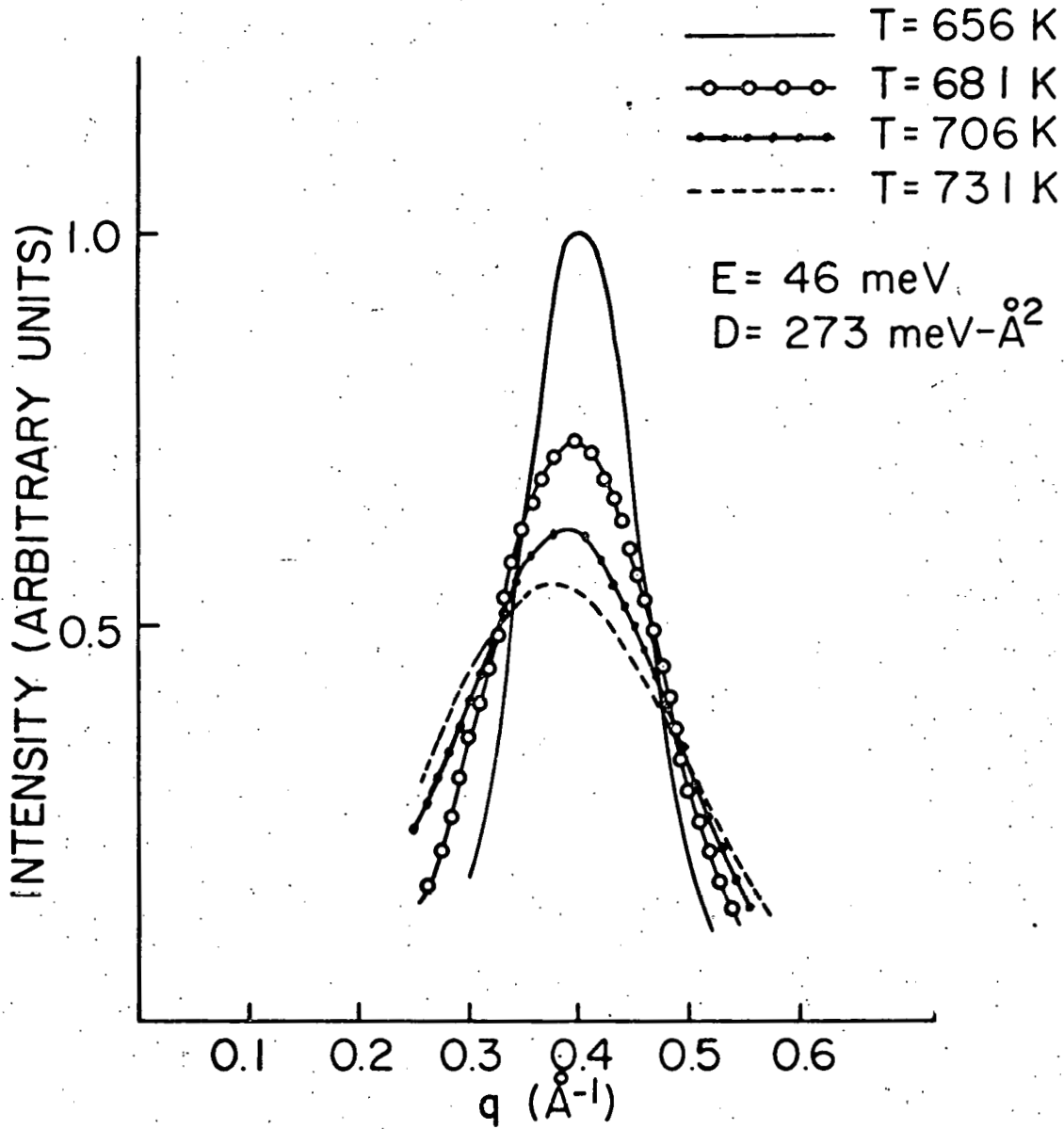


Figure 9. Temperature dependence of the theoretical line shape.

