Correlation theory of the Heisenberg antiferromagnet and ferromagnet: Dependence on dimension, field, and temperature

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The correlation theory is applied to a Heisenberg antiferromagnet in a magnetic field. Special cases covered are the ferromagnet and an anisotropic Heisenberg model. The theory includes self-consistently correlation effects in static and dynamic properties. It is a generalization of the random-phase approximation and is applicable to the quantum spin case for any dimension and temperature. The static susceptibilities and the excitation spectrum are calculated. Besides the spin-wave excitations a central peak is found which can be understood as coming from local longitudinal fluctuations. The results of the theory are exemplified by numerical calculations for the one-dimensional $S=1$ quantum antiferromagnetic chain. Qualitative agreement is found with computer simulations on a classical chain.

I. INTRODUCTION

An antiferromagnet in an applied field was discussed in considerable detail by Lovesey and Loveluck[1] using the Mori[2] theory. In this approach the dynamical variables are usually taken to be the operators (and derivatives thereof), which in the hydrodynamic limit satisfy conservation laws. The rationale is that these modes should be slowly varying with time and should therefore dominate the dynamical behavior. For the antiferromagnet these variables are the magnetization density $M_{\vec{q}}$ and the energy density $E_{\vec{q}}$. A coupling between these should occur for finite external fields. Clearly, this basis is designed to treat the long-wavelength limit $q\rightarrow 0$ and $\omega\rightarrow 0$. However, this limit is not easily observable by either neutron scattering or in numerical simulation studies. In the correlation theory[3] an alternative set of dynamical variables is chosen, namely dynamical variables, which allows the local or short-range properties to be calculated exactly. This provides a description of the normal modes at high q and ω. It is therefore a theory designed to obtain properties which can be tested by the above-mentioned measurements. This basis turns out to be the same as is convenient for the description of the ordered phase for $T<T_N$. Consequently, all temperatures can be treated in the same framework. The correlation theory does not use the hydrodynamic concept energy modes, but gives a rather simple picture of the dynamics determined by spin fluctuations on different sublattices. If no approximations were made the choice of dynamical variables is only a matter of taste and convenience. However, since approximations are necessary, the most physical basis (i.e., the one giving the best noninteracting normal modes) should be the most adequate. It is therefore of interest to compare the results of the correlation theory with the complementary theory by Lovesey and Loveluck. Another motivation for this study is a comparison with the extensive computer-simulation studies on a classical one-dimensional antiferromagnet by Balcar *et al.*[4]

The correlation theory here presented is valid for any dimension and lattice structure. The two-dimensional antiferromagnet on a hexagonal lattice is particularly interesting because it may represent a frustrated ground state.[5,6] The present theory has some bearings on this question, but this will be discussed in a separate paper. The theoretical frame work is also applicable to the ferromagnet for any temperature and field. The present work therefore extends the discussion on the Heisenberg magnet in the paramagnetic phase previously published.[3] As stated in Ref. 3 the correlation theory is not intended to go beyond the many existing theories on the Heisenberg model addressing special questions.[7] In particular, for one-dimensional systems, where the static properties can be exactly calculated and frequency moments up to $\langle\omega^6_{\vec{q}}\rangle$ are known,[8] the correlation theory is bound to be less accurate. However, the reason for studying this case is to test the approximate theory in the presence of strong correlation effects. The correlation theory is easily generalized to higher dimensions, small spin values, and other regions, where the particular theories do not apply and where the correlation theory may be expected to be more accurate than in the test case. The discussion of an antiferromagnet in a field turns out to cover the case of a quite general anisotropic spin Hamiltonian as well.

The organization of the paper is as follows. Firstly, we discuss the solution of the normal modes for a locally correlated ground state. Secondly, we list a number of exact moments or sum rules. Then the static susceptibility tensor is calculated including pair-correlation effects using a mode-mode—coupling approximation. Explicit results are given for the ferromagnet and the antiferromagnet for different temperatures and fields. Some exact results on the dynamics are given. The two-pole approximation, which was successfully used for the paramagnetic phase,[3] is generalized to the present case with several dynamical variables and finite first-moment components. The results of the theory are computed numerically, in particular for the antiferromagnetic quantum $S=1$ chain.

Moreover, results on the S dependence and the dimensionality are also discussed. Finally, the results are summarized and compared with other theories.

II. ANTIFERROMAGNET IN A MAGNETIC FIELD

Let us discuss the correlation theory for an antiferromagnet in a field with a nearest-neighbor exchange interaction. The Hamiltonian is

$$\mathcal{H}=\tfrac{1}{2}J\sum_{i,j}\tilde{\vec{S}}_i\cdot\tilde{\vec{S}}_j-H\sum_i\tilde{S}_i^z\ . \tag{1}$$

The basic idea is to attempt to solve the local properties exactly and calculate the interaction effects as corrections to these. The major effect of the field in combination with the effective antiferromagnetic exchange field is to orient the spins nearly perpendicular to the external field with a small tilt angle θ towards it. The minimum classical energy is obtained if the spins are tilted in the same plane, say the xz plane. For a one-dimensional chain this order is destroyed at long distances due to torsions of the plane, which perhaps can be described by solitons. However, the local or the short-range behavior should be well described by the above picture even in the disordered phase at not too high temperatures. Let us therefore generally assume that the physics is most clearly described in this two-sublattice framework, which is necessary for the theory of the ordered phase. The assumed ground state is shown in Fig. 1. The use of this framework for the theory implies no approximations if the correlation effects are accurately calculated. The classical internal energy per spin is

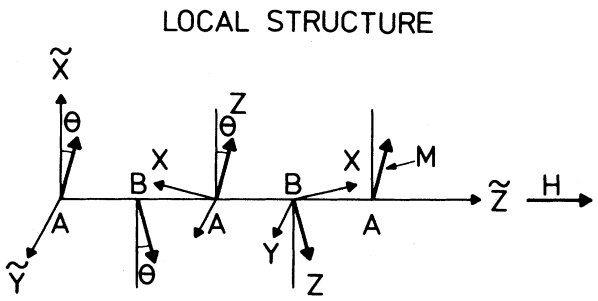

FIG. 1. Classical local ground state for intermediate fields for an antiferromagnetic chain.

$$E=\tfrac{1}{2}J_0M^2\cos(2\theta)-HM\sin\theta\ .$$

The tilt angle θ obtained from $\partial E/\partial\theta=0$ is given by

$$\sin\theta=H/(2J_0M)\ . \tag{2}$$

For ρ nearest neighbors, we define

$$J_0=\rho J$$

and

$$J_{\vec{q}}=\gamma_{\vec{q}}J_0\ \text{ with }\gamma_{\vec{q}}=\frac{1}{\rho}\sum_{\vec{R}}e^{i\vec{q}\cdot\vec{R}}\ .$$

M is the local average moment, which may be close to S at low temperatures. The tilt angle θ, Eq. (2), will be modified by correlation effects; see (84). By the following transformation we obtain the local coordinate system for sublattices A and B such that the z axis is the quantization axis:

$$\begin{aligned}
&(\tilde{S}_A^x)_{\vec{R}}=(S_A^x)_{\vec{R}}\sin\theta+(S_A^z)_{\vec{R}}\cos\theta,\ \ (\tilde{S}_B^x)_{\vec{R}}=(S_B^x)_{\vec{R}}\sin\theta-(S_B^z)_{\vec{R}}\cos\theta\ ,\\
&(\tilde{S}_A^y)_{\vec{R}}=(S_A^y)_{\vec{R}},\ \ (\tilde{S}_B^y)_{\vec{R}}=(S_B^y)_{\vec{R}}\ ,\\
&(\tilde{S}_A^z)_{\vec{R}}=-(S_A^x)_{\vec{R}}\cos\theta+(S_A^z)_{\vec{R}}\sin\theta,\ \ (\tilde{S}_B^z)_{\vec{R}}=(S_B^x)_{\vec{R}}\cos\theta+(S_B^z)_{\vec{R}}\sin\theta\ .
\end{aligned} \tag{3}$$

Let us define $\vec{R}_{AB}$ as the vector connecting the two sublattices and the Fourier transforms by

$$(S_A^x)_{\vec{q}}=\sum_{\vec{R}}e^{i\vec{q}\cdot\vec{R}}(S_A^x)_{\vec{R}}\ \text{ and }(S_B^x)_{\vec{q}}=\sum_{\vec{R}}e^{i\vec{q}\cdot(\vec{R}+\vec{R}_{AB})}(S_B^x)_{\vec{R}}\ . \tag{4}$$

The index $\vec{q}$ will sometimes be suppressed for simplicity. The total relaxation functions are expressed in terms of the sublattice relaxation functions

$$\begin{aligned}
&\tilde{R}^{\parallel}_{\vec{q}\omega}=(\tilde{S}^z_{\vec{q}}\tilde{S}^z_{-\vec{q}})_\omega=\{[(S_A^xS_A^x)_{\vec{q}\omega}-(S_A^xS_B^x)_{\vec{q}\omega}]\cos^2\theta+[(S_A^zS_A^z)_{\vec{q}\omega}+(S_A^zS_B^z)_{\vec{q}\omega}]\sin^2\theta\}\ ,\\
&\tilde{R}^{\perp}_{\vec{q}\omega}=(\tilde{S}^x_{\vec{q}}\tilde{S}^x_{-\vec{q}})_\omega=\{[(S_A^xS_A^x)_{\vec{q}\omega}+(S_A^xS_B^x)_{\vec{q}\omega}]\sin^2\theta+[(S_A^zS_A^z)_{\vec{q}\omega}-(S_A^zS_B^z)_{\vec{q}\omega}]\cos^2\theta\}\ ,\\
&\tilde{R}^{\perp}_{\vec{q}\omega}=(\tilde{S}^y_{\vec{q}}\tilde{S}^y_{-\vec{q}})_\omega=[(S_A^yS_A^y)_{\vec{q}\omega}+(S_A^yS_B^y)_{\vec{q}\omega}]\ ,
\end{aligned} \tag{5}$$

A relaxation function in (5) is defined[2] as the Fourier transform of

$$[\vec{S}(t)\vec{S}^\dagger(0)]=\int_0^\beta d\lambda\langle\vec{S}(t-i\lambda)\vec{S}^\dagger(0)\rangle-\beta\langle\vec{S}\rangle\langle\vec{S}^\dagger\rangle\ ,$$

where $\vec{S}$ is the dynamical variable vector.

The total susceptibility components are given by $\tilde{\chi}^{\alpha\alpha}_{\vec{q}}=\int_\infty^\infty(\tilde{S}^\alpha_{\vec{q}}\tilde{S}^\alpha_{-\vec{q}})_\omega d\omega$. For symmetry reasons, $(\tilde{S}^x_{\vec{q}}\tilde{S}^x_{-\vec{q}})_\omega$ and $(\tilde{S}^y_{\vec{q}}\tilde{S}^y_{-\vec{q}})_\omega$ must be identical in the disordered state, but they differ in the symmetry-broken (antiferromagnetic) state.

The degree to which the symmetry is obtained serves as a check on the theory. The exact Hamiltonian in the transformed systems is expressed in terms of the Fourier-transformed variables (4),

$$\mathcal{H}=J_0\sum_{\vec{k}}\{-\phi_{\vec{k}}[(S_A^x)_{\vec{k}}(S_B^x)_{-\vec{k}}+(\delta S_A^z)_{\vec{k}}(\delta S_B^z)_{-\vec{k}}]+\gamma_{\vec{k}}(S_A^y)_{\vec{k}}(S_B^y)_{-\vec{k}}+\sigma_{\vec{k}}[(\delta S_A^z)_{\vec{k}}(S_B^x)_{-\vec{k}}-(S_A^x)_{\vec{k}}(\delta S_B^z)_{-\vec{k}}]\}$$

$$-J_0Mh\sum_{\vec{k}}[(S_A^z)_{\vec{k}}+(S_B^z)_{\vec{k}}]+\tfrac{1}{2}J_0M^2\,, \tag{6}$$

where $M=\langle S_A^z\rangle=\langle S_B^z\rangle$, $\delta S_{\vec{q}}^z=S_{\vec{q}}^z-M\delta(\vec{q})$, $\phi_{\vec{k}}=\gamma_{\vec{k}}\cos(2\theta)$, and $\sigma_{\vec{k}}=\gamma_{\vec{k}}\sin(2\theta)$. We have $h=\cos(2\theta)+H\sin\theta/J_0M$, the molecular field in units of J_0M. If we use (2), $h=1$ for $\theta<90°$ and $h=H/J_0M-1$ for large fields. The Hamiltonian (6) is clearly a quite general anisotropic Hamiltonian, for which the following discussion also applies directly. The dynamical variables to be used in the correlation theory are the orthogonal $(S_N^x)_{\vec{q}}$,$(S_N^y)_{\vec{q}}$, and $(\delta S_N^z)_{\vec{q}}$ (for $N=A$ and B), which are also necessary to describe the properties of the ordered phase.

