Spin Dynamics of Linear Heisenberg Magnetic Chains*

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The self-consistent theory of Blume and Hubbard is used to calculate the time-dependent spin-pair correlation functions for the Heisenberg linear chain at finite temperatures. Numerical solutions of the approximate coupled integro-differential equations are presented for both ferromagnetic and antiferromagnetic chains with nearest-neighbor interactions. The classical static correlation functions are used as initial conditions for the spin dynamics. The validity of the results is not restricted to classical systems, however, as it is argued that in the quantum regime the thermodynamically important correlations appearing in the kinetic equations are those of sufficiently long wavelength that classical statistics are applicable. For high temperatures the theory of Blume and Hubbard is applied directly. In the low-temperature "spin-wave" regime, however, the theory is generalized to include the presence of long-range "short-range-order." An order parameter is introduced which describes the local spin magnetization against which the spin waves oscillate. Expressions are derived for the temperature and q dependence of the magnon lifetimes as well as for the temperature renormalization of their frequencies. Within the framework of the theory the principal mode of spin-wave damping is attributed to scattering from fluctuations in the local magnetization. The theory is shown to be internally consistent in that it predicts that well-defined spin-wave excitations exist only with wavelengths less than the correlation length of the local order, and further, the magnon lifetimes are limited by the characteristic times associated with changes in the local order. The generalized structure factor for inelastic neutron scattering is obtained from the Fourier transform of the spin-pair correlation function and is compared in some detail with recent experimental results on the one-dimensional antiferromagnet $(CD_3)_4NMnCl_3$ (TMMC). The calculated structure factors show well-defined spin-wave peaks at low temperature, the shape and position of which—both as a function of temperature and of wave vector—agree quite well with the experimental data. There is some discrepancy in the predicted intensities at the lowest temperatures; this may be a manifestation of quantum effects but more likely is due to limitations imposed on the data by the instrumental resolution function.

I. INTRODUCTION

In a recent paper[1] Blume and Hubbard presented an approximate self-consistent method of calculating the time-dependent spin correlation functions of a Heisenberg spin system in the high-temperature limit. They found good agreement with the results of computer-simulation calculations[2] for the case of a simple cubic lattice with nearest-neighbor interactions. Hubbard[3] later generalized the method to include the entire paramagnetic phase ($T > T_C$), the principal additional feature being the requirement that the static correlation functions be known as functions of temperature to serve as initial conditions for the kinetic equations. He derived approximate expressions for the static correlation functions by the application of certain sum rules, and he reported the results of the time-dependent calculations for the case of a simple cubic lattice with nearest-neighbor ferromagnetic interactions. (In the following we will refer to Refs. 1 and 3 collectively as BH.)

It is natural to consider the application of this technique to the study of the spin dynamics of linear magnetic chains. In the first place, one-dimensional systems traditionally serve as testing grounds for theories designed to treat real three-dimensional systems. Next, one-dimensional magnetic systems are paramagnetic for all temperatures and thus may be useful in understanding certain aspects of spin dynamics of systems of higher-order dimensionality in which the persistence of magnonlike modes has been observed at and above the critical temperature.[4,5] Further, for the case of the classical Heisenberg magnetic chain with nearest-neighbor interactions only, certain static properties can be calculated exactly. In particular, the static correlation functions are known exactly,[6] and hence the initial conditions for the time-dependent correlations are known exactly. Finally, but not least, there recently has been a great deal of interest in real magnetic systems which approximate one-dimensional behavior.[7-10] In such systems the magnetic interactions between magnetic ions are highly anisotropic, being directed predominately along chains of magnetic ions. The coupling between magnetic ions located on adjacent chains is very weak in comparison. Thus, as far as magnetic properties are concerned, the one-dimensional model serves as a good first approximation, the precision of which is better the weaker the transverse (cross-chain) coupling compared with the intrachain coupling.

A typical system satisfying these conditions is

the linear-chain antiferromagnet $(CD_3)_4NMnCl_3$ (henceforth called TMMC).[8] Recently, Birgeneau *et al.*[9] and Hutchings *et al.*[10] conducted a comprehensive experimental investigation of the magnetic properties of this compound. From quasielastic neutron scattering[9] they observed the static magnetic correlations of TMMC and found that the $MnCl_3$ chains exhibit purely one-dimensional paramagnetic behavior down to 1.1 °K. They found that both the spatial and thermal variations of the static correlations can be quantitatively accounted for by using Fisher's theory[6] for the classical Heisenberg chain. They also studied the time-dependent correlations using inelastic-neutron-scattering techniques.[10] The principal result was their observation of well-defined spin waves at low temperature, which fit the pattern of the usual spin-wave theory over much of the one-dimensional Brillouin zone. As the temperature increased, the spin-wave peaks weakened in intensity and broadened asymmetrically, with the scattering increasing on the low-energy side.

Except for some recent calculations[11] using a simple Green's-function theory[11(a)] and interpolation techniques,[11(b)] theoretical work on the spin dynamics of linear magnetic chains has tended to focus on the case of spin-$\frac{1}{2}$ at either of two temperature limits, $T=\infty$ or $T=0$. Carboni and Richards[12(a)] and Gersch[12(b)] have performed calculations at infinite temperature which indicate that the scattering function is of the Lorentzian form for long wavelengths; but at shorter wavelengths it has a broad profile with a peak at nonzero frequency which may be a remnant of the $T=0$ spin wave. Lieb, Schultz, and Mattis[13] derived the spectrum of the first excited state for the X-Y model at absolute zero, and that for the Heisenberg model has been calculated exactly by Des Cloizeaux and Pearson.[14] Both take the form of magnonlike states with a simple sine dispersion law, although the coefficients differ for the two models. Kawasaki[15] and Kuramoto[16] have considered the spin-$\frac{1}{2}$ case at finite temperature, but the situation is not entirely clear. Virtually no theoretical work has been done on higher-spin systems at finite temperature. The present work, in conjunction with a computer-simulation study of the classical spin system at arbitrary temperature by Blume, Watson, and Vineyard,[17] was initiated as a comprehensive effort aimed at filling this gap. The computer-simulation studies may be viewed as further experimental data—on purely classical systems—with which theory may be compared, in addition to the experimental data of Hutchings *et al.*[10] on a real finite-spin system.

In this paper we undertake a study of the spin dynamics of linear Heisenberg magnetic chains using the technique introduced by BH as a point of departure. The primary goal is to calculate the spin correlation function $C_q(t)=\langle \frac{1}{2}[\vec{S}_{-q}\cdot\vec{S}_q(t)+\vec{S}_q(t)\cdot\vec{S}_{-q}]\rangle$, or equivalently, the relaxation shape function $F_q^\alpha(t)=\{S_{-q}^\alpha, S_q^\alpha(t)\}/\{S_{-q}^\alpha, S_q^\alpha\}$, where $\{A, B(t)\}$ is the Kubo relaxation function. Once either is known the other may be obtained by the fluctuation-dissipation theorem. We, in fact, derive equations for the $F_q^\alpha(t)$, which via a sum over the whole Brillouin zone, involve the correlation functions $C_{q'}^{\alpha'}(t)$ for all wave vectors q'. As initial conditions on the $C_{q'}^{\alpha'}(t)$ we use the static correlations of the classical chain. However, the validity of our results is not restricted to the classical system. At low temperatures, where the consequences of the fluctuation-dissipation theorem are most important, we are able to argue that only the correlations of the long-wavelength fluctuations are significant in contributing to the dynamics of $F_q^\alpha(t)$. It is generally believed, and supported by a good deal of experimental evidence, that classical-spin systems represent good approximations to finite-spin (quantum-mechanical) systems of sufficiently large spin (e.g., $S=\frac{5}{2}$), at least for the long-wavelength fluctuations. Hence, we use the classical limit of the fluctuation-dissipation theorem, $C_{q'}^{\alpha'}(t)=C_{q'}^{\alpha'}F_{q'}^{\alpha'}(t)$, where $C_{q'}^{\alpha'}$ are the classical static correlation functions, to close the system of equations for $F_q^\alpha(t)$. The $F_q^\alpha(t)$ that we derive in this manner are thus not restricted to the classical case, as long as only those correlations $C_{q'}^{\alpha'}(t)$ for which the classical approximation is valid are significant in the kinetic equations for $F_q^\alpha(t)$.

Once we have determined the $F_q^\alpha(t)$, the quantity of greatest experimental interest is the generalized structure factor $S_q^{\alpha\beta}(\omega)$. For Heisenberg systems having axial symmetry $S_q^{\alpha\beta}(\omega)=S_q^\alpha(\omega)\,\delta^{\alpha\beta}$, and $S_q^\alpha(\omega)$ is related to $F_q^\alpha(\omega)$, the Fourier transform of $F_q^\alpha(t)$, by[18]

$$S_q^\alpha(\omega)=\frac{x_q^\alpha}{\beta}\,\frac{\beta\omega}{1-e^{-\beta\omega}}\,F_q^\alpha(\omega)\,, \tag{1}$$

where x_q^α is the q-dependent static susceptibility, $\beta=1/T$, and T is the absolute temperature. (We take the Boltzmann constant k_B and $\hbar$ equal to unity throughout this paper.) We note that the fluctuation-dissipation theorem is accounted for in Eq. (1) by the presence of the detailed balance factor.

When we apply the BH technique to the linear magnetic chain in a straightforward manner, we find quite reasonable results for the high-temperature regime $T>JS(S+1)$, where J is the exchange constant; there is good agreement with the computer-simulation studies of Blume *et al.*[17] For low temperature $T\lesssim\frac{1}{3}JS(S+1)$; on the other hand, there is a dramatic failure of the theory, namely, spin waves are not predicted by the theory. This is in sharp contrast to actual fact as the experi-

mental results of Hutchings *et al.*[10] clearly indicate. We then realize that even though true long-range magnetic order is absent for the linear chain, there is considerable correlation among spins in a region of the order of the correlation length $l_c \sim \pi JS(S+1)/T$, which becomes very large as $T \to 0$.

We are, therefore, led to introduce the notion of a short-range-order parameter which, for the short-wavelength high-frequency excitations (i.e., spin waves), serves as an effective local magnetization. An important requirement for the validity of this concept is that the local ordering changes slowly—both spatially and temporally—on the scale of spin-wave oscillations. The idea is very similar to that in the conventional spin-wave theory of three-dimensional Heisenberg systems: that spin-wave energies renormalize with temperature not as the macroscopic magnetization but as the energy.[19] In effect spin waves do not exist sufficiently long to see the true macroscopic time-averaged background. Instead they see an instantaneous nonequilibrium snapshot of the background, and it is from the instantaneous background that the spin-waves oscillate. Lines[20] utilized very similar arguments in his treatment of the two-dimensional Heisenberg magnet, particularly in connection with his introduction of a wave-vector-dependent magnon renormalization.

When we generalize the theory to incorporate the idea of a local order parameter, spin waves appear, of course, in a natural fashion. The usefulness of the theory is then judged mainly by how well it describes the damping of the magnon modes as a function of temperature. In particular, we can compare the line shapes calculated from the theory for inelastic neutron scattering with the experimental results. We find that the agreement is remarkably good and also that the theory is internally consistent in that it yields well-defined spin waves only for wavelengths less than the coherence length l_c. Further, the lifetimes of the spin waves are limited by the characteristic times necessary for appreciable changes to occur in the local order.

In Sec. II we describe how the BH theory may be generalized to include magnetic order. We do not specify whether the order is local in the sense described above or is a true long-range magnetic order. Thus, the generalized theory may also be applied in principle to two- and three-dimensional systems below the critical temperature. The principal effect of including magnetic order in the theory is that instead of a single correlation or relaxation function, we must consider both longitudinal and transverse correlations with respect to the axis of quantization. We derive a system of coupled equations connecting the longitudinal and transverse functions, as well as the different wave vectors of the zone. We employ essentially the same approximations used by BH without attempting to justify them or to ascertain their validity. Rather, their appropriateness will be determined by comparing the results of the theory with experiment.

The case of the antiferromagnet in the ordered state is rather more complicated in detail than for the ferromagnet, and we relegate this case to Appendix A. Also, the theory developed in Sec. II is easily extended to include the presence of an external magnetic field or to apply to the cases of the anisotropic-Heisenberg Hamiltonian and of the "truncated-dipolar" Hamiltonian at infinite temperature. For the sake of completeness those extensions are discussed in Appendix B.

In Sec. III we apply the theory to one-dimensional systems in the conventional manner, using macroscopic ensemble averages and hence setting the order parameter equal to zero. The resulting equations are applicable at high temperatures $T \gtrsim JS(S+1)$. We then demonstrate the failure of these equations at lower temperatures. We go on in Sec. IV to discuss the spin-wave region making use of the concept of local order. We address ourselves to the question of the proper choice of the short-range order parameter as well as to the related questions of (i) the appropriate breakdown of the static correlation functions into longitudinal and transverse components with respect to the local axis of magnetization, and (ii) the appropriate spatial-averaging process necessary to describe the macroscopic sample. We study in some detail the damping of the magnon modes within our approximations, and we examine the internal consistency of the theory.

It is always advantageous to have an exact result with which to compare the results of an approximate theory. For the classical linear chain with nearest-neighbor interactions only, we have already mentioned that the static correlation functions are known and that we make use of these as initial conditions for $C_q(t)$. In principle, all the frequency moments of $C_q(\omega)$ can also be calculated exactly for this case, as they all can be reduced to equal-time, multiple-spin correlation functions which can then be calculated by the transfer-matrix technique.[21] In Appendix C we present the calculation of the second-frequency moment. (In practice, the labor involved in calculating higher-order moments is prohibitive.) We make considerable use of this exact result in discussing our theory in Secs. III and IV.

We present the results of the numerical solutions of the kinetic equations in Sec. V considering various regions of interest in turn: (i) the long-

wavelength fluctuations at all temperatures, (ii) the high-temperature region, and (iii) the spin-wave (low-temperature) regime. For the spin-wave region, we compare the generalized structure factor, Eq. (1), for inelastic neutron scattering predicted by our theory for an antiferromagnetic spin system of spin-$\frac{5}{2}$ with the experimental results of Hutchings *et al.*[10] on TMMC.

II. FORMAL THEORY

In this section we indicate how the theory developed by Blume and Hubbard[1,3] may be extended to include systems having an axis of quantization. In particular, we are interested in systems described by an isotropic Heisenberg Hamiltonian with magnetic ordering present. However, essentially the same treatment applies to all cases where the total component of spin in the z direction commutes with the interaction Hamiltonian. The principal added difficulty over the paramagnetic case is that instead of a single relaxation function (or correlation function) we must now calculate three relaxation functions for each wave vector q, corresponding to the longitudinal and two transverse directions with respect to the quantization axis. The set of coupled integro-differential equations that we derive couple the longitudinal and transverse correlation functions as well as the various q values of the Brillouin zone.

Our primary goal is to calculate certain symmetrized correlation functions $C^{AB}(t)=\langle \frac{1}{2}[AB(t)+B(t)A]\rangle$. We do this by deriving equations for the corresponding relaxation shape functions

$$F^{AB}(t)=\frac{\{A, B(t)\}}{\{A, B\}}=\frac{\langle \delta B(t)\rangle_A}{\langle \delta B\rangle_A} , \tag{2}$$

where $t>0$, $\{A, B(t)\}$ is Kubo's relaxation function[22]

$$\{A, B(t)\}=\int_{-\infty}^{0} dt' \frac{1}{i}\langle [A(t'), B(t)]\rangle$$

$$=\int_0^{\beta} d\lambda \langle e^{\lambda H} A e^{-\lambda H} B(t)\rangle - \beta\langle A^0 B^0\rangle \tag{3}$$

and $\langle \delta B(t)\rangle_A$ is the change[1] in the expectation value of $B(t)$ for $t>0$ due to a constant perturbation proportional to A applied between $t=-\infty$ and $t=0$. In (3), $[A, B]=AB-BA$, $\langle A\rangle=\mathrm{Tr}(Ae^{-\beta H})/\mathrm{Tr}(e^{-\beta H})$, and

$$\beta\langle A^0 B^0\rangle=\lim_{t\to\infty}\int_0^{\beta} d\lambda\langle e^{\lambda H} A e^{-\lambda H} B(t)\rangle . \tag{4}$$

The fluctuation-dissipation theorem provides the connection between the relaxation shape function and the corresponding symmetrized correlation function. We shall return to this point in more detail below. For the moment, we simply note that for classical systems and for quantum-mechanical systems in the limit $T\to\infty$, $\{A, B(t)\}=\beta[\langle AB(t)\rangle-\langle A^0 B^0\rangle]=\beta[C^{AB}(t)-C^{AB}(0)]$.

In the absence of the perturbation A, the operator B evolves with time in the usual way according to $B(t)=e^{iHt}Be^{-iHt}$. However, as shown in Ref. 1, the perturbation

$$H'(t)=-Ae^{\epsilon t}\theta(-t) , \tag{5}$$

where $\epsilon=0+$ and

$$\theta(z)=\begin{cases}1 & z>0\\ 0 & z<0\end{cases}$$

disturbs this evolution, causing B to evolve instead into the operator $\hat{B}(t)=B(t)+\delta B(t)$, where $\hat{B}(t)$ satisfies the equation of motion

$$i\frac{d\hat{B}(t)}{dt}=[\hat{B}(t), \hat{H}(t)] . \tag{6}$$

Here, $\hat{H}(t)=H+\delta H(t)$ is the perturbed Hamiltonian operator. Since $i\,dB(t)/dt=[B(t), H]$, we have

$$i\frac{d}{dt}[\delta B(t)]=[B(t), \delta H(t)]+[\delta B(t), H] . \tag{7}$$

The basic procedure is to solve this equation for the relaxation of $\delta B(t)$ after the perturbation is turned off. Then by (2) we obtain the relaxation shape function and thence the correlation function via the fluctuation-dissipation theorem.

Before proceeding to the details, we first comment on the meaning of the averaging brackets in our expressions. As mentioned in Sec. I, true statistical-ensemble averages may be inappropriate for certain fluctuations, namely, those whose spatial extent and lifetimes are sufficiently short that they do not "sample" a macroscopically averaged background. For these excitations some sort of local-averaging process is in order. As we develop the formal theory in this section, we shall leave the meaning of the averages open. This question will be elaborated upon in some detail in Sec. IV.

We consider the isotropic Heisenberg spin Hamiltonian

$$H=-\frac{1}{2}\sum_{i,j} J_{ij}\vec{S}_i\cdot\vec{S}_j$$

with $J_{ii}=0$. Since we wish to treat the case of magnetic order, the direction of which we take to be along the z axis, we must consider the three total-spin-conserving relaxation shape functions:

$$F_q^z(t)=\{S_{-q}^z, S_q^z(t)\}/\{S_{-q}^z, S_q^z\} ,$$

$$F_q^+(t)=\{S_{-q}^-, S_q^+(t)\}/\{S_{-q}^-, S_q^+\} , \tag{8}$$

$$F_q^-(t)=\{S_{-q}^+, S_q^-(t)\}/\{S_{-q}^+, S_q^-\} .$$

Here

$$S_q^\alpha(t)=(1/N)\sum_i S_i^\alpha(t)e^{i\vec{q}\cdot\vec{R}_i} ,$$

where the sum is over the N sites of the lattice and $S_q^{\pm}=S_q^x\pm iS_q^y$. From the equation of motion $\dot{S}_i^{\alpha}=i[H,S_i^{\alpha}]$ one finds [see Eq. (5) of Ref. 3]

$$\dot{\vec{S}}_q(t)=-\frac{1}{2}\sum_{q'}J_{qq'}\vec{S}_{q'}(t)\times\vec{S}_{q-q'}(t)\,, \tag{9}$$

where

$$J_{qq'}=J_{q'}-J_{q-q'}\,, \tag{10}$$

which shows explicitly that $\dot{S}_{q=0}=0$; and from (7), we have

$$\delta\dot{\vec{S}}_q(t)=-\frac{1}{2}\sum_{q'}J_{qq'}[\delta\vec{S}_{q'}(t)\times\vec{S}_{q-q'}(t)-\vec{S}_{q-q'}(t)\times\delta\vec{S}_{q'}(t)]\,. \tag{11}$$

Following Hubbard[3] we introduce the idea of S ordering, where in products of spin operators $[S_1(t_1)S_2(t_2)\cdots S_i(t_i)\bar{S}_j(t_j)\cdots\bar{S}_n(t_n)]$ barred operators $\bar{S}$ are to go to the right of all unbarred operators S. Then the unbarred operators are rearranged according to the usual time-ordering convention (latest times to the left), and the barred operators are rearranged in inverse time order (latest times to the right). Further, we define

$$\hat{S}_q^{\alpha}(t)=\tfrac{1}{2}[S_q^{\alpha}(t)+\bar{S}_q^{\alpha}(t)]\,. \tag{12}$$

Then we rewrite (11) as

$$\delta\dot{\hat{S}}_q^{\alpha}(t)=\sum_{\alpha'q'}ih_{qq'}^{\alpha\alpha'}(t)\,\delta S_{q'}^{\alpha'}(t)\,, \tag{13}$$

where, for the usual Cartesian coordinates,

$$ih_{qq'}^{\alpha\alpha'}(t)=J_{qq'}\,\epsilon_{\alpha\nu\alpha'}\,\hat{S}_{q-q'}^{\nu}(t) \tag{14}$$

(sum over repeated indices). With an axis of quantization, however, it is most convenient to use the (S^+,S^z,S^-) representation. In this representation the equation of motion (13) has the same form, but the components of the matrix $h_{qq'}(t)$ are given by

$$h_{qq'}(t)=J_{qq'}\begin{pmatrix}\hat{S}_{q-q'}^{z}(t) & -\hat{S}_{q-q'}^{+}(t) & 0\\ -\tfrac{1}{2}\hat{S}_{q-q'}^{-}(t) & 0 & \tfrac{1}{2}\hat{S}_{q-q'}^{+}(t)\\ 0 & \hat{S}_{q-q'}^{-}(t) & -\hat{S}_{q-q'}^{z}(t)\end{pmatrix} \tag{15}$$

[the rows and columns are labeled by $(+\,z\,-)$ indices]. To solve (13) in compact fashion we introduce the $3N$ component vector operator $\delta S(t)=(\delta S_{q_1}^{+}\,\delta S_{q_1}^{z}\,\delta S_{q_1}^{-}\,\delta S_{q_2}^{+}\cdots\delta S_{q_N}^{z}\,\delta S_{q_N}^{-})$. Then (13) may be rewritten

$$\delta\dot{S}(t)=ih(t)\,\delta S(t)\,, \tag{16}$$

where $ih(t)$ is now to be considered a superoperator as in BH (a tensor in both q space and spin space and an operator in Hilbert space). With the aid of the S-ordering convention, the solution of (16) is found by the usual iterative process

$$\delta S(t)=\{\exp_+[i\int_0^t h(t')\,dt']\,\delta S\}_S\,, \tag{17}$$

where $\exp_+$ means the ordinary time-ordered exponential—the spin operators being subsequently reordered by $\{\cdots\}_S$.

