Spin correlations in a Heisenberg system within the paramagnetic region*

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The two-spin correlation function for a Heisenberg system is investigated, in the paramagnetic region, using a new approach through which one is able to reveal the dynamics more clearly than previously. The conventional mode-mode coupling theory is obtained as a lowest-order approximation; this entails ignoring all third- and higher-order irreducible spin correlations. It is argued that the mechanism for sloppy spin waves is lost in the mode-mode theory and that this is the reason for some of the most noticable discrepancies between theory and experiments.

I. INTRODUCTION

In the last decade a wealth of new experimental data has been collected on the spin dynamics for various magnetic systems of different dimensionalities.[1-7] The most detailed information was obtained through inelastic-neutron-scattering experiments.[3-7] On the theoretical side we have to confess that some of the most striking observations are still rather poorly understood.

At temperatures far below the magnetic transition point one normally observes well-defined collective magnetic excitations, magnons, whose life time decreases as T approaches the critical temperature. Above T_c no long-range order exists and ordinary spin waves disappear. The mechanism behind the propagation and damping of spin waves is quite well understood. For insulators, in particular, the calculations are normally based on the Heisenberg model. Unfortunately, we cannot solve exactly the equations of motion for this model and we have to resort to approximations. The random-phase approximation (RPA) has been found to describe most of the qualitative features correctly. By including some corrections to RPA, one also understands some more detailed questions.[8] Above the critical point, RPA gives meaningful results for the static properties, but it gives no meaningful dynamics. Experimentally, one finds in some cases that short-wavelength "sloppy spin waves"[9] exist beyond the critical point. This has been most clearly demonstrated for the antiferromagnetic $RbMnF_3$ through neutron scattering experiments.[4] The effect is even more dramatic in some one-dimensional systems. There no long-range order can exist for any finite temperature.[10] In spite of this one observes in $(CO_3)_4NMnCl_3$ (TMMC) at 4 K, for instance, extremely sharp spin-wave resonances which disappear at higher temperatures.[5] At the lower temperature, the spins are very strongly correlated over several hundred Ångstroms and the system appears to have a certain long-range order in a local sense.

So far a three-peak structure in the neutron scattering spectrum has been found only for systems with antiferromagnetic coupling. For three-dimensional ferromagnetic systems one has only observed a single quasielastic peak, which broadens as the temperature is increased.[6,11] Recent computer simulations, based on the classical Heisenberg model, have given similar results.[12] At present no theory is capable of explaining this difference between ferro- and antiferromagnetic systems, nor can one explain the observed three-peak structure in a satisfactory way. McLean and Blume[13] were quite successful in analyzing the experimental data on TMMC, but they introduced from the beginning a finite order parameter, which we cannot accept in a theory based on first principles. Also, one has been rather successful in getting agreement with experiments using Mori's formulation and determining various parameters through low-order frequency moments. In this way one can reproduce a three-peak structure for antiferromagnets[14] and one gets sharp spin-wave peaks in the one-dimensional case.[15] However, lacking a theory for the memory function in the Mori formulation, this cannot be considered as fully satisfactory.

Very close to the critical point, strong critical fluctuations dominate the behavior of the magnetic system and RPA, and various improved versions of it are known to fail in the critical region.[16] So, for instance, they do not provide any explanation of the physics behind the critical exponents. For calculating static quantities, the problem has been approached very differently and the assumption of certain scaling properties[16,17] has given a means of correlating various critical exponents. The most recent progress was made by Wilson[18] by introducing a new renormalization procedure. He clearly demonstrated that extremely strong renormalization processes occur near the critical point and that long-wavelength fluctuations are responsible for these. The dynamics involved here are not well understood at present. The only existing microscopic theory for analyzing the dynamics of the critical fluctuations is the so-called mode-mode coupling theory.[19-22] In certain respects, this theory is in excellent agreement with experiments. It does, for instance, yield the dynamical scaling, first suggested on an intuitive basis by Halperin and Hohenberg[23] and by Ferrell *et al.*,[24] which seems to be well supported by experiments.[6,7] It has also given some details of linewidths which are in good agreement with experiments.[25]