III. RANDOM-PHASE APPROXIMATION

In terms of the sublattice Green's function[10] $G_{NM}^{\alpha\beta}=\langle\langle S_N^\alpha S_M^\beta\rangle\rangle$ it is a straightforward procedure to derive the random-phase approximation (RPA). This approximation means that we assume a spin S_R and its neighbors participating in a certain spin wave with wave vector and frequency q and ω_q, respectively. If the neighboring spins participate in other spin waves one assumes that these have arbitrary phase, wave vector $\vec{k}$, and frequency $\omega_{\vec{k}\,(\neq\vec{q})}$. To a first approximation, $S_{\vec{R}}$ therefore only sees the local average of its neighboring spins, $\langle\vec{S}_{\vec{R}+\vec{\delta}}\rangle=M\hat{z}$. If there are couplings between the modes, this picture is modified. Using the equation of motion,

$$\omega\langle\langle AB\rangle\rangle=\frac{1}{2\pi}\langle[A,B]\rangle+\langle\langle[A,\mathcal{H}]B\rangle\rangle\,,$$

and RPA decoupling, one finds, assuming that the z axes are along the average direction of the moments,

$$\begin{aligned}
G_{AA}^{xx}&=G_{BB}^{xx}=\frac{iM}{2\pi}J_0Mh\,[\omega^2-(J_0M)^2(h^2-\gamma_{\vec{q}}^2)]/D(\omega)\,,\\
G_{AB}^{xx}&=G_{BA}^{xx}\\
&=\frac{iM}{2\pi}J_0M\,[\gamma_{\vec{q}}\omega^2-(J_0M)^2\phi_{\vec{q}}(h^2-\gamma_{\vec{q}}^2)]/D(\omega)\,,\\
G_{AA}^{yy}&=G_{BB}^{yy}=\frac{iM}{2\pi}J_0Mh\,[\omega^2-(J_0M)^2(h^2-\phi_{\vec{q}}^2)]/D(\omega)\,,\\
G_{AB}^{yy}&=G_{BA}^{yy}\\
&=\frac{-iM}{2\pi}J_0M\,[\phi_{\vec{q}}\omega^2-(J_0M)^2\gamma_{\vec{q}}(h^2-\phi_{\vec{q}}^2)]/D(\omega)\,,
\end{aligned} \tag{7}$$

where

$$D(\omega)=(\omega^2-\omega_{\|}^2)(\omega^2-\omega_{\perp}^2)\,.$$

The system has therefore, in the RPA, two pairwise nondegenerate frequencies,

$$\begin{aligned}
\omega_{\|}&=\pm J_0M[(h-\gamma_{\vec{q}})(h+\phi_{\vec{q}})]^{1/2}\,,\\
\omega_{\perp}&=\pm J_0M[(h+\gamma_{\vec{q}})(h-\phi_{\vec{q}})]^{1/2}\,.
\end{aligned} \tag{8}$$

If we use $M=S$ and $h=1$ these frequencies are identical to those derived by Lovesey[9] using simple spin-wave theory. They also agree with the classical results.[8]

Using the exact relation

$$\langle AB\rangle=\frac{1}{2\pi i}\int\frac{\mathrm{Im}\langle\langle AB\rangle\rangle_\omega}{1-e^{-\beta\omega}}d\omega\,,$$

where $\beta=1/k_BT$, one finds in the RPA from (7),

$$\begin{aligned}
\langle S_A^xS_A^x\rangle_{\vec{q}}&=\langle S_B^xS_B^x\rangle_{\vec{q}}=(W+V\phi_{\vec{q}})/(h^2-\phi_{\vec{q}}^2)\,,\\
\langle S_A^xS_B^x\rangle_{\vec{q}}&=\langle S_B^xS_A^x\rangle_{\vec{q}}=(W\phi_{\vec{q}}+V)/(h^2-\phi_{\vec{q}}^2)\,,\\
\langle S_A^yS_A^y\rangle_{\vec{q}}&=\langle S_B^yS_B^y\rangle_{\vec{q}}=(W-V\gamma_{\vec{q}})/(h^2-\gamma_{\vec{q}}^2)\,,\\
\langle S_A^yS_B^y\rangle_{\vec{q}}&=\langle S_B^yS_A^y\rangle_{\vec{q}}=-(W\gamma_{\vec{q}}-V)/(h^2-\gamma_{\vec{q}}^2)\,,
\end{aligned} \tag{9}$$

where, in terms of ω_n from (8),

$$W+V=\frac{k_BT}{2J_0}\frac{\beta\omega_{\|}\sinh(\beta\omega_{\|})}{1-\cosh(\beta\omega_{\|})}$$

and

$$W-V=\frac{k_BT}{2J_0}\frac{\beta\omega_{\perp}\sinh(\beta\omega_{\perp})}{1-\cosh(\beta\omega_{\perp})}\,. \tag{10}$$

The RPA solution for the z part is more trivial and is the same as the mean-field result $\chi_{AA}^{zz}=1/J_0$ and $\chi_{AB}^{zz}=0$. The RPA-type susceptibility including the interaction for the zz part will be given in (45).

The original relaxation functions (5), $\omega\tilde{R}_{\vec{q}\omega}=-2\pi\,\mathrm{Im}\tilde{G}(\vec{q},\omega)$, and spin-correlation functions,

$$\langle\tilde{S}_{\vec{q}}\tilde{S}_{-\vec{q}}\rangle_\omega=\omega\tilde{R}_{\vec{q}\omega}/(1-e^{-\beta\omega})\,,$$

are now easily derived. We find, using (5), (7), and (45),

$$\langle\tilde{S}_{\vec{q}}^x\tilde{S}_{-\vec{q}}^x\rangle_\omega=M\left[\frac{h+\gamma_{\vec{q}}}{h-\phi_{\vec{q}}}\right]^{1/2}\tfrac{1}{2}[n_\perp\delta(\omega-\omega_\perp)+(n_\perp+1)\delta(\omega+\omega_\perp)]\sin^2\theta+\frac{k_BT}{2J_0}\frac{1}{1+\phi_{\vec{q}}}\cos^2\theta\,\delta(\omega)\,, \tag{11}$$

$$\langle \tilde{S}^y_{\vec{q}} \tilde{S}^y_{-\vec{q}} \rangle_\omega = M \left[\frac{h-\phi_{\vec{q}}}{h+\gamma_{\vec{q}}} \right]^{1/2} \tfrac{1}{2}[n_\perp \delta(\omega-\omega_\perp)+(n_\perp+1)\delta(\omega+\omega_\perp)] , \tag{12}$$

$$\langle \tilde{S}^z_{\vec{q}} \tilde{S}^z_{-\vec{q}} \rangle_\omega = M \left[\frac{h-\gamma_{\vec{q}}}{h+\phi_{\vec{q}}} \right]^{1/2} \tfrac{1}{2}[n_\parallel \delta(\omega-\omega_\parallel)+(n_\parallel+1)\delta(\omega+\omega_\parallel)]\cos^2\theta + \frac{k_B T}{2J_0} \frac{1}{1-\phi_{\vec{q}}} \sin^2\theta\, \delta(\omega) , \tag{13}$$

where $n_\perp = (e^{\beta\omega_\perp}-1)^{-1}$ and $n_\parallel = (e^{\beta\omega_\parallel}-1)^{-1}$.

In the paramagnetic phase we must symmetrize between $\tilde{x}$ and $\tilde{y}$ in order to obtain the transverse correlation function,

$$\langle \tilde{S}^\perp_{\vec{q}\perp} \tilde{S}^\perp_{-\vec{q}} \rangle_\omega = \frac{J_0 M}{2\omega_\perp}[h(1+\sin^2\theta)+\gamma_{\vec{q}}(3\sin^2\theta-1)][n_\perp \delta(\omega-\omega_\perp)+(n_\perp+1)\delta(\omega+\omega_\perp)] + \frac{k_B T}{4J_0} \frac{1}{1+\phi_{\vec{q}}} \cos^2\theta\, \delta(\omega) . \tag{14}$$

The interpretation of this result is that the transverse part measures the antiferromagnetic mode $\omega_\perp$, which vanishes for $\gamma_{\vec{q}} = -1$ or at the zone boundary $q=\pi$; the longitudinal part $\langle \tilde{S}^\parallel_{\vec{q}} \tilde{S}^\parallel_{-\vec{q}} \rangle_\omega = \langle \tilde{S}^z_{\vec{q}} \tilde{S}^z_{-\vec{q}} \rangle_\omega$ measures the ferromagnetic mode $\omega_\parallel$, which vanishes at $q=0$, for small fields where $\theta < 90°$. Both parts have, in addition, a central-peak component, which in the present theory can be understood simply as a mixing in of locally longitudinal spin fluctuations χ^{zz}. Since $|\phi_{\vec{q}}| = |\gamma_{\vec{q}} \cos(2\theta)|$ is less than 1, the central peak does not diverge and becomes significant only when the respective spin-wave modes approach low frequencies. It may therefore be difficult to separate or detect a central peak, especially since all peaks must have a finite width. The width will be calculated subsequently by means of the correlation theory. In the RPA theory the spectrum is simply represented by a set of δ functions. In the next section we shall show that the position and width of these are such that the first-moment matrix of the relaxation function is correctly obtained, when the RPA susceptibility is correct. The RPA susceptibility is expected to be accurate when $|\gamma_{\vec{q}}|$ is not too close to 1 and for temperatures $k_B T < h$. Using (7) and $\chi = -2\pi i G(\omega=0)$ plus the local susceptibility approximation $\chi^{zz}_{AA} = 1/J_0$ and $\chi^{zz}_{AB} = 0$, one finds the total longitudinal susceptibility (5),

$$\tilde{\chi}^{zz}_{\vec{q}} = \frac{1}{J_0} \left[\frac{\cos^2\theta}{1+\phi_{\vec{q}}} + \sin^2\theta \right] . \tag{15}$$

In the present notation, Lovesey and Loveluck[1] found, near $\theta = \pi/4$, χ^z_t, which should be equal to $\chi^{zz}_{\vec{q}}$,

$$\chi^z_t = \frac{1}{J_0} \left[\frac{\cos^2\theta}{1+\phi_{\vec{q}}} + \frac{1}{\sin(2\theta)} \sin^2\theta \right] . \tag{16}$$

The two expressions agree exactly at $\theta = \pi/4$. Using the susceptibilities including the interactions (45), one finds

$$\tilde{\chi}^\parallel_{\vec{q}} = \frac{1}{J_0} \left[\frac{\cos^2\theta}{1+\phi_{\vec{q}}} + \frac{\sin^2\theta}{1-\phi_{\vec{q}}} \right] \to \frac{1}{J_0} \quad \text{when } \theta = \pi/4 . \tag{17}$$

This reduces to the noninteracting susceptibility $1/J_0$ for $\theta = \pi/4$, as would be expected when the two sublattices are perpendicular to each other. This implies a vanishing correlation length (or diverging inverse correlation length as formulated by Lovesey and Loveluck). In addition $\tilde{\chi}^\parallel_{\vec{q}}$ changes from antiferromagnetic to ferromagnetic behavior for fields larger than those corresponding to $\theta = \pi/4$. This was also emphasized by Lovesey and Loveluck.