We are now in a position to derive expressions for the relaxation shape functions. For the moment we restrict our attention to the case of the ferromagnet; we will return to the case of antiferromagnetic ordering below. Combining Eqs. (2) and (17), we have

$$F_q^{\alpha}(t)=\langle(\{\exp_+[i\int_0^t h(t')\,dt']\,\delta S\}_S)_q^{\alpha}\rangle/\langle\delta S_q^{\alpha}\rangle\,, \tag{18}$$

where $\alpha=(+,z,-)$ and $(\cdots)_q^{\alpha}$ means the $\alpha-q$ component of the vector $(\cdots)$. This expression is exact within linear-response theory. To proceed further, it is necessary to make simplifying approximations. First, we note that the h operators in (18) are linear in the spin operators. To account for magnetic order we write $\vec{S}=\langle\vec{S}\rangle+\delta\vec{S}$, where $\langle\delta\vec{S}\rangle=0$. Then the principal approximation that we make, following BH, is the spin-pair decoupling

$$\langle\delta S_1\,\delta S_2\cdots\delta S_N\rangle\cong\sum_{\text{pairing}}\langle\delta S_1\,\delta S_2\rangle\times\langle\delta S_3\,\delta S_4\rangle\cdots\langle\delta S_{N-1}\,\delta S_N\rangle\,, \tag{19}$$

where the sum is over all possible spin pairs. The first consequence of the approximation (19) is that in (18) we can replace δS by $\langle\delta S\rangle$, giving

$$F_q^{\alpha}(t)\cong\langle(\{\exp_+[i\int_0^t h(t')\,dt']\}_S)_{qq'}^{\alpha\alpha'}\langle\delta S_{q'}^{\alpha'}\rangle/\langle\delta S_q^{\alpha}\rangle\,. \tag{20}$$

Next, since the applied field was proportional to $S_{-q}^{-\alpha}$ [see Eqs. (2) and (8)], we have[23] $\langle\delta S_{q'}^{\alpha'}\rangle=0$ unless $\alpha'=\alpha$ and $q'=q$. (Note that $\langle\delta S_{q'}^{\alpha'}\rangle=\{S_{-q}^{-\alpha},S_{q'}^{\alpha'}\}$.) Hence, we have

$$F_q^{\alpha}(t)\cong\langle(\{\exp_+[i\int_0^t h(t')\,dt']\}_S)_{qq}^{\alpha\alpha}\rangle\equiv V_{qq}^{\alpha\alpha}(t)\,, \tag{21}$$

where $V(t)=\langle\{\exp_+[i\int_0^t h(t')\,dt']\}_S\rangle$. We now use the cumulant expansion[1,24] for the statistical average in (21) and use the approximation (19) to neglect all but the first two terms of the expansion, i.e., all cumulants of higher order than second vanish in the spin-pair decoupling approximation. We obtain the result

$$F_q^{\alpha}(t)\cong(\exp_+\{i\int_0^t dt'\,\langle h(t')\rangle-\int_0^t dt'\int_0^t dt''\,\langle[h(t')\,h(t'')]_S\rangle_c\})_{qq}^{\alpha\alpha}\,. \tag{22}$$

Here $\langle[h(t')h(t'')]_S\rangle_C=\langle[h(t')h(t'')]_S\rangle-\langle h(t')\rangle\times\langle h(t'')\rangle$. Note that the exponential must still be time ordered since the $h(t)$ are matrices, but the S ordering is taken into the $\langle\cdots\rangle$ average, since it only rearranges the spin operators. We also note the obvious fact that the first term does not vanish when magnetic order is present.

We now differentiate (22) with respect to time to obtain an equation of motion for $F_q^{\alpha}(t)$. The

basic idea is to obtain an equation for the relaxation shape function in terms of the correlation function of the fluctuating internal field:

$$\dot{F}_q^\alpha(t) = (\{i\langle h(t)\rangle - \int_0^t dt' \langle [h(t)\,h(t')]_S\rangle_c\}$$
$$\times \exp\{i\int_0^t dt_1 \langle h(t_1)\rangle$$
$$- \int_0^t dt_1 \int_0^{t_1} dt_2 \langle [h(t_1)\,h(t_2)]_S\rangle_c\})_{+\,qq}^{\alpha\alpha}$$
$$= (\{i\langle h(t)\rangle - \int_0^t dt' \langle [h(t)\,h(t')]_S\rangle_c\}\, V(t)_{+qq}^{\alpha\alpha} . \tag{23}$$

For the ferromagnet the matrix $V(t)$ is diagonal, [25]

$$V_{qq'}^{\alpha\alpha'}(t) = V_{qq}^{\alpha\alpha}(t)\,\delta^{\alpha\alpha'}\,\delta_{qq'} \tag{24}$$

which is simply a consequence of assuming linear-response theory as well as a statement that correlations not conserving total spin vanish. [This can also be shown by direct expansion of $V(t)$ and taking matrix elements term by term.] The first term in (23) thus reduces to $i\langle h_{qq}\rangle F_q(t)$. In the second term we apply the "disentangling" approximation $V(t) = V(t-t')V(t')$ to account for the time-ordering in the lowest-order nontrivial manner, and, again using (24), the second term becomes

$$-\int_0^t dt' \sum_{q'\,\alpha'} \langle [h_{qq'}^{\alpha\alpha'}(t)\,h_{q'q}^{\alpha'\alpha}(t')]_S\rangle_c\, F_{q'}^{\alpha'}(t-t')\,F_q^\alpha(t') . \tag{25}$$

The terms in $\langle\cdots\rangle$ brackets are the correlation functions of the internal magnetic field. Magnetic order is accounted for by letting

$$\langle S_q^z\rangle = S\Delta\delta_{q,0} , \tag{26}$$

where Δ is the order parameter ($\Delta = 1$ in the completely ordered state and $\Delta = 0$ in the completely disordered state). Passing over the details, except to note that upon making use of (15) for the components of the h matrix the correlation functions of the internal field can be expressed in terms of the symmetrized spin correlation functions, we have as the results

$$\dot{F}_q^z(t) = -\frac{1}{2}\sum_{q'} K_{qq'}\int_0^t dt' \,[C_{q'}^-(t-t')\,F_{q-q'}^+(t-t')$$
$$+ C_{q'}^+(t-t')\,F_{q-q'}^-(t-t')]\,F_q^z(t') ,$$

$$\dot{F}_q^+(t) = iJ_{qq}S\Delta F_q^+(t)$$
$$-\sum_{q'} K_{qq'}\int_0^t dt' \{\tfrac{1}{2}C_{q'}^+(t-t')\,F_{q-q'}^z(t-t')$$
$$+ [C_{q'}^z(t-t') - \delta_{q',0}S^2\Delta^2]F_{q-q'}^+(t-t')\}\,F_q^+(t') , \tag{27}$$

$$\dot{F}_q^-(t) = [\dot{F}_q^+(t)]^* ,$$

where

$$K_{qq'} = J_{q,q-q'}\,J_{q-q',q} = (J_{q'} - J_q)\,(J_{q'} - J_{q-q'}) , \tag{28}$$

and

$$C_q^z(t) = \langle \tfrac{1}{2}[S_{-q}^z\,S_q^z(t) + S_q^z(t)\,S_{-q}^z]\rangle ,$$

$$C_q^+(t) = \langle \tfrac{1}{2}[S_{-q}^-\,S_q^+(t) + S_q^+(t)\,S_{-q}^-]\rangle , \tag{29}$$

$$C_q^-(t) = [C_q^+(t)]^* .$$

In order to close the system of equations we must now express the correlation functions $C_q^\alpha(t)$ in terms of the relaxation shape functions via the fluctuation-dissipation theorem [22]

$$C_q^\alpha(t) = x_q^\alpha\left(\frac{i}{2}\frac{d}{dt}\right)\coth\left(\frac{i\beta}{2}\frac{d}{dt}\right)F_q^\alpha(t) , \tag{30}$$

where $x_q^\alpha = \{S_{-q}^{-\alpha},\ S_q^\alpha\}$ is the q-dependent static susceptibility. The right-hand side of (30) is an infinite series in $F_q(t)$ and all its even integer derivatives, the first few terms of which are

$$C_q^\alpha(t) = (x_q^\alpha/\beta)\,[F_q^\alpha(t) - \tfrac{1}{12}\beta^2\ddot{F}_q^\alpha(t) + \cdots] . \tag{31}$$

Thus, although in principle the fluctuation-dissipation theorem provides closure to the system of Eqs. (27), in practice the solution of (27) is still a formidable task in general. However, if we take the Fourier transform of (30)

$$C_q^\alpha(\omega) = (x_q^\alpha/\beta)\,(\tfrac{1}{2}\beta\omega)\coth(\tfrac{1}{2}\beta\omega)\,F_q^\alpha(\omega)$$
$$= (x_q^\alpha/\beta)\,[1 - \tfrac{1}{3}(\tfrac{1}{2}\beta\omega)^2 + \ldots]\,F_q^\alpha(\omega) , \tag{32}$$

we see that in those cases where the frequencies of interest are small, $(\frac{1}{2}\beta\omega)^2 \ll 1$, we can make the approximation

$$C_q^\alpha(t) \cong C_q^\alpha\,F_q^\alpha(t) , \tag{33}$$

where $C_q^\alpha = x_q^\alpha/\beta$ is the static correlation function in this case.

The condition $(\frac{1}{2}\beta\omega)^2 \ll 1$ holds for a number of practical cases. It is obviously true for all systems at sufficiently high temperatures, and it holds for classical-spin systems at all temperatures. [26] Hubbard[3] has shown that the condition is applicable with good accuracy for the case of three-dimensional Heisenberg magnets down to the critical temperature. In the present paper we are primarily concerned with one-dimensional Heisenberg systems at low temperatures. For the classical linear chain we know from Fisher's work[6] that the static susceptibility is peaked very sharply around $q = 0$ at low temperatures with a width of the order $\Delta q \sim 1/K$, where $K = J_{c1}/T \gg 1$ and q is measured in units of inverse lattice spacing. Therefore, in this case the major contribution to the q' sums in Eqs. (27) comes from the region $|q'| \lesssim 1/K$. If we now assume that the classical susceptibility is a good approximation to the true quantum-mechanical susceptibility, at least in the

long-wavelength region for systems of relatively large spin,[27] then we assert that for the quantum-mechanical case also the important region of q space in the sums of (27) is the region $|q'| \lesssim 1/K$, where now $K = JS(S+1)/T$. Hence, the frequencies of interest in application of the fluctuation-dissipation theorem are those associated with the long-wavelength excitations of the system. It then follows that if these frequencies are sufficiently low, approximation (33) is valid. The sums in (27) are in essence thermodynamic sums; thus at low temperatures one expects that only the low-frequency excitations will make a contribution. In fact, as will be discussed in further detail in later sections, if we assume that the long-wavelength fluctuations are primarily longitudinal and diffusive in nature, $F_q(t) \approx e^{-Dq^2 t}$ for $q \to 0$, then the characteristic frequencies are of the order $Dq^2 \sim D/K^2$, and hence $(\frac{1}{2}\beta\omega)^2 \sim (\beta D/2K^2)^2 \ll 1$, as long as the diffusion constant D is well behaved as $T \to 0$, i.e., $D \sim T^\alpha$, where $\alpha \gtrless 0$. The point that we are emphasizing is that in order for us to close the system of Eqs. (27) via the approximation (33), it is *not* necessary that the condition $(\frac{1}{2}\beta\omega)^2 \ll 1$ be true for *all* wave numbers, but rather it is necessary for it to hold *only* for the region of q space making the major contribution to the sums. This, we assert, is the case for one-dimensional systems for all accessible temperatures.

Finally, for those cases where the approximation $C_q^\alpha(t) \cong C_q^\alpha F_q^\alpha(t)$ is valid, it remains to determine the static correlation functions C_q^α. In the limit of infinite temperature, $\langle S_i^\alpha S_j^\beta \rangle = \frac{1}{3}S(S+1) \times \delta^{\alpha\beta}\delta_{ij}$ and hence $C_q^{\alpha\beta} = \langle S_{-q}^\alpha S_q^\beta \rangle = [S(S+1)/3N]\delta^{\alpha\beta}$. For the three-dimensional Heisenberg magnet in the paramagnetic regime, Hubbard[3] derived approximate expressions for C_q^α by using various sum rules on the frequency moments. For the classical Heisenberg linear chain with nearest-neighbor interactions only, the C_q^α are known exactly (for the macroscopic ensemble average), and as discussed above, we argue that the classical approximation to C_q^α is valid with the transcription of $J_{cl} \to JS(S+1)$ for the long-wavelength fluctuations of the quantum-mechanical chain of relatively large spin. Now, knowing C_q^α,[28] providing the order parameter Δ is known as a function of T—by perhaps some self-consistent calculation, then the solution of the coupled set of integro-differential equations (27) is obtained by integrating forward in time from $t = 0$ using the initial conditions $F_q(t=0) = 1$.

Up to this point we have limited our discussion to ferromagnetic systems. In the case of the antiferromagnet below the Neél temperature, magnetic order occurs with the characteristic wave vector $Q = \pi$. This fact requires modification of the theory developed for the ferromagnet. The principal modification is that the matrix elements of the quantity $V(t) = \langle \{\exp_+[i\int_0^t h(t')\,dt']\}_S \rangle$ are no longer diagonal in q space, as was the case for the ferromagnet. Now, with the magnetic ordering at $Q = \pi$, the matrix elements $V^{\alpha\alpha}_{q,q\pm\pi}$ and $V^{\alpha\alpha}_{q\pm\pi',q}$ are also nonzero in general. As a consequence, instead of the coupled system of Eqs. (27), we are led to a more complicated system of equations in which the off-diagonal matrix elements of $V(t)$ as well as the diagonal elements are coupled. A more detailed treatment of the antiferromagnet is relegated to Appendix A. Here, it suffices to say that for the one-dimensional antiferromagnet at low temperature, one can make a linear combination of diagonal and off-diagonal elements for given q, which essentially is the Holstein–Primakoff transformation, and one again for all practical purposes arrives at a system of equations similar to (27) for the transformed functions.

We note that it is a simple matter to extend our treatment of isotropic Heisenberg systems to include an applied external field. Further, very similar treatments can be applied to the cases of the anisotropic Heisenberg model in the absence of an external field and of the reduced-dipolar Hamiltonian at high temperatures. These extensions are discussed in Appendix B.

Before concluding this section we return to the fluctuation-dissipation theorem and examine the relationship between the static correlation function C_q^α and the corresponding static susceptibility x_q^α for the case where the frequencies of interest in (32) are large, i.e., $\frac{1}{2}\beta\omega \gg 1$. Setting $t = 0$ in (30) gives

$$C_q^\alpha = (x_q^\alpha/\beta)\,[1 + \tfrac{1}{12}\beta^2 \langle \omega_q^2 \rangle_\alpha + \cdots]\,, \tag{34}$$

where we use the definition of the nth frequency moments of $F_q^\alpha(\omega)$:

$$\langle \omega_q^n \rangle_\alpha = \int_{-\infty}^{\infty} d\omega\, \omega^n F_q^\alpha(\omega) = \left(-\frac{1}{i}\right)^n \frac{d^n}{dt^n}\,[F_q^\alpha(t)]_{t=0}\,. \tag{35}$$

Using (35) we rewrite (34) in a form analogous to (32):

$$C_q^\alpha = (x_q^\alpha/\beta)\langle \tfrac{1}{2}\beta\omega \coth \tfrac{1}{2}\beta\omega \rangle_\alpha\,. \tag{36}$$

This is the required expression linking C_q^α and x_q^α. Note that if $(\frac{1}{2}\beta)^n \langle \omega_q^n \rangle_\alpha \ll 1$ for all n, (36) reduces simply to $C_q^\alpha = x_q^\alpha/\beta$, a result we have already utilized.

III. APPLICATION TO ONE-DIMENSIONAL SYSTEMS: HIGH-TEMPERATURE REGIME

We now apply the theory sketched in Sec. II to the linear magnetic chain with nearest-neighbor interactions only. For simplicity and without loss of generality we consider first the classical chain, for

which the C_q are known exactly.[6] As discussed earlier, for the calculation of the relaxation shape functions, the classical system is a good approximation to real quantum-mechanical systems of sufficiently large spin provided only the small wave numbers make the major contribution to the q' sums in the equations of motion (27).

The Hamiltonian is

$$H = -J_{c1} \sum_l \hat{S}_l \cdot \hat{S}_{l+1} , \tag{37}$$

where the classical spin variable $\hat{S}_l$ is a unit vector. The equal time-spatial correlation functions are given by

$$C_l = \langle \hat{S}_i \cdot \hat{S}_{i+l} \rangle = [u(K)]^{|l|} , \tag{38}$$

where

$$u(K) = \operatorname{sgn} J_{c1} (\coth K - 1/K) \tag{39}$$

and $K = |J_{c1}|/T$. Our convention here is that K is a positive parameter for both ferromagnetic ($J_{c1} > 0$) and antiferromagnetic ($J_{c1} < 0$) coupling, but $\operatorname{sgn} J_{c1}$ ensures the proper sign for $u(K)$. We note that the high- and low-temperature limits of $u(K)$ are

$$\begin{aligned} u(K) &\cong \operatorname{sgn} J_{c1} (\tfrac{1}{3} K - \tfrac{1}{45} K^3 + \cdots) \quad (T \to \infty) , \\ u(K) &\cong \operatorname{sgn} J_{c1} \left(1 - \frac{1}{K} + 2e^{-2K} + 2e^{-4K} + \cdots\right) \\ &\qquad (T \to 0) . \end{aligned} \tag{40}$$

The transcription to the case of finite spin is made simply by replacing J_{c1} with $JS(S+1)$. The q-dependent static correlations are given by the Fourier transforms of (38):

$$\begin{aligned} C_q = \langle \hat{S}_{-q} \cdot \hat{S}_q \rangle &= \frac{1}{N} \sum_l \langle \hat{S}_i \cdot \hat{S}_{i+l} \rangle e^{iql} \\ &= \frac{1}{N} \frac{1 - u^2}{1 + u^2 - 2u \cos q} , \end{aligned} \tag{41}$$

where the units of q are a^{-1}, the inverse lattice parameter. Finally, the fluctuation-dissipation theorem, providing the relationship between the time-dependent correlation function $C_q(t) = \langle \hat{S}_{-q} \cdot \hat{S}_q(t) \rangle$ and the relaxation shape function, reduces simply to $C_q(t) = C_q F_q(t)$.

The second frequency moment[29] of $C_q(\omega)$, the temporal Fourier transform of $C_q(t)$, can be calculated in a straightforward manner by the transfer matrix method along the lines used by Fisher in calculating the C_q. The details of this calculation are presented in Appendix C; the final result is (for all temperatures)

$$\begin{aligned} \langle \omega_q^2 \rangle_{\text{exact}} = -\frac{\ddot{C}_q(0)}{C_q} = -\ddot{F}_q(0) &= 4 J_{c1}^2 \frac{|u|}{K(1-u^2)} \\ &\times (1 - \cos q)(1 + u^2 - 2u \cos q) . \end{aligned} \tag{42}$$

This is an exact result for the classical model and as such provides a useful test for the validity of approximate theories of spin dynamics.

The equations of motion (27) for the relaxation shape functions are partitioned into equations for the longitudinal and transverse components with respect to an axis of quantization. But true long-range order cannot be presented in the linear chain in the absence of an applied magnetic field; i.e., one-dimensional systems cannot exhibit spontaneous magnetization at any finite temperature. (The Curie temperature is absolute zero.) Hence, in a strict sense using the usual ensemble averages, the order parameter Δ is zero, $C_q^x = C_q^y = C_q^z = \frac{1}{3} C_q$, and $F_q^x(t) = F_q^y(t) = F_q^z(t) = F_q(t)$. Then Eqs. (27) reduce to a single equation for $F_q(t)$:

$$\dot{F}_q(t) = -\frac{2}{3} \sum_{q'} K_{qq'} C_{q'} \int_0^t dt' F_{q'}(t - t') F_{q-q'}(t - t') F_q(t') . \tag{43}$$

For high temperatures $K \ll 1$, (43) apparently provides a good description of the spin dynamics. First, if we expand the exact second moment (42) in powers of K to order K^2 we have

$$\langle \omega_q^2 \rangle_{\text{exact}} \cong \tfrac{4}{3} J_{c1}^2 (1 - \cos q)(1 \mp \tfrac{2}{3} K \cos q + \tfrac{7}{45} K^2) , \tag{44}$$

where the sign of the second term in parentheses is negative for the ferromagnet and positive for the antiferromagnet. The corresponding result calculated from our approximate theory, Eq. (43), is

$$\begin{aligned} \langle \omega_q^2 \rangle_{\text{approx}} &= \frac{2}{3} \sum_{q'} K_{qq'} C_{q'} \\ &\cong \tfrac{4}{3} J_{c1}^2 (1 - \cos q)(1 \mp \tfrac{2}{3} K \cos q + \tfrac{1}{9} K^2) . \end{aligned} \tag{45}$$

Thus, $\langle \omega_q^2 \rangle_{\text{approx}}$ is exact to first order in $1/T$ and yields an error of the order of 25% only in the coefficient of $(1/T)^2$. Further, as will be pointed out in Sec. V, where we present the results of the numerical calculations, Eq. (43) produces good agreement with the computer-simulation results of Blume *et al.*[17] at high temperatures. We conclude that (43) provides a good description of the spin dynamics in the high-temperature regime and that no significant local magnetic order is present.