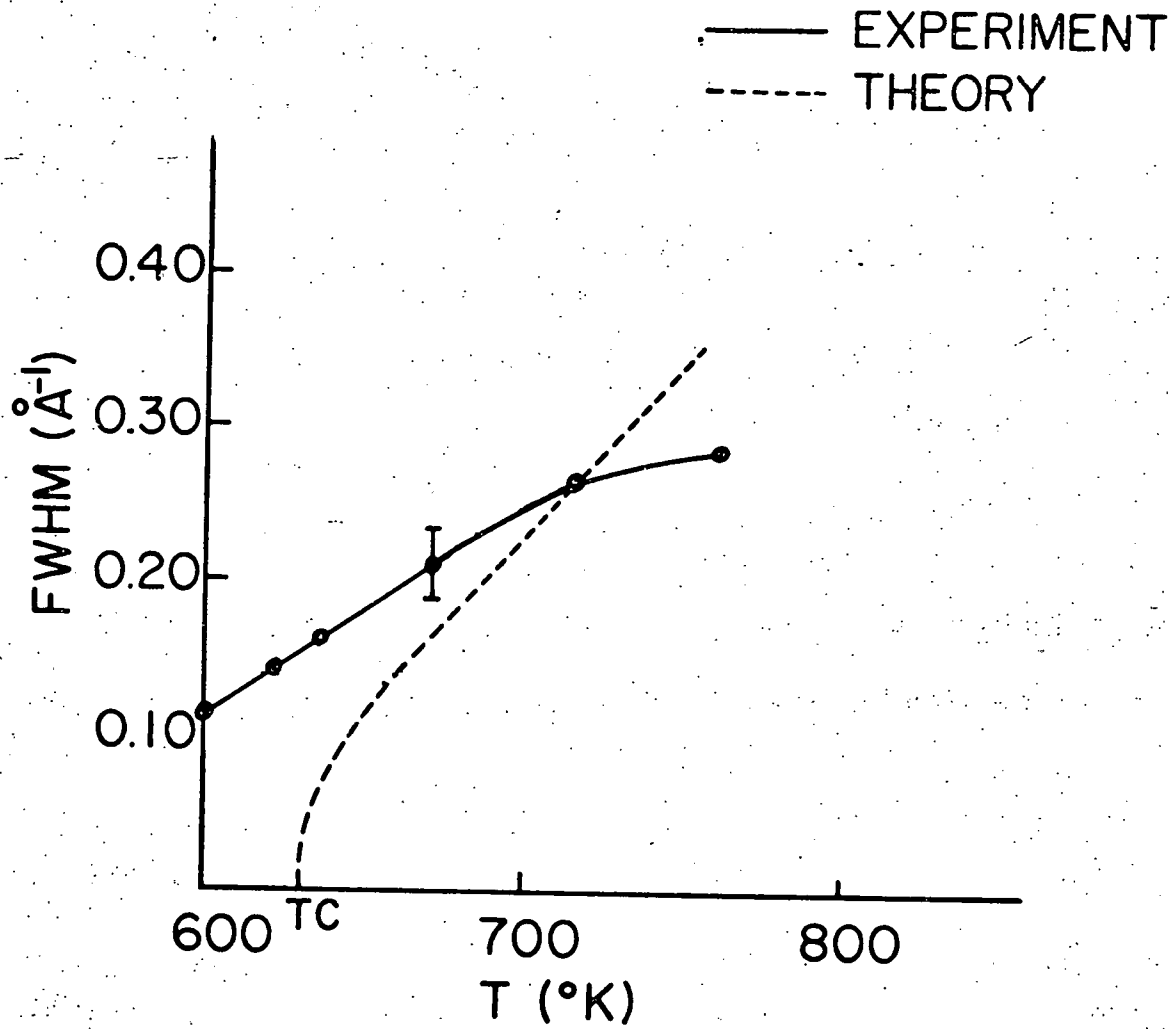


Figure 10. Comparison between the experimental and theoretical line widths in q as a function of temperature for nickel at an energy transfer of $\Delta E = 12.4$ meV.

has been subtracted out. In the region from $656^{\circ}\text{K}(1.03T_c)$ to 706°K ($1.12T_c$) the slope of the curve produced by our theory compares favorably with the experimental curve. However, below these temperatures the line width of the theoretical peak approaches zero as $T \rightarrow T_c$. This is not surprising because in applying spin wave theory to evaluate the spin correlation functions involving spins within a correlated cluster, we have included only the lowest order linear and nonlinear terms in the Holstein-Primakoff spin operator expansions. By doing this we have constructed a theory in which the spins within a cluster are strongly correlated and thus they tend to be aligned in one particular direction defined to be the local z axis of the cluster. Consequently, at $T = T_c$ the spin alignment in our model corresponds to that which actually exists near $T = 0^{\circ}\text{K}$. This observation implies that our theory should work better in the case of one dimensional magnetic systems where $T_c = 0^{\circ}\text{K}$.