IV. EXACT RELATIONS FOR THE FIRST AND SECOND MOMENTS

Let us consider a dynamical variable vector

$$\underline{A}^\dagger = \{(S^x_A)_{\vec{q}}, (S^x_B)_{\vec{q}}, (S^y_A)_{\vec{q}}, (S^y_B)_{\vec{q}}\}$$

and the transverse response matrix $(\underline{A}\, \underline{A}^\dagger)^\perp_{\vec{q}\omega}$. By the exact relations $\langle \underline{\omega} \rangle_\perp \underline{\chi}_\perp = \langle [\underline{A}, \underline{A}^\dagger] \rangle$ and

$$\langle \underline{\omega}^2 \rangle_\perp \underline{\chi}_\perp = \langle [\underline{A}, \mathscr{H}], \underline{A}^\dagger \rangle ,$$

one finds, straightforwardly from (6),

$$\langle \underline{\omega} \rangle_\perp \underline{\chi}_\perp \equiv iM \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ -1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{bmatrix} , \tag{18}$$

$$\langle \underline{\omega}^2\rangle_\perp \underline{\chi}_\perp \equiv J_0 M^2 \begin{bmatrix} h & \gamma_{\vec{q}} & 0 & 0 \\ \gamma_{\vec{q}} & h & 0 & 0 \\ 0 & 0 & h & -\phi_{\vec{q}} \\ 0 & 0 & -\phi_{\vec{q}} & h \end{bmatrix} + J_0 \begin{bmatrix} a_x' & b_x'\gamma_{\vec{q}} & 0 & 0 \\ b_x'\gamma_{\vec{q}} & a_x' & 0 & 0 \\ 0 & & cb_y' & -\phi_{\vec{q}} b_y' \\ 0 & 0 & -\phi_{\vec{q}} b_y' & cb_y' \end{bmatrix} . \tag{19}$$

By (19) we have proved rigorously that $\chi^{xy}_{NM} = \chi^{yx}_{NM} = 0$ for any $N,M = A,B$. Similarily, for a dynamical vector $\underline{B}^\dagger = \{(S^z_A)_{\vec{q}}, (S^z_B)_{\vec{q}}\}$, one finds for the longitudinal response matrix $(\underline{B}\,\underline{B}^\dagger)_{\vec{q}\omega}$,

$$\langle \underline{\omega}\rangle_{||} \underline{\chi}_{||} \equiv \begin{bmatrix} 0 & 0 \\ 0 & 0 \end{bmatrix} , \tag{20}$$

$$\langle \underline{\omega}^2\rangle_{||} \underline{\chi}_{||} \equiv \begin{bmatrix} a_z' & b_z'\gamma_{\vec{q}} \\ b_z'\gamma_{\vec{q}} & a_z' \end{bmatrix} . \tag{21}$$

The results involve a number of pair-correlation terms which will be defined as

$$c_{xx}' = \frac{1}{N}\sum_{\vec{k}} \gamma_{\vec{k}} \langle S^x_A S^x_B\rangle_{\vec{k}}, \quad c_{xx} = \frac{1}{N}\sum_{\vec{k}} \gamma^2_{\vec{k}} \langle S^x_A S^x_A\rangle_{\vec{k}} ,$$

$$c_{yy}' = \frac{1}{N}\sum_{\vec{k}} \gamma_{\vec{k}} \langle S^y_A S^y_B\rangle_{\vec{k}}, \quad c_{yy} = \frac{1}{N}\sum_{\vec{k}} \gamma^2_{\vec{k}} \langle S^y_A S^y_A\rangle_{\vec{k}} , \tag{22}$$

$$c_{zz}' = \frac{1}{N}\sum_{\vec{k}} \gamma_{\vec{k}} \langle \delta S^z_A\, \delta S^z_B\rangle_{\vec{k}}, \quad c_{zz} = \frac{1}{N}\sum_{\vec{k}} \gamma^2_{\vec{k}} \langle \delta S^z_A\, \delta S^z_A\rangle_{\vec{k}} .$$

They represent essentially nearest-neighbor correlations between different sublattices (conventionally denoted by a prime) and within the same sublattices. They appear in the following combinations:

$$\begin{aligned} &a_x = s^2 c_{xx} + c_{yy} + c^2 c_{zz}, \quad a_x' = -c_{yy}' + c c_{zz}' , \\ &b_x = c(c_{yy} + c_{zz}), \quad b_x' = -c c_{yy}' + c_{zz}' , \\ &a_z = c^2 c_{xx} + c_{yy} + s^2(c_{zz} + M^2/\rho), \quad a_z' = -c_{yy}' + c c_{xx}' , \\ &b_z = c(c_{yy} + c_{xx}), \quad b_z' = -c c_{yy}' + c_{xx}' , \\ &a_y = (c^2 + s^2)(c_{xx} + c_{zz}) + s^2 M^2/\rho, \quad b_y' = c_{xx}' + c_{zz}' , \\ &c = \cos(2\theta), \quad s = \sin(2\theta) . \end{aligned} \tag{23}$$

The susceptibility tensors at low temperatures, neglecting correlation effects, may be obtained using (7),

$$\underline{\chi}^{\mathrm{RPA}}_\perp = \frac{1}{J_0} \begin{bmatrix} h & \phi_{\vec{q}} & 0 & 0 \\ \phi_{\vec{q}} & h & 0 & 0 \\ 0 & 0 & h & -\gamma_{\vec{q}} \\ 0 & 0 & -\gamma_{\vec{q}} & h \end{bmatrix} \begin{bmatrix} (h^2-\phi^2_{\vec{q}})^{-1} & 0 & 0 & 0 \\ 0 & (h^2-\phi^2_{\vec{q}})^{-1} & 0 & 0 \\ 0 & 0 & (h^2-\gamma^2_{\vec{q}})^{-1} & 0 \\ 0 & 0 & 0 & (h^2-\gamma^2_{\vec{q}})^{-1} \end{bmatrix} , \tag{24}$$

$$(\underline{\chi}_\perp^{-1})^{\mathrm{RPA}} = J_0 \begin{bmatrix} h & -\phi_{\vec{q}} & 0 & 0 \\ -\phi_{\vec{q}} & h & 0 & 0 \\ 0 & 0 & h & \gamma_{\vec{q}} \\ 0 & 0 & \gamma_{\vec{q}} & h \end{bmatrix} . \tag{25}$$

We notice that $\chi^{\mathrm{RPA}}_\perp$ is independent of the temperature. From (18) and (19), neglecting the correlation term, one finds, by multiplication with (25),

$$\langle \underline{\omega}\rangle^{\mathrm{RPA}}_\perp = i J_0 M \begin{bmatrix} 0 & 0 & h & \gamma_{\vec{q}} \\ 0 & 0 & \gamma_{\vec{q}} & h \\ -h & \phi_{\vec{q}} & 0 & 0 \\ \phi_{\vec{q}} & -h & 0 & 0 \end{bmatrix} , \tag{26}$$

$$\langle \underline{\omega}^2\rangle^{\mathrm{RPA}}_\perp = \langle \underline{\omega}\rangle^2_\perp = (J_0 M)^2 \begin{bmatrix} A_{\vec{q}} & B_{\vec{q}} & 0 & 0 \\ B_{\vec{q}} & A_{\vec{q}} & 0 & 0 \\ 0 & 0 & A_{\vec{q}} & B_{\vec{q}} \\ 0 & 0 & B_{\vec{q}} & A_{\vec{q}} \end{bmatrix} , \tag{27}$$

where $A_{\vec{q}} = h^2 - \gamma_{\vec{q}}\phi_{\vec{q}}$ and $B_{\vec{q}} = \gamma_{\vec{q}} h[1-\sin(2\theta)]$. The diagonalization of (26) or (27) gives the RPA frequencies $\omega_{||}$ and $\omega_\perp$ [Eq. (8)]. Clearly, the RPA frequency for $\chi_{||}(\vec{q},\omega)$ is zero according to (20). By investigating the

remaining terms in (19) and (21) one observes that the fluctuation corrections of the second moment are quite similar for the various variables.

At high temperatures the susceptibility may be obtained by the high-temperature expansion. One finds

$$\chi_\perp \to \frac{1}{J_0 t}\begin{bmatrix} 1 & \phi_{\vec{q}}/t & 0 & 0 \\ \phi_{\vec{q}}/t & 1 & 0 & 0 \\ 0 & 0 & 1 & -\gamma_{\vec{q}}/t \\ 0 & 0 & -\gamma_{\vec{q}}/t & 1 \end{bmatrix} \text{ as } T\to\infty \ , \tag{28}$$

where $t=T/\Theta_{\rm CW}$, and $\Theta_{\rm CW}=J_0 S(S+1)/3k_B$ is the Curie-Weiss constant. It is straightforward, but very laborious, to derive the expansion to higher order in $1/t$. Here, for simplicity, we already neglect the $1/t^3$ terms in the diagonal. The structure of (28) is the same as (24), and the first-moment matrix can be diagonalized to yield the eigenvalues for large T,

$$\begin{aligned} \omega_{\parallel} &= \pm J_0 M[(t-\gamma_{\vec{q}})(t+\phi_{\vec{q}})]^{1/2} \ , \\ \omega_\perp &= \pm J_0 M[(t+\gamma_{\vec{q}})(t-\phi_{\vec{q}})]^{1/2} \ . \end{aligned} \tag{29}$$

Since $t=1$ for $T=\Theta_{\rm CW}$ we conclude, by comparing with (8), that the RPA theory might be adequate for $T\sim\Theta_{\rm CW}$. For $T\gg\Theta_{\rm CW}$ one has $M=H\chi_\parallel\to H/J_0 t$ and the frequencies (29) reduce to the Larmor frequencies in the applied field H. In this limit a δ-function representation of the spectrum is only relevant for $H\gg k_B T$.

V. CALCULATION OF THE SUSCEPTIBILITY TENSOR INCLUDING PAIR-CORRELATION EFFECTS

We now need to calculate the static susceptibility more accurately, including correlation effects. This is done in the correlation theory by going to second order in the equation of motion for the Green's functions, which give the identity

$$(\ddot{A}B)=\langle[[A,\mathscr{H}],B]\rangle \ . \tag{30}$$

Using a Hartree-Fock–type decoupling of the four-spin relaxation function

$$(S^\alpha S^\beta S^\alpha S^\beta)\sim\langle S^\alpha S^\alpha\rangle(S^\beta S^\beta) \ ,$$

which is equivalent to the mode-mode decoupling to be used for the dynamic properties, one finds, from $(\ddot{S}^\alpha S^\alpha)$ for $\alpha=x$, y, and z, the equations

$$\begin{bmatrix} A^\alpha_{\vec{q}} & B^\alpha\gamma_{\vec{q}} \\ B^\alpha\gamma_{\vec{q}} & A^\alpha_{\vec{q}} \end{bmatrix}\begin{bmatrix} \chi^{\alpha\alpha}_{AA} \\ \chi^{\alpha\alpha}_{AB} \end{bmatrix} = \begin{bmatrix} P^\alpha \\ Q^\alpha \end{bmatrix} \ , \tag{31}$$

where

$$\begin{aligned} A^x_{\vec{q}} &= J_0^2[M^2(h^2-\gamma_{\vec{q}}\phi_{\vec{q}})+a_x-\gamma^2_{\vec{q}}cb'_x+s^2\gamma^2_{\vec{q}}c'_{yy}] \ , \\ P^x &= J_0(M^2h+a'_x) \\ B^x &= J_0^2[M^2h(1-c)+b_x-ca'_x+s^2(2c'_x+c'_{zz})] \ , \\ Q^x &= J_0(M^2+b'_x) \\ A^y_{\vec{q}} &= J_0^2[M^2(h^2-\gamma_{\vec{q}}\phi_{\vec{q}})+a_y-\gamma^2_{\vec{q}}cb'_y] \ , \\ P^y &= J_0(M^2h+cb'_y) \\ B^y &= J_0^2[M^2h(1-c)-a_y+cb'_y] \ , \quad Q^y=-cJ_0(M^2+b'_y) \\ A^z_{\vec{q}} &= J_0^2(a_z-\gamma^2_{\vec{q}}cb'_z+s^2\gamma^2_{\vec{q}}c'_{yy}) \ , \quad P^z=J_0a'_z \\ B^z &= J_0^2[b_z-ca'_z+s^2(c'_{xx}+2c'_{zz})] \ , \quad Q^z=J_0b'_z \ . \end{aligned} \tag{32}$$

The solutions of (31) for the matrix elements $\chi^{\alpha\alpha}_{NM}$ of the susceptibility tensor are given by

$$\chi^\alpha_\pm=\chi^{\alpha\alpha}_{AA}\pm\chi^{\alpha\alpha}_{AB}=\frac{P^\alpha\pm Q^\alpha\gamma_{\vec{q}}}{A^\alpha_{\vec{q}}\pm B^\alpha\gamma_{\vec{q}}}=(\bar{\chi}^{\alpha\alpha}_{AA}\pm\bar{\chi}^{\alpha\alpha}_{AB})^{-1} \ , \tag{33}$$

where $\bar{\chi}^{\alpha\alpha}_{NM}$ is an element of the inverse susceptibility matrix. χ^α_+ and χ^α_- represent the physically important components of the uniform and staggered susceptibility matrices, which are diagonal. It is clear from (33) that $\chi^\alpha_+(\gamma_{\vec{q}})=\chi^\alpha_-(-\gamma_{\vec{q}})$.

The results (32) and (33) reduce to the exact high-temperature results (28) for $T\to\infty$, and to the RPA results (27) and (24) if the pair-correlation terms $c_{\alpha\alpha}$ and $c'_{\alpha\alpha}$ are neglected. Let us discuss a number of limiting cases of the quite general solutions (33).