For lower temperatures, $K > 1$, on the other hand, the situation is not good. The theory does in fact predict critical slowing down for the antiferromagnet for $q \sim \pi$. But spin waves do not appear in the theory as T decreases, in sharp contrast to both the experimental work of Hutchings *et al.*[10] and the computer-simulation results of Blume *et al.*[17] Rather, the theory yields broadened peaks for $C_q(\omega)$ centered around $\omega = 0$ and $C_q(t)$ displays highly damped oscillatory behavior, all completely unlike spin-wave behavior. Further, the exact second moment as given by (42) is obviously indicative of usual spin-wave behavior at low tempera-

ture. In fact, in the limit $T \to 0$, (42) yields

$$\langle \omega_q^2 \rangle_{\text{exact}} = 4 J_{c1}^2 \begin{cases} (1-\cos q)^2 & \text{(ferro)} \\ \sin^2 q & \text{(antiferro)} . \end{cases} \quad (46)$$

As just noted, the solution of (43) produces broad peaks around $\omega = 0$ even in the limit $T \to 0$, with a second moment given by

$$\langle \omega_q^2 \rangle_{\text{approx}} = \tfrac{2}{3} \langle \omega_q^2 \rangle_{\text{exact}} \quad (T = 0) . \quad (47)$$

A first attempt to patch up the theory[30] was the application of the so-called semiclassical approximation first used by Schofield[31] in neutron scattering from fluids. This approximation makes the correspondence

$$S_q^{\text{qm}}(\omega) = e^{\beta\omega/2} S_q^{\text{cl}}(\omega) \quad (48)$$

between the true quantum-mechanical generalized structure factor $S_q^{\text{qm}}(\omega)$ and the idealized classical structure factor $S_q^{\text{cl}}(\omega)$. In essence, this correspondence forces the classical function to satisfy the condition of detailed balance at low T. However, as shown by Siggia and Blume,[32] the semiclassical approximation applied to spin systems is valid only to first order in $\frac{1}{2}\beta\omega$. But the region of spin-wave behavior is precisely the region $\frac{1}{2}\beta\omega > 1$. ($\frac{1}{2}\beta\omega \sim \beta J_{c1} = K > 1$ for spin waves.) In applying the semiclassical approximation to the spin-wave region we found something like spin waves were produced which behaved in rough qualitative agreement with experiment, but first, the value of the exchange constant calculated from the "spin-wave" dispersion was too large by a factor of $\gtrsim 50\%$ at $T = 0$; second, the "spin-wave" frequencies renormalized downward with temperature far too rapidly; third, the linewidths were too large by a factor of 2–3. Finally, one feels that the purely classical spin system should exhibit spin waves at low temperature without being forced by an approximation like (48). This feeling is strongly reinforced by the form of the exact second moment (46) at $T = 0$.

It is apparent that some essential physics is lacking in the application of (27) at low temperatures. Any theory which would properly describe the spin-wave region would have to include this physics as input to the kinetic equations. In Sec. IV we discuss the modification of the theory necessary to treat the spin dynamics at low temperature.

IV. SPIN-WAVE REGION

A. Local Order Parameter

At low but finite temperature—even though true long-range order does not exist for one-dimensional systems—considerable order does exist over distances of the order of the correlation length $l_c \sim \pi K a$. This is described by the fact that correlations between spins l lattice spacings apart go like $[u(K)]^{|l|}$, and as $T \to 0$ ($K \to \infty$), $|u(K)| \to 1 - (1/K)$. For the short-wavelength high-frequency excitations, i.e., spin waves, the system does in fact appear essentially ordered at low temperature. The physical picture is that of spin waves propagating in locally ordered regions.[33] This picture is valid as long as the wavelengths of the spin waves are small compared with l_c and the periods of the spin-wave oscillations are small compared with the characteristic times required for appreciable changes to occur in the local ordering.

We are thus led to introduce the notion of a short-range order parameter, which for the high-frequency, short-wavelength fluctuations describes a local magnetization at low temperature. Into the theory we write

$$\langle\langle \hat{S}(r,t) \rangle\rangle = \Delta \hat{i}(r,t) , \quad (49)$$

where $\langle\langle \cdots \rangle\rangle$ denotes a local average in some sense to be described below, Δ is the order parameter, and $\hat{i}(r,t)$ is a unit vector giving the direction of the local magnetization. (We continue to confine our attention to the classical case without loss of generality.) The basic underlying assumption is that $\hat{i}(r,t)$ changes slowly with both r and t in comparison with the spin-wave oscillations, so that to the spin waves $\hat{i}(r,t)$ appears to be a constant vector to a good approximation. As $T \to 0$ the order parameter approaches unity and $\hat{i}(r,t)$ approaches a constant vector in the macroscopic sense, as the scale of variation in the local order becomes longer, both spatially and temporally. For T large, of course, $\Delta \to 0$ and we revert to the theory of Sec. III.

We are in essence characterizing the fluctuations of the system as being either one of two types. The long-wavelength fluctuations $q \lesssim 1/K$ are characterized as being predominantly diffusive in nature and longitudinal with respect to the direction of the local magnetization. These fluctuations decay slowly and are associated with the slow changes in the order parameter. The shorter-wavelength oscillations $q \gg 1/K$ are primarily spin-wave-like and transverse in nature; they decay much more rapidly. Within the framework of these ideas it is doubtful that long-wavelength ($q < 1/K$) spin waves can be said to exist. Further, we note the obvious fact that the size of the spin-wave region shrinks as the temperature is increased.

These arguments are quite similar to those used by Lines[21] in his treatment of the two-dimensional Heisenberg magnet, in particular, his introduction of wave-vector-dependent magnon renormalization. He argues that magnons with wavelengths greater than some coherence length L have a frequency small compared with the reciprocal relaxation time of the background disorder and can therefore be described in first approximation as displacements

from the time-averaged background. This leads to a renormalization of magnon energy according to the magnetization $\langle \vec{S} \rangle$ (which for the case of the linear chain is zero). Lines then argues, however, that such an approximation is clearly inappropriate for short-wavelength spin waves $\lambda \ll L$. These have a frequency large compared to the background reciprocal relaxation time, and can therefore, to a first approximation, be described as displacements from the instantaneous background. Using an analogy first introduced by Keffer and Loudon, [34] the additional magnon excitations are like ripples superimposed on the instantaneous nonequilibrium position of the existing waves. For nearest-neighbor exchange this concept leads to a magnon energy renormalization as $(\langle \vec{S}_0 \cdot \vec{S}_a \rangle)^{1/2}$, where $\vec{S}_0$ and $\vec{S}_a$ are nearest neighbors. The principal distinction between the present treatment and that of Lines is that we claim that long-wavelength magnons are not present in the linear chain; the long-wavelength fluctuations are to be associated with variations in the local order.

Now we introduce the idea of a local coordinate system with the z axis in the direction of the local magnetization. The orientation of the local coordinate system changes slowly with time and with distance along the chain. In order for the kinetic equations that we derived for the relaxation shape functions to describe the spin-wave excitations, they must be considered applicable in the local sense, with the longitudinal and transverse components taken relative to the (slowly varying) local coordinate system. Then to describe the results of experiments on macroscopic samples, we must perform a spatial average at the end of the calculation.

In the limit $T = 0$, these ideas are certainly correct and yield exact results. At absolute zero we have complete alignment of spins; hence $\Delta = 1$, $C_q^z = \delta_{q,0} C_q$, and $C_q^x = C_q^y = 0$. (For the antiferromagnet with magnetic ordering at wave vector $Q = \pi$, the only change is $C_q^z = \delta_{q,\pi} C_q$.) The first moment of the spectral line is given by

$$\langle \omega_q \rangle = -(1/i)\dot{F}_q^{\pm}(0) = \begin{cases} -J_{qq} = 2J_{c1}(1 - \cos q) & \text{(ferro)} \\ (J_{q,q+\pi} J_{q+\pi,q})^{1/2} = 2|J_{c1} \sin q| & \text{(antiferro)} . \end{cases} \tag{50}$$

[See Eq. (27) for the ferromagnet and Eq. (A8) for the antiferromagnet.] The second moment is given by

$$\langle \omega_q^2 \rangle = -\ddot{F}_q^{\pm}(0) = \langle \omega_q \rangle^2 , \tag{51}$$

and hence the linewidth Γ_q, which is proportional to $(\langle \omega_q^2 \rangle - \langle \omega_q \rangle^2)^{1/2}$, is zero. The theory thus produces infinitely sharp spin-wave peaks over the whole of the Brillouin zone with the usual dispersion. The real test of the theory comes as we look at finite temperatures and consider both the renormalization of the spin-wave energies and the damping of the spin waves as a function of temperature.

As we extend these ideas to finite temperatures we have several problems to consider. First, we must make a proper choice of the "order parameter" as a function of temperature, and, secondly, we must determine the appropriate breakdown of the static correlation functions into longitudinal and transverse components. We then study the damping of the spin waves; in particular, by calculating the first and second frequency moments of the transverse fluctuations, we arrive at expressions for the magnon lifetimes (or linewidths). Next, we examine the internal consistency of the theory by looking at the long-wavelength fluctuations, and checking to ensure that the characteristic times associated with changes in the local order are indeed long compared with the time scale of the spin-wave oscillations. Finally, we consider the appropriate spatial averaging process necessary to describe the macroscopic sample.

We have defined the order parameter in terms of a local average of the magnetization $\langle\langle \hat{S}(r, t) \rangle\rangle = \Delta \hat{i}(r, t)$. Physically it is clear that Δ is associated with some sort of weighted average of the spin density over a region of the order of l_c centered around the point r. To arrive at an operational definition of Δ we note from the form of the dynamic equations (27) that Δ also gives directly the renormalization of the spin-wave energies. That the local average of the magnetization and the magnon renormalization are directly related follows immediately from our earlier discussion in which we picture the short-wavelength excitations of the system as oscillating against the instantaneous local background, the configuration of which changes only slowly with space and time. It is then natural to make use of the result of conventional spin-wave theory [20,34] for the renormalization of the spin-wave energies, namely, that Δ is given by the square root of the near-neighbor correlation. (This result follows when, in considering exchange effects between neighboring spins $\vec{S}_1$ and $\vec{S}_2$, those components of each which are perpendicular to $\vec{S}_1 + \vec{S}_2$ are averaged to zero.) Thus, for the classical linear chain we have

$$\Delta = |u(K)|^{1/2} \cong 1 - 1/2K \quad (\text{as } T \to 0) . \tag{52}$$

As we shall see in Sec. V this renormalization agrees very well with the experimental data on $(CH_3)_4NM_nCl_3$, (TMMC), clear evidence that the usual concepts of spin-wave theory are applicable to one-dimensional systems—as long as the wavelengths are shorter than the coherence length.

B. Static Correlation Functions

We consider the static correlation functions defined by

$$C_q = \langle \hat{S}_{-q} \cdot \hat{S}_q \rangle = \frac{1}{N} \sum_l \langle \hat{S}_i \cdot \hat{S}_{i+l} \rangle e^{iql} . \tag{53}$$

This definition is the macroscopic ensemble average, which has been calculated exactly [see Eq. (41)]. The connection with the local averaging process is made by expressing the spins in (53) in terms of the local magnetization and the fluctuations from the local order. We write the spin at site i as

$$\hat{S}_i = \langle\langle \hat{S}_i \rangle\rangle + \hat{S}_i - \langle\langle \hat{S}_i \rangle\rangle = \vec{\Delta}_i + \delta\vec{S}_i \quad , \tag{54}$$

where $\vec{\Delta}_i = \Delta \hat{i}(r_i)$ and $\delta\vec{S}_i = \hat{S}_i - \vec{\Delta}_i$. Substituting into (53) we have (noting that $\langle \vec{\Delta}_i \cdot \delta\vec{S}_{i+l} \rangle = 0$)

$$C_q = \frac{1}{N} \sum_l \left(\langle \vec{\Delta}_i \cdot \vec{\Delta}_{i+l} \rangle + \langle \delta\hat{S}_i \cdot \delta\hat{S}_{i+l} \rangle \right) e^{iql} . \tag{55}$$

Clearly, the first term in this expression is associated with the (long-wavelength) fluctuations of the order parameter; these fluctuations stem from the fact that the local order varies slowly in direction with position along the chain. They are longitudinal in nature with respect to the local coordinate system. The second term of (55), which corresponds to transverse fluctuations around the local mean field, is then associated with the shorter-wavelength spin-wave modes. With these remarks in mind we rewrite (55) as

$$C_q = C_q^L + 2C_q^T , \tag{56}$$

with the obvious partitioning into longitudinal and transverse components:

$$C_q^L = \frac{1}{N} \sum_l \langle \vec{\Delta}_i \cdot \vec{\Delta}_{i+l} \rangle e^{iql} ,$$

$$C_q^T = \frac{1}{2N} \sum_l \langle \delta\vec{S}_i \cdot \delta\vec{S}_{i+l} \rangle e^{iql} . \tag{57}$$

We assume that this correspondence is valid at sufficiently low temperature. As the temperature is raised the concept of local order becomes hazy and therefore so does the correspondence (57).

In the limit $T = 0$ we have noted that C_q is a δ function at $q = 0$ (or at $q^* = \pi - q = 0$ for the antiferromagnet). At low but finite temperature, C_q is peaked very sharply around $q = 0$ with a spread in q of the order $\Delta q = 1/K$. Therefore, the important thermal fluctuations are those of long wavelength ($q \lesssim 1/K$) and are associated primarily with the spatial variations of the order parameter. It is this region of wave-vector space that dominates the q' sums in the kinetic equations (27). Consequently, as a first approximation in solving the kinetic equations (for the spin-wave relaxation functions), we assume that all q dependence of the static correlation functions is accounted for by the spatial variations in the order parameter. We neglect the transverse static correlations altogether and write

$$C_q^L \cong C_q \quad (q \lesssim 1/K) ,$$
$$C_q^T \cong 0 . \tag{58}$$

It is to be stressed that this approximation is valid only in the long-wavelength region. In fact, for $q \gg 1/K$ the opposite condition of (58) prevails: The transverse fluctuations dominate (on a relative basis) and we have $C_q^L \cong 0$, $C_q^T \cong \frac{1}{2} C_q$. Though this region of q space is negligible in thermodynamic sums at low temperature,[35] it *is* important in calculating the generalized structure factor (1) for neutron scattering in the spin-wave region. Specifically, the C_q^T determines the intensity scale of the spectral line for momentum transfer q (see Sec. V).

The intermediate region $q \gtrsim 1/K$ presents a much more difficult problem, because the partitioning of C_q into longitudinal and transverse components is obviously strongly q dependent as one goes from (58) for $q \lesssim 1/K$ to the opposite condition for $q \gg 1/K$. In order to render the calculations tractable and as simple as possible, we will apply the approximation (58) in solving the kinetic equations, assuming that the q' sums are sufficiently weighted for small q' that only a minor error is introduced. This assumption will be checked (and apparently borne out) by our comparison of the calculations with experimental data in Sec. V.

C. Spin-Wave Damping

We now calculate the first and second frequency moments of the transverse correlation functions utilizing the ideas and approximations we have introduced. This will enable us to derive quantitative expressions for the damping of the spin waves as the temperature is increased from $T = 0$. From the dynamic equations (27) for $F_q^{\pm}(t)$, we have, for the first moment,

$$\langle \omega_q \rangle_T = -\frac{1}{i} \dot{F}_q^{\pm}(0)$$
$$= \begin{cases} 2J_{c1}\Delta(1 - \cos q) & \text{(ferro)} \\ 2\Delta |J_{c1} \sin q| & \text{(antiferro)} . \end{cases} \tag{59}$$

From the definition of the order parameter, Eq. (52), we have the result that the spin-wave energies renormalize downward linearly with increasing temperature. The second moment is given by

$$\langle \omega_q^2 \rangle_T = -\ddot{F}_q^{\pm}(0) = \sum_{q'} K_{qq'} (C_{q'}^L + C_{q'}^T) = \sum_{q'} K_{qq'} C_{q'} \tag{60}$$

on making use of approximation (58). After a straightforward calculation, we find the results

$$\langle \omega_q^2 \rangle_T = 4 J_{c1}^2 \begin{cases} (1-\cos q)^2 \left(1-\frac{1}{K}\right) + \frac{1}{2K^2}(1-\cos q) & \text{(ferro)} \\ \sin^2 q \left(1-\frac{1}{K}\right) + \frac{1}{2K^2}(1-\cos q) & \text{(antiferro)} \end{cases} \tag{61}$$

to order[36] $1/K^2$. The first term in each of these expressions is the square of the first moment; thus, the leading-order temperature effect in $\langle \omega_q^2 \rangle_T$ is simply a shift in the peak position. In a certain sense this result corroborates our choice of Δ with the approximation (58) that went into the derivation of (61). Further, we can rewrite (61) as

$$\langle \omega_q^2 \rangle_T = \langle \omega_q \rangle_T^2 + \Gamma_q^2 , \tag{62}$$

where the temperature-dependent linewidth Γ_q, defined by $\Gamma_q = (\langle \omega_q^2 \rangle_T - \langle \omega_q \rangle_T^2)^{1/2}$, is given by

$$\Gamma_q = \frac{\sqrt{2}\, J_{c1}}{K}(1-\cos q)^{1/2} = \sqrt{2}\, T (1-\cos q)^{1/2} \tag{63}$$

for both the ferromagnet and the antiferromagnet. Hence, the theory predicts a linear broadening of the spin-wave spectral lines as T increases, as well as a q dependence of the linewidths of the form $(1-\cos q)^{1/2}$. Thus, Γ_q varies appreciably with q—for both the ferromagnet and antiferromagnet—in the region of momentum transfer $0 < q < \frac{1}{2}\pi$, but relatively little for the region $\frac{1}{2}\pi < q < \pi$; in fact, $\partial \Gamma_q / \partial q = 0$ at $q = \pi$.

The most useful way of presenting the q dependence of the linewidths, however, is to look at the quantity σ_q, defined to be the ratio of linewidth to peak energy, i.e., $\sigma_q = \Gamma_q / \langle \omega_q \rangle_T$. From (59) and (63) we have to lowest order in temperature

$$\sigma_q = \begin{cases} \frac{1}{2K}\left(\frac{2}{1-\cos q}\right)^{1/2} & \text{(ferro)} \\ \frac{1}{2K}\left(\frac{2}{1+\cos q}\right)^{1/2} & \text{(antiferro)} . \end{cases} \tag{64}$$

For the ferromagnet then, σ_q diverges as $q \to 0$, and furthermore, $\sigma_q > 1$ for $q < 1/K$ ($\sigma_q = 1$ at $q = 1/K$). This is simply a statement of the fact that well-defined spin-wave excitations do not exist for $q < 1/K$, and it provides a check of the internal consistency of the theory. In the spin-wave region $q > 1/K$, σ_q is less than unity, attaining its smallest value $\sigma_q = 1/2K$ at the zone boundary $q = \pi$. For the antiferromagnet these same remarks apply if we replace q by $q^* = \pi - q$. In this case σ_q diverges at $q^* = 0$ and well-defined ($\sigma_q < 1$) spin waves exist only in the region $q^* > 1/K$.

One may inquire as to what processes give rise to the finite spin-wave lifetimes in the approximation $C_q^L \cong C_q$, $C_q^T \cong 0$. We attribute the scattering in this approximation to the fact that the local magnetization $\vec{\Delta}$ does in reality vary, both in time and with position along the chain. Thus, a spin-wave fluctuation—even of short wavelength—does not see truly long-range order, and this leads to decay of the spin waves. By attributing the long-wavelength fluctuations of the system to the variations in the local order, we have been able to incorporate this mode of scattering into the theory.

The other mode of spin-wave decay that one might suspect to be operating is, of course, scattering from other spin waves. However, as is by now clear from our arguments, spin waves with wave vectors $q \lesssim 1/K$ cannot be said to exist, and it is precisely this region which makes the major contribution to the q' sums in the kinetic equations. Thermal spin waves—in the sense of spin-wave modes being thermally populated—do not exist for all practical purposes in the linear chain. Spin waves exist only above the thermodynamically important region, and the boundary ($q \sim 1/K$) between the spin wave and thermodynamic regions increases with increasing temperature.

D. Fluctuations in Local Order

We have associated the long-wavelength fluctuations of the system with spatial variations in the order parameter. We now examine briefly the time dependence of these fluctuations. In particular, we are interested in the characteristic times required for appreciable variations in $\vec{\Delta}(r, t)$ and the possible relations to the spin-wave lifetimes; we must verify that $\vec{\Delta}(r, t)$ changes sufficiently slowly that our procedure for calculating spin-wave dynamics is justified.

Thus, we study the time dependence of the relaxation shape functions for wave vectors $q \lesssim 1/K$. For these fluctuations the concept of local order obviously has no meaning. True macroscopic averages must be used from the outset [$F_q^x(t) = F_q^y(t) = F_q^z(t)$], and the appropriate dynamical equation is (43) of Sec. III. First, note that since $q \lesssim 1/K$ and C_q is peaked sharply at low T around $q = 0$ with a spread in q of the order $1/K$, all the F functions appearing in the time integral have small wave-vector arguments and hence presumably a slow time dependence. Thus, to obtain very quickly an idea of the characteristic time scale for the long-wavelength fluctuations, we iterate (43) once, making the zero-order approximation $F_q = 1$ for the relaxation shape functions on the right-hand side. We write the result as

$$F_q(t) \approx 1 - \frac{1}{2}\left(\frac{t}{\tau_q}\right)^2 \qquad (t < \tau_q)\,, \tag{65}$$

where

$$\frac{1}{\tau_q^2} = \frac{2}{3}\sum_{q'} K_{qq'}\, C_{q'}$$

$$\cong \tfrac{8}{3} J_{\mathrm{cl}}^2 \begin{cases} \frac{1}{4} q^2(q^2 + 1/K^2) & \text{(ferro)} \\ q^{*2} + 1/K^2 & \text{(antiferro)}\,. \end{cases} \tag{66}$$

[Note that $F_q(t)$ is essentially Gaussian for small times.] As $q \lesssim 1/K$ (or $q^* = \pi - q \lesssim 1/K$ for the antiferromagnet) we have

$$\tau_c \approx \begin{cases} K^2/J_{\mathrm{cl}} & \text{(ferro)} \\ K/J_{\mathrm{cl}} & \text{(antiferro)} \end{cases} \tag{67}$$

as the characteristic time scale of the long-wavelength fluctuations and hence of $\vec{\Delta}(r, t)$. We point out that the corresponding results for a real system of spin S are $\tau_c = K^2/J\,[S(S+1)]^{1/2}$ for the ferromagnet and $\tau_c = K/J\,[(S+1)]^{1/2}$ for the antiferromagnet.