The other deviation from the experimental results is observed above 706°K . In this high temperature region the theoretical curve has the same slope as it did when $T < 706^{\circ}\text{K}$ because the linewidths of the theoretical peaks continue to increase at the same rate. However, the experimental curve bends over, because the experimental linewidths are observed to remain almost constant up to the highest temperature used ($2T_c$). A possible reason for this discrepancy lies in our choice of a static spin correlation function for the measurement of the probability that the spins within a spin cluster are correlated. Consequently, in

our simple theory, line broadening is only affected by one parameter, the spin correlation length λ . As the temperature increases, λ decreases, and this tends to broaden the line because the clusters become smaller. The peaks will continue to broaden as the temperature increases. However, in a more refined calculation a time dependent spin correlation function must be used to specify the probability because above T_c spin diffusion causes the spin clusters to decay with time. Thus as the temperature increases the tendency for the line to broaden due to a decrease in the correlation length is balanced by the increase in spin diffusion which effectively allows only the large clusters to sustain a spin wave excitation while in the small clusters this cannot occur. Hence the line should not broaden very much as the temperature increases.

VI. CONCLUSION

The single most important result obtained from the foregoing calculation is that above the critical temperature when $r < q^{-1} < \lambda$ is satisfied, positional correlations between spins must be considered because these correlations directly effect the shape of the neutron scattering cross section. Within this wave vector region we have found that when the localized spin system is approximated by clusters of correlated spins whose degree of order is measured by a time-independent order parameter, the two spin correlation function $\rho(\vec{q})$, then the Fourier transform, $G(\vec{q}, \omega)$, of the dynamical spin correlation function, obtained through the use of this approximation, exhibited distinct spin wave like peaks. The full-width-at-half-maximum of these peaks and their position compare qualitatively with the experimental results obtained by Mook, Lynn, and Nicklow.¹⁷ A more detailed comparison reveals that we can only achieve a quantitative agreement between experiment and theory by refining the approximations that have been used. These include the following.

1. Magnon-magnon interactions have been neglected. This causes our line-widths to approach zero as $T \rightarrow T_c$.
2. The higher order coefficients in the time Fourier series expansion of the dynamical spin correlation function involve progressively larger numbers of spins within the cluster. Hence our approximation that $r < \lambda$ must breakdown at some order.

3. The temperature within a cluster has been assumed to be the same as that of the whole crystal. This approximation is valid as long as there are no strong temperature fluctuations over small regions in the system and the clusters are large.

4. The local order parameter used in this theory was the static spin-pair correlation function. This approximation is one of the major causes for the continued increase in the theoretical line-widths as the temperature increases. As we pointed out in Chapter V this approximation can be refined through the use of a spin diffusion correlation function.

It is also important to note that the theory developed here is applicable to simple Heisenberg ferromagnets. Consequently, it is not surprising that only quantitative agreement with the inelastic neutron scattering data is obtained with Ni because although nickel does exhibit characteristics of localized spin systems, it is well known that the magnetic moment per atom in Ni is not an integral number of Bohr magnetons which implies the d-electrons are itinerant. Furthermore, neutron scattering experiments on Ni have found a sharp cutoff in spin wave intensities above a certain energy which is predicted by the Stoner model of itinerant ferromagnetism in which a sharp decrease in intensity occurs when the spin wave dispersion curve intersects a continuum band of Stoner excitations. Our model in its present form can not predict such a sharp cutoff. These findings indicate that a complete theory dealing with spin fluctuations in metals must explicitly consider the itinerant nature of the conduction electrons.