A. Ferromagnet at all T with or without a magnetic field

For this case $\theta=90°$ or $c=-1$, and $s=0$. The ferromagnetic Hamiltonian is obtained by replacing $J_0\to-(J_0)$ and $-h$ by $h'=H/|J_0M|+1$. We then obtain, from (23) and (33),

$$\begin{aligned} \chi^x_+ &= \chi^y_+=\frac{1}{|J_0|}\frac{M^2(h'-\gamma_{\vec{q}})+b'_x(1-\gamma_{\vec{q}})}{M^2(h'-\gamma_{\vec{q}})^2+(a_x-\gamma_{\vec{q}}b'_x)(1-\gamma_{\vec{q}})} \ , \\ \chi^z_+ &= \frac{1}{|J_0|}\frac{1}{a_z/b'_z-\gamma_{\vec{q}}} \ . \end{aligned} \tag{34}$$

The total susceptibility components (5) are $\tilde{\chi}^{xx}_{\vec{q}}=\tilde{\chi}^{yy}_{\vec{q}}=\chi^x_+$, and $\tilde{\chi}^{zz}_{\vec{q}}=\chi^z_+$. For the external field $H=0$ we find

$$\chi^x_+=\chi^y_+=\frac{1}{|J_0|}\frac{1}{R_y-\gamma_{\vec{q}}} \ , \tag{35}$$

where

$$R_y=(M^2+a_y)/(M^2+b'_y) \ .$$

Equations (34) and (35) also apply in the ordered phase. The transition temperature is given by $R_y=1$, or $c_{xx}+c_{zz}=c'_{xx}+c'_{zz}$, which is the same condition as found previously for the Heisenberg paramagnet.[3]

B. Antiferromagnet in no magnetic field for any T

For this case, $\theta=0°$ or $c=1$, and $s=0$ and $h=1$. In the paramagnetic case, $M=0$. From (23) and (33) we find

$$\chi^x_+=\frac{1}{J_0}\frac{1}{R_x-\gamma_{\vec q}},\quad \chi^y_+=\frac{1}{J_0}\frac{1}{R_y+\gamma_{\vec q}},$$
$$\chi^z_+=\frac{1}{J_0}\frac{1}{R_z-\gamma_{\vec q}}, \tag{36}$$

where

$$R_\alpha=(M^2+a_\alpha)/(M^2+b'_\alpha)$$

for $\alpha=x$, y, and z. By symmetry, $R_x=R_y$. The Néel temperature is obtained by the condition $R_y=1$, or when $c_{xx}+c_{zz}=c'_{xx}+c'_{zz}$. There is full symmetry between the x and z components. The χ^α_- components are obtained by changing the sign of $\gamma_{\vec q}$. The total susceptibility components, Eq. (5), are $\tilde\chi^{xx}_{\vec q}=\tilde\chi^{zz}_{\vec q}=\chi^x_-$, and $\tilde\chi^{yy}_{\vec q}=\chi^y_+$. In the paramagnetic phase they are all equal and diverge at T_N for $q=\pi$ or $\gamma_{\vec q}=-1$.

C. Antiferromagnet in a large magnetic field

When the induced moment along the field is large, the angle $\theta=90°$ or $c=-1$, and $s=0$, as for the ferromagnetic case, but now $h=H/J_0M-1$. Using (23) and (33) we find

$$\chi^x_+=\chi^y_+=\frac{1}{J_0}\frac{M^2(h+\gamma_{\vec q})-b'_y(1-\gamma_{\vec q})}{M^2(h+\gamma_{\vec q})^2+(a_y-\gamma_{\vec q}b'_y)(1-\gamma_{\vec q})},$$
$$\chi^z_+=\frac{1}{J_0}\frac{1}{\gamma_{\vec q}-a_z/b'_z}. \tag{37}$$

The total susceptibility components (5) are $\tilde\chi^{xx}_{\vec q}=\tilde\chi^{yy}_{\vec q}=\chi^x_+$, and $\tilde\chi^{zz}_{\vec q}=\chi^z_+$.

D. Antiferromagnet in a small magnetic field for any temperature

Near T_N and below, the angle θ is between 0° and 90°. In this case both c and s are finite and θ is determined such that $h=1$ if we use (2). We find

$$\chi^y_+=\frac{1}{J_0}\frac{1}{R^y_{\vec q}+\gamma_{\vec q}},\quad R^y_{\vec q}=\frac{M^2(1-\phi_{\vec q})+a_y(1-\gamma_{\vec q})}{M^2(1-\phi_{\vec q})+cb'_y(1-\gamma_{\vec q})}. \tag{38}$$

When $a_y\geq cb'_y$ or $R^y_{\vec q}\geq 1$, the ordering wave vector at which the χ^y_+ diverge is $\vec q=\vec q_{\rm zone}$. The Néel temperature is given by

$$a_y=cb'_y \text{ or } c_{xx}+c_{zz}+s^2M^2/\rho=c(c'_{xx}+c'_{zz}).$$

For small fields we may write the equation as

$$c^0_{xx}+c^0_{zz}=(1-KH^2)(c^{0\prime}_{xx}+c^{0\prime}_{zz})$$

where K is a constant and the superscript 0 indicates functions calculated in zero field. This gives the classical result $T_N(0)-T_N(H)\sim H^2$ if the short-range correlation functions are assumed to depend linearly on T near $T_N(0)$. However, the self-consistent calculation may modify both the exponent 2 and the sign of the constant K. By expanding near $\vec q=\vec q_{\rm zone}$ and writing $\gamma_{\vec q}=-1+q^2$ we have

$$\chi^y_+=\frac{1}{J_0}\frac{1}{\kappa_1^2+q^2},\quad \kappa_1^2=\frac{a_y-cb'_y}{M^2(1+c)+2cb'_y}, \tag{39}$$

where κ_1 is the inverse correlation length.

In the case when $a_y\leq cb'_y$, $R^y_{\vec q}$ is less or equal to 1 and the ordering may occur at an incommensurate wave vector. This will be discussed further in the numerical section. For the x and z components we may write

$$\chi^x_+=\frac{1}{J_0}\frac{1}{R^x_{\vec q}-\phi_{\vec q}},\quad \chi^z_-=\frac{1}{J_0}\frac{1}{R^z_{\vec q}+\phi_{\vec q}}, \tag{40}$$

where

$$R^x_{\vec q}=\frac{(M^2+a_x+\gamma_{\vec q}s^2c'_{yy})(1+\gamma_{\vec q})-c\gamma_{\vec q}[b_x-a_x+s^2(2c'_{xx}-c'_{yy}+c'_{zz})]}{(M^2+a'_x)(1+\gamma_{\vec q})+\gamma_{\vec q}(b'_x-a'_x)}, \tag{41}$$

$$R^z_{\vec q}=\frac{(a_z+\gamma_{\vec q}s^2c'_{yy})(1+\gamma_{\vec q})-\gamma_{\vec q}[b_z+a_z+s^2(c'_{xx}-c'_{yy}+2c'_{zz})]}{a'_z(1+\gamma_{\vec q})-\gamma_{\vec q}(b'_z+a'_z)}. \tag{42}$$

Here $R^x_{\vec q=\pi}=-c$ and $R^z_{\vec q=\pi}=c$ for $\gamma_{\vec q}=-1$, when the following relations between the correlation functions hold at $T=T_N$:

$$\begin{aligned}
c_{yy}&=(c'_{xx}-c'_{zz})/2,\\
c_{xx}&=2c'_{yy}\cos^2\theta+(1+\cos^2\theta)(c'_{xx}+c'_{zz})\\
&\quad-(2\cos^2\theta-1)M^2_{||}/\rho,\\
c_{zz}&=-2c'_{yy}\cos^2\theta-(1+\sin^2\theta)(c'_{xx}+c'_{zz})\\
&\quad-(1+2\cos^2\theta)M^2_{||}/\rho,
\end{aligned} \tag{43}$$

where $M_{||}=M\sin\theta$ is the moment induced along the applied field. The susceptibilities (40) and the relations (43) ensure that the symmetry relation

$$\tilde\chi^{yy}_{\vec q}=\tilde\chi^{xx}_{\vec q}=\sin^2\theta\chi^x_++\cos^2\theta\chi^z_-=\frac{1}{J_0}\frac{1}{1+\gamma_{\vec q}}$$

holds at T_N for $\vec q$ close to the zone boundary. Since χ^x_- and χ^z_+ are obtained from (40)–(42) by changing the sign of $\gamma_{\vec q}$ we find, at T_N,

$$\tilde{\chi}^{zz}_{\vec{q}}=\cos^2\theta\chi^x_-+\sin^2\theta\chi^z_+=-\frac{1}{J_0}\frac{1}{1-\gamma_{\vec{q}}}\ . \tag{44}$$

This shows that the longitudinal susceptibility is simultaneously divergent for $q=0$. The negative sign means that the induced moment along the field increases with a vertical slope at T_N with increasing temperatures. The symmetry requirement that $\tilde{\chi}^{yy}_{\vec{q}}=\tilde{\chi}^{xx}_{\vec{q}}$ for all $\vec{q}$ and T gives (if at all possible to fulfill in the present theory) even more strict relations between the correlation functions than (43), which contains the weaker condition $a'_y=cb'_y$ for $T=T_N$. At low temperatures $T\to 0$, where the correlation corrections vanish, $R^{\alpha}_{\vec{q}}\to 1$ and $R^{y}_{\vec{q}}\to 1$, and we recover the RPA solutions for χ^x_+ and χ^y_+. The RPA χ^z_+ is, however, modified to take into account the interactions. Let us call this an improved RPA result,

$$\chi^z_+=\frac{1}{J_0}\frac{1}{1-\phi_{\vec{q}}},\quad \chi^z_-=\frac{1}{J_0}\frac{1}{1+\phi_{\vec{q}}}\ . \tag{45}$$

It is interesting that we incorrectly would obtain the RPA result even if the correlation corrections were not assumed to be small, but just assumed to obey the relations found in the RPA theory (9) and (23),

$$(c'_{xx})_{\rm RPA}=cc_{xx},\quad (c'_{yy})_{\rm RPA}=-c_{yy},\quad c'_{zz}=cc_{zz}\ . \tag{46}$$

If we now neglect only terms proportional to s^2, all primed and unprimed quantities in (23) are equal, and in (38), and (41) and (42), $R^x_{\vec{q}}=R^y_{\vec{q}}=R^z_{\vec{q}}=1+\sigma(s^2)$. What we should learn from this is that the introduction of RPA relations and simplifying assumptions in a theory which includes correlation effects may seriously impair the obtainable results.

VI. DYNAMICAL PROPERTIES

Let us list a number of exact results. The exact first-moment matrix $\langle\underline{\omega}\rangle$ is obtained from (18) by multiplying with an exact $\underline{\chi}_\perp^{-1}$. The diagonalization of $\langle\underline{\omega}\rangle$ gives the exact eigenvalues in terms of the diagonal susceptibility components defined in (33),

$$\begin{aligned}\omega_{||}&=\omega_-=\pm M[(\bar{\chi}^{xx}_{AA}-\bar{\chi}^{xx}_{AB})(\bar{\chi}^{yy}_{AA}-\bar{\chi}^{yy}_{AB})]^{1/2}\\&=\pm M(\chi^x_-\chi^y_-)^{-1/2}\ ,\\ \omega_\perp&=\omega_+=\pm M[(\bar{\chi}^{xx}_{AA}+\bar{\chi}^{xx}_{AB})(\bar{\chi}^{yy}_{AA}+\bar{\chi}^{yy}_{AB})]^{1/2}\\&=\pm M(\chi^x_+\chi^y_+)^{-1/2}\ .\end{aligned} \tag{47}$$

It is sometimes convenient in the following to replace the index $||$ by $-$ and $\perp$ by $+$. If we insert the RPA susceptibility components (25) we recover the RPA frequencies (8). The exact first-moment matrix squared is simply

$$\langle\underline{\omega}\rangle^2=\begin{bmatrix} w & w' & 0 & 0\\ w' & w & 0 & 0\\ 0 & 0 & w & w'\\ 0 & 0 & w' & w\end{bmatrix}, \tag{48}$$

$$\begin{aligned}w&=M^2(\bar{\chi}^{xx}_{AA}\bar{\chi}^{yy}_{AA}+\bar{\chi}^{xx}_{AB}\bar{\chi}^{yy}_{AB})=(\omega_\perp^2+\omega_{||}^2)/2\ ,\\ w'&=M^2(\bar{\chi}^{xx}_{AB}\bar{\chi}^{yy}_{AA}+\bar{\chi}^{xx}_{AA}\bar{\chi}^{yy}_{AB})=(\omega_\perp^2-\omega_{||}^2)/2\ .\end{aligned} \tag{49}$$

The RPA expression (27) again checks with this.