The mode-mode theory has also been used at high temperatures,[20,26-28] and the results agree well with those obtained from computer simulations.[12] This theory does not give any three-peak structure in the energy spectrum for antiferromagnetic systems,[28] nor does it give any spin waves in the one-dimensional case.[13] Therefore it does not provide any explanation of the observed difference between ferro- and antiferromagnetic systems and it gives no explanation for the occurrence of "sloppy spin waves." Whether it gives the correct line shape within the critical region is not yet possible to test experimentally. One is at present only able to measure the half-width of the lines. Our investigation of the mode-mode theory indicates that the mechanism for the sloppy spin waves has been lost. Whether this has any implications on the critical fluctuations is not clear to us at present.

It has been stated that the mode-mode theory gives the spin diffusion equation for fluctuations of wavelength large compared to the spin correlation length and for long times.[27-29] One of us has, however, shown that this is not true for asymptotic times.[30] It was concluded that the mode-mode theory gives approximately simple diffusion only for some intermediate and finite time region. Considering the conservation of the total magnetization for an isotropic magnet, one would expect spin diffusion to be the correct mechanism for asymptotic times.[31,32] If this is true, we seem to have here a contradiction to the prediction of the mode-mode theory. This may not be of any practical importance, but it raises some conceptual questions.

What has been said above calls for a critical discussion of the mode-mode theory and a search for the most important corrections. The basic equations have been derived in many different ways.[20,26-28,33-36] Actually somewhat different expressions have been obtained. None of these derivations seemed suitable for an investigation of corrections. We have therefore worked out a new approach which, in a more straightforward and systematic way, leads to the conventional mode-mode theory in lowest-order approximation. It provides us with a means of going beyond this approximation even though we have not as yet reached any definite conclusion concerning an improved theory. In this paper we will give a general discussion of the equation of motion for the two-spin correlation function, based on the Heisenberg model. We show how the usual mode-mode theory results if we ignore entirely all higher-order irreducible spin correlations. In the conclusion, we will make some remarks which are partly based on results that we hope to be able to present in a later publication.

The contents of the paper are the following. In Sec. II, we give definitions of various relevant correlation functions and we also outline the conventional mode-mode theory. Section III contains the basic mathematical formulation and, in Sec. IV, we derive a formally exact expression for the memory function which enters the theory. Then, in Sec. V, we show how the conventional mode-mode theory is obtained as a lowest-order approximation. In Sec. VI, we conclude by making some general remarks concerning the mode-mode theory. The main text is followed by three Appendixes, where some clarifying details are given.

II. GENERAL RELATIONS AND THE MODE-MODE COUPLING THEORY

The quantity we will be mainly interested in is the retarded spin response function ($\hbar=1,\ \mu_B=1$)

$$\chi_r^{\alpha\alpha}(\vec{q},t)=(i/N)\langle[S^\alpha(\vec{q},t),S^\alpha(-\vec{q},0)]\rangle\,\theta(t). \tag{2.1}$$

The notations are the conventional ones; $\langle\ \rangle$ denotes an average over the equilibrium ensemble, $[\ \ ]$ is a commutator, $\theta(t)$ is the unit step function, N is the total number of spins, and

$$S^\alpha(\vec{q},t)=\sum_{\vec{R}}e^{i\vec{q}\cdot\vec{R}}S^\alpha(\vec{R},t) \tag{2.2}$$

is the spatial Fourier transform of α component of the spin operator. The real part of the spin correlation function

$$C^{\alpha\alpha}(\vec{q},t)=(1/2N)\times\langle S^\alpha(\vec{q},t)S^\alpha(-\vec{q},0)+S^\alpha(-\vec{q},0)S^\alpha(\vec{q},t)\rangle \tag{2.3}$$