Returning to Eq. (63) we see that the spin-wave lifetimes $\tau_q^{sw} = \Gamma_q^{-1}$ are of the order of τ_c or less (much less for ferromagnetic magnons of wave vector $q \gg 1/K$). Equality, $\tau_q^{sw} = \tau_c$, is obtained at $q = 1/K$ for the ferromagnet and at $q^* = 1/K$ for the antiferromagnet. In addition for the well-defined spin waves $q \gg 1/K$, for which $\alpha_q \ll 1$, we obviously have the reciprocal spin-wave frequencies $\omega_q^{-1} \ll \tau_c$. Consequently, the theory is consistent in that the time scale for the (well-defined) spin-wave oscillations is indeed much less than the characteristic time for changes in the local order, and further we have the result that the spin-wave lifetimes are limited by τ_c. This corroborates our conjecture that spin-wave decay results primarily from the fluctuations of the order parameter; i.e., our model predicts that spin waves exist for a time not greater than the time over which the local order remains relatively constant. We have therefore demonstrated that the physical picture of our model is valid and consistent, in the temporal sense as well as in the spatial sense. We will have more to say in Sec. V concerning the long-time ($t \gg \tau_c$) behavior of the long-wavelength fluctuations.

E. Spatial Averaging Process

First, we compare the result for the transverse second moment in the spin-wave region $q > 1/K$ computed from our approximate theory, Eq. (61), with the exact second moment (42). At low temperature $\langle \omega_q^2 \rangle_{\mathrm{exact}}$ is to order $1/K^2$,

$$\langle \omega_q^2 \rangle_{\mathrm{exact}} = 4 J_{\mathrm{cl}}^2 \begin{cases} (1-\cos q)^2 (1 - 3/2K + 1/4K^2) + (1/2K^2)(1-\cos q) & \text{(ferro)} \\ \sin^2 q (1 - 3/2K + 1/4K^2) + (1/2K)(1-\cos q) & \text{(antiferro)}\,. \end{cases} \tag{68}$$

On comparison with (61) we see that the temperature factors multiplying the term $(1-\cos q)^2$ for the ferromagnet (and $\sin^2 q$ for the antiferromagnet) are different for $\langle \omega_q^2 \rangle_{\mathrm{exact}}$ and $\langle \omega_q^2 \rangle_T$, the primary difference being that the coefficient of the linear term in $1/K$ is $-\frac{3}{2}$ for the exact second moment, whereas it is -1 for our approximate expression. The discrepancy, however, does not necessarily indicate that the approximate theory is incorrect. For we have only calculated the second moment of the transverse correlation function from our theory; we have said nothing about the moments of the longitudinal correlation functions. The exact second moment, on the other hand, certainly contains contributions from both. This point leads to the discussion of the appropriate averaging process that should be performed in order to describe experiments on macroscopic samples.

We have argued that for the spin-wave excitations the kinetic equations (27) are applicable in the local sense, and the results of Sec. IV D, indicating that the local order changes slowly on the time scale of the magnons, substantiate our assertions. Clearly, then, the macroscopic observable time-dependent correlation function $C_q(t)$ $= \langle \hat{S}_{-q} \cdot \hat{S}_q(t) \rangle$ is given by

$$C_q(t) = C_q^L F_q^L(t) + 2 C_q^T F_q^T(t)\,, \tag{69}$$

where $F_q^L(t)$ and $F_q^T(t) = \mathrm{Re}[F_q^+(t)]$ are the local relaxation shape functions obtained by the solution of (27) and C_q^L and C_q^T are the local static correlations [see Eqs. (56) and (57)]. Hence, as long as we are in the regime of well-defined local order (i.e., low T) and if the local longitudinal and transverse components C_q^L and C_q^T are known, (69) contains all the information necessary to describe the dynamics of the macroscopic system.

Consider, for example, the second moment of the macroscopically observable spectral line,

$$\langle \omega_q^2 \rangle = -\frac{\dot{C}_q(0)}{C_q} = \alpha_q \langle \omega_q^2 \rangle_L + 2\beta_q \langle \omega_q^2 \rangle_T\,, \tag{70}$$

where $\langle \omega_q^2 \rangle_L = -\dot{F}_q^L(0)$, $\langle \omega_q^2 \rangle_T = -\dot{F}_q^T(0)$, $\alpha_q = C_q^L/C_q$, and $\beta_q = C_q^T/C_q$. (Note that $\alpha_q + 2\beta_q = 1$.) We have argued previously, at least implicitly, that in the spin-wave region $q \gg 1/K$ the major contribution to the spectral line is due to the transverse fluctuations, or spin waves, and therefore $\alpha_q \cong 0$ and $\beta_q \cong \frac{1}{2}$. Furthermore, using the long-wavelength approximation (58) in the "thermodynamic" q' sums

in (27), we have $\langle \omega_q^2 \rangle_L = 0$ and $\langle \omega_q^2 \rangle_T$ given by (61). If we now assume that the long-wavelength approximation (58) is good to a high degree of precision at low T (it is rigorously true in the limit $T=0$), we can determine the lowest-order temperature corrections to α_q and β_q in the spin-wave region by simply forcing the equality of the second moment computed by (70) with the exact second moment (68). We find the results

$$\alpha_q = \frac{1}{2K} \quad , \quad \left(q \gg \frac{1}{K} \; ; \quad K \gg 1 \right)$$
$$\beta_q = \frac{1}{2}\left(1 - \frac{1}{2K}\right) . \tag{71}$$

Thus, the relative proportion of spin-wave component decreases linearly with increasing temperature, with a corresponding increase in the relative amount of longitudinal component. In this description the spectral function $C_q(\omega)$ for the macroscopic system would exhibit two strong spin-wave peaks at $\omega = \pm \omega_q$, where ω_q is given by (59), and a weak central component. The spin-wave components shift downward and broaden as T increases. The intensity of the central component, which is a δ function at $\omega = 0$ in the present model [$\langle \omega_q^2 \rangle_L = 0$ within the long-wavelength approximation (58)], increases linearly with T relative to the spin-wave components.

We expect, of course, that the central component also broadens with temperature. In order to account for this with our theory, however, it would be necessary to improve upon the approximation (58) for the long-wavelength region $q \lesssim 1/K$. In particular, we would in some way have to incorporate q dependence into the partitioning of C_q into longitudinal and transverse components in this (thermodynamically important) region. Then $\langle \omega_q^2 \rangle_L$ is in general nonzero. However, the experimental work on the antiferromagnetic linear chain TMMC [10] shows no evidence of a central component at low T. In fact, to anticipate somewhat the discussion of Sec. V, the essential features of the experimental scattering are described very well with the present theory using the simple approximation (58). In this connection it should be pointed out that TMMC is a "real" quantum system of spin-$\frac{5}{2}$, and hence the neutron scattering is related to the generalized susceptibility, Eq. (1), which contains the factor of detailed balance $\beta\omega/(1+e^{-\beta\omega})$. For low temperatures in the spin-wave region, $\beta\omega \gg 1$, and this factor is very important, serving to amplify greatly the spin-wave emission peak relative to the central component (and to the magnon absorption peak). Thus, even if a central component is present in real systems, it would be difficult to discern experimentally. Therefore, even if we attempted theoretically to treat the central component with more sophistication (i.e., by including the q dependence of the partitioning of C_q), we still would be unable to compare theory with experiment. This is sufficient grounds for not going beyond the simple approximation (58) at this time.

V. RESULTS AND DISCUSSION

A. Numerical Calculations

Returning to the dynamical equations (27) we rewrite them in the "memory kernel" form

$$\dot{F}_q^z(t) = -\int_0^t dt' \, k_q^z(t-t') \, F_q^z(t') ,$$
$$\dot{F}_q^{\pm}(t) = \mp i\omega_q F_q^{\pm}(t) - \int_0^t dt' \, k_q^{\pm}(t-t') \, F_q^{\pm}(t') , \tag{72}$$

where ω_q is the transverse first moment given by (59) and the memory functions $k_q^{\alpha}(t)$ are defined by

$$k_q^z(t) = \sum_{q'} K_{qq'} \, C_{q'} \, \beta_{q'} \, [F_{q'}^-(t) F_{q-q'}^+(t) + F_{q'}^+(t) F_{q-q'}^-(t)] ,$$
$$k_q^{\pm}(t) = \sum_{q'} K_{qq'} \, C_{q'} \Big[\tfrac{1}{2} \beta_{q'} \, F_{q'}^{\pm}(t) F_{q-q'}^z(t)$$
$$+ \left(\alpha_{q'} F_{q'}^z(t) - \frac{\Delta^2}{C_{q'}} \delta_{q',0} \right) F_{q-q'}^{\pm}(t) \Big] . \tag{73}$$

Here $K_{qq'}$ is defined by (28) and $C_{q'}$ is given by (41); for generality we have written $C_{q'}^z = \alpha_{q'} C_{q'}$ and $C_{q'}^T = \beta_{q'} C_{q'}$ where $\alpha_q + 2\beta_q = 1$. For the antiferromagnet with local order occurring at wave vector $Q = \pi$ these same equations are applicable except that the $F_q^{\pm}(t)$ are replaced by the transformed functions $f_q^{\pm}(t)$ (see Appendix A) and $\delta_{q,0} \to \delta_{q,\pi}$. We solve these equations for high ($K \lesssim 1$) and low ($K \gg 1$) temperatures. We are unable to say much quantitatively about intermediate temperatures $K \sim 1$, but this is presently the least interesting region physically.

As discussed in Sec. III, at high temperatures we have $\Delta = 0$, $\alpha_q = \beta_q = \frac{1}{3}$, and $F_q^z(t) = F_q^{\pm}(t) = F_q(t)$. Then Eqs. (72) reduce to the single equation

$$\dot{F}_q(t) = -\int_0^t dt' \, k_q(t-t') \, F_q(t') , \tag{74}$$

where

$$k_q(t) = \frac{2}{3} \sum_{q'} K_{qq'} \, C_{q'} \, F_{q'}(t) F_{q-q'}(t) . \tag{75}$$

We note that for $K \ll 1$,

$$C_q \cong \frac{1}{N}\left(1 \pm \frac{2K}{3} \cos q + \frac{2K^2}{9} \, (2\cos q - 1) \right) , \tag{76}$$

where the plus sign is for ferromagnetic coupling and the minus sign for antiferromagnetic coupling.

At low temperatures we distinguish between the long- and short-wavelength fluctuations. For long wavelengths (74) is in fact the appropriate equation at all temperatures, as the concept of local order is not applicable. Further, as only the F functions with wave vectors $q \lesssim 1/K$ contribute significantly to the sum in (75), there is no coupling to the spin-

wave modes in the first approximation. For the spin-wave region $q \gg 1/K$ we apply the discussion of Sec. IV: $\Delta^2 = |u| \cong 1 - 1/K$ and $\alpha_q \cong 1$, $\beta_q \cong 0$. Then $k_q^z(t) \cong 0$ and consequently $F_q^z(t) \cong 1$ on the time scale of the spin waves. For the transverse fluctuations we make the *Ansatz*

$$F_q^{\pm}(t) = e^{\mp i \omega_q t} F_q^T(t) \tag{77}$$

and we take advantage of the fact that $C_{q'}$ is peaked sharply around $q' = 0$ by setting $F_{q-q'}^T(t) \cong F_q^T(t)$ and $\omega_{q-q'} \cong \omega_q$ in $k_q^{\pm}(t)$. Then we find that the envelope function $F_q^T(t)$ for the spin-wave oscillations satisfies the equation

$$\dot{F}_q^T(t) = -\int_0^t dt' \, k_q^T(t-t') F_q^T(t') , \tag{78}$$

where

$$k_q^T(t) = \sum_{q'} K_{qq'} C_{q'} \left(F_{q'}^z(t) - \frac{\Delta^2}{C_{q'}} \delta(q') \right) F_q^T(t). \tag{79}$$

[$\delta(q') = \delta_{q',0}$ for the ferromagnet and $\delta(q') = \delta_{q',\pi}$ for the antiferromagnet.] The $F_{q'}^z(t)$ appearing in (79) must be obtained from the long-wavelength solutions of the kinetic equations, i.e., from the solution of (74). We note that (79) may be rewritten

$$k_q^T(t) = [A_q(t) - \omega_q^2] F_q^T(t) , \tag{80}$$

where

$$A_q(t) = \sum_{q'} K_{qq'} C_{q'} F_{q'}^z(t) . \tag{81}$$

The equations of motion—either Eqs. (74) and (75) or (78) and (79)—are solved numerically by integrating forward in time from $t = 0$ using the initial conditions $F_q^{\alpha}(0) = 1$. Time is measured in units of $2J_{c1}$ for the classical case or $2J[S(S+1)]^{1/2}$ upon making the transcription to the quantum-mechanical case; temperature is given by the values of the parameter $K = J_{c1}/T$ (or $JS(S+1)/T$); and the wave vectors q are measured in units of a^{-1}, the inverse lattice spacing. Sometimes we will refer to normalized wave vectors defined by $Q = q/2\pi$ so that $Q = 0.5$ corresponds to the Brillouin-zone boundary. The computations were performed on a CDC 6600 computer using for the high-temperature solutions a 25-point grid of q values evenly spaced between 0 and π and a time step in the time integration of 0.04. For the low-temperature solutions a 201-point q grid and a time step of 0.1 were used. The finer q mesh was necessary in this case due to the sharpness of the q' integrands around $q' = 0$.

Once the relaxation shape functions $F_q^{\alpha}(t)$ are determined, the time-dependent correlation functions are given (for the classical system) simply by $C_q^{\alpha}(t) = C_q^{\alpha} F_q^{\alpha}(t)$ and the spatial time-dependent correlations $C_l(t) = \langle \hat{S}_i \cdot \hat{S}_{i+l}(t) \rangle$ by

$$C_l(t) = \sum_{q,\alpha} C_q^{\alpha}(t) e^{iql} . \tag{82}$$

The neutron-scattering cross section is related by Eq. (1) to the Fourier transform of $F_q^{\alpha}(t)$, which can be computed directly by

$$F_q^{\alpha}(\omega) = \int_{-\infty}^{\infty} dt \, e^{-i\omega t} F_q^{\alpha}(t) = 2 \int_0^{\infty} dt \cos\omega t \, F_q^{\alpha}(t) , \tag{83}$$

or, for the cases where $F_q^{\alpha}(t)$ goes to zero slowly, by Eq. (93) of Ref. 1[37]:

$$F_q^{\alpha}(\omega) = \frac{2 k_q^{\alpha'}(\omega)}{[k_q^{\alpha'}(\omega)]^2 + [\omega + k_q^{\alpha''}(\omega)]^2} . \tag{84}$$

Here

$$k_q^{\alpha}(\omega) = k_q^{\alpha'}(\omega) + i k_q^{\alpha''}(\omega) ,$$

where

$$\begin{aligned} k_q^{\alpha'}(\omega) &= \int_0^{\infty} dt \cos\omega t \, k_q^{\alpha}(t) , \\ k_q^{\alpha''}(\omega) &= -\int_0^{\infty} dt \sin\omega t \, k_q^{\alpha}(t) , \end{aligned} \tag{85}$$

and $k_q^{\alpha}(t)$ is computed either by (75) or (79).

B. Long-Time Long-Wavelength Solutions

Before presenting the results of the numerical solutions in detail, we look more closely at the time behavior of $F_q(t)$ for small q. We examined this question briefly in Sec. IV where we essentially made a small-time expansion of the equations at low temperatures in order to determine the characteristic time τ_c over which the order parameter varies significantly. We found the result that $\tau_c \sim K^2/J_{c1}$ {$\Rightarrow K^2/J[S(S+1)]^{1/2}$ for a real system} for the ferromagnet and $\tau_c \sim K/J_{c1}$ {$\Rightarrow K/J[S(S+1)]^{1/2}$} for the antiferromagnet. Now we ascertain the behavior of $F_q(t)$ in the limits $t \to \infty$ and $q \to 0$ (or $q^* = \pi - q \to 0$ for the antiferromagnet), using Eqs. (74) and (75), which are valid at all temperatures for long wavelengths.

We follow the arguments of BH. As $t \to \infty$ the memory function $k_q(t)$ goes to zero much faster than $F_q(t)$, as $k_q(t)$ is quadratic in the F functions. Thus, we can replace $F_q(t')$ in the time integral of (74) by $F_q(t)$ and remove it outside the integral. The solution of (74) as $t \to \infty$ is then

$$F_q(t) \cong e^{-\psi_q t} , \tag{86}$$

where

$$\psi_q = \frac{2}{3} \sum_{q'} K_{qq'} C_{q'} \int_0^{\infty} dt' \, F_{q'}(t') F_{q-q'}(t') . \tag{87}$$

We now expand ψ_q in a power series in q (or q^* for the antiferromagnet) and retain only the leading-order nonzero term. We consider the cases of ferromagnetic and antiferromagnetic coupling separately.

For the ferromagnet the leading-order nonzero term is of order q^2. A straightforward calculation leads to the diffusion-law result $\psi_q = Dq^2$, where the diffusion constant is given by

$$D=\frac{2}{3N}\sum_{q'} W_{q'}(K)\sin^2 q'\int_0^\infty dt'\,F_{q'}^2(t')\,, \tag{88}$$

with

$$W_{q'}(K)=\frac{(1+u)(1-u)^3}{(1+u^2-2u\cos q')^2}\,. \tag{89}$$

As $T\to\infty$, $u\to 0$ and (88) reduces simply to

$$D=\frac{1}{3N}\sum_{q'} \sin^2 q'\int_0^\infty dt'\,F_{q'}^2(t')\,, \tag{90}$$

which is the analog of Eq. (83) of Ref. 1. For this case all wave vectors q' contribute to the sum and we need the full time-dependent solutions of the $F_q(t)$. The result from the numerical solutions is $D=0.345$, or in terms of conventional units for a system of spin S the result is $D=0.69Ja^2\times[S(S+1)]^{1/2}$. We note that at high temperatures there is no distinction between the ferromagnet and antiferromagnet, and hence these results apply equally to the antiferromagnet.

At low temperatures $K\gg 1$ the function $W_{q'}(K)$ is peaked very sharply around $q'=0$ with a spread of order $1/K$. In this case only the long-wavelength fluctuations are thermally excited and contribute to the diffusion process. Making the approximations $u(K)\cong 1-1/K$, $\sin q'\cong q'$, and $\cos q'\cong 1-\frac{1}{2}q'^2$, we have

$$D\cong\frac{2}{3K^3}\int_{-\pi}^{\pi}\frac{dq'}{2\pi}\,\frac{q'^2}{[q'^2+(1/K^2)]^2}\int_0^\infty dt'\,F_{q'}^2(t')\,. \tag{91}$$

Further, since only the small wave vectors are important, we can also approximate $F_{q'}(t)\cong e^{-Dq'^2t}$. Carrying out the time integration we find an explicit expression for D:

$$D^2=\frac{1}{3K^3}\int_{-\pi}^{\pi}\frac{dq'}{2\pi}\,\frac{1}{[q'^2+(1/K^2)]^2}\,. \tag{92}$$

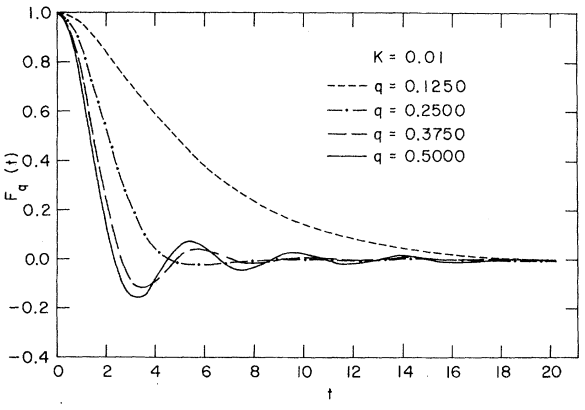

FIG. 1. $F_q(t)$ vs t for several values of q for $K=J_{c1}/T=0.01$ ($T=100\,J_{c1}$). In all figures the time t is measured in units of $2|J_{c1}|$.

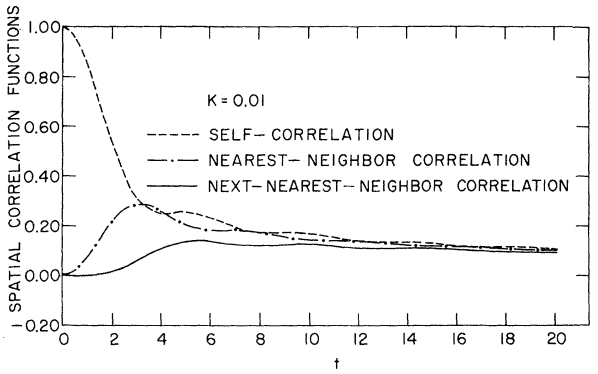

FIG. 2. Correlation functions $C_l(t)=\langle S_i(0)\cdot S_{i+l}(t)\rangle$ for the self-correlation function ($l=0$), the nearest-neighbor ($l=1$), and next-nearest-neighbor ($l=2$) correlation function. $T=100$.

The remaining integration over q' is easily accomplished and in the limit $K\to\infty$ we find the result $D=1/2\sqrt{3}=0.289$, or in conventional units, $D=0.578Ja^2\,[S(S+1)]^{1/2}$. We have the perhaps somewhat surprising result that the diffusion constant approaches a nonzero value at $T=0$. But note that the characteristic time that follows from the diffusion-law behavior is of the order $(Dq^2)^{-1}\approx K^2/J\times[S(S+1)]^{1/2}$, which is the same as obtained from the small-time expansion. Thus we conclude that the result for D is consistent and reasonable. Also, the region of diffusive behavior, $q\lesssim 1/K$, becomes very small in the limit of low temperature. The diffusive process governs the dynamics of only those fluctuations with wavelengths greater than the coherence distance.