In the notation introduced in the preceding section, (23), (38), and (40), we can write the exact second-moment matrix (19) as

$$\langle\underline{\omega}^2\rangle=\begin{bmatrix} w_x & w'_x & 0 & 0\\ w'_x & w_x & 0 & 0\\ 0 & 0 & w_y & w'_y\\ 0 & 0 & w'_y & w_y\end{bmatrix}, \tag{50}$$

where

$$\begin{aligned}w_x&=J_0(M^2h+a'_x)\bar{\chi}^{xx}_{AA}+J_0(M^2+b'_x)\gamma_{\vec{q}}\bar{\chi}^{xx}_{AB}\ ,\\ w'_x&=J_0(M^2h+a'_x)\bar{\chi}^{xx}_{AB}+J_0(M^2+b'_x)\gamma_{\vec{q}}\bar{\chi}^{xx}_{AA}\ ,\\ w_y&=J_0(M^2h+cb'_y)\bar{\chi}^{yy}_{AA}-J_0(M^2+b'_y)\phi_{\vec{q}}\bar{\chi}^{yy}_{AB}\ ,\\ w'_y&=J_0(M^2h+cb'_y)\bar{\chi}^{yy}_{AB}-J_0(M^2+b'_y)\phi_{\vec{q}}\bar{\chi}^{yy}_{AA}\ .\end{aligned} \tag{51}$$

The exact result for the dynamical response matrix for the variable vector $\underline{A}$, introduced in Sec. IV, is formally solved by the Mori theory.[2] The Laplace transform is

$$(\underline{A}\,\underline{A}^\dagger)_{\vec{q},z}=\underline{\chi}_\perp[z\underline{1}-i\langle\underline{\omega}\rangle+(\underline{A}_1\underline{A}_1^\dagger)^{\mathscr{L}^{-1}}_{\vec{q},z}\underline{\chi}_\perp^{-1}]^{-1}\ , \tag{52}$$

where $z=i\omega+\epsilon$ (we use the notation z' if there is a possibility for confusion with the coordinate z) and $\underline{A}_1$ is the random-force variable. $\underline{A}_1$ is the part of $-i\dot{\underline{A}}=[\mathscr{H},\underline{A}]=\mathscr{L}\underline{A}$ which is orthogonal to $\underline{A}$. Using the Mori projection operator $\mathscr{P}$, which acts as $\mathscr{P}\underline{B}=(\underline{B}\,\underline{A}^\dagger)(\underline{A}\,\underline{A}^\dagger)^{-1}\underline{A}$, one writes $\underline{A}_1=(1-\mathscr{P})\mathscr{L}\underline{A}=\mathscr{L}\underline{A}-\langle\underline{\omega}\rangle\underline{A}$. The random-force response matrix can be written as follows:

$$(\underline{A}_1\underline{A}_1^\dagger)^{\mathscr{L}^{-1}}_{\vec{q},z}\underline{\chi}_\perp^{-1}=\begin{bmatrix}\Delta_xF_x(z) & \Delta'_xF_x(z) & 0 & 0\\ \Delta'_xF_x(z) & \Delta_xF_x(z) & 0 & 0\\ 0 & 0 & \Delta_yF_y(z) & \Delta'_yF_y(z)\\ 0 & 0 & \Delta'_yF_y(z) & \Delta_yF_y(z)\end{bmatrix}. \tag{53}$$

The spectral weight functions $F_\alpha(z)$ are normalized; i.e., $\int_{-\infty}^{\infty}F_\alpha(\omega)d\omega=1$. By integration of (53) one therefore has by definition, that the coefficients Δ^ν_α are the matrix elements of $\langle\underline{\omega}^2\rangle-\langle\underline{\omega}\rangle^2$: i.e., $\Delta_\alpha=w-w_\alpha$ and $\Delta'_\alpha=w'-w'_\alpha$ for $\alpha=x,y$. As carefully discussed by Mori, the time evolution of the random forces is not governed by the total Hamiltonian, but must be considered in a rotating coordinate system, which follows the regular spin procession. This ensures that the orthogonality $(\underline{A}_1(t)\underline{A}^\dagger)=0$ holds at all times. Therefore $F_\alpha(z)$ must be calculated using the modified Liouville operator $\mathscr{L}_1=(1-\mathscr{P})\mathscr{L}$, where the regular precession is projected out from the original $\mathscr{L}$. The $(\underline{A}_1\underline{A}_1^\dagger)$ matrix is 2×2 block diagonal for x,y (and also z). Let us consider only one such block and write, for $\alpha=x,y$ (and also z),

$$(\underline{A},\underline{A}_1^\dagger)_z\,|\,_\alpha=\begin{bmatrix}X_\alpha(z) & X'_\alpha(z)\\ X'_\alpha(z) & X_\alpha(z)\end{bmatrix} \text{ and } (\underline{A}_1\underline{A}_1^\dagger)^{-1}=\frac{1}{X_\alpha^2-(X'_\alpha)^2}\begin{bmatrix}X_\alpha & -X'_\alpha\\ -X'_\alpha & X_\alpha\end{bmatrix}, \tag{54}$$

where

$$X_\alpha(z)=([1-\mathscr{P}](\dot{S}_A^\alpha)_{\vec{q}}\,|\,[1-\mathscr{P}](\dot{S}_A^\alpha)_{\vec{q}}^\dagger)_z^{\mathscr{L}^{-1}},\quad X'_\alpha(z)=([1-\mathscr{P}](\dot{S}_A^\alpha)_{\vec{q}}\,|\,[1-\mathscr{P}](\dot{S}_B^\alpha)_{\vec{q}}^\dagger)_z^{\mathscr{L}^{-1}}. \tag{55}$$

By definition, $X_\alpha=\int_{-\infty}^{\infty}X_\alpha(\omega)d\omega$ and $X'_\alpha=\int_{-\infty}^{\infty}X'_\alpha(\omega)d\omega$. Then,

$$(\underline{A}_1\underline{A}_1^\dagger)_z\underline{\chi}^{-1}=(\underline{A}_1\underline{A}_1^\dagger)_z(\underline{A}_1\underline{A}_1^\dagger)^{-1}(\underline{A}_1\underline{A}_1^\dagger)(\underline{A}\,\underline{A}^\dagger)^{-1}=(\underline{A}_1\underline{A}_1^\dagger)_z(\underline{A}_1\underline{A}_1^\dagger)^{-1}(\langle\underline{\omega}^2\rangle-\langle\underline{\omega}\rangle^2)$$
$$=\frac{1}{X_\alpha^2-(X'_\alpha)^2}\begin{bmatrix}X_\alpha(z) & X'_\alpha(z)\\ X'_\alpha(z) & X_\alpha(z)\end{bmatrix}\begin{bmatrix}X_\alpha & -X'_\alpha\\ -X'_\alpha & X_\alpha\end{bmatrix}\begin{bmatrix}\Delta_\alpha & \Delta'_\alpha\\ \Delta'_\alpha & \Delta_\alpha\end{bmatrix}. \tag{56}$$

By comparing this with (53), we find that

$$F_\alpha(z)=[X_\alpha X_\alpha(z)-X'_\alpha X'_\alpha(z)]/[X_\alpha^2-(X'_\alpha)^2], \tag{57}$$

since $X_\alpha X'_\alpha(z)-X'_\alpha X_\alpha(z)=0$ because $\int_{-\infty}^{\infty}\omega F(\omega)d\omega=0$ and

$$\int_{-\infty}^{\infty}X_\alpha X'_\alpha(\omega)-X'_\alpha X_\alpha(\omega)d\omega=0.$$

The frequency dependence of $F_\alpha(z)$ may be quite complicated, and must be such that all moments of $(\underline{A}\,\underline{A}^\dagger)_{\vec{q},z}$ are finite.

A. Simplifying approximation for the frequency dependence

It was found for the Heisenberg magnet that it is a great simplification and a good approximation for the frequency interval of interest to parametrize $F_\alpha(z)$ as a single Lorentzian.[3] This corresponds to assuming an exponential decay of the random-force correlation functions (for intermediate times) with the decay time $1/K_\alpha$. We write

$$F_\alpha(z)=1/(z+K_\alpha), \tag{58}$$

where K_α is assumed to be nearly independent of z for frequencies $0<|\omega|<\max(\omega_{||},\omega_\perp)$. Inserting (58) in (53) gives a solution for each component with eight poles and such that the second moment is finite. The spectrum could be cut off at high frequencies to yield the fourth moment and to make all higher moments finite. As discussed for the Heisenberg paramagnet,[3] this procedure does not give any qualitatively different results from the simple pole approximation (58), but may give up to a 40% increase of the calculated linewidths. The consequences of a cutoff will not be considered further in the present case.

It is convenient to consider the diagonal uniform and staggered susceptibility components χ_+^α and χ_-^α. The matrices are obtained by transforming each 2×2 matrix $\underline{M}$ in the preceding section to $\underline{M}_d=\underline{T}^{-1}\underline{M}\,\underline{T}$, where

$$\underline{T}=\begin{bmatrix}1 & -1\\ 1 & 1\end{bmatrix}\Big/\sqrt{2}.$$

The first-moment matrix $\langle\underline{\omega}\rangle\underline{\chi}$ does not change by this transformation, but remains as (18).

The frequency dependence for the z,z susceptibility components is then, using (52) and (56),

$$\chi_\pm^z(\vec{q},z')=\chi_\pm^z\frac{1}{z'+(\Delta_z\pm\Delta'_z)/(z'+K_z)}, \tag{59}$$

where, according to (20) and (21), $\Delta_z\pm\Delta'_z=a'_z\pm b'_z\gamma_{\vec{q}}$. If $\Delta_z\pm\Delta'_z\ll K_z$, (59) simplifies to

$$\chi_\pm^z(\vec{q},z')=\chi_\pm^z(q)\frac{1}{z'+\Gamma_\pm^z}\quad\text{where } \Gamma_\pm^z=(\Delta_z\pm\Delta'_z)/K_z. \tag{60}$$

The frequency dependence for the transverse, diagonal susceptibility components is obtained by transforming (52) and (53) by $\underline{T}$. Explicitly, we find, for $(\underline{A}\,\underline{A}^\dagger)_{\vec{q},z}$ a transverse, diagonal

$$\begin{bmatrix}\chi_+^x(\vec{q}) & 0 & 0 & 0\\ 0 & \chi_-^x(\vec{q}) & 0 & 0\\ 0 & 0 & \chi_+^y(\vec{q}) & 0\\ 0 & 0 & 0 & \chi_-^y(\vec{q})\end{bmatrix}\begin{bmatrix}z+\Sigma_+^x & 0 & iV_+^y & 0\\ 0 & z+\Sigma_-^x & 0 & iV_-^y\\ -iV_+^x & 0 & z+\Sigma_+^x & 0\\ 0 & -iV_-^x & 0 & z+\Sigma_-^y\end{bmatrix}^{-1}, \tag{61}$$

where for $\alpha=x,y$,

$$\Sigma_\pm^\alpha=\frac{\Delta_\alpha\pm\Delta'_\alpha}{z+K_\alpha}\quad\text{and}\quad V_\pm^\alpha=M/\chi_\pm^\alpha(\vec{q}). \tag{62}$$

If we, for simplicity, use the RPA static $\chi_\pm^\alpha(\vec{q})$, we find from (19) and (27) that

$$\Delta_x\pm\Delta'_x=a'_x\pm b'_x\gamma_{\vec{q}}\quad\text{and}\quad\Delta_y\pm\Delta'_y=cb'_y(1\mp\gamma_{\vec{q}}). \tag{63}$$

For a more accurate determination of the weight factors for $\Sigma_\pm^\alpha$ in (61) one should, of course, use the correlation theory susceptibilities $\chi_\pm^\alpha(\vec{q})$ from (33). We notice that (61) can be block-diagonalized and can represent two

independent families of modes belonging to $\omega_\perp$ and $\omega_\parallel$, each described by four poles. Another independent family of modes is the longitudinal $\alpha=z$, which is also described by four poles. A line shape described by four poles is clearly still quite general. Let us therefore discuss some limiting cases, which also are useful for setting up the self-consistency equations for determining the damping constant K_α.

B. Limiting cases

In the case when the amplitude of the random forces is small compared to the mean forces, $\langle\omega^2\rangle-\langle\omega\rangle^2\ll\langle\omega\rangle^2$, the spectrum is only slightly modified from the δ-function spectrum at $\pm\omega_\perp$ and $\pm\omega_\parallel$. Consequently, one finds that the poles of (52) simplify to four complex poles and, in addition, one central, purely imaginary pole at

$$\omega=i\left[\sum_\alpha[2-(\Delta_\alpha+\Delta'_\alpha)/\omega_\perp^2-(\Delta_\alpha-\Delta'_\alpha)/\omega_\parallel^2]/K_\alpha\right]^{-1}\cong iK\ , \tag{64}$$

where $1/K=2(1/K_x+1/K_y)$. The central peak is approximately Lorentzian with a half width $\sim K$. If we introduce only this average decay constant, the renormalized and damped regular precession modes are given by the complex poles:

$$\begin{aligned}\tilde\omega_\perp&=\pm\omega_\perp+(\pm\omega_\perp+iK)\sum_\alpha(\Delta_\alpha+\Delta'_\alpha)/2(\omega_\perp^2+K^2)\\&=\pm\hat\omega_\perp+i\Gamma_\perp\ ,\\\tilde\omega_\parallel&=\pm\omega_\parallel+(\pm\omega_\parallel+iK)\sum_\alpha(\Delta_\alpha-\Delta'_\alpha)/2(\omega_\parallel^2+K^2)\\&=\pm\hat\omega_\parallel+i\Gamma_\parallel\ .\end{aligned} \tag{65}$$

We notice that the half widths $\Gamma_\perp$ and $\Gamma_\parallel$ are smaller than that of the central peak, since by assumption, $\Delta_\alpha^\nu<\langle\min(\omega_\parallel^2,\omega_\perp^2)$. The relative spectral weight of these modes will be discussed below in (69)–(71).