Finally, because of the simplicity of the theory we have developed we should be able to readily modify it so that we can study the magnetic excitations of systems in which the crystal field only ground state is a singlet. In these systems it has been found⁴⁹⁻⁵¹ that the existence of a magnetic phase transition is dependent upon the magnitude of the ratio of the exchange interaction to the crystal field interaction which are usually of comparable size. When the exchange interaction is small this ratio is less than a certain threshold value⁵¹ and no magnetic ordering occurs, however when the ratio exceeds this value a magnetic phase transition is expected where the ordering is due to a polarization (induced moment) instability in the ground state wave function. In the theories⁵²⁻⁵⁵ that have been proposed this phase transition is driven by the softening of that magnetic exciton mode whose wave vector \vec{q} corresponds to the wave vector of the ordered structure. This soft-mode behavior has, however, not been observed experimentally.⁵⁶⁻⁵⁸ It is important to note that in these inelastic neutron scattering experiments the scattering wave vector used was $q > 0.5\text{\AA}^{-1}$, and thus only short wavelength magnetic exciton modes were being excited. Consequently it would be quite interesting to examine the effect of positional correlations between the induced moments on the line shape of the neutron scattering cross section through the use of our theory.

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IX. APPENDIX A: CALCULATION OF $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ AND $\langle G_{\vec{q}}^{(3)} \rangle^{+-}$

From Eq. (III.40) we know that

$$\langle G_{\vec{q}}^{(0)} \rangle = \langle S_{\vec{q}}^+ S_{-\vec{q}}^- \rangle. \quad (\text{A.1})$$

We can approximate this expansion coefficient in the manner discussed in Chapter IV by taking the positional correlations of the spins explicitly into consideration:

$$\langle G_{\vec{q}}^{(0)} \rangle^{+-} = \sum_j [A_{ij}^{+-}(0) + B_{ij}^{+-}(0)] e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{A.2})$$

where

$$A_{ij}^{+-}(0) = \langle S_i^+ S_j^- \rangle_{sw} \rho_{ij} \quad (\text{A.3})$$

and

$$B_{ij}^{+-}(0) = \langle S_i^+ S_j^- \rangle (1 - \rho_{ij}), \quad (\text{A.4})$$

in which ρ_{ij} is defined by Eq. (IV.46) in Chapter IV. The correlation function $\langle S_i^+ S_j^- \rangle$ in $A_{ij}^{+-}(0)$ can be evaluated through the use of spin wave theory since the spins at sites i and j lie within a correlated cluster. In $B_{ij}^{+-}(0)$ the spin correlation function is evaluated in terms of an equilibrium spin pair correlation function, Eq. (IV.47). When the Holstein-Primakoff spin operator expansion, Eqs. (IV.65) and (IV.66) are employed we obtain

$$\begin{aligned}
A_{ij}^{+-}(0) &= \left\{ \frac{2S}{N} \sum_{\vec{k}, \vec{k}_1} e^{i\vec{k} \cdot \vec{R}_i} e^{-i\vec{k}_1 \cdot \vec{R}_j} \langle a_{\vec{k}}^+ a_{\vec{k}_1}^+ \rangle \right. \\
&\quad - \frac{1}{2N^2} \sum_{\substack{\vec{k}, \vec{k}_1 \\ \vec{k}_2, \vec{k}_3}} e^{i\vec{k} \cdot \vec{R}_i} e^{-i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3) \cdot \vec{R}_j} \\
&\quad \left. \langle a_{\vec{k}}^+ a_{\vec{k}_1}^+ a_{\vec{k}_2}^+ a_{\vec{k}_3} \rangle \right. \\
&\quad - \frac{1}{2N^2} \sum_{\substack{\vec{k}, \vec{k}_1 \\ \vec{k}_2, \vec{k}_3}} e^{-i(\vec{k}_1 - \vec{k}_2 - \vec{k}_3) \cdot \vec{R}_i} e^{-i\vec{k} \cdot \vec{R}_j} \\
&\quad \left. \langle a_{\vec{k}_1}^+ a_{\vec{k}_2} a_{\vec{k}_3} a_{\vec{k}}^+ \rangle \right\} \rho_{ij}. \tag{A.5}
\end{aligned}$$