is then given by the fluctuation-dissipation theorem[37,38]

$$C^{\alpha\alpha}(\vec{q},\omega)=\coth(\tfrac{1}{2}\beta\omega)\,\chi''(\vec{q},\omega), \tag{2.4}$$

where $\beta=1/k_BT$ is the inverse temperature,

$$\chi_r^{\alpha\alpha}(\vec{q},\omega)=\int_{-\infty}^{\infty}dt\,e^{i\omega t}\chi_r^{\alpha\alpha}(\vec{q},t), \tag{2.5}$$

and $C^{\alpha\alpha}(\vec{q}\omega)$ is similarly the time Fourier transform of $C^{\alpha\alpha}(\vec{q}t)$; $\chi'(\vec{q}\omega)$ and $\chi''(\vec{q}\omega)$ are the real and imaginary parts of $\chi_r^{\alpha\alpha}(\vec{q}\omega)$. The Kramers-Kronig relations give further

$$\chi'(\vec{q},\omega)=P\int_{-\infty}^{\infty}\frac{\chi''(\vec{q},\omega')}{\omega'-\omega}\,\frac{d\omega'}{\pi}\,. \tag{2.6}$$

The Kubo relaxation function,[38] which is often used in this connection, is defined through[39]

$$F^{\alpha\alpha}(\vec{q},t)=\frac{1}{N}\int_0^\beta d\lambda\,\langle e^{\lambda H}S^\alpha(\vec{q},t)e^{-\lambda H}S^\alpha(-\vec{q},0)\rangle \tag{2.7a}$$

$$=\frac{i}{N}\int_{-\infty}^{0}dt'\,\langle[S^\alpha(\vec{q},t),S^\alpha(-\vec{q},t')]\rangle, \tag{2.7b}$$

where H is the Hamiltonian of the system. It follows from the definition that $F^{\alpha\alpha}(\vec{q},t)$ is an even and real function of time and that

$$\frac{d}{dt}F^{\alpha\alpha}(\vec{q}t)=-\frac{i}{N}\langle[S^\alpha(\vec{q}t),S^\alpha(-\vec{q},0)]\rangle. \tag{2.8}$$

This implies that

$$F^{\alpha\alpha}(\vec{q},\omega)=2\chi''(\vec{q},\omega)/\omega \tag{2.9}$$

and that

$$F^{\alpha\alpha}(\vec{q},t=0)=\chi_{is}(\vec{q})=P\int_{-\infty}^{\infty}\frac{\chi''(\vec{q},\omega')}{\omega'}\,\frac{d\omega'}{\pi}\,, \tag{2.10}$$

where $\chi_{is}(\vec{q})$ is the wave-vector-dependent isothermal susceptibility.

If we were able to obtain an appropriate equation of motion for $\chi_r^{\alpha\alpha}(\vec{q}t)$ and were also able to solve this equation, we would rather easily obtain all the other related quantities, including the static susceptibility, through the above relations.

For mathematical convenience we shall consider a time-ordered response function, defined along a certain path L in time, and we denote this quantity by

$$\chi_L^{\alpha\alpha}(\vec{q},t)=(i/N)\langle T_L S^\alpha(\vec{q},t)S^\alpha(-\vec{q},0)\rangle_c, \tag{2.11}$$

where $\langle AB\rangle_c$ stands for $(\langle AB\rangle-\langle A\rangle\langle B\rangle)$. We shall subsequently proceed to the retarded response function.