The above case is clearly not relevant at the transition temperature near $q=0$ and $q=\pi$, where $\omega_\perp$ and $\omega_\parallel$ vanish. When $\omega_\perp^2$ and Δ_α^ν are small compared to $\omega_\parallel^2$, one finds the same high-frequency pole $\tilde\omega_\parallel$ as in (65), but the low-frequency part of the spectrum is described by the following two poles:

$$\tilde\omega_\perp=K\frac{\pm[K^2\omega_\perp^2-(\Delta_x-\Delta_y+\Delta'_x-\Delta'_y)^2/4]^{1/2}+i\left[\sum_\alpha(\Delta_\alpha+\Delta'_\alpha)/2-\omega_\perp^2\right]}{\sum_\alpha(\Delta_\alpha+\Delta'_\alpha)-K^2-\omega_\perp^2(1+K^2/\omega_\parallel^2)}=\pm\hat\omega_\perp+i\Gamma_\perp\ . \tag{66}$$

When $\omega_\parallel^2$ and Δ_α^ν are small compared to $\omega_\perp^2$, one finds, equivalently, the $\tilde\omega_\perp$ of (65) and the low-frequency poles at

$$\tilde\omega_\parallel=K\frac{\pm[K^2\omega_\parallel^2-(\Delta_x-\Delta_y-\Delta'_x+\Delta'_y)^2/4]^{1/2}+i\left[\sum_\alpha(\Delta_\alpha-\Delta'_\alpha)/2-\omega_\parallel^2\right]}{\sum_\alpha(\Delta_\alpha-\Delta'_\alpha)-K^2-\omega_\parallel^2(1+K^2/\omega_\perp^2)}=\pm\hat\omega_\parallel+i\Gamma_\parallel\ . \tag{67}$$

These solutions are expected to be relevant at T_N near $q=\pi$ and $q=0$, respectively. We notice that the potentially soft modes $\omega_\perp$ or $\omega_\parallel$ merge with $-\omega_\perp$ or $-\omega_\parallel$, respectively, and go into an overdamped-oscillator form with two purely imaginary poles. One of the poles vanishes at T_N, i.e., $\omega\to i0$. In other words, the soft modes are being absorbed into the previously discussed central peak. The expected critical slowing down at T_N is thereby obtained.

It is instructive to consider these solutions of (52) in the real time space (i.e., the inverse Laplace transform). First, let us assume that the description with two damped modes given by (65)–(67) is sufficient. We then find from (52), for $\alpha=x,y$,

$$(S_A^\alpha S_B^\alpha)_{\vec q,t}=\tfrac12[\chi_+^\alpha e^{-\Gamma_\perp t}\cos(\omega_\perp t)\pm\chi_-^\alpha e^{-\Gamma_\parallel t}\cos(\omega_\parallel t)]\ , \tag{68}$$

where the partial susceptibility components χ_+^α and χ_-^α are defined in (33). If we include the central peak (64) it contributes to a first approximation simply by an additional term $\exp(-Kt)$, with the appropriate relative spectral weight. The result is

$$\begin{aligned}&(S_A^\alpha S_A^\alpha)_{\vec q,t}+(S_A^\alpha S_B^\alpha)_{\vec q,t}\\&\quad=\chi_+^\alpha[P_\perp^\alpha e^{-Kt}+(1-P_\perp^\alpha)e^{-\Gamma_\perp t}\cos(\omega_\perp t)]\ ,\end{aligned} \tag{69}$$

$$\begin{aligned}&(S_A^\alpha S_A^\alpha)_{\vec q,t}-(S_A^\alpha S_B^\alpha)_{\vec q,t}\\&\quad=\chi_-^\alpha[P_\parallel^\alpha e^{-Kt}+(1-P_\parallel^\alpha)e^{-\Gamma_\parallel t}\cos(\omega_\parallel t)]\ .\end{aligned} \tag{70}$$

The weight factors for the central peak are

$$\begin{aligned}P_\perp^\alpha&=(\Delta_\alpha+\Delta'_\alpha)/(\omega_\perp^2+\Delta_\alpha+\Delta'_\alpha)\ ,\\P_\parallel^\alpha&=(\Delta_\alpha-\Delta'_\alpha)/(\omega_\parallel^2+\Delta_\alpha-\Delta'_\alpha)\ .\end{aligned} \tag{71}$$

Equations (69) and (70) comprise the generalization of the RPA result (11) and (12). The correlation theory gives the damping and also a central peak for the y,y component, which is required by symmetry.

Finally, we need to calculate the decay constants K_α. This is done using the mode-mode–coupling approximation, which was found to be excellent for the pure Heisenberg paramagnet.[3] In the present case there is the complication that we have the regular precession modes (47) which must be projected out. This is difficult to do exactly and we shall use the following approximate scheme. Consider, for example, the complete equation of motion for $(S_A^x)_{\vec q}$ using (6) for $h=1$,

$$(\dot{S}^x_A)_{\vec{q}}=iJ_0\sum_{\vec{k}}[\phi_{\vec{k}}(S^y_A)_{\vec{k}+\vec{q}}(\delta S^z_B)_{\vec{k}}+\gamma_{\vec{k}}(\delta S^x_A)_{\vec{k}+\vec{q}}(S^y_B)_{\vec{k}}-\sigma_{\vec{k}}(S^y_A)_{\vec{k}+\vec{q}}(S^x_B)_{\vec{k}}]+iJ_0M[(S^y_A)_{\vec{q}}+\gamma_{\vec{q}}(S^y_B)_{\vec{q}}]\ . \quad (72)$$

The last part shows the terms which are explicitly proportional to the components of $\underline{A}^\dagger=\{S^x_A)_{\vec{q}},(S^x_B)_{\vec{q}},(S^y_A)_{\vec{q}},(S^y_B)_{\vec{q}}\}$. This is the dominant part to be projected out, and we shall neglect the (in a sense, higher-order) contributions from the rest. The first part of (72) therefore yields an approximation for the first component of $\underline{A}_1$. In other words, $\underline{A}_1$ is obtained from $\dot{\underline{A}}$ by neglecting the terms proportional to M (the RPA part). Inserting this into (55) and using the mode-mode decoupling, we then find

$$X_x(z')=k_BTJ_0^2\sum_{\vec{k}}\int_0^\infty\{[(\phi^2_{\vec{k}}+\gamma^2_{\vec{k}-\vec{q}})(\delta S^z_A\,\delta S^z_A)^{\mathscr{L}_1}_{\vec{k},t}+\sigma^2_{\vec{k}}(S^x_AS^x_A)^{\mathscr{L}_1}_{\vec{k},t}](S^y_AS^y_A)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}$$
$$+2\phi_{\vec{k}}\gamma_{\vec{k}-\vec{q}}(\delta S^z_A\,\delta S^z_B)^{\mathscr{L}_1}_{\vec{k},t}(S^y_AS^y_B)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}\}e^{-z't}dt\ , \quad (73)$$

$$X'_x(z')=k_BTJ_0^2\sum_{\vec{k}}\int_0^\infty\{[(\phi^2_{\vec{k}}+\gamma^2_{\vec{k}-\vec{q}})(\delta S^z_A\,\delta S^z_B)^{\mathscr{L}_1}_{\vec{k},t}-\sigma^2_{\vec{k}}(S^x_AS^x_B)^{\mathscr{L}_1}_{\vec{k},t}](S^y_AS^y_B)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}$$
$$+2\phi_{\vec{k}}\gamma_{\vec{k}-\vec{q}}(\delta S^z_A\,\delta S^z_A)^{\mathscr{L}_1}_{\vec{k},t}(S^y_AS^y_A)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}\}e^{-z't}dt\ . \quad (74)$$

The z parts $X_z(z')$ and $X'_z(z')$ are identical with (73) and (74) if δS^z and S^x are interchanged. The y part is a little more complicated:

$$X_y(z')=k_BTJ_0^2\sum_{\vec{k}}\int_0^\infty\{[(\phi^2_{\vec{k}}+\phi^2_{\vec{k}-\vec{q}})(\delta S^z_A\,\delta s^z_A)^{\mathscr{L}_1}_{\vec{k},t}+\sigma^2_{\vec{k}}(S^x_AS^x_A)^{\mathscr{L}_1}_{\vec{k},t}](S^x_AS^x_A)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}$$
$$+\sigma^2_{\vec{k}}(\delta S^z_A\,\delta S^z_A)^{\mathscr{L}_1}_{\vec{k},t}(\delta S^z_A\,\delta S^z_A)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}-2\phi_{\vec{k}}\phi_{\vec{k}-\vec{q}}(\delta S^z_A\,\delta S^z_B)^{\mathscr{L}_1}_{\vec{k},t}(S^x_AS^x_B)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}$$
$$+\sigma_{\vec{k}}\sigma_{\vec{k}-\vec{q}}[(S^x_AS^x_B)^{\mathscr{L}_1}_{\vec{k},t}(S^x_AS^x_B)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}+(\delta S^z_A\,\delta S^z_B)^{\mathscr{L}_1}_{\vec{k},t}(\delta S^z_A\,\delta S^z_B)^{\mathscr{L}_1}_{\vec{q}-\vec{k},t}]\}e^{-z't}dt\ . \quad (75)$$

In analogy with $X'_x(z')$, the $X'_y(z')$ part is obtained by interchanging the AA and AB terms. Under the sum and integral in (73)—(75), the precise time dependence of $(S^\alpha_NS^\alpha_M)^{\mathscr{L}_1}_{\vec{k},t}$ is not too important. Let us therefore use (68) to estimate the time dependence under $\mathscr{L}_1$. Consider one of the diagonal, dynamical susceptibility components (33), $\chi(z)=\chi^\alpha_\pm(z)/\chi^\alpha_\pm$, and the corresponding memory function (54), $\mathscr{M}^{\mathscr{L}_1}(z)=X^\alpha_\pm(z)=X_\alpha(z)\pm X'_\alpha(z)$. Then one has exactly[2] that

$$\mathscr{M}^{\mathscr{L}}(z)=\mathscr{M}^{\mathscr{L}_1}(z)-\mathscr{M}^{\mathscr{L}_1}(z)\chi(z)\mathscr{M}^{\mathscr{L}_1}(z)/\chi^\alpha_\pm\ . \quad (76)$$

Using the full $\mathscr{L}$ [or (68)] introduces erroneously long-lived (oscillatory) behavior from $\chi(z)$ in the memory function, which is not present when $\mathscr{M}(z)$ is calculated in the correct, rotating coordinate system. However, for $\Delta_\alpha\pm\Delta'_\alpha<\omega^2_\pm(q)$ the relative weight of this contribution is $P^\alpha_\pm$ [Eq. (71)]. Thus, if the central peak can be neglected, it is consistent to also neglect the last term in (76). It can be included in a complete calculation.

C. Self-consistent determination of the damping constants K_α

For the diagonal susceptibility components we can write the self-consistency equations (57) and (58) for $\alpha=x,y,z$ simply as

$$\frac{1}{z+K_\alpha}=\frac{1}{2}\left[\frac{X^\alpha_+(z)}{X^\alpha_+}+\frac{X^\alpha_-(z)}{X^\alpha_-}\right]\Bigg|_{z=z_0}\ , \quad (77)$$

where $X^\alpha_\pm(z)=X_\alpha(z)\pm X'_\alpha(z)$, and z_0 is a characteristic frequency. For the transverse damping with $\alpha=x,y$ we shall use $z_0=i\omega_+(\vec{q})$ and equate the real parts of (77). One clearly obtains the same result by using $z_0=-i\omega_+(\vec{q})$ or $z_0=\pm i\omega_-(\vec{q})$. For $\alpha=z$ the characteristic frequency is $z_0=\Gamma^z_+(\vec{q})$. From (61), (62), and (68), we then find the damping constant $\Gamma_+(\vec{q})$ for the frequency $\omega_+(\vec{q})$ to be

$$\Gamma_+(\vec{q})=\tfrac{1}{2}\sum_{\alpha=x,y}(\Delta_\alpha+\Delta'_\alpha)\mathrm{Re}\left[\frac{1}{2}\left[\frac{X^\alpha_+(i\omega_+)}{X^\alpha_+}+\frac{X^\alpha_-(i\omega_+)}{X^\alpha_-}\right]\right]\ , \quad (78)$$

and similarily for the longitudinal central peak,

$$\Gamma^z_+(\vec{q})=(a'_z+b'_z\gamma_{\vec{q}})\mathrm{Re}\left[\frac{1}{2}\left[\frac{X^z_+(\Gamma^z_+)}{X^z_+}+\frac{X^z_-(\Gamma^z_+)}{X^z_-}\right]\right]. \tag{79}$$