We now consider the antiferromagnet at low temperatures. In this case we expand (87) in powers of $q^*=\pi-q$, and we find that the leading-order nonzero term is the first, or q^*-independent term, $\psi_{q^*}\cong\psi_0$. It is found simply by setting $q^*=0$ in (87),

$$\psi_0=\frac{2}{3}\sum_{q'} K_{\pi q'}\,C_{q'}\int_0^\infty dt'\,F_{q'}^2(t')\,. \tag{93}$$

On making the approximation $F_{q'}(t')\cong e^{-\psi_0 t'}$ we find the result $\psi_0=1/\sqrt{3}K$ in reduced units. Again, we find agreement with the small-time expansion as to the time scale of variation. Our result indicates that ordinary diffusive behavior—as obtained for the ferromagnet—does not apply to the antiferromagnet at low temperature. However, the phenomenon of critical slowing down at the zone boundary $q\sim\pi$ is clearly predicted by our result $F_{q=\pi}=e^{-(t/\sqrt{3}K)}$.

C. High-Temperature Solutions

In Figs. 1–3, we show the results of the numerical solutions of (74) for $T=100$ (in units of J), which is effectively the infinite-temperature limit. Figure 1 shows the time dependence of the functions

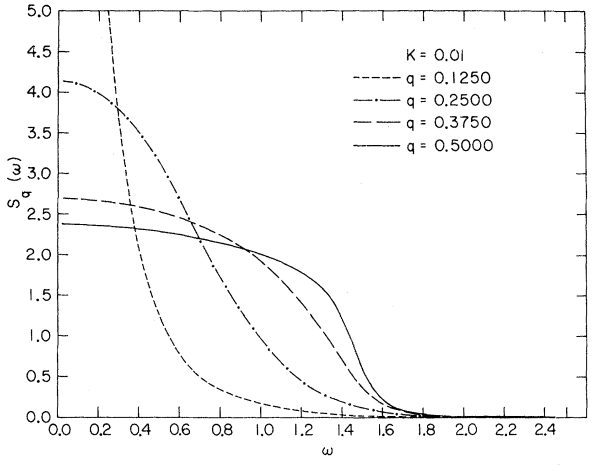

FIG. 3. $F_q(\omega)$ vs ω for several values of q at $T=100$.

$F_q(t)$ for several values of q. The characteristic behavior is a relatively slow decrease for small q, becoming more rapid for larger q, with highly damped oscillations setting in for $q > \frac{1}{2}\pi$. In Fig. 2 we plot the spatial-correlation functions—the self-correlations nearest-neighbor, and next-nearest-neighbor correlations—against time. We note the appearance of plateaus, especially noticeable in $C_0(t)$. Further, we note the slow decay for all three functions at long times. We attribute this to the slow decay of the long-wavelength diffusive modes. Indeed, converting (82) to an integral and substituting $C_q^{\alpha}(t) = (1/3N)\, e^{-Dq^2t}$ for long times—such that only the long-wavelength fluctuations remain—we have

$$C_l(t) \underset{t\to\infty}{\rightarrow} \int_{-\pi}^{\pi} \frac{dq}{2\pi}\, e^{-Dq^2t} \cos ql$$

$$= 2\int_0^{\infty} \frac{dq}{2\pi}\, e^{-Dq^2t} \cos ql$$

$$= \frac{1}{2(\pi Dt)^{1/2}}\, e^{-l^2/4Dt}\,. \tag{94}$$

In the limit $t\to\infty$ $C_l(t) \to 1/2(\pi Dt)^{1/2}$ for all values of l. Substituting $D = 0.345$ we find excellent agreement with the long time behavior of Fig. 2. The Fourier transforms of $F_q(t)$ are shown in Fig. 3. The point of note here is that $F_q(\omega)$ for the larger q values exhibits a broad flat profile with a relatively sharp shoulder at $\omega \sim 2J$.

As we lower the temperature nothing very striking occurs. For practical purposes the results are virtually the same down to $T \sim 2$. For $T \lesssim 1$ the oscillations in the $F_q(t)$ increase somewhat in frequency but they remain highly damped. There is a corresponding broadening of the Fourier transforms, but the broad flat profiles with the sharp shoulders persist. As we pointed out in Sec. III, even as T becomes very small this same pattern continues, and no real evidence of spin waves appears. This, of course, served as the impetus to introduce the idea of local order at low temperatures. For $T \sim 1$ differences do appear between the ferromagnet and the antiferromagnet, primarily the manifestation of "slowing down" in the antiferromagnet for wave vectors in the interval $\frac{1}{2}\pi < q < \pi$. The differences are quite evident in the spatial correlations $C_l(t)$, which we show plotted in Figs. 4 and 5 for the ferromagnet and antiferromagnet, respectively, at $T = 1$. As expected, the nearest-neighbor correlation is initially negative in the antiferromagnet. We note the continued hint of plateaus for the ferromagnet, whereas definite oscillatory behavior occurs in the $C_l(t)$ for the antiferromagnet.

We point out in passing that the high-temperature solutions—as well as the low-temperature solutions to be discussed below, agree quite well with the results of the computer simulation studies of Blume

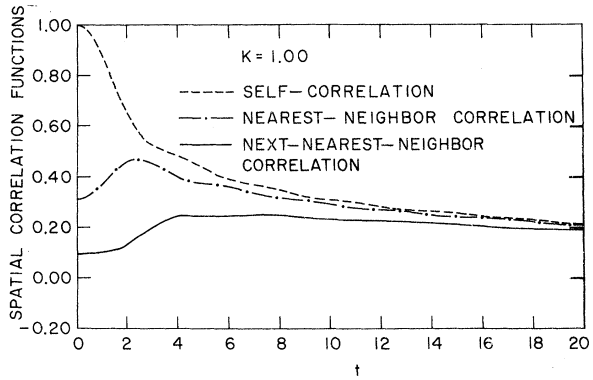

FIG. 4. $C_l(t)$ vs t ($l = 0, 1, 2$) for a ferromagnet at $T = 1.0$.

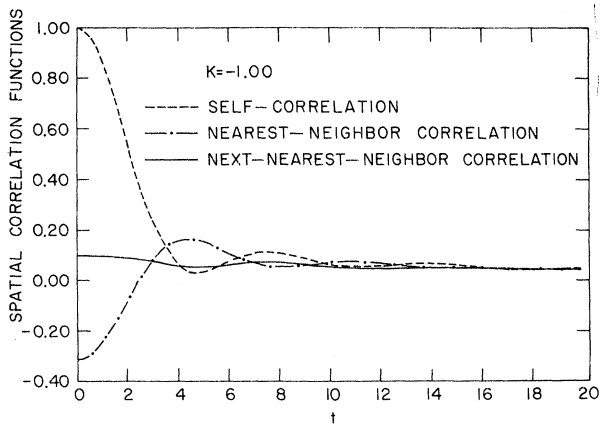

FIG. 5. $C_l(t)$ vs t ($l = 0, 1, 2$) for an antiferromagnet at $T = 1.0$.

et al.[17] Detailed comparisons will be presented in a future publication.

D. Time-Dependent Solutions–Spin-Wave Region

Here we present the time-dependent solutions $F_q^T(t)$ for the case of the classical chain at low temperatures. The envelope functions $F_q^T(t)$ of the spin-wave oscillations satisfies Eqs. (78) and (79), and the full relaxation functions $F_q^{\pm}(t)$ are then related to $F_q^T(t)$ via (77). In particular, $F_q^x(t) = F_q^y(t) = \langle S_{-q}^x S_q^x(t)\rangle / \langle S_{-q}^x S_q^x\rangle$ is related to $F_q^T(t)$ by

$$F_q^x(t) = \cos\omega_q t\, F_q^T(t) , \tag{95}$$

where ω_q, the first transverse moment is $\Delta \cong 1 - 1/2K$ in reduced units. In Fig. 6 we plot the spin-wave relaxation functions $F_q^x(t)$ for the wave vectors $Q = 0.25$ and $Q = 0.50$ for the ferromagnet at $T = 0.1$. This figure simply serves to illustrate that well-defined spin waves exist and that their lifetimes are sufficiently long to contain a number of oscillations.

Figures 7 and 8 show the envelope functions $F_q^T(t)$ for a series of Q values at two temperatures $T = 0.3$ and 0.1 for the ferromagnet and antiferromagnet, respectively. These figures depict both the q dependence of the fluctuations at fixed T as well as the variations with temperature. If we define the spin-wave lifetimes as $\tau_q = \Gamma_q^{-1}$, where Γ_q is given by (63) [so that $\tau_q = \sqrt{2}K/(1-\cos q)^{1/2}$ in reduced units], then we find that in general $F_q^T(t) \approx 0.6$ at $t = \tau_q$ and at $t \approx 2\tau_q$, $F_q^T(t)$ is reduced to approximately 0.1. We find $F_q^T(t)$ generally goes negative in the vicinity of $t \sim 2.5\tau_q$ and that the behavior for longer times is one of a slowly oscillating small-amplitude damped motion. However, for long times, say for $t \gtrsim 2\tau_q$, we do not expect our equations to be valid, since variations in the local order become appreciable for $t > \tau_c$, where τ_c is given by (67). Hence, the long time behavior of $F_q^T(t)$ is very likely meaningless. We note that the

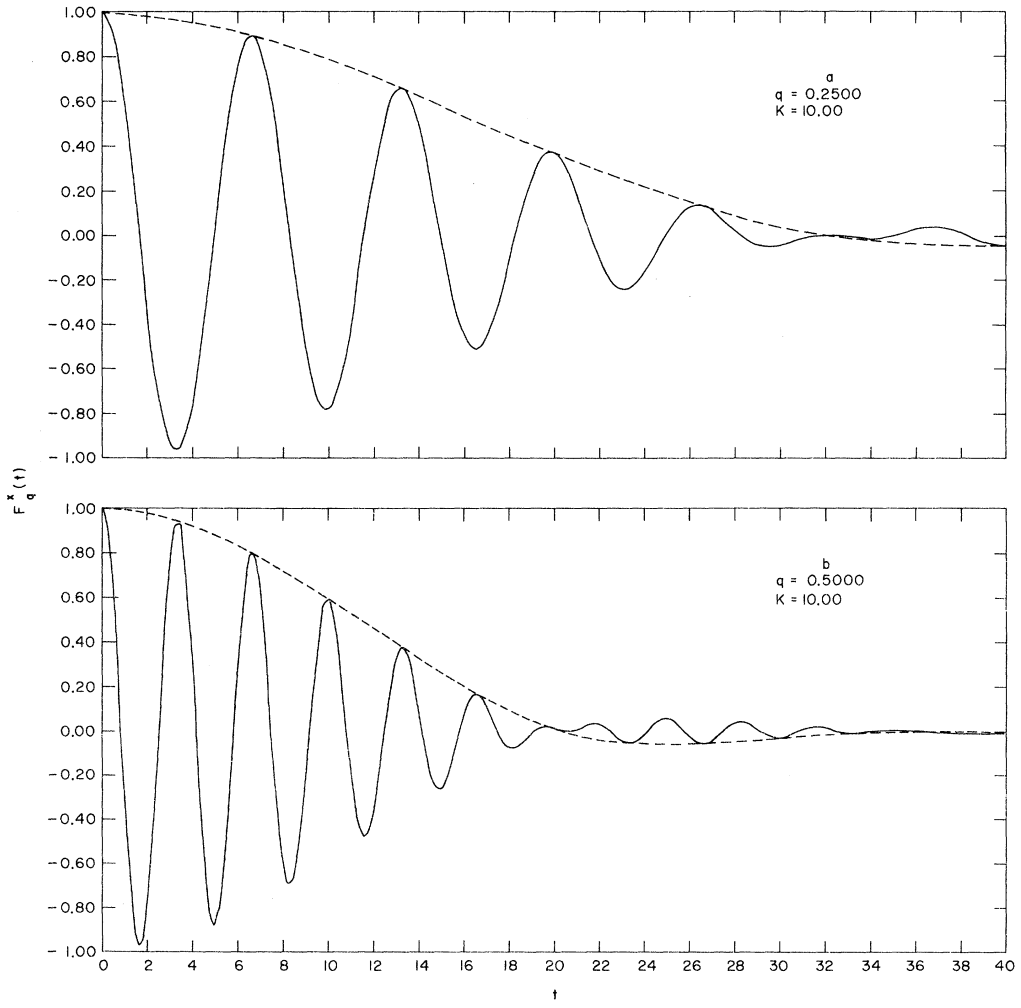

FIG. 6. $F_q^x(t)$ and its envelope vs t for a ferromagnet at $T = 0.1$: (a) $q = 0.25$; (b) $q = 0.5$.

spin-wave oscillations are quite heavily damped at the higher temperature $T = 0.3$; we do not expect that our simplified theory of the spin-wave region can be pushed any higher in temperature.

E. Neutron Scattering: Application to TMMC

1. Generalized Structure Factor at Low T

For comparison with experimental inelastic-neutron scattering data, the quantity of interest is the generalized structure factor $S_q^\alpha(\omega)$. For classical systems this quantity is simply $S_q^\alpha(\omega) = C_q^\alpha F_q^\alpha(\omega)$. But for real quantum systems of finite spin the fluctuation-dissipation theorem plays an important role at low temperatures. It may be useful to briefly recapitulate the situation here. For isotropic-Heisenberg systems $S_q^\alpha(\omega)$ is given by Eq. (1), where for linear-magnetic chains the directions x, y, z are all equivalent for macroscopic samples as true long-range order is absent. In our simplified theory developed in Sec. IV, we calculate the spin-wave contribution to the scattering. This is done by calculating the transverse fluctuations taken with respect to the direction of the local magnetization. The results are macroscopic averages as the spin-wave contribution is in effect an average over all directions. However, our theory neglects the longitudinal fluctuations of short wavelength and therefore we cannot make any predictions about any longitudinal—or central—component that may be present in the scattering. The factor of detailed balance $\beta\omega(1-e^{-\beta\omega})^{-1}$ serves to repress the magnon absorption peak and to enhance the magnon emission peak. In fact, if $S_q^-(\omega)$ and $S_q^+(\omega)$ are the structure factors corresponding to magnon absorption and emission, respectively, we have

$$\frac{S_q^-(-\omega)}{S_q^+(\omega)} = e^{-\beta\omega} \quad . \tag{96}$$

For the spin wave region, $\beta\omega \gg 1$, and this ratio is much less than unity. We will present the results for the emission line $S_q^+(\omega)$, then the absorp-

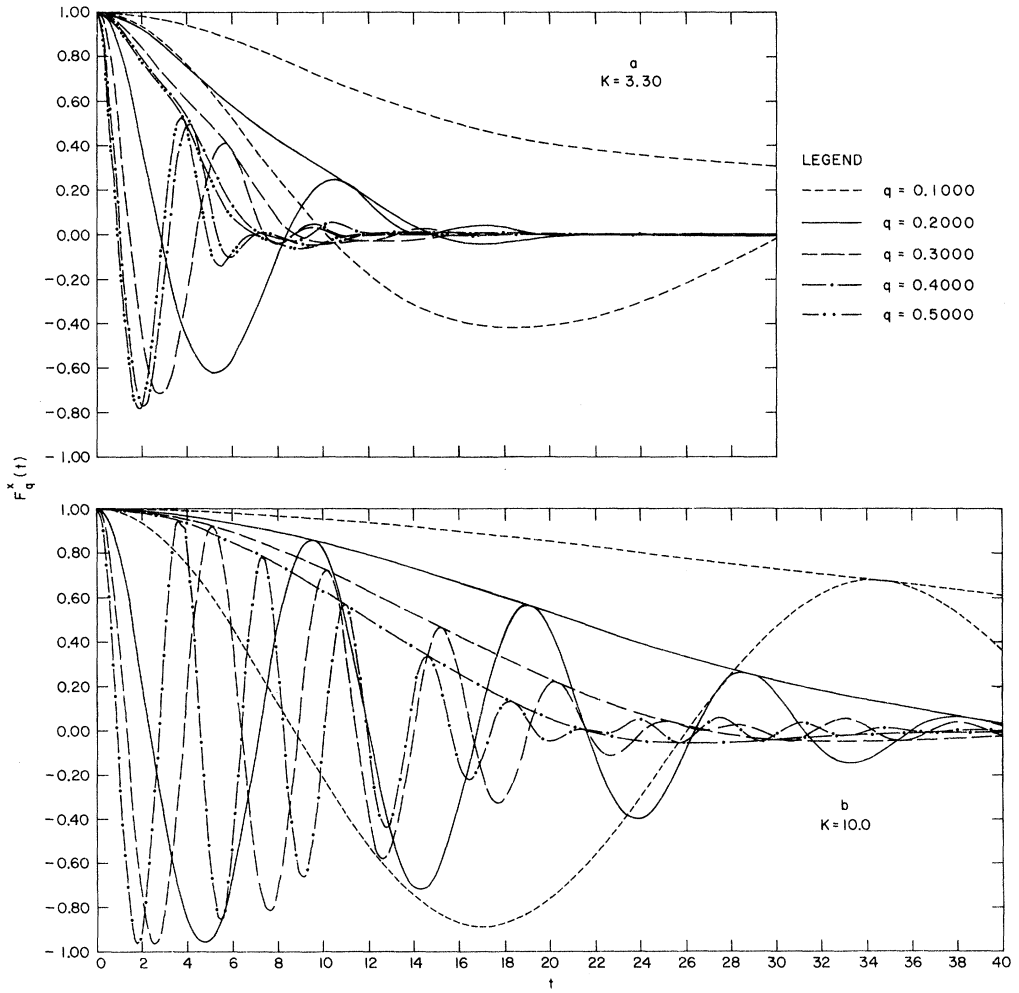

FIG. 7. $F_q^x(t)$ vs t for a ferromagnet at various values of q: (a) $T = 0.3$; (b) $T = 0.1$.

tion peak is found simply by applying the ratio (96). (But experimentally the absorption peak is not seen as it is down in the noise.)

We rewrite Eq. (1) for spin-wave emission in the following way:

$$S_q^+(\omega) = \frac{x_q^T}{\beta} R_q^+(\omega) , \tag{97}$$

where x_q^T is the transverse-static susceptibility with respect to the local magnetization and all the frequency dependence of the scattering is contained in $R_q^+(\omega)$ defined by

$$R_q^+(\omega) = \frac{\beta\omega}{1 - e^{-\beta\omega}} \operatorname{Re}\{F_q^+(\omega)\} . \tag{98}$$

Thus, $R_q^+(\omega)$ contains all the essential information concerning the line shape (peak positions, linewidths, etc.), i.e., it described the contribution to the structure factor originating from the dynamics. On the other hand, the static (but q-dependent) factor x_q^T/β can be regarded as an over-all scaling factor for the intensities of neutrons scattered with momentum loss q. It is determined solely by the equal-time correlations of the system.

The main thrust of this paper has been to calculate the spin dynamics of linear chains, i.e., to calculate $F_q^\alpha(\omega)$. Indeed, we did make use of the static correlations as initial conditions for the kinetic equations. However, at low temperature we argued that only the long-wavelength ($q \lesssim 1/K$) correlations play a significant role, and for these correlations the classical approximation to the fluctuation-dissipation theorem is valid, $C_q^\alpha(t) \cong (x_q^\alpha/\beta) F_q^\alpha(t) = C_q^\alpha F_q^\alpha(t)$. Then we used the exact calculations of C_q for the classical chain as input to the spin dynamics. These approximations are in essence verified by the experimental results of Birgeneau *et al.*[9] on the static correlations in TMMC, where they found the static susceptibility to be well described by Fisher's equal-time cor-

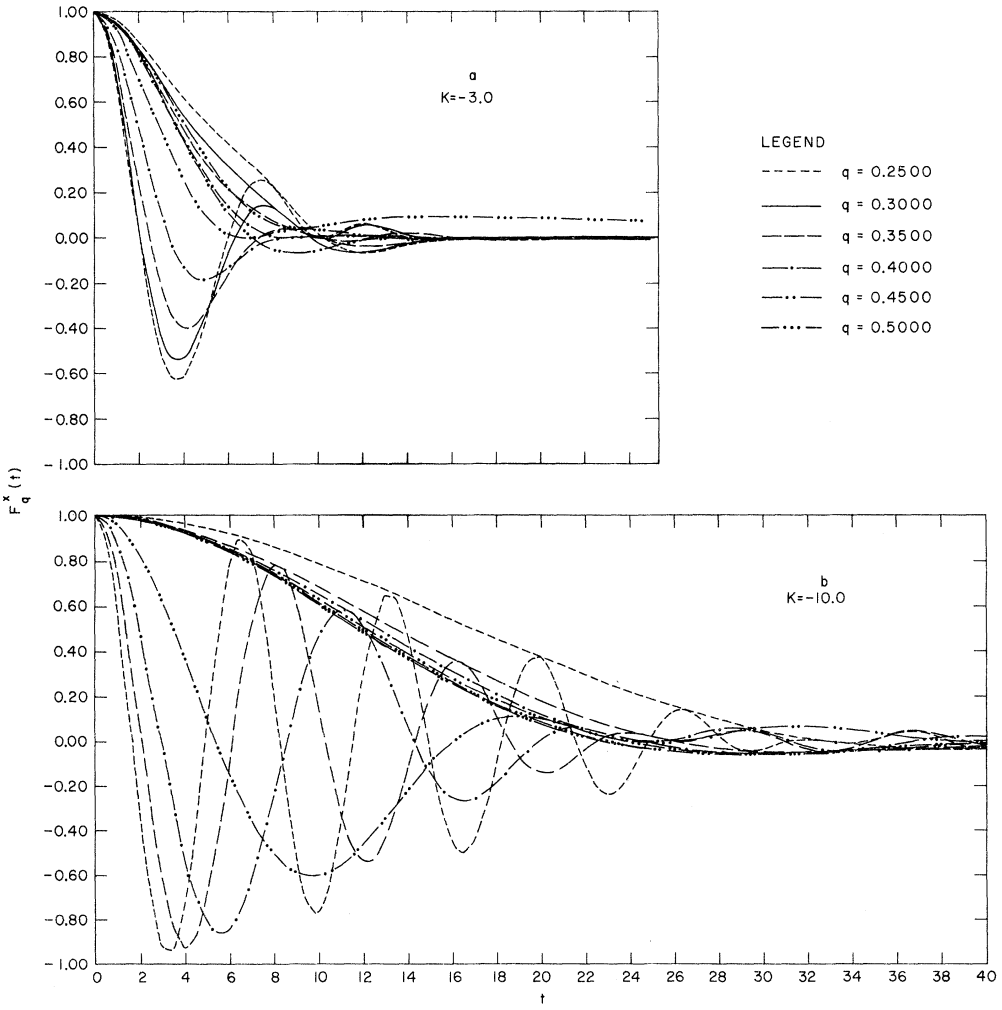

FIG. 8. $F_q^x(t)$ vs t for an antiferromagnet: (a) $T = 0.3$; (b) $T = 0.1$.

relation function for $q \lesssim 3/K$.