Thus when the sum in j in Eq. (A.1) is carried out the following expression for $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ results:

$$\begin{aligned}
\langle G_{\vec{q}}^{(0)} \rangle^{+-} &= \frac{2}{N} (S - \frac{1}{N} \sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle) \sum_{\vec{q}'} (\langle n_{\vec{q}'} \rangle + 1) \rho(\vec{q} - \vec{q}') \\
&\quad + \left[\frac{2}{3} S(S+1) \right] (\rho(\vec{q}) - \frac{1}{N} \sum_{\vec{q}'} \rho(\vec{q}') \rho(\vec{q} - \vec{q}')) \tag{A.6}
\end{aligned}$$

which contains those terms which make a non-zero contribution to the series expansion of the time dependent correlation function. The last term in the above expression, as discussed in Appendix B, approaches zero as $T \rightarrow T_c$, so when $T \gtrsim T_c$ and $q > \lambda^{-1}$ the first term dominates.

The expansion coefficient $\langle G_{\vec{q}}^{(3)} \rangle^{+-}$ can also be approximated in the same manner, i.e.

$$\langle G_{\vec{q}}^{(3)} \rangle^{+-} = \sum_j [A_{ij}^{+-}(3) + B_{ij}^{+-}(3)] e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (\text{A.7})$$

where

$$A_{ij}^{+-}(3) = \langle [S_i^+, H][H, [H, S_j^-]] \rangle_{sw} \rho_{ij} \quad (\text{A.8})$$

and

$$B_{ij}^{+-}(3) = 0, \quad (\text{A.9})$$

by symmetry. The reason why $B_{ij}^{+-}(3)$ is zero is discussed in detail in Chapter IV. Since H is the Heisenberg Hamiltonian, then after some manipulation, spin operator commutator algebra yields

$$\begin{aligned} A_{ij}^{+-}(3) = & \{ 8 \sum_{\ell p r} J_{i\ell} J_{jp} J_{jr} \langle (s_{\ell}^+ s_i^z - s_i^+ s_{\ell}^z) (s_r^- s_j^z - s_j^- s_r^z) s_p^z \rangle, \\ & - 4 \sum_{\ell p r} J_{i\ell} J_{jp} J_{pr} \langle (s_{\ell}^+ s_i^z - s_i^+ s_{\ell}^z) s_j^- (s_p^+ s_r^- - s_r^+ s_p^-) \rangle \\ & - 8 \sum_{\ell p r} J_{i\ell} J_{pr} \langle (s_{\ell}^+ s_i^z - s_i^+ s_{\ell}^z) (s_r^- s_p^z - s_p^- s_r^z) s_j^z \rangle \\ & - 4 \sum_{\ell p r} J_{i\ell} J_{jp} J_{jr} \langle (s_{\ell}^+ s_i^z - s_i^+ s_{\ell}^z) s_p^- (s_j^+ s_r^- - s_r^+ s_j^-) \rangle \} \rho_{ij} \end{aligned} \quad (\text{A.10})$$

2

Then using the spin operator expansions given by Eqs. (IV.37), (IV.65), (IV.66) and completing the sums on j , l , p , and r , we obtain

$$\langle G_{\vec{q}}^{(3)} \rangle^{+-} = \frac{2}{N} \left(S - \frac{1}{N} \sum_{\vec{q}''} \langle n_{\vec{q}} \rangle \right) \sum_{\vec{q}'} \omega(\vec{q}')^3 \times (\langle n_{\vec{q}'} \rangle + 1) \rho(\vec{q} - \vec{q}') \quad (A.11)$$

which again contains those terms which make a finite contribution to the Taylor series expansion of the dynamical spin correlation function. It is quite evident that an explicit calculation of the higher order expansion coefficients becomes extremely tedious due to the number of terms contained in the multiple commutator expressions.