Using the property that any quantity $F_+(\gamma_{\vec{q}})=F_-(-\gamma_{\vec{q}})$, we find explicitly the mode-mode coupling contributions for the one-dimensional case, where $\gamma_q=-\gamma_{q-\pi}$:

$$X^x_\pm(z)=k_BTJ_0^2\sum_{\vec{k}}\left[(\phi_{\vec{k}}\pm\gamma_{\vec{k}-\vec{q}})^2\chi^z_+(\vec{k})\chi^y_\pm(\vec{q}-\vec{k})\tfrac{1}{2}\sum_s\frac{1}{z+\Gamma^z_+(\vec{k})+D^s_\pm(\vec{q}-\vec{k})}\right.$$
$$\left.+\sigma^2_{\vec{k}}\chi^x_+(\vec{k})\chi^y_\mp(\vec{q}-\vec{k})\tfrac{1}{4}\sum_{s,s'}\frac{1}{z+D^{s'}_+(\vec{k})+D^s_\mp(\vec{q}-\vec{k})}\right], \tag{80}$$

where the sums over s run over $s=1$ and 2, and $D^s_\pm(\vec{k})=\Gamma_\pm(\vec{k})+(-1)^s i\omega_\pm(\vec{k})$. For the z component the result is the same, when x and z indices are interchanged and the corresponding frequencies inserted. Finally, we find

$$X^y_\pm(z)=k_BTJ_0^2\sum_{\vec{k}}\left[(\phi_{\vec{k}}\mp\phi_{\vec{q}-\vec{k}})^2\chi^z_+(\vec{k})\chi^x_\pm(\vec{q}-\vec{k})\tfrac{1}{2}\sum_s\frac{1}{z+\Gamma^z_+(\vec{k})+D^s_\pm(\vec{q}-\vec{k})}\right.$$
$$+(\sigma_{\vec{k}}\pm\sigma_{\vec{q}-\vec{k}})^2\chi^x_+(\vec{k})\chi^x_\pm(\vec{q}-\vec{k})\tfrac{1}{4}\sum_{s,s'}\frac{1}{z+D^{s'}_+(\vec{k})+D^s_\pm(\vec{q}-\vec{k})}$$
$$\left.+(\sigma_{\vec{k}}\pm\sigma_{\vec{q}-\vec{k}})^2\chi^z_+(\vec{k})\chi^z_\pm(\vec{q}-\vec{k})\frac{1}{z+\Gamma^z_+(\vec{k})+\Gamma^z_\pm(\vec{q}-\vec{k})}\right]. \tag{81}$$

The interpretation of (78) and (79) is quite simply that there is a contribution to the damping from the coupling at all wave vectors of the various normal modes associated with the susceptibilities $\chi^\alpha_\pm(\vec{k})$. In general, when $\gamma_{\vec{q}}\neq-\gamma_{\vec{q}-\pi}$ we must separately also consider the contribution to (78) and (79) from $\chi^\alpha_-\chi^\beta_-$ and $\chi^\alpha_-\chi^\beta_+$. When solving the self-consistent equations (78)—(81) it is convenient to assume a functional form for $\Gamma(\vec{q})$. As was done for EuO, let us assume a Fourier expansion,

$$\Gamma_+(q)=\sum_{n=0,1,\ldots}\tau_n\cos(nq)$$

and (82)

$$\Gamma^z_+(q)=\sum_{n=0,1,\ldots}\lambda_n\cos(nq),$$

where the coefficients τ_n and χ_n are to be determined.

D. Numerical results

The numerical evaluation of the results falls in two parts. First, the static properties, the correlation functions (27), the local moment M, and the angle θ must be determined. For a classical one-dimensional magnet one could obtain these properties exactly by the transfer-matrix method. However, here we shall test the self-consistent correlation theory and make a comparison with the known results for the classical chain. The self-consistent calculation of the eight static quantities is quite tricky, but can be done easily for not too small magnetic fields by iteration. The results are independent of the start values for the iteration. In the evaluation of the correlation functions we have used the self-consistent first-moment frequencies (47) and the susceptibilities (33). The local moment is obtained from the exact relation

$$M^2=S(S+1)-\langle S^x_0S^x_0\rangle-\langle S^y_0S^y_0\rangle-\langle\delta S^z_0\,\delta S^z_0\rangle. \tag{83}$$

The theory is therefore dependent on the spin value S. The angle θ is determined by minimizing the free energy, which gives $\langle\partial\mathcal{H}/\partial\theta\rangle=0$. Using (6) we find the exact relation

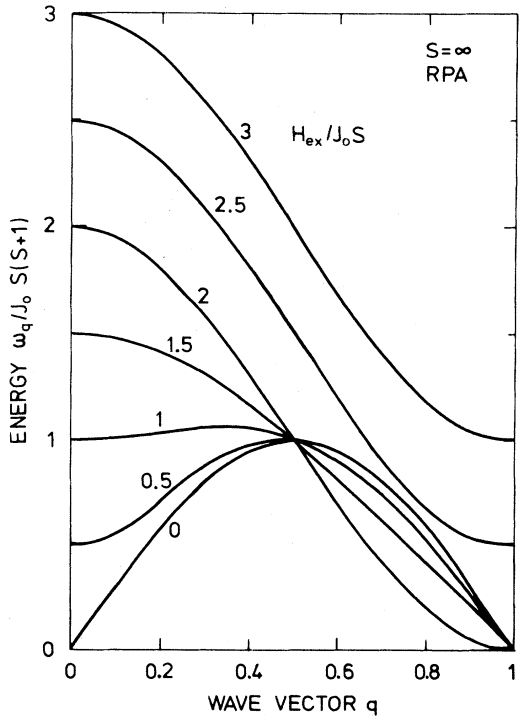

FIG. 2. Classical spin-wave dispersion for an antiferromagnetic chain in a magnetic field. It is identical to the RPA solution (8).

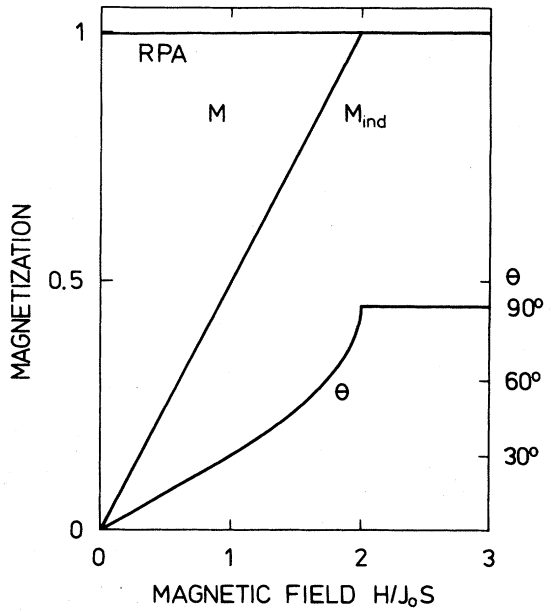

FIG. 3. Induced moment M_{ind}, the local moment M, and the canting angle θ calculated by the RPA as a function of the magnetic field for the antiferromagnetic chain.

$$\sin\theta = HM/[2J_0(M^2+\langle S_0^x S_1^x\rangle+\langle \delta S_0^z\,\delta S_1^z\rangle)] \ . \quad (84)$$

This expression is more accurate than the classical expression (2). If one uses (2) one finds $h=1$ in the canted region where $\theta\neq 90°$; this is no longer exactly true in the presence of nearest-neighbor correlations (84).

For $S=\infty$ (here we have used $S=1000$) the magnitude of the correlations vanishes compared to M^2, and one obtains the RPA results (8)—(17). The plot of RPA frequency versus wave vector is shown for different fields in Fig. 2. It agrees with the exact classical result and the computer simulations. The canting angle and the induced moment is shown in Fig. 3. When the external field H equals $2J_0S$ the angle θ becomes smoothly 90°. For $H=\sqrt{2}J_0S$ the canting angle is $\theta=45°$.

Here, we shall present results for $S=1$ for two temperatures $T/J_0S(S+1)=0.025$ and 0.3. The first-moment frequency is shown in Fig. 4. Because of the pair-correlation terms in (84), it shows an energy gap, which is not present in the RPA result, Fig. 2, in the canted phase. Figure 5 shows the local and induced moment M,M_{ind}, and the canting angle θ. The transition to the $\theta=90°$ structure now occurs abruptly for fields lower than $2J_0S$, although it approaches this at low temperatures. It is interesting to follow the correlation functions in the canted phase. In Fig. 6 we plot, for $T/J_0S(S+1)=0.025$, the normalized, total correlation functions for the same site, nearest and next-nearest neighbors,

$$\tilde{c}^0_{\alpha\alpha}=\langle \tilde{S}^\alpha_0\tilde{S}^\alpha_0\rangle/S(S+1),\quad \tilde{c}'_{\alpha\alpha}=\langle \tilde{S}_0\tilde{S}^\alpha_1\rangle/S(S+1)$$

and

$$\tilde{c}_{\alpha\alpha}=\langle \tilde{S}^\alpha_0\tilde{S}^\alpha_2\rangle/S(S+1)\ .$$

First, we notice that we have not obtained full symmetry between $\tilde{x}$ and $\tilde{y}$ as required by symmetry. The same

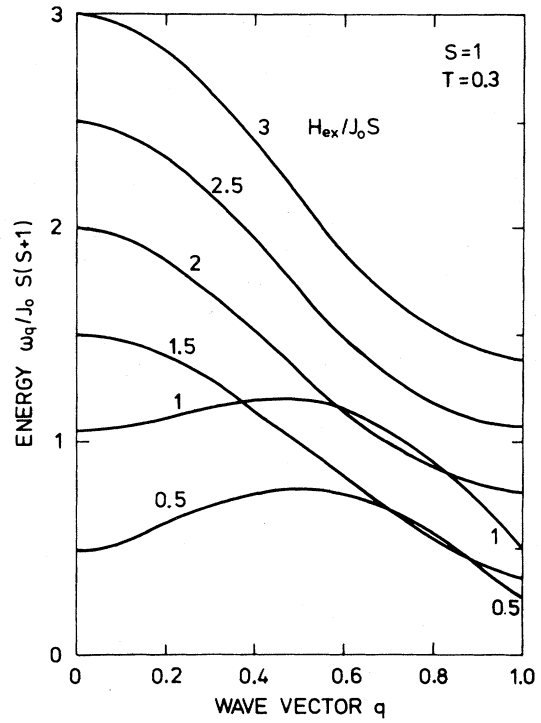

FIG. 4. First-moment frequency calculated by correlation theory for an antiferromagnetic quantum $S=1$ chain for $T=0.3J_0S(S+1)$ and different magnetic fields.

was the case with the RPA in (11)—(13). By symmetrizing, we find the dashed curves for $\tilde{c}^0_\perp$, $\tilde{c}'_\perp$, and $\tilde{c}_\perp$. These together with the z components agree qualitatively with the exact classical correlation functions calculated by Lovesey and Loveluck.[1] The obtained asymmetry be-

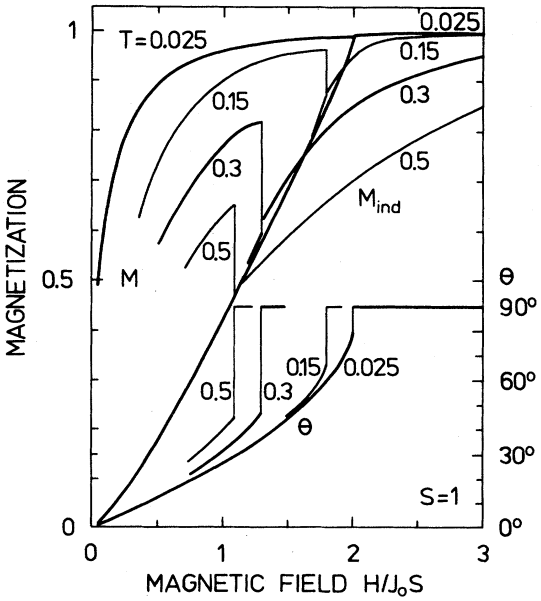

FIG. 5. Induced moment M_{ind}, the local moment M, and the canting angle θ calculated for an antiferromagnetic quantum $S=1$ chain for various temperatures $T/J_0S(S+1)$ as a function of a magnetic field H/J_0S.

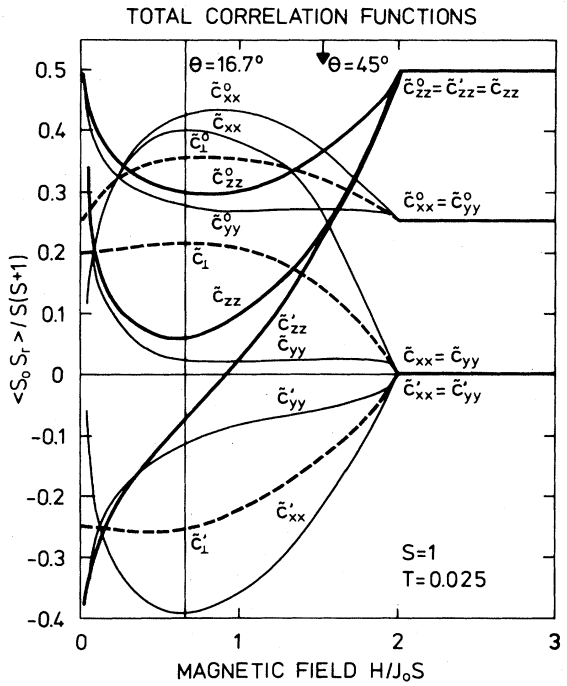

FIG. 6. Total correlation functions in the original $(\tilde{x},\tilde{y},\tilde{z})$ coordinate system normalized by $S(S+1)$. They are defined as on site, $\tilde{c}^0_{\alpha\alpha}=\langle\tilde{S}^\alpha_0\tilde{S}^\alpha_0\rangle/S(S+1)$; nearest neighbor, $\tilde{c}'_{\alpha\alpha}=\langle\tilde{S}^\alpha_0\tilde{S}^\alpha_1\rangle/S(S+1)$; and next-nearest neighbor, $\tilde{c}_{\alpha\alpha}=\langle\tilde{S}^\alpha_0\tilde{S}^\alpha_2\rangle/S(S+1)$. $\tilde{c}^\gamma_\perp$ represent the average of $\tilde{c}^\gamma_{xx}$ and $\tilde{c}^\gamma_{yy}$. All are calculated for the antiferromagnetic quantum $S=1$ chain for $T=0.025J_0S(S+1)$.