On the other hand, in the spin-wave region $q \gg 1/K$ we do not expect the classical approximation to x_q^T to be valid. In fact, for systems of finite spin the frequencies of interest in the spin-wave region are such that $\frac{1}{2}\beta\omega \gg 1$. Then, from our general discussion of the fluctuation-dissipation theorem in Sec. II [see Eq. (36)], we have that x_q^T is related by C_q^T by

$$\frac{x_q^T}{\beta} = \frac{C_q^T}{\langle \frac{1}{2}\beta\omega \coth \frac{1}{2}\beta\omega \rangle_T} , \tag{99}$$

where the averaging brackets refer to the frequency moments of $F_q^{\pm}(t)$ as defined by (35). [Note that only even order moments appear, so that (99) is true for both magnon emission and absorption.] But we are in no better shape with (99) as the unknown static correlation C_q^T still appears.

Summarizing, the dynamics of the long-wavelength fluctuations (i.e., of the local-order parameter) is classical in nature, but the same need not be true for spin waves, the quantum-mechanical aspects being manifested, however, only in the static intensity factor in (97). We are therefore at an apparent impasse if we attempt to predict the intensities of the neutron-scattering lines for finite spin systems unless the C_q are known for all q. However, we find that the simple classical approximation[38] $x_q^T/\beta \cong C_q^T \cong \frac{1}{2}C_q$, where we use the classical result[39] for C_q with $K = JS(S+1)/T$, provides remarkably good agreement with the experimental intensities, except at very low temperatures $K \lesssim 30$, where there are also additional difficulties in making comparisons with experiment (see Sec. V E. 2). This is a rather surprising result. It seems to imply that the classical correlation function is a good approximation in TMMC to the static susceptibility x_q^T/β, but *not* to the correlation function C_q^T in (99).

2. *Comparison with Experiment*

We make a direct comparison of our theoretical calculations with the experimental data of Hutchings *et al.*[10] on TMMC. Thus, we take $S = \frac{5}{2}$ and use the value of J they[40] found from the spin-wave dispersion, namely, $J = 14.1$ °K. Consequently, our reference temperature is $JS(S+1) = 124$ °K. We make comparisons at the K values 3, 6, 10, and 28, corresponding to the temperatures 41, 21, 12, and 4.4 °K, respectively. We recall that for finite spin time is measured in our calculations in units of $2J[S(S+1)]^{1/2}$ and frequency in units of $2J[S(S+1)]^{-1/2}$, the factor $\beta\omega$ in (98) becomes $2K\omega/[S(S+1)]^{1/2}$, where ω is measured in the reduced units.[41]

One difficulty in making a detailed comparison of theory with experiment is that the experimental results do not give directly the scattering function $S_q^{\alpha}(\omega)$. Rather, the observed intensity at momentum transfer q and energy transfer ω is given by the convolution of the true scattering function with the instrumental resolution function.[10] For the lowest temperature at which experimental measurements were made, $T = 1.9$ °K corresponding to $K = 65$, the spin-wave linewidths are quite obviously limited by the resolution function, the intrinsic linewidths evidently being smaller. The theoretical calculations at this temperature do in fact produce very sharp spin-wave peaks with widths of the order $1/K$ in reduced units. The theoretical linewidths are considerably smaller than the observed linewidths. At 4.4 °K, however, the intrinsic linewidths have broadened to the point that they are becoming manifested in the experimental results, and at higher temperatures the observed linewidths are probably very close to the intrinsic linewidths. Thus, we compare the calculated line shapes with experiment for temperatures $T \gtrsim 4.4$ °K, keeping in mind that the instrumental resolution function may still be fairly important at 4.4 °K but much less so for $T = 12$ °K and higher.

In Fig. 9 we reproduce the experimental results for the scattering intensity for several wave vectors q at 4.4 °K and in Fig. 10 we give the corre-

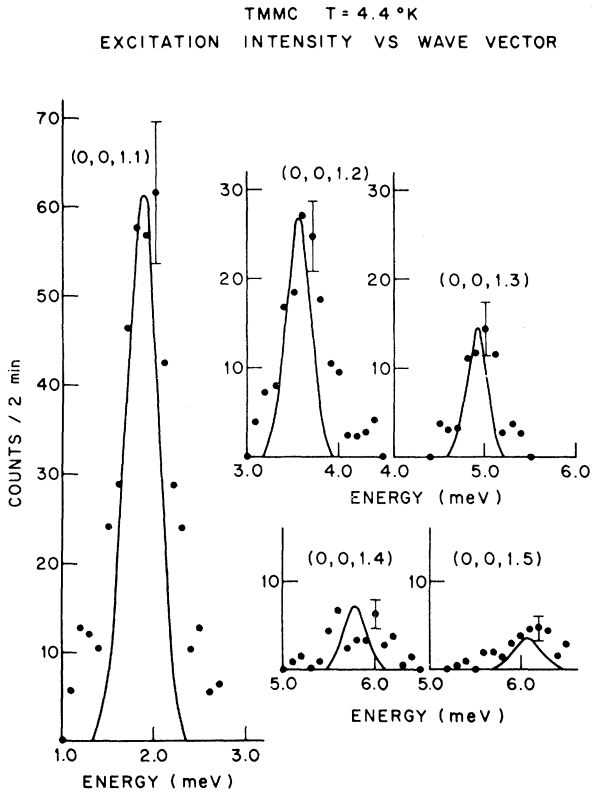

FIG. 9. $S_q^{\text{expt}}(\omega)$ vs ω at $T = 4.4$ °K for TMMC (from Fig. 8 of Ref. 10).

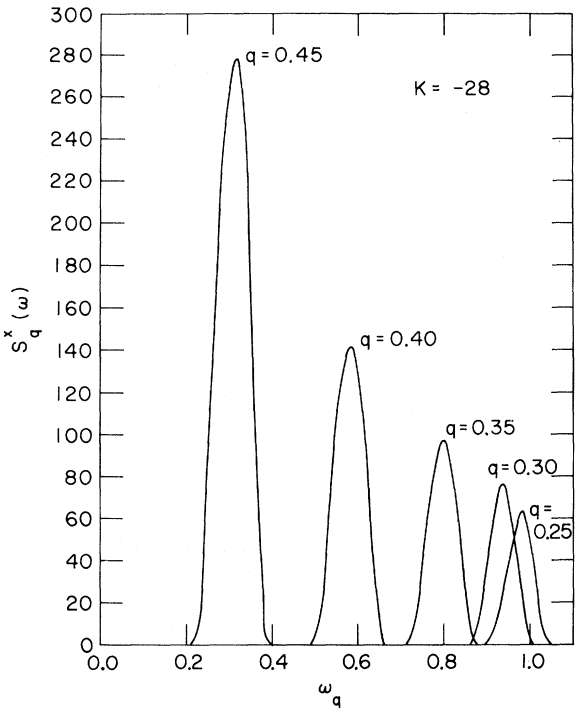

FIG. 10. $S^x_q(\omega)$ vs ω at 4.4 °K (theoretical).

sponding results of the theory. The curves are labeled in terms of the reduced wave vector $Q^* = (\pi - q)2\pi$ (so that $Q^* = 0.25$ corresponds to the antiferromagnetic zone boundary). The energy coordinates for the experimental plots are in meV units, whereas the theoretical curves employ the reduced frequency units. As is apparent, there is good qualitative agreement between theory and experiment. We note that even at this temperature a small asymmetry is discernible in both the theoretical and experimental line shapes, the intensity being slightly larger on the low-frequency side of the peaks.

To compare the line shapes on a quantitative basis, we determine σ_q, which we define[39] here as the ratio of the linewidth at half maximum to the peak energy, for both the experimental and theoretical curves. In Table I the values of σ_q are listed for several values of Q^*. We consider the agreement between the experimental and theoretical σ_q values to be quite good; even with the uncertainty associated with σ_q^{expt} for the larger values of Q^*, it is clear that the theory gives a good representation of the line shapes for all wave vectors. We point out that the theory predicts a slight decrease in the actual linewidth Γ_{q^*} for increasing Q^*. The variation occurs primarily for the larger Q^* values ($Q^* = 0.20$ and 0.25) and is of the order of 30%. On the other hand, the experimental linewidths are approximately equal for all the Q^* values shown in Fig. 9. While it is apparent that there is a good deal of uncertainty associated with the experimental data for the larger values of Q^*, it is in fact very likely that at $T = 4.4$ °K any decrease in the linewidth may not show up due to the limitations imposed by the instrumental-resolution function. This would be particularly true if the line-widths for smaller Q^* are just at the verge of the resolution width.

Also listed in Table I are the peak intensities I_q, where for the sake of comparison we have normalized both the experimental and theoretical intensities to 100 at $Q^* = 0.05$. There is a good match between I_q^{expt} and I_q^{th} for $Q^* = 0.10$ and 0.15, but a significant discrepancy exists for the larger values $Q^* = 0.20$ and 0.25. This discrepancy may be due, at least in part, to the limitation of the instrumental resolution. Indeed, if the real line shapes are sharper than can be resolved, then the resolution function serves to broaden the lines as well as to smooth the peaks, and hence reduce the peak intensities. However, we cannot dismiss the possibility that the discrepancy may be due in part to quantum-mechanical effects in the static correlations for large q^*.

For completeness we plot in Fig. 11 the dispersion curve obtained from the theory at $T = 4.4$ °K. The agreement with the experimental dispersion curve is essentially exact. That there is agreement is not really surprising in view of the manner in which we formulated the theory and since we used the experimental value of the exchange constant to set the temperature scale. But that the agreement is exact would seem to confirm the correctness of our approach to calculating spin-wave properties of linear chains.

Next, we consider the temperature dependence of the scattering. In Fig. 12 we show the temperature variation of the $Q^* = 0.05$ line, part (a) of the figure reproducing the experimental results for the temperatures 4.4, 12, 20, and 40 °K, and part (b) showing the corresponding theoretical curves. Figure 13 shows a similar comparison for the line $Q^* = 0.125$. Again, it is clear that the theory pro-

TABLE I. Linewidth-to-peak energy ratios and peak intensities at $T = 4.4$ °K.

Q^* [a]	σ_q^{expt}	σ_q^{th}	I_q^{expt}	I_q^{th}
0.05	0.30	0.30	100	100
0.10	0.14	0.16	44	39
0.15	0.10	0.12	23	27
0.20*	0.1	0.085	11	21
0.25*	0.1	0.065	7	18

[a]The scatter in the experimental data for these values of Q^* enable one only to make a rough estimate of σ_q^{expt} and I_q^{expt}.

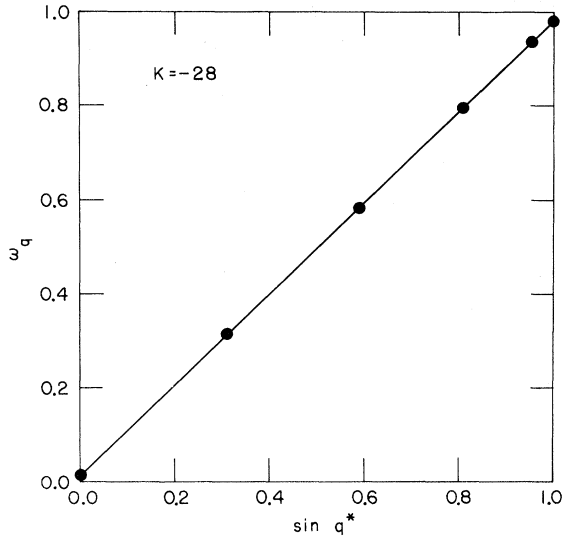

FIG. 11. Dispersion curve ω_q vs sin q at $T=4.4$ °K (theoretical). The points are determined from the peaks in the theoretical calculations of $F_q(\omega)$.

vides a good description of the temperature dependence of the scattering, both in the broadening of the lines as well as the increasing asymmetry of the line shapes with increasing temperature. As before, to make a quantitative comparison between theory and experiment we list in Table II the σ_q values for temperatures 4.4, 12, and 20 °K. (It is obvious that it is meaningless to ascribe a value to σ_q at $T=40$ °K for either experimental or theoretical curves.) There is excellent agreement between σ_q^{th} and σ_q^{expt} for the $Q^*=0.125$ line. We note the very nearly linear increase in $\sigma_{0.125}$ with temperature in accordance with the theoretical prediction [Eq. (64)]. For the $Q^*=0.05$ line there is (exact) agreement at $T=4.4$ °K, but a sizeable discrepancy develops at the higher temperatures, with the theory predicting larger damping of the magnons on the order of 30% at these temperatures. This is not surprising, however, as our physical model of the spin-wave region is strictly valid only for wave vectors $q^* \gg 1/K$, and hence the theory is expected to begin breaking down first for the smaller values of q^* as the temperature is raised. As can be seen from Table II, $\sigma_{0.05}$ is

TABLE II. Temperature dependence of the linewidths for $Q^*=0.05$ and 0.125.

	$Q^*=0.05$		$Q^*=0.125$	
T	σ^{expt}	σ^{th}	σ^{expt}	σ^{th}
4.4	0.30	0.30	0.12	0.13
12	0.53	0.74	0.28	0.32
20	1.0	1.22	0.54	0.53

TABLE III. Temperature dependence of the peak intensities normalized to their values at $T=12$ °K.

	$Q^*=0.05$		$Q^*=0.125$	
T	I^{expt}	I^{th}	I^{expt}	I^{th}
4.4	140	253	147	282
12	100	100	100	100
20[a]	65	65	60	62.5
40[a]	30	28	37	33

[a]Noise levels subtracted from experimental intensities (10 for $Q^*=0.05$ and 25 for $Q^*=0.125$).

becoming of order unity for $T>12$ °K and the spin-wave mode is no longer a well-defined excitation for this wave vector. We cannot say, though, why the observed broadening is less than the theoretical prediction.

Table III shows the temperature variation of the peak intensities, where we have normalized I_q^{expt} and I_q^{th} to 100 at $T=12$ °K. This particular normalization was chosen because, as apparent from the Table III, there is quite close agreement between the experimental and theoretical intensities for the temperature range $T \gtrsim 12$ °K. But there is a large discrepancy between I_q^{expt} and I_q^{th} at 4.4 °K for both wave vectors. Again, if the resolution of the measuring instrument is sufficiently important at 4.4 °K, the experimental peak intensities may be chopped to the extent of the discrepancy in Table III. Also, it may be that quantum effects are playing an important role in the statics at low temperatures, whereas classical statis are adequate at the higher temperatures. The really surprising result is that the simple classical approximation $x_q^T/\beta=\frac{1}{2}C_q$ works so well.

Finally, we consider the renormalization of the peak energies with temperature. In Table IV we list the positions $\omega_q(T)$ of the peaks for the spectral lines shown in Figs. 12 and 13. The $\omega_q(T)$ are normalized to their positions at 1.9 °K. (At 40 °K we take the shoulders of the spectral lines to be the peak position.) For the $Q^*=0.125$ line, the theory is essentially in exact agreement with the experimental values for all temperature values. For $Q^*=0.05$ we have the same pattern as

TABLE IV. Renormalization of peak frequencies with temperature normalized to their values at $T=1.9$ °K.

	$Q^*=0.05$		$Q^*=0.125$		
T	ω^{expt}	ω^{th}	ω^{expt}	ω^{th}	ω^{el}
1.9	1.0	1.0	1.0	1.0	1.0
4.4	1.02	1.02	0.98	0.99	0.98
12	1.0	1.09	0.99	0.99	0.95
20	1.1	1.24	0.96	0.97	0.92
40	1.4	1.6	0.94	0.93	0.84

for the spin wave damping, while there is agreement at low temperature, a sizeable discrepancy exists for higher temperature. Again, this is not unexpected as discussed above. Further, the scattering profiles are so broad for $T \gtrsim 20$ °K that the placement of the peak positions is ambiguous.

At first glance there is one surprising point concerning the peak renormalizations. That is that the peak frequency actually renormalizes upward with increasing temperature for $Q^* = 0.05$, both experimentally and theoretically. For $Q^* = 0.125$ there is essentially no renormalization with T until $T \sim 20$ °K after which ω_q definitely decreases. These results may be surprising in view of our general discussion of the order parameter in Sec. IV in which we argued that the spin-wave energies should renormalize down with T according to $\Delta \cong 1 - 1/2K = 1 - T/2JS(S+1)$. But this is how the *first moment of* $F_q^{\pm}(\omega)$ renormalizes according to our arguments, and *not* how the peak position of the generalized structure factor $S_q^+(\omega)$ should renormalize. In fact, the frequency dependence of $S_q^+(\omega)$ involves the product of $F_q^+(\omega)$ with the factor of detailed balance [see Eq. (98)]. It is this factor, $\beta\omega(1 - e^{-\beta\omega})^{-1}$, which accounts for the renormalization described by Table IV. In the last column of this table we give the renormalization of ω_q for $F_q^+(\omega)$ that results from our choice of Δ; this would also be the renormalization of the scattering peaks for a classical system where the detailed balance factor is unity.

The reason that the actual renormalization of the scattering peak, where the detailed balance factor is included, is significantly less—or of opposite sign—than for ω_q^{cl} is not hard to determine. For $\beta\omega \gg 1$, $S_q^+(\omega)$ becomes

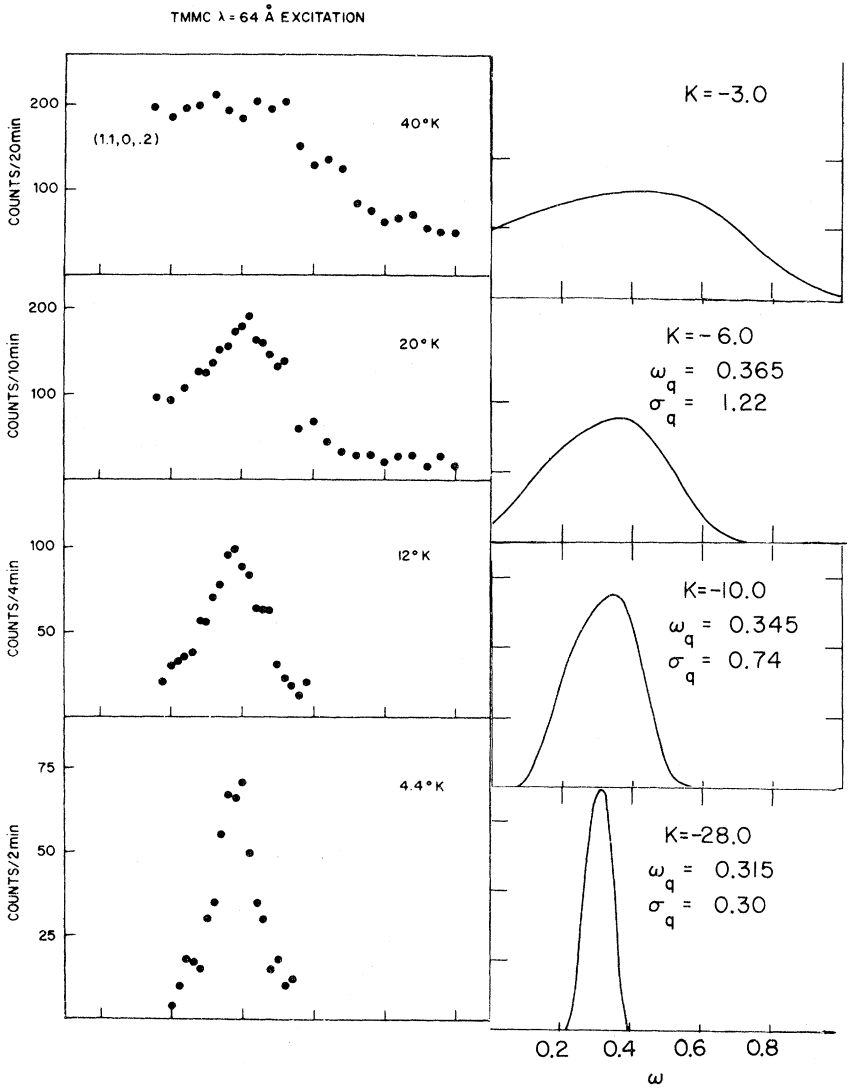

FIG. 12. Temperature dependence of the $q = 0.04$ line; experimental results from Ref. 10 on the left-hand side; theoretical calculations on the right-hand side.