In the conventional mode-mode theory, one considers the relaxation function $F^{\alpha\alpha}(\vec{q},t)$ as the basic quantity and one writes for this the Mori equation[40]

$$\frac{d}{dt}F^{\alpha\alpha}(\vec{q},t)+\int_0^t dt'\,M_r^{\alpha\alpha}(\vec{q},t-t')F^{\alpha\alpha}(\vec{q},t')=0. \tag{2.12}$$

The explicit expression for the memory function $M_r(\vec{q},t)$ is[27,35]

$$M_r^{\alpha\alpha}(\vec{q},t)=[\beta\chi_{is}(\vec{q})]^{-1}\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2 \times F^{\alpha\alpha}(\vec{q}',t)F^{\alpha\alpha}(\vec{q}-\vec{q}',t). \tag{2.13}$$

The integration goes over the first Brillouin zone, of volume v, and $J(\vec{q})$ is the Fourier transform of the exchange integral. Different derivations of the memory function have led to slightly different expressions, but these differences are of no essential importance for us here. More essential is the somewhat different attitude taken by Kawasaki[35] relative to others (see, e.g., Ref. 27). He employs Eqs. (2.12) and (2.13) only for the long-wavelength critical fluctuations and he also includes certain renormalizations of the exchange integral. In his case, he cannot determine $\chi_{is}(\vec{q})$ within his theory, and he suggests using the susceptibility as obtained either from experiments or from some static theory. Others, like Blume and Hubbard,[26,27] use the above equations for all wave vectors and they are then able to obtain a self-consistent $\chi_{is}(\vec{q})$, which turns out to give the same result as the spherical model.[41]

Differentiating Eq. (2.12), performing a partial integration, and using Eq. (2.8), we obtain the corresponding equation for $\chi_r^{\alpha\alpha}(\vec{q},t)$,

$$\frac{d}{dt}\chi_r^{\alpha\alpha}(\vec{q},t)+\int_0^t dt'\,M_r^{\alpha\alpha}(\vec{q},t-t')\chi_r^{\alpha\alpha}(\vec{q},t') =M_r^{\alpha\alpha}(\vec{q},t)\chi_{is}(\vec{q}). \tag{2.14}$$

The reason we write down this equation is that in our derivation we will first arrive at an equation of this form and we can then, if desired, go back to Eq. (2.12).

III. MATHEMATICAL FORMULATION

This section serves several purposes. It explains the philosophy behind our procedure and contains the necessary mathematical preparations. The main aim is, however, to present the formal mathematical procedure leading to an equation of the form (2.14).

Our derivation will be based on the Heisenberg model Hamiltonian ($\hbar=1$, $\mu_B=1$)

$$H=-\frac{1}{2}\sum_{\vec{R}\vec{R}'}J(\vec{R}-\vec{R}')\vec{S}(\vec{R})\cdot\vec{S}(\vec{R}') -\sum_{\vec{R}}\vec{S}(\vec{R})\cdot\vec{h}(\vec{R},t), \tag{3.1}$$

where $\vec{h}(\vec{R},t)$ is an arbitrary external magnetic field and where $J(\vec{R}=0)=0$. This is supplemented by the commutation relations

$$[S^\alpha(\vec{R}),S^{\alpha'}(\vec{R}')]=i\delta_{\vec{R},\vec{R}'}S^{\alpha''}(\vec{R}). \tag{3.2}$$

Throughout this paper we use unprimed, singly primed, and doubly primed indices (like α, α', α'' above) for the Cartesian components when they are placed in cyclic order; e.g., if $\alpha = x$ then $\alpha' = y$ and $\alpha'' = z$.

The time evolution of the spin operators is governed by the equation

$$\frac{\partial}{\partial t} S^\alpha(\vec{R}, t) + \sum_{\vec{R}'} J(\vec{R} - \vec{R}') \times [S^{\alpha'}(\vec{R}', t) S^{\alpha''}(\vec{R}, t) - S^{\alpha''}(\vec{R}', t) S^{\alpha'}(\vec{R}, t)] = -h^{\alpha'}(\vec{R}, t) S^{\alpha''}(\vec{R}t) + h^{\alpha''}(\vec{R}, t) S^{\alpha'}(\vec{R}, t). \quad (3.3)$$