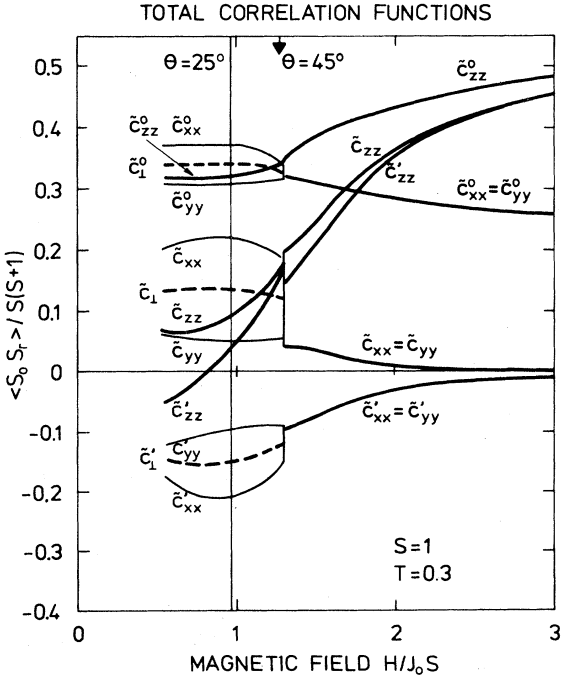

FIG. 7. Same correlation functions as in Fig. 6 for $T=0.3J_0S(S+1)$.

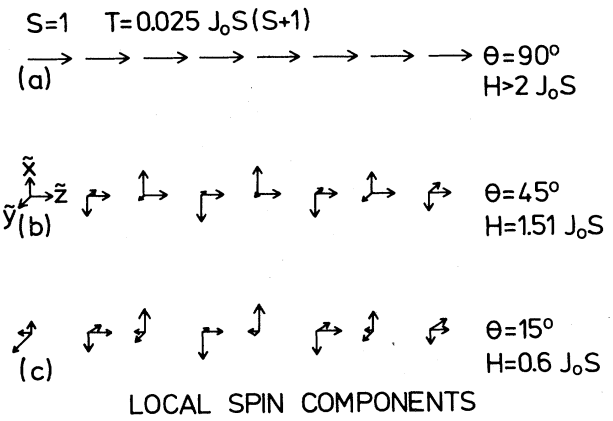

FIG. 8. Arrows represent the components of the local-moment structure (schematic) deduced from the correlation functions on Fig. 6. (a) is the fully aligned high-field state. (b) shows that an antiferromagnetic $\tilde{y}$ component is present at $\theta=45°$. (c) At still lower fields an antiferromagnetic component is also induced for the $\tilde{z}$ component. The general structure for $\theta<90°$ is most likely a complex cone structure with an incommensurate spiral vector.

tween $\tilde{x}$ and $\tilde{y}$ is, however, very instructive. It shows that the assumed structure, Fig. 1 (which is correct for the classical case $S=\infty$), is modified by twisting the various sublattices. Analyses of the correlation functions show that the structure is a complex cone structure, as shown in Fig. 8. Such a structure automatically has full symmetry between $\tilde{x}$ and $\tilde{y}$ and will also make the frequency on Fig. 4 vanish at an incommensurate wave vector. If the cone structure has a long wavelength, the correlation functions calculated using Fig. 1 are still valid. However, difficulties appear at small fields. For $H<0.7J_0S$, $\chi^{zz}_{\vec{q}}$ develops a pole for $0<q<\pi$; this also happens for $\chi^{xx}_{\vec{q}}$, and $\chi^{yy}_{\vec{q}}$ for $H<0.5J_0S$, indicating that a complex cone structure is needed. The results for low fields are therefore not reliable before a full self-consistent cone situation has been considered. Figure 7 shows the equivalent results for $T=0.3J_0S(S+1)$. In this case there is better symmetry between $\tilde{x}$ and $\tilde{y}$—until $\chi^{\alpha\alpha}_{\vec{q}}$ develops, the finite-$\vec{q}$ poles for $H<J_0S$ and $H<0.6J_0S$. Notice that $\theta=45°$ is now very close to the field at which the abrupt transition to $\theta=90°$ occurs. This is very different from the RPA, Fig. 3. The indication that the structure of an antiferromagnetic chain in a field for small S is a kind of cone structure might have been guessed from the exact calculation by Ishimura and Shiba[11] for $S=\frac{1}{2}$. This shows that the spin waves develop zero-frequency modes for field-dependent q values between 0 and π. The present calculation brings some physical insight into this interesting result which should be investigated experimentally. It is not just the case for $S=\frac{1}{2}$, but holds also for other small spin values.

The results for other small values of S are qualitatively similar to those reported for $S=1$, when plotted on the relative field and temperature scales H/J_0S and $T/J_0S(S+1)$. The relative importance of the correlation effects is largest for $S=\frac{1}{2}$ and decreases with increasing S. For $S=\frac{1}{2}$ the fact that $\langle S^2_\alpha\rangle=\frac{1}{4}$ at all fields and

temperatures is exactly fulfilled at high fields, but at low fields deviations between $\langle S_\alpha^2 \rangle$ and $\frac{1}{4}$ are found of the order of 10%. For all values of S, the relative local moment M/S approaches 1 for large fields. The quantum effects are therefore quite accurately obtained by the theory. The result that the theory reduces to the RPA theory for $S \to \infty$, which is temperature independent and has no damping on the relative field and temperature scale, may be a problem with the numerical calculation of the limit $S \to \infty$. Certainly, if we use the RPA results for a classical spin of formal length 1, one would calculate a temperature dependence and damping rather similar to that found for the quantum spin $S=1$.

The correlation theory is not restricted to the one-dimensional case and it is easy to extend it to higher dimensions, where it actually should be more reliable. As an example in Fig. 9 we show the induced moments and canting angle for the two-dimensional square case. An analysis of the correlation effects in the frustrated triangular lattice will be considered in a separate paper. It is found that the correlation effects introduce local ordering with incommensurate wave vectors.

After having investigated the static properties, the second part, the dynamic properties going beyond the first-moment frequencies, remains to be calculated. Equations (59) and (61) show that there are three independent families of excitations, two transverse with first-moment frequencies $\omega_+(\vec{q})$ and $\omega_-(\vec{q})$, and one longitudinal with zero frequency. Furthermore, $\omega_+(\vec{q})$ is equivalent to $\omega_-(q-\pi)$. The Fourier coefficients of the expansion (82) of the damping constants $\Gamma_+(\vec{q})$ and $\Gamma_+^z(\vec{q})$, Eqs. (78) and (79), can now be calculated using (80) and (81). The result depends strongly on the static properties and the accuracy with which these are obtained. For $S=1$ and

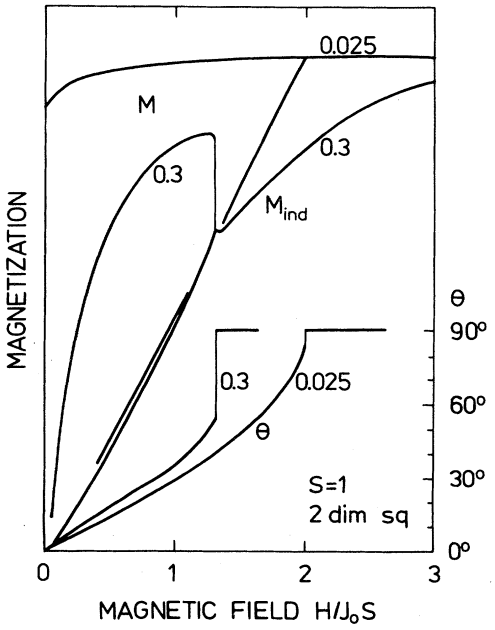

FIG. 9. Induced moment M_{ind}, the local moment M, and the canting angle θ for a two-dimensional square lattice for $S=1$ for $T=0.025$ and $0.3J_0S(S+1)$.

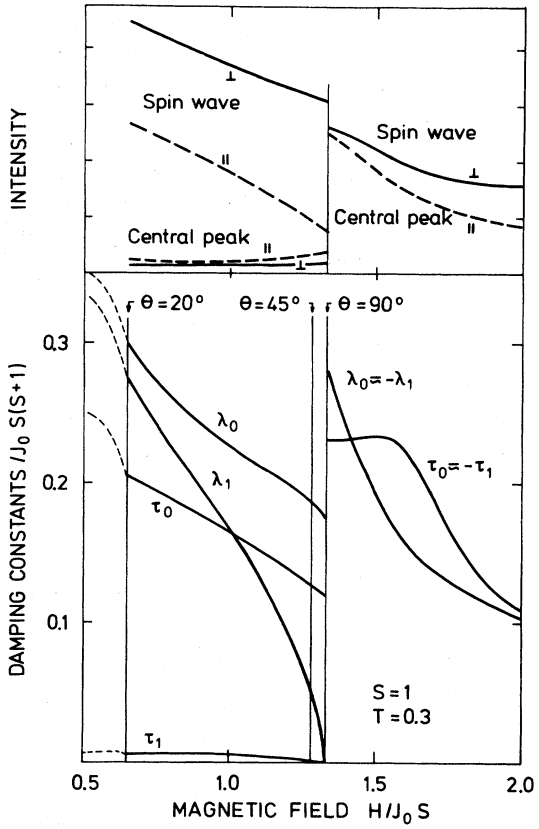

FIG. 10. Lower part shows the Fourier coefficients for the damping constants $\Gamma_\pm(\vec{q})$ and $\Gamma_\pm^z(\vec{q})$ for the spin-wave mode $\omega_{\vec{q}}$ (Fig. 4) and the central peak as a function of the magnetic field. The upper part shows the intensity of the spin wave and the central peak for $[\tilde{\chi}^{xx}(\vec{q},\omega)+\tilde{\chi}^{yy}(\vec{q},\omega)]/2$ (solid line) and $\tilde{\chi}^{zz}(\vec{q},\omega)$ (dashed line) for $q=\pi$.

$T=0.3J_0S(S+1)$, on Fig. 10, we show the damping constants and the intensity of the spin-wave and central-peak contributions to the total transverse and longitudinal response functions (5), $[X^{xx}(\vec{q},\omega)+\tilde{\chi}^{yy}(\vec{q},\omega)]/2$ and $\tilde{\chi}^{zz}(\vec{q},\omega)$. The corresponding spin-wave frequency is given in Fig. 4. In the $\theta=90°$ region the spin waves and the central peak have widths which vanish for large fields, but grow strongly near the transition to $\theta \neq 90°$. In the field range for $20° < \theta < 90°$ we find damping constants of the order of 10% of the spin-wave frequencies. For $\theta < 16°$ we are in the region where $\chi_{\vec{q}}^{\alpha\alpha}$ has poles for $0 < q < \pi$. The calculated damping is strong in this region, indicating that the assumed structure shown in Fig. 1 is not adequate in this region. The magnitude and field dependence is in qualitative agreement with the computer-simulation studies for the classical ($S=\infty$) chain. We were unable to make a direct comparison because in the present numerical calculation the correlation effect, and thereby the damping, vanishes when $S \to \infty$.

VII. SUMMARY

The static and dynamic properties of a Heisenberg magnet in a magnetic field was derived using the correlation

theory. This gives results including correlation effects for various lattice dimensions, spin values, temperatures, and signs of the interaction constant. As an illustration of the results, the one-dimensional antiferromagnet, when correlation effects are expected to be large, was treated, particularly for $S=1$. The results agree qualitatively with the computer-simulation results on the classical chain. However, indications are also found of a complex cone structure similar to that expected, leading to the exact results for the $S=\frac{1}{2}$ chain. By choosing dynamical variables which diagonalize the local properties, we found a central-peak component, which can be understood as coming from the local longitudinal susceptibility. This provides an alternative, simpler description of the central peak than the hydrodynamic energy-mode concept. It would be interesting to calculate the dynamics using the exact static properties known for the one-dimensional system to test the mode-mode—coupling approximation. For two dimensions the present theory allows a calculation of both static and dynamic properties which have not previously been obtained.

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