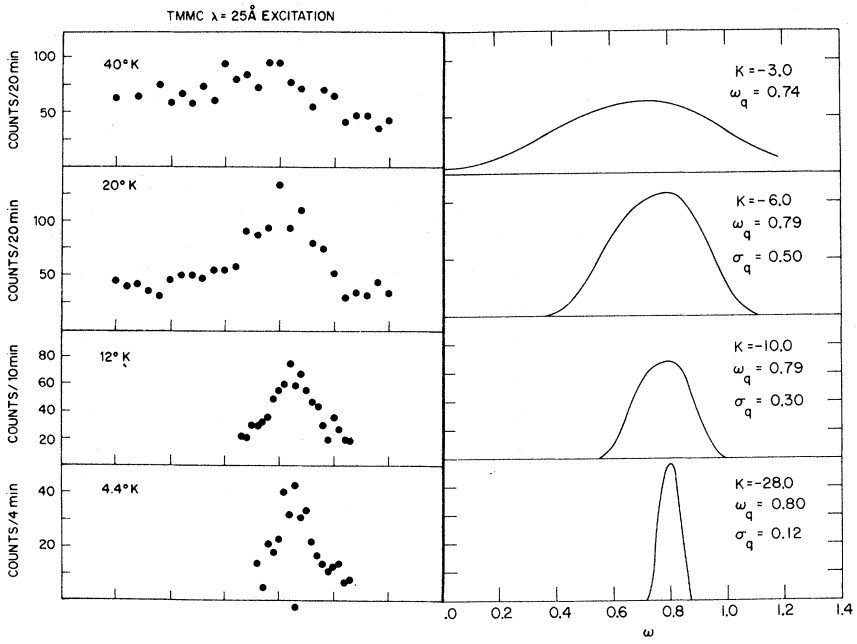

FIG. 13. Temperature dependence of the $q = 0.125$ line; experimental results on the left-hand side, theoretical results on the right-hand side.

$$S_q^+(\omega) \cong \frac{\chi_q^T}{\beta} \beta\omega F_q^+(\omega) , \tag{100}$$

so we have a situation where the observed peak is given by the maximum of a function $xf(x)$, where $f(x)$ has a characteristic resonance maximum at $x = x_0$ and goes to zero rapidly for $|x - x_0| > \delta x$. It is not hard[42] to see that the peak of $xf(x)$ occurs at $x > x_0$. We conclude that the almost precise agreement between the renormalization of ω_q^{expt} and ω_q^{th} for the larger wave vector $Q^* = 0.125$ effectively confirms our choice of the order parameter as being the correct one physically.

3. *Discussion*

We have seen that the frequency dependence predicted by the theory compares very favorably with experiment, both as to the q dependence and the temperature dependence. The only real discrepancy that we found is in the behavior of the small q^* line ($Q^* = 0.05$) as the temperature is raised. And this discrepancy itself serves to confirm our criterion for the existence of propagating spin-wave modes, namely, that $q^* \gg 1/K$. In fact, we may use the comparisons of theory with experiment to sharpen our criterion for well-defined spin-wave behavior. From the comparisons of Tables II and IV we see that an appreciable discrepancy develops for the $Q^* = 0.05$ line between theory and experiment for $T \gtrsim 12$ °K (or $K \lesssim 10$). Since $q^* = 0.31$ we are led to the conclusion that our theory of one-dimensional spin waves is valid to fairly good precision for wave vectors $q \gtrsim 3/K$, and that it is only qualitatively valid for $1/K < q^* < 3/K$. For $q^* < 1/K$ it is meaningless to speak of spin waves.

Along with these remarks it is understandable that the experimental results did not indicate the existence of two-magnon scattering in TMMC. As pointed out in Ref. 10, two-magnon scattering, if significant, would manifest itself in one dimension as a logarithmic singularity in the longitudinal-response function $S_q^z(\omega)$ at the single spin-wave position. This singularity occurs if one assumes the existence of spin waves for all wave vectors, because it becomes infinitely easy to create pairs of magnons, one at $[q^*, \omega(q^*)]$ and the other at $[q^* \to 0, \omega(q^*) \to 0]$. But as we have again just emphasized, spin waves with wave vectors $q^* \to 0$ and corresponding energies $\omega(q^*) \to 0$ do not exist for the linear chain, and hence the singularity in $S_q^z(\omega)$ does not exist. The only possible magnon-magnon scattering processes are those in which the wave vectors of both magnons are greater than $1/K$. Since these fluctuations are not thermally excited, the resulting intensity is expected to be extremely weak. The spin-wave modes are of course coupled to the long wavelength longitudinal fluctuations of the system, i.e., those associated with the variations of the order parameter. This coupling is precisely what gives rise to the damping of the magnons, and is quite well described by our theory.

Finally, the classical approximation to the transverse susceptibility $\chi_q^T/\beta \cong \frac{1}{2} C_q$ works surprisingly well. It certainly yields the correct temperature dependence of the peak intensities for $T \gtrsim 12$ °K. To be sure, at lower temperatures there are some significant discrepancies between experi-

ment and theory. We have argued that the discrepancies may be traced to the limitations imposed on the experimental measurements by the instrumental resolution function. But we cannot rule out the possibility that quantum-mechanical effects may be important in the static correlations at low temperatures for the shorter wavelengths $q^* \gg 1/K$.

In summary, our simplified theory of the spin dynamics of linear chains provides quite satisfactory agreement with experiment. Once the notion of a local-order parameter was introduced and one realized that the kinetic equations should be solved in a local sense for the spin-wave excitations, the essential feature of the treatment is the approximation (58)—$C^L_{q*} \cong C_q$ and $C^T_{q*} \cong 0$—for the long-wavelength static correlations appearing in the kinetic equations. This simple approximation led to the analytic expressions for magnon damping presented in Sec. IV and, as we have just seen, provides an adequate description of the experimental results for spin waves of wave vector $q^* \gtrsim 3/K$. The adequacy of this approximation implies that to a good approximation the thermodynamically important fluctuations at low temperature are indeed those associated with variations in the local order and that these fluctuations are predominantly responsible for the damping of the shorter-wavelength magnon modes. It is possible that the present treatment may be extended within the same theoretical framework by improving upon the approximation (58), in particular by including q dependence of the partitioning of C_q into longitudinal and transverse components. This might be necessary in order to quantitatively describe fluctuations of intermediate wave vectors $1/K < q^* < 3/K$, particularly at higher temperature where this wave-vector region covers an appreciable portion of the zone.

ACKNOWLEDGMENTS

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APPENDIX A: EXTENSION OF THEORY TO ORDERED ANTIFERROMAGNET SYSTEMS

We briefly sketch here how the theory developed in Sec. II may be modified in order to treat the antiferromagnet below the ordering temperature, in which case magnetic order sets in with the characteristic wave vector $Q = \pi$. The principal modification is that the quantity

$$V(t) = \langle \{ \exp_+ [i \int_0^t dt' \, h(t')] \}_S \rangle \tag{A1}$$

is no longer diagonal in q space as in the case of the ferromagnet. In addition to the diagonal elements $F^\alpha_q(t) = V^{\alpha\alpha}_{qq}(t)$, we must also consider the off-diagonal elements $G^\alpha_q(t) = V^{\alpha\alpha}_{q\pm\pi,q}(t)$ [and $G^\alpha_{q+\pi}(t) = V^{\alpha\alpha}_{q,q+\pi}(t)$]. In what follows we are mainly interested in the classical antiferromagnet, though we let the magnitude of S be arbitrary for the sake of generality.

First, consider the limit $T = 0$, where the ground state is precisely the Neél state. This case will give us insight into the problem as well as provide us with a point of departure for the case of finite temperature. By direct expansion of (A1) and taking matrix elements term by term, where we take $\langle S^z_q \rangle = S\delta_{q,\pi}$ in the Neél state and we use Eq. (15) of the main text for the matrix elements of $h_{qq'}$, we find for the diagonal elements

$$F^z_q(t) = 1 \,, \quad F^\pm_q(t) = \cos\omega_q t \,, \tag{A2}$$

where $\omega_q = 2JS|\sin q|$. The nonzero off-diagonal matrix elements in this limit are

$$\begin{aligned} G^z_q(t) &= 0 \,, \\ G^\pm_q(t) &= \pm i \left(\frac{1+\cos q}{|\sin q|} \right) \sin\omega_q t \,. \end{aligned} \tag{A3}$$

Clearly, all the dynamic information of the system is contained in the set of functions $F^z_q(t)$, $F^+_q(t)$, and $G^+_q(t)$, so we can restrict our study to this set. Now we form the transverse linear combinations $f^\pm_q(t)$, of the functions $F^+_q(t)$ and $G^+_q(t)$ such that

$$\dot{f}^\pm_q(t) = \pm i\omega_q f^\pm_q(t) \,. \tag{A4}$$

Then the $f^\pm_q(t)$ represent the elementary excitations of the system, i.e., antiferromagnetic magnons. From (A2) and (A3) it is clear that the required combinations are

$$f^\pm_q(t) = F^+_q(t) \pm \frac{|\sin q|}{1+\cos q} \, G^+_q(t) \,; \tag{A5}$$

for them we have simply $f^\pm_q(t) = e^{\pm i\omega_q t}$. This is essentially the Holstein–Primakoff transformation. We note that it is but a simple matter to go a step further and include the presence of an external magnetic field of Zeeman frequency ω_0 and an anisotropy field ω_A of wave vector $Q = \pi$. We can then make solid contact with the usual $T = 0$ spin-wave theory of antiferromagnets. The diagonalizing transformation is

$$f^\pm_q(t) = F^+_q(t) \pm \frac{[(1+\omega_A)^2 - \cos^2 q]^{1/2}}{1+\cos q+\omega_A} \, G^+_q(t) \tag{A6}$$

and the eigenfrequencies are

$$\tilde{\omega}^\pm_q = \omega_0 \pm [(1+\omega_A)^2 - \cos^2 q]^{1/2} \,, \tag{A7}$$

where the frequencies are measured here in units of $(2JS)^{-1}$. In the limits $\omega_0 \to 0$ and $\omega_A \to 0$, (A6) reduces to (A5) and $\omega_q^{\pm} \to \pm\omega_q$.

We now make the extension to finite temperature (for simplicity, without external and anisotropy fields). First we derive equations of motion for the functions $F_q^z(t)$, $F_q^+(t)$, and $G_q^+(t)$ in a manner similar to the development in Sec. II. In particular, we take $\langle S_q^z \rangle = S\Delta\delta_{q,\pi}$, where Δ is the (local) antiferromagnetic ordering parameter; we make the cumulant expansion of (A1); and we use the spin-pair decoupling and the "disentangling" approximations. The results are [corresponding to the system (27) of the text]

$$\dot{F}_q^z(t) = -\frac{1}{2}\sum_{q'} K_{qq'} \int_0^t dt' \left[C_{q'}^-(t-t')F_{q-q'}^+(t-t') + C_{q'}^+(t-t')F_{q-q'}^-(t-t')\right] F_q^z(t') \,,$$

$$\dot{F}_q^+(t) = iJ_{q,q+\pi}S\Delta G_q^+(t) - \sum_{q'} \int_0^t dt' \left\{\tfrac{1}{2}K_{qq'} C_{q'}^+(t-t')F_{q-q'}^z(t-t')F_q^+(t')\right.$$

$$\left. + \left[C_{q'}^z(t-t') - \delta_{q',\pi}S^2\Delta^2\right]\left[K_{qq'}F_{q-q'}^+(t-t')F_q^+(t') + K_{q,q'+\pi}G_{q-q'+\pi}^+(t-t')G_q^+(t')\right]\right\}, \quad \text{(A8)}$$

$$\dot{G}_q^+(t) = iJ_{q+\pi,q}S\Delta F_q^+(t) - \sum_{q'} \int_0^t dt' \left\{\tfrac{1}{2}K_{q+\pi,q'} C_{q'}^+(t-t')F_{q-q'+\pi}^z(t-t')G_q^+(t')\right.$$

$$\left. + \left[C_{q'}^z(t-t') - \delta_{q',\pi}S^2\Delta^2\right]\left[K_{q+\pi,q'+\pi}G_{q-q'}^+(t-t')F_q^+(t') + K_{q+\pi,q'}F_{q-q'+\pi}^+(t-t')G_q^+(t')\right]\right\},$$

where we recall

$$K_{qq'} = (J_{q'} - J_{q-q'})(J_{q'} - J_q) \,. \quad \text{(A9)}$$

The corresponding equations for $\dot{F}_q^-(t)$ and $\dot{G}_q^-(t)$ are the complex conjugates of the equations for $F_q^+(t)$ and $G_q^+(t)$, respectively. Thus, we have a highly coupled system of equations linking the F- and G-functions as well as the different wave vectors of the zone. We note that the initial conditions in (A8) are $F_q^\alpha(0) = 1$ and $G_q^\alpha(0) = 0$. We note further that in the limit $T = 0$ ($\Delta = 1$) we recover the solutions (A2) and (A3), and in the limit $T \to \infty$ ($\Delta = 0$), we have $G_q^\pm(t) \to 0$ and Eqs. (A8) reduce to the single equation (43) of Sec. III. For intermediate temperatures we must in general solve the full system of Eqs. (A8). Even though we are interested in the time-dependent correlation functions $C_q^\alpha(t)$, which are related to the F functions simply by $C_q^\alpha(t) = C_q^\alpha F_q^\alpha(t)$ for the classical case, we must also solve for the G functions along with the F functions.

In this regard it is instructive to consider the moments of the spectral functions $C_q^\pm(\omega)$. From (A8) and the initial conditions on $F_q^\alpha(t)$ and $G_q^\alpha(t)$, we find that the first and all odd order moments of $C_q^\pm(\omega)$ vanish, which simply implies that the spectral functions are even functions in ω. For the second moment, given by $\langle\omega_q^2\rangle_\pm = -\ddot{F}_q^\pm(0)$, we find

$$\langle\omega_q^2\rangle_\pm = \sum_{q'} K_{qq'}(C_{q'}^z + \tfrac{1}{2}C_{q'}^+) \,. \quad \text{(A10)}$$

The sole effect of the G functions is to produce $(2JS\Delta \sin q)^2$ when we differentiate the first term on the right-hand side of the equation for $F_q^+(t)$, which cancels the contribution arising from the term $\delta_{q',\pi}S^2\Delta^2$ in the second expression on the right-hand side. The result (A10) is precisely of the same form as for the ferromagnet, for which case the G functions are identically zero. (The second moments are of course not the same for the ferromagnet and antiferromagnet as the static correlations C_q differ in the two cases.) Thus, except for the cancellation mentioned above, the G functions do not enter explicitly into the calculation of the second moment. However, they do enter explicitly in the fourth- and higher-order moments, which may be seen easily by iterating Eqs. (A8) once.

At low but finite temperatures such that $K = |J_{c1}|/T \gg 1$, matters simplify to a great extent. In fact, we can derive equations for the antiferromagnet which are of exactly the same form as the Eqs. (77)–(81) of Sec. V, which were derived there for the ferromagnet at low temperature. We make use of the fact that at low temperature the system does not depart far from the Neél state (in a local sense). Therefore, we begin by assuming that the diagonalizing transformation (A5) for the $T = 0$ case is also a good first approximation at low but finite temperature. We take advantage of the fact that the static correlation function $C_q = \langle \hat{S}_{-q} \cdot \hat{S}_q \rangle$ for the antiferromagnet is peaked sharply around $q = \pi$ with a spread of the order $\Delta q \sim 1/K$, and analogous to the ferromagnet at low temperature, we take $C_q^\pm \cong 0$ and $C_q^z \cong C_q$ for the long-wavelength fluctuations $|q - \pi| \lesssim 1/K$. Making the *Ansatz*

$$f_q^\pm(t) = e^{\pm i\omega_q t}F_q^T(t) \,, \quad \text{(A11)}$$

where $\omega_q = 2JS\Delta|\sin q|$, we find the resulting equation for the envelop function $F_q^T(t)$, subject to the requirement that the approximations we make preserve the second moment (A10). [Note that the

second moment of $F_q^+(t)$ is also the second moment of $f_q^\pm(t)$.] After a rather lengthy but straightforward calculation, we find in a manner similar to the corresponding derivation for the ferromagnet that $F_q^T(t)$ satisfies the equation

$$\dot{F}_q^T(t) = -\int_0^t dt'\, k_q^T(t-t')F_q^T(t') \,, \tag{A12}$$

where

$$k_q^T(t) = \left(\sum_{q'} K_{qq'} C_{q'} F_{q'}^z(t) - \omega_q^2\right) F_q^T(t) \,. \tag{A13}$$

We note again that our diagonalizing procedure for finite temperature correctly produces the first two moments of the spectral function [correct within the framework of the general theory Eqs. (A8)]. Thus, at low temperature where the magnon lines are sharp and are adequately characterized by the first two moments, our procedure is justified. Only as the temperature is raised and the spectral lines broaden sufficiently do the higher order moments play a significant role in determining the line shape. Then errors arise from using (A12) rather than the general system of Eqs. (A8). But then the whole concept of local order as discussed in this paper begins to break down also. It is probably safe to conclude that the use of (A12) is justified as long as the concept of local order is valid, particularly in view of the agreement between our theory and experiment.

APPENDIX B: OTHER EXTENSIONS OF THEORY

The formal theory of Sec. II was developed explicitly for the case of an isotropic-Heisenberg exchange Hamiltonian with magnetic ordering present. However, essentially the same formalism applies with only minor modification to any system having an axis of quantization. Here, for the sake of completeness, we discuss briefly the modification necessary to treat three such cases of practical interest.

a. Isotropic Heisenberg system with applied external magnetic field. In addition to the spin interaction Hamiltonian $H = -\frac{1}{2}\sum_{i,j} \vec{S}_i \cdot \vec{S}_j$, we have the Zeeman term $H_z = -\omega_0 \sum_i S_i^z$, where $\omega_0 = g\mu_B h^{\text{ext}}$, with h^{ext} being the magnitude of the applied external field, g the gyromagnetic ratio, and μ_B the Bohr magneton. The direction of the applied field is taken to be the positive z axis, as is, of course, the direction of any magnetic order present in this case. It is easily found that the effect of H_z is to simply add the contributions $\mp i\omega_0 \delta S_q^\pm(t)$ to the equations of motion for $\delta \dot{S}_q^\pm(t)$. [See Eq. (11).] These additions are incorporated without change in the formal structure of the theory by letting

$$J_{qq'} S_{q-q'}^z(t) \to -\omega_0 \delta_{q,q'} + J_{qq'} S_{q-q'}^z(t) \tag{B1}$$

in the h matrix [Eq. (15)] in the final results for the coupled equations of motion for the F functions [Eqs. (27)], the equation for $F_q^z(t)$ remains the same but the terms $\mp i\omega_0 F_q^\pm(t)$ are added to the right-hand sides of the equations for $F_q^\pm(t)$.

b. Anisotropic Heisenberg Hamiltonian. For the case of axial symmetry around the z axis the Hamiltonian is

$$H = -\frac{1}{2}\sum_{i,j}[J_{ij}^L S_i^z S_j^z + J_{ij}^T (S_i^x S_j^x + S_i^y S_j^y)] \,. \tag{B2}$$

The only difference now in the development is that we must keep track of the components of the exchange interaction. Again, we can write the equation of motion for $\delta \dot{S}_q^\alpha(t)$ in the same manner as Eq. (13), but where the components of the matrix $h_{qq'}(t)$ are now

$$h_{qq'} = \begin{pmatrix} J_{qq'}^{TL} \hat{S}_{q-q'}^z(t) & -J_{qq'}^{LT} \hat{S}_{q-q'}^+(t) & 0 \\ -\frac{1}{2} J_{qq'}^{TT} \hat{S}_{q-q'}^-(t) & 0 & \frac{1}{2} J_{qq'}^{TT} \hat{S}_{q-q'}^+(t) \\ 0 & J_{qq'}^{LT} \hat{S}_{q-q'}^-(t) & -J_{qq'}^{TL} \hat{S}_{q-q'}^z(t) \end{pmatrix}, \tag{B3}$$

where

$$J_{qq'}^{\alpha\beta} = J_{q'}^\alpha - J_{q-q'}^\beta \,. \tag{B4}$$

The resulting equations of motion for $F_q^\alpha(t)$ are

$$\begin{aligned} \dot{F}_q^z(t) &= -\frac{1}{2}\sum_{q'} K_{qq'}^{(1)} \int_0^t dt' \,[C_{q'}^-(t-t')F_{q-q'}^+(t-t') \\ &\qquad + C_{q'}^+(t-t')F_{q-q'}^-(t-t')]\, F_q^z(t') \,, \\ \dot{F}_q^+(t) &= iJ_{qq}^{TL} S\Delta F_q^+(t) \\ &\quad -\sum_{q'} \int_0^t dt' \,\{\tfrac{1}{2} K_{qq'}^{(2)} C_{q'}^+(t-t')F_{q-q'}^z(t-t') \\ &\qquad + K_{qq'}^{(3)}[C_{q'}^z(t-t') - \delta_{q',0} S^2\Delta^2] \\ &\qquad \times F_{q-q'}^+(t-t')\} F_q^+(t') \,, \\ \dot{F}_q^-(t) &= [\dot{F}_q^+(t)]^* \,, \end{aligned} \tag{B5}$$

where

$$\begin{aligned} K_{qq'}^{(1)} &= J_{q,q-q'}^{TT} J_{q-q',q}^{LT} \,, \\ K_{qq'}^{(2)} &= J_{q,q-q'}^{LT} J_{q-q',q}^{TT} \,, \\ K_{qq'}^{(3)} &= J_{q,q-q'}^{TL} J_{q-q',q}^{TL} \,. \end{aligned} \tag{B6}$$

c. Reduced dipolar Hamiltonian. For a system of pure interacting dipoles, it has been shown[43,44] that with the application of an external magnetic field the Hamiltonian can be put into the form

$$H = H_z + H_d + H' \,, \tag{B7}$$

where H_z is the Zeeman Hamiltonian, H_d is the so-called truncated or reduced dipolar Hamiltonian, and H' contains the truncated off-diagonal terms of the dipolar interaction. The reduced Hamiltonian is of the anisotropic Heisenberg form (B2) with

$$J^L_{ij} = -\gamma^2\left(\frac{1-3\cos^2\theta_{ij}}{r^3_{ij}}\right) J^T_{ij} = \frac{\gamma^2}{2}\left(\frac{1-3\cos^2\theta_{ij}}{r^3_{ij}}\right), \tag{B8}$$

where $\gamma = g\mu_B$ and θ_{ij} is the angle $\vec{r}_{ij}$ makes with the direction of the applied field. If the strength of the dipole interaction is weak compared with the Zeeman energy splitting, then the effect of H_d is to broaden the transitions between adjacent Zeeman levels (as induced for example in magnetic resonance experiments with an alternating rf magnetic field of frequency $\sim\omega_0$ applied perpendicular to the static field). The effect of H' enters only in second order in perturbation theory and leads primarily to "forbidden" transitions between the zero-order Zeeman states; it is seen in resonance experiments as weak satellite lines separated by ω_0 from the primary Zeeman transition at $\omega = \omega_0$. Thus, if we are interested only in calculating the shape of the primary resonance line, we can omit H' from the Hamiltonian (B7). We are then led to a combination of the two previous cases. In particular, in Eqs. (B5), we need only add the terms $\mp i\omega_0 F^{\pm}_q(t)$ to the right-hand sides of the equations for $\dot{F}^{\pm}_q(t)$. We note that an isotropic-exchange Hamiltonian may be added to (B7) without changing the discussion, and hence exchange narrowing can be included.