X. APPENDIX B: TEMPERATURE DEPENDENCE OF THE BACKGROUND TERMS IN $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$

AND $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$

In the expression Eq. (A.6) we obtained for $\langle G_{\vec{q}}^{(0)} \rangle^{+-}$ the term that was neglected when we determined the Taylor series expansion of the dynamical spin correlation function has the form

$$\frac{2}{3} s(s+1) [\rho(\vec{q}) - \frac{1}{N} \sum_{\vec{q}'} \rho(\vec{q}') \rho(\vec{q} - \vec{q}')].$$

This is the same, except for a multiplicative constant, as the term that was neglected in $\langle G_{\vec{q}}^{(0)} \rangle^{zz}$. The reason for this comes from the fact that above T_c , when the spins lie outside the correlated cluster, the correlation between pairs of spins satisfies the following equality:

$$\langle S_i^x S_j^x \rangle = \langle S_i^y S_j^y \rangle = \langle S_i^z S_j^z \rangle.$$

In order to examine the size of Eq. (B.1) and its temperature dependence a specific model will be used. For simplicity we take $\rho(\vec{q})$ to have a Gaussian form so $\rho(\vec{q}) \propto e^{-\lambda^2 q^2}$ where λ is the two-spin correlation length. Then the integral that results when the summation on \vec{q}' is converted to an integration can be solved analytically. The dominant term [see Eq. (A.6)] can also be evaluated using this model, however the value for this term must be determined through the use of numerical integration techniques. When these calculations are carried

out we find that at $T_c + 25\text{K}$ (656 K) the ratio of the background term to the term we have retained is on the order of 0.01. However, the background is very temperature sensitive because at $T_c + 40\text{K}$ (671 K) this ratio is 0.15, and as the temperature increases to $T_c + 100\text{K}$ the background term is no longer negligible. In general as long as $q\lambda \gtrsim 4.0$ and $\lambda^{-1} \leq 0.10$ this term makes only a small contribution which can be neglected.

The temperature dependence of the background term in $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$ can also be examined through the use of the above method. The integrations that arise in this analysis are again greatly simplified by the use of a Gaussian spin correlation function and the approximation, $J(0) - J(\vec{q}) = \alpha q^2$. When this is done the value of the background term can be determined via numerical integration, and the ratio of this term to the dominant contribution to $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$ is found to be 0.02 at $T_c + 25\text{K}$. Thus at temperatures near T_c , the contribution of the background term is small and may be neglected, however it should be noted that this term is also very strongly temperature dependent, and as the temperature increases the background contribution becomes significant. At $T_c + 40\text{K}$ the background term is on the order of the term we have retained in the expression for $\langle G_{\vec{q}}^{(2)} \rangle^{+-}$. Consequently, the expressions we have used for the expansion coefficients in the Taylor series expansion of the dynamical spin correlation function are very good approximations only at temperatures near the critical temperature.

XI. APPENDIX C: CALCULATION OF $\langle G_{\vec{q}}^{(0)} \rangle^{zz}$ AND $\langle G_{\vec{q}}^{(2)} \rangle^{zz}$

The zeroth order coefficient in the time Taylor series expansion of the longitudinal dynamical spin correlation function is approximated in the usual way:

$$\langle G_{\vec{q}}^{(0)} \rangle^{zz} \approx \sum_j [(A_{ij}^{zz}(0) + B_{ij}^{zz}(0))] e^{-i\vec{q} \cdot \vec{R}_{ij}}, \quad (C.1)$$

where

$$A_{ij}^{zz}(0) = \langle S_i^z S_j^z \rangle_{sw} \rho_{ij} \quad (C.2)$$

and

$$B_{ij}^{zz}(0) = \langle S_i^z S_j^z \rangle (1 - \rho_{ij}). \quad (C.3)$$

Spin wave theory is again used to evaluate the spin correlation function in $A_{ij}^{zz}(0)$ and $B_{ij}^{zz}(0)$ is evaluated in terms of an equilibrium correlation function, Eqs. (IV.46) and (IV.47). Thus when the Holstein-Primakoff spin operator expansion for S_i^z is employed we obtain

$$\begin{aligned} A_{ij}^{zz}(0) = & \left\{ S^2 - \frac{2S}{N} \sum_{\vec{k}} \langle n_{\vec{k}} \rangle + \frac{1}{N^2} \sum_{\vec{k}, \vec{k}'} \langle n_{\vec{k}} \rangle \langle n_{\vec{k}'} \rangle \right. \\ & + \frac{1}{N^2} \sum_{\vec{k}, \vec{k}'} e^{-i(\vec{k} - \vec{k}') \cdot \vec{R}_{ij}} (1 + \langle n_{\vec{k}'} \rangle) \\ & \left. \times \langle n_{\vec{k}} \rangle \right\} \rho_{ij}. \end{aligned} \quad (C.4)$$