We now consider a situation where, at $t = -\infty$, the spin system was in thermal equilibrium at temperature T with no external field present. The external field is then turned on and induces a certain mean magnetization, $\langle \vec{S}(\vec{R}t) \rangle$, at finite times. $\vec{S}(\vec{R}t)$ is the spin operator in the Heisenberg picture with the external field included in the time evolution, and the bracket $\langle \;\rangle$ denotes an averaging over the initial ($t = -\infty$) canonical ensemble with no external field present. In principle, the magnetization is obtained from Eq. (3.3) after averaging over the initial equilibrium ensemble. We may write the equation in the somewhat more compact form

$$\frac{\partial}{\partial t_1} \langle S^\alpha(1) \rangle + \int d(1') d(1'') J(1; 1'1'') \langle S^{\alpha'}(1') S^{\alpha''}(1'') \rangle = -h^{\alpha'}(1) \langle S^{\alpha''}(1) \rangle + h^{\alpha''}(1) \langle S^{\alpha'}(1) \rangle \quad (3.4)$$

by letting 1, 1′, etc. stand for $(\vec{R}_1 t_1)$, $(\vec{R}_1' t_1')$, etc., and introducing

$$J(1; 1'1'') = \delta(t_1 - t_1') \delta(t_1 - t_1'') \times [\delta_{\vec{R}_1 \vec{R}_1''} J(\vec{R}_1 - \vec{R}_1') - \delta_{\vec{R}_1 \vec{R}_1'} J(\vec{R}_1 - \vec{R}_1'')], \quad (3.5)$$

and further letting the integration symbol include both the integration over time and the summation over the lattice points.

Another way of writing the induced magnetization would be

$$\langle S^\alpha(\vec{R}, t) \rangle = \langle \mathcal{S}(-\infty, t) S_0^\alpha(\vec{R}, t) \mathcal{S}(t, -\infty) \rangle, \quad (3.6)$$

where $S_0^\alpha(\vec{R}t)$ is the Heisenberg spin operator in zero external field and $\mathcal{S}(t, t')$ is the time-evolution operator originating from the external field. If we associate the two $t = -\infty$ with the starting point and the end point, respectively, of a certain time path, we can formally introduce a time ordering along this path and make use of all of the resulting mathematical convenience. For this purpose, we define a time path L stretching from $t = -\infty$ to $t = +\infty$ and further to $t = -\infty$ as in Fig. 1. We associate the $t = -\infty$ standing to the right in Eq. (3.6) with the starting point on L and the $t = -\infty$ standing to the left with the end point. We introduce a time ordering along this path and denote this by a T_L, and we let the magnetic field run along this time path. This means that we formally distinguish $\vec{h}(\vec{R}t)$ for times lying to the left and to the right of $t = +\infty$. Through this formal trick, we can write

$$\langle S^\alpha(\vec{R}, t) \rangle = \langle T_L S_0^\alpha(\vec{R}, t) \mathcal{S}_L \rangle / \langle \mathcal{S}_L \rangle, \quad (3.7)$$

and

$$\mathcal{S}_L = T_L \exp\left(i \sum_{\vec{R}_1} \int_L dt_1 \vec{S}(\vec{R}_1, t_1) \cdot \vec{h}(\vec{R}_1, t_1) \right). \quad (3.8)$$

The time integration here goes over the path L. One consequence of Eqs. (3.7) and (3.8) is that we can write for arbitrary nonequilibrium situations

$$\delta \langle S^\alpha(\vec{R}_1, t_1) \rangle = \sum_\beta \sum_{\vec{R}_2} \int_L dt_2 \chi_L^{\alpha\beta}(\vec{R}_1 t_1; \vec{R}_2 t_2) \delta h^\beta(\vec{R}_2, t_2), \quad (3.9)$$

where $\chi_L^{\alpha\beta}$