APPENDIX C: CALCULATION OF EXACT SECOND MOMENT FOR CLASSICAL CHAIN

We compute the second moment of the spin-fluctuation spectrum of the classical chain directly from its definition

$$\langle\omega_q^2\rangle = -\left(\frac{d^2F_q(t)}{dt^2}\right)_{t=0} = -\frac{\ddot{C}_q(0)}{C_q}, \tag{C1}$$

where $C_q = \langle\hat{S}_{-q}\cdot\hat{S}_q\rangle$ is given by Eq. (41). We consider only the case of the isotropic-Heisenberg chain with nearest-neighbor interactions, $H = -J_{c1}\sum_l \hat{S}_l\cdot\hat{S}_{l+1}$. As all directions are equivalent for the macroscopic-ensemble average, we write (C1) as

$$\langle\omega_q^2\rangle = -\ddot{C}^z_q(0)/C^z_q, \tag{C2}$$

where $C^z_q = \frac{1}{3}C_q$ and $\ddot{C}^z_q$ is given in terms of spatial correlation functions as

$$\ddot{C}^z_q = \frac{1}{N}\sum_l \langle S^z_i\ddot{S}^z_{i+l}\rangle e^{-iql} = -\frac{1}{N}\sum_l \langle S^z_i[[S^z_{i+l}, H], H]\rangle e^{-iql}, \tag{C3}$$

where we have utilized the equations of motion $i\dot{S}^z_k = [S^z_k, H]$. From the equal-time spin commutation relations, we find that the quantities $\langle S^z_i\ddot{S}^z_k\rangle$ can be expressed in terms of certain four-spin correlation functions of the general form

$$C^{(4)}_{\alpha\beta\gamma\delta}(r, r', r'') = \langle S^\alpha_1 S^\beta_{1+r} S^\gamma_{1+r+r'} S^\delta_{1+r+r'+r''}\rangle, \tag{C4}$$

where α, β, γ, δ denote the indices $+$, z, $-$ and r, r', r'' are non-negative integers denoting the relative spin locations along the chain. Without loss of generality we have labeled the first spin in (C4) as one, and we close the chain with periodic boundary conditions so that $S_{N+1} = S_1$.

The quantities (C4) can be computed exactly—for the classical case only—using the transfer matrix technique[45,46] along the lines used by Fisher[6] in calculating the two spin-static correlations. Using the abbreviated notation $C^{(4)} = C^{(4)}_{\alpha\beta\gamma\delta}(r, r', r'')$ we write (C4)

$$\begin{aligned} C^{(4)} &= \frac{1}{Z}\int d\Omega_1\int d\Omega_2\cdots \\ &\quad\times\int d\Omega_N\, e^{-\beta H} S^\alpha_1 S^\beta_{1+r} S^\gamma_{1+r+r'} S^\delta_{1+r+r'+r''} \\ &= \frac{1}{Z}\int d\Omega_1\int d\Omega_2\cdots \\ &\quad\times\int d\Omega_N\, A(S_1, S_2)A(S_2, S_3)\cdots A(S_N, S_1) \\ &\quad\times S^\alpha_1 S^\beta_{1+r} S^\gamma_{1+r+r'} S^\delta_{1+r+r'+r''}, \end{aligned} \tag{C5}$$

where Ω_i is the solid angle corresponding to the direction of $\hat{S}_i$, Z is the partition function

$$\begin{aligned} Z &= \int d\Omega_1\int d\Omega_2\cdots\int d\Omega_N\, e^{-\beta H} \\ &= \int d\Omega_1\int d\Omega_2\cdots \\ &\quad\times\int d\Omega_N\, A(S_1, S_2)A(S_2, S_3)\cdots A(S_N, S_1) \end{aligned} \tag{C6}$$

and $A(S, S')$ is the transfer matrix defined by

$$\begin{aligned} A(S, S') &= e^{K\hat{S}\cdot\hat{S}'} \\ &= \sum_{l,m}\lambda_l Y^m_l(\hat{S})Y^{m*}_l(\hat{S}'). \end{aligned} \tag{C7}$$

Here, $K = \beta J_{c1}$, $Y^m_l(\hat{S})$ are the spherical harmonics, $l \geq 0$ and m is summed from $-l$ to $+l$, and the eigenvalues λ_l are given by $\lambda_l = (-1)^l j_l(iK)$, where $j_l(iK)$ are the spherical Bessel functions having purely imaginary arguments. (For convenience we differ somewhat in notation in this appendix from the main text by letting K be a signed parameter, the sign being that of J_{c1}.) We can carry out immediately all angular integrations in (C5), except those over $d\Omega_1$, $d\Omega_{1+r}$, $d\Omega_{1+r+r'}$, and $d\Omega_{1+r+r'+r''}$, using the orthonormality of the Y^m_l's

$$\int d\Omega\, Y^{m*}_l(\Omega)Y^{m'}_{l'}(\Omega) = \delta_{ll'}\delta_{mm'} \tag{C8}$$

to obtain

$$\begin{aligned} C^{(4)} &= \frac{1}{Z}\sum_{\substack{l_1 l_2 l_3 l_4 \\ m_1 m_2 m_3 m_4}} \lambda^r_{l_1}\lambda^{r'}_{l_2}\lambda^{r''}_{l_3}\lambda^{N-r-r'-r''}_{l_4} \\ &\quad\times\int d\Omega\, Y^{m_4*}_{l_4} S^\alpha Y^{m_1}_{l_1} \end{aligned}$$

$$\times\int d\Omega\, Y_{l_1}^{m_1*} S^\beta Y_{l_2}^{m_2} \int d\Omega\, Y_{l_2}^{m_2*} S^\gamma Y_{l_3}^{m_3} \times\int d\Omega\, Y_{l_3}^{m_3*} S^\delta Y_{l_4}^{m_4} \quad \text{(C9)}$$

and

$$Z=\textstyle\sum_l \lambda_l^N\ . \quad \text{(C10)}$$

As $N\to\infty$, the ratio $(\lambda_l/\lambda_0)^N\to 0$ for $l\geq 1$, and we have $Z=\lambda_0^N$ and $\lambda_{l4}\to\lambda_0$ in (C9). Hence $C^{(4)}$ becomes

$$C^{(4)}=\sum_{\substack{l_1 l_2 l_3\\ m_1 m_2 m_3}}\left(\frac{\lambda_{l1}}{\lambda_0}\right)^r\left(\frac{\lambda_{l2}}{\lambda_0}\right)^{r'}\left(\frac{\lambda_{l3}}{\lambda_0}\right)^{r''}\int d\Omega\, Y_0^{0*} S^\alpha Y_{l_1}^{m_1} \times\int d\Omega\, Y_{l_1}^{m_1*} S^\beta Y_{l_2}^{m_2}\int d\Omega\, Y_{l_2}^{m_2*} S^\gamma Y_{l_3}^{m_3} \times\int d\Omega\, Y_{l_3}^{m_3*} S^\delta Y_0^0\ . \quad \text{(C11)}$$

Making the correspondence $+\to 1$, $z\to 0$, and $-\to -1$ we note that S^α is given in terms of Y_1^α as

$$S^\alpha = b^\alpha\sqrt{\tfrac{4}{3}\pi}\, Y_1^\alpha(\Omega)\ , \quad \text{(C12)}$$

where $b^1=-\sqrt{2}$, $b^0=1$, $b^{-1}=\sqrt{2}$. Substituting (C12) into (C11) and using $Y_0^0=1/\sqrt{4\pi}$, we can carry out two more of the angular integrations, again using the orthonormality condition (C8), obtaining

$$C^{(4)}=\frac{4\pi}{9}B^{\alpha\beta\gamma\delta}\textstyle\sum_{l_2 m_2}(-1)^{m_2}\left(\frac{\lambda_1}{\lambda_0}\right)^{r+r''}\left(\frac{\lambda_{l2}}{\lambda_0}\right)^{r'} \times\int d\Omega\, Y_1^\alpha Y_1^\beta Y_{l_2}^{m_2}\int d\Omega\, Y_1^\gamma Y_1^\delta Y_{l_2}^{-m_2}\ , \quad \text{(C13)}$$

where $B^{\alpha\beta\gamma\delta}=b^\alpha b^\beta b^\gamma b^\delta$ and l_2 is summed from zero to two.

The two remaining angular integrals can be expressed in terms of the 3 n-j symbols of Wigner in the following manner[46–48]:

$$\int d\Omega\, Y_{l_1}^{m_1} Y_{l_2}^{m_2} Y_{l_3}^{m_3}=\left(\frac{(2l_1+1)(2l_2+1)(2l_3+1)}{4\pi}\right)^{1/2} \times\begin{pmatrix} l_1 & l_2 & l_3\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} l_1 & l_2 & l_3\\ m_1 & m_2 & m_3\end{pmatrix}\ . \quad \text{(C14)}$$

We then have

$$C^{(4)}=B^{\alpha\beta\gamma\delta}\left(\frac{\lambda_1}{\lambda_0}\right)^{r+r''}\textstyle\sum_{l,m}(-1)^m(2l+1)\left(\frac{\lambda_l}{\lambda_0}\right)^{r'} \times\begin{pmatrix} 1 & 1 & l\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} 1 & 1 & l\\ \alpha & \beta & m\end{pmatrix}\begin{pmatrix} 1 & 1 & l\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} 1 & 1 & l\\ \gamma & \delta & -m\end{pmatrix}\ . \quad \text{(C15)}$$

But

$$\begin{pmatrix} 1 & 1 & 1\\ 0 & 0 & 0\end{pmatrix}=0$$

(corresponding to the fact that $\int d\Omega\, Y_1^{m_1} Y_1^{m_2} Y_1^{m_3}=0$), and therefore the $l=1$ terms drop out. The $l=0$ contribution reduces to

$$\tfrac{1}{9}B^{\alpha\beta\gamma\delta}\left(\frac{\lambda_1}{\lambda_0}\right)^{r+r''}\delta_{\alpha,-\beta}\delta_{\gamma,-\delta}(-1)^{\alpha+\gamma}\ . \quad \text{(C16)}$$

Upon making use of the facts

$$\begin{pmatrix} 1 & 1 & 2\\ 0 & 0 & 0\end{pmatrix}^2=\frac{2}{15}\ ,$$
$$\begin{pmatrix} 1 & 1 & 2\\ \alpha & \beta & m\end{pmatrix}=0 \text{ unless } m=-(\alpha+\beta)\ , \quad \text{(C17)}$$

we have as our result for $C^{(4)}$

$$C^{(4)}=\tfrac{1}{9}B^{\alpha\beta\gamma\delta}\left(\frac{\lambda_1}{\lambda_0}\right)^{r+r''}\left[(-1)^{\alpha+\gamma}\delta_{\alpha,-\beta}\delta_{\gamma,-\delta} +6(-1)^{\alpha+\beta}\left(\frac{\lambda_2}{\lambda_0}\right)^{r'}\delta_{\alpha+\beta,-\gamma-\delta}\begin{pmatrix} 1 & 1 & 2\\ \alpha & \beta & -(\alpha+\beta)\end{pmatrix} \times\begin{pmatrix} 1 & 1 & 2\\ \gamma & \delta & -(\gamma+\delta)\end{pmatrix}\right]. \quad \text{(C18)}$$

To express our result in a more compact form, we introduce a 3×3 matrix D whose elements $D_{\alpha\beta}$ are defined by

$$D_{\alpha\beta}=\begin{pmatrix} 1 & 1 & 2\\ \alpha & \beta & -(\alpha+\beta)\end{pmatrix}\ . \quad \text{(C19)}$$

With this definition and evaluating the corresponding Wigner symbols, the matrix D is given explicitly by

$$D=\frac{1}{\sqrt{5}}\begin{pmatrix} 1 & -\sqrt{\frac{1}{2}} & \sqrt{\frac{1}{6}}\\ -\sqrt{\frac{1}{2}} & \sqrt{\frac{2}{3}} & -\sqrt{\frac{1}{2}}\\ \sqrt{\frac{1}{6}} & -\sqrt{\frac{1}{2}} & 1\end{pmatrix}\ , \quad \text{(C20)}$$

with the rows and columns labeled in the order +, 0, −. Our final result for $C^{(4)}$ is

$$C^{(4)}_{\alpha\beta\gamma\delta}(r,r',r'')=\tfrac{1}{9}B^{\alpha\beta\gamma\delta}\left(\frac{\lambda_1}{\lambda_0}\right)^{r+r''}\left[(-1)^{\alpha+\gamma}\delta_{\alpha,-\beta}\delta_{\gamma,-\delta} +6(-1)^{\alpha+\beta}\left(\frac{\lambda_2}{\lambda_0}\right)^{r'}D_{\alpha\beta}D_{\gamma\delta}\delta_{\alpha+\beta,-\gamma-\delta}\right], \quad \text{(C21)}$$

where, from the definition of the eigenvalues λ_l, we have

$$\lambda_1/\lambda_0=u(K)=\coth K-1/K\ ,$$
$$\lambda_2/\lambda_0=1-3u(K)/K\ . \quad \text{(C22)}$$

We note in passing that (C21) applies equally well when one or more of the r's are zero.

It is now quite straightforward to combine Eq. (C3) with the result (C21) to produce the correlation function $\ddot{C}_q^z$:

$$\ddot{C}_q^z=-\frac{4J_{01}^2}{3N}\frac{u(K)}{K}(1-\cos q)\ . \quad \text{(C23)}$$

We note that the contribution from the $l\geq 2$ terms of the series in (C3) cancel exactly to zero, i.e.,

(C23) arises solely from the terms $\langle S_i^z \ddot{S}_i^z \rangle$ and $\langle S_i^z \ddot{S}_{i\pm1}^z \rangle$ in (C3). Finally, from (C2) we obtain the exact second moment of the classical chain

$$\langle \omega_q^2 \rangle_{\text{exact}} = 4J_{\text{cl}}^2 \frac{u}{K(1-u^2)} (1-\cos q)(1+u^2-2u\cos q) . \tag{C24}$$

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[1]M. Blume and J. Hubbard, Phys. Rev. B **1,** 3815 (1970).

[2]C. G. Windsor, *Inelastic Scattering of Neutrons* (International Atomic Energy Agency, Vienna, 1968), Vol. 2, p. 83.

[3]J. Hubbard, J. Phys. Chem. **4,** 53 (1971). See also,P. Resibois and M. de Leener, Phys. Rev. **152,** 305 (1966); K. Kawasaki, Prog. Theor. Phys. **39,** 285 (1968); F. Wegner, Z. Phys. **216,** 433 (1968); G. Reiter, Phys. Rev. B **5,** 222 (1972).

[4]R. Nathans, F. Menzinger, and S. J. Pickart, J. Appl. Phys. **31,** 1237 (1968).

[5]J. Skalyo, Jr., G. Shirane, R. J. Birgeneau, and H. J. Guggenhein, Phys. Rev. Lett. **23,** 1394 (1969).

[6]M. E. Fisher, Am. J. Phys. **32,** 343 (1964).

[7]J. Skalyo, Jr., G. Shirane, S. A. Friedberg, and H. Kobayashi, Phys. Rev. B **2,** 4632 (1970).

[8]R. Dingle, M. E. Lines, and S. L. Holt, Phys. Rev. **187,** 643 (1969).

[9]R. J. Birgeneau, R. Dingle, M. T. Hutchings, G. Shirane, and S. L. Holt, Phys. Rev. Lett. **26,** 718 (1971).

[10]M. T. Hutchings, G. Shirane, R. J. Birgeneau, and S. L. Holt, Phys. Rev. B **5,** 1999 (1972).

[11](a) P. M. Richards, Phys. Rev. Lett. **27,** 1800 (1971); (b) D. McFadden and R. A. Tahir-Kheli, Phys. Rev. B **1,** 3671 (1970); S. W. Lovesey and R. A. Meserve, Phys. Rev. Lett. **28,** 614 (1972); K. Tomita (unpublished).

[12](a) F. Carboni and P. M. Richards, Phys. Rev. **177,** 889 (1969); (b) H. A. Gersch, Phys. Rev. B **1,** 2270 (1970).

[13]E. Lieb, T. Schultz, and D. C. Mattis, Ann. Phys. (N.Y.) **16,** 407 (1961).

[14]J. Des Cloizeaux and J. J. Pearson, Phys. Rev. **128,** 2131 (1962).

[15]K. Kawasaki, Ann. Phys. (N.Y.) **37,** 142 (1966).

[16]Y. Kuramoto, Prog. Theor. Phys. **40,** 36 (1968).

[17]M. Blume, R. E. Watson, and G. H. Vineyard, Bull. Am. Phys. Soc. **16,** 629 (1971); and (unpublished).

[18]For a general review of neutron scattering from magnetic systems see W. Marshall and R. Lowde, Rep. Prog. Phys. **37,** 705 (1968).

[19]For example, see F. Keffer, in *Handbuch der Physik,* edited by S. Flugge (Springer, Berlin, 1966), Vol. 18, Pt. 2, p. 1.

[20]M. E. Lines, Phys. Rev. B **3,** 1749 (1971).

[21]G. H. Wannier, Rev. Mod. Phys. **17,** 50 (1945).

[22]R. Kubo, J. Phys. Soc. Jap. **12,** 570 (1957).

[23]We make the association $+\to 1$, $z\to 0$, and $-\to -1$. Then the meaning of $S^{-\alpha}$ becomes clear.

[24]R. Kubo, J. Phys. Soc. Jap. **17,** 1100 (1962).

[25]For the case of antiferromagnetic order, the change in the Brillouin zone leads to off-diagonal elements of $V(t)$ in the q indices.

[26]The classical spin system is the large spin limit of "real" finite-spin quantum-mechanical systems. The transcription from the quantum-mechanical case to the classical case as S becomes large is made by setting $JS(S+1)=J_{\text{cl}}$. Hence, $\frac{1}{2}\beta\omega \sim \beta JS \to \beta JS_{\text{cl}}/(S+1)$, which goes to zero in the limit $S\to\infty$ if βJ_{cl} remains finite.

[27]The same discussion applies as well to the antiferromagnet, except that the wave vector region of importance is $|q^*|=|\pi-q| \gtrsim 1/K$. The experimental evidence of Birgeneau *et al.*[9] on the static correlation functions of TMMC, for which $S=5/2$, indicates that the classical approximation is clearly valid for the wave vector region $q^* < 3/K$.

[28]When we apply the concept of a local averaging process to the linear chain, we must know how $C_q = \langle S_{-q} \cdot S_q \rangle$ is partitioned into longitudinal and transverse components. This is discussed in Sec. IV.

[29]For the classical chain, all the odd moments vanish.

[30]F. B. McLean and M. Blume, J. Appl. Phys. **42,** 1380 (1971).

[31]P. Schofield, Phys. Rev. Lett. **4,** 239 (1960).

[32]E. D. Siggia and M. Blume (unpublished).

[33]See W. Marshall, Conference on Critical Phenomena (NBS Miscellaneous Publication No. 273, 1965) (unpublished); J. L. Beeby and J. Hubbard, Phys. Lett. A **26,** 376 (1968).

[34]F. Keffer and R. Loudon, J. Appl. Phys. Suppl. **32,** 2 (1961); J. Appl. Phys. **33,** 250 (1962). See also Ref. 20, p. 50.

[35]It follows from Eq. (41) that $C(q \gg 1/K) \sim (1/K^2) C(q \sim 0)$.

[36]The exact result as calculated from Eq. (60) is $\langle \omega_q^2 \rangle_T = \frac{4}{3} J_{\text{cl}} (1-\cos q)(1+u^2-2u\cos q)$ but, of course, we expect the approximations that went into deriving Eq. (6) are valid only in the temperature region where the expansion of $\langle \omega_q^2 \rangle_T$ to order $1/K^2$ is valid.

[37]The reasoning behind this is that $k_q^\alpha(t)$ is quadratic in the F-functions and hence approaches zero more rapidly than $F_q(t)$.

[38]In Sec. IV we argued that in the spin wave region $C_q^T \cong \frac{1}{2} C_q$ where $C_q = \langle S_{-q} \cdot S_q \rangle$. We expect this to be true for finite spins as well as for classical systems.

[39]There is a factor of 2 difference in the definition of J between our papers.

[40]Our system of units may be clearer if we let τ and Ω be the time and frequency in reduced units. Then, in terms of the time t and frequency ω measured in the usual units, we have $\tau = 2J[S(S+1)]^{1/2} t$ and $\Omega = \omega/2J[S(S+1)]^{1/2}$.

[41]In Eq. (64) we defined a theoretical σ_q in terms of the first and second moments of $F_q^{\pm}(t)$. The present definition is convenient in a practical sense.

[42]Take $f(x)>0$ for all x, $f'(x)=0$ at $x=x_0$, and $f'(x<x_0)>0$ and $f'(x>x_0)<0$. Then $(d/dx)[xf(x)] = f(x)+xf'(x)=0$ for $x=-f(x)/f'(x)$. This can only occur (for x positive) for $x=x_m>x_0$.

[43]J. H. Van Vleck, Phys. Rev. **74,** 1168 (1948).

[44]Also see, for example, A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, England, 1960), p. 97.

[45]G. H. Wannier, Rev. Mod. Phys. **17,** 50 (1945).

[46]G. S. Joyce, Phys. Rev. **155,** 478 (1967).

[47]A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton U. P., Princeton, N. J., 1957), p. 63.

[48]M. Rotenberg, R. Bivens, N. Metropolis, and J. K. Wooten, *The 3-j and 6-j Symbols* (MIT, Cambridge, Mass., 1959).