When this is substituted into Eq. (C.1) and the sum on j is carried out we find that

$$\begin{aligned}
 \langle G_{\vec{q}}^{(0)} \rangle^{zz} &= [s - \frac{1}{N} \sum_{\vec{q}'} \langle n_{\vec{q}'} \rangle]^2 \rho(\vec{q}) \\
 &+ \frac{1}{N^2} \sum_{\vec{q}', \vec{q}''} \langle n_{\vec{q}'} \rangle \langle n_{\vec{q}''} \rangle + 1) \rho(\vec{q} + \vec{q}' - \vec{q}'') \\
 &+ [\frac{2}{3}s(s+1)] (\rho(\vec{q}) - \frac{1}{N} \sum_{\vec{q}'} \rho(\vec{q}') \rho(\vec{q} - \vec{q}')) \quad (C.5)
 \end{aligned}$$

where the last term in this equation makes only a small contribution when $T \gtrsim T_c$ and $q > \lambda^{-1}$. (See Appendix B.)

In like manner we can write

$$\langle G_{\vec{q}}^{(2)} \rangle^{zz} \cong \sum_j [A_{ij}^{zz}(2) + B_{ij}^{zz}(2)] e^{-i\vec{q} \cdot \vec{R}_{ij}} \quad (C.6)$$

where

$$A_{ij}^{zz}(2) = \langle [s_{ij}^z, H] [H, s_j^z] \rangle_{sw} \rho_{ij} \quad (C.7)$$

and

$$B_{ij}^{zz}(2) = \langle [s_i^z, H] [H, s_j^z] \rangle (1 - \rho_{ij}) . \quad (C.8)$$

Since

$$[s_i^z, H] = \sum_n J_{in} (s_i^+ s_n^- - s_n^+ s_i^-) \quad (C.9)$$

then

$$A_{ij}^{zz}(2) = \sum_{nm} J_{in} J_{jm} \{ \langle S_i^+ S_n^- S_m^+ S_j^- \rangle + \langle S_n^+ S_i^- S_j^+ S_m^- \rangle - \langle S_i^+ S_n^- S_j^+ S_m^- \rangle - \langle S_n^+ S_i^- S_m^+ S_j^- \rangle \}, \quad (C.10)$$

where the Holstein-Primakoff operator expansions for S_n^+ and S_m^- , Eqs. (IV.35) and (IV.36), are now used to rewrite the above four-spin correlation functions. In order to simplify $B_{ij}^{zz}(2)$ we use the cumulant expansion method⁴⁰ so that above T_c the four spin correlation functions can be written in terms of pairs of 2-spin equilibrium correlation functions, Eq. (IV.56). When this is done and the sums on j , n , and m are carried out we obtain

$$\begin{aligned} \langle G_{\vec{q}}^{(2)} \rangle^{zz} &= \frac{1}{N} \sum_{\vec{q}' \vec{q}''} (\omega(\vec{q}') - \omega(\vec{q}''))^2 \\ &\quad \times (1 + \langle n_{\vec{q}'} \rangle) \langle n_{\vec{q}''} \rangle \rho(\vec{q} + \vec{q}' - \vec{q}'') \\ &\quad + \left[\frac{2}{3} s(s+1) \right]^2 \left\{ \frac{4}{N} \sum_{\vec{q}'} [J(\vec{q}') - J(\vec{q} + \vec{q}')]^2 \right. \\ &\quad \times \rho(\vec{q}') \rho(\vec{q} + \vec{q}') \\ &\quad \left. - \frac{4}{N} \sum_{\vec{q}' \vec{q}''} [J(\vec{q}') - J(\vec{q}' + \vec{q}'')]^2 \rho(\vec{q}') \rho(\vec{q}' + \vec{q}'') \rho(\vec{q} - \vec{q}'') \right\}, \end{aligned} \quad (C.11)$$

where the last two terms tend to cancel each other as $T \rightarrow T_c$ and

$$q > \lambda^{-1}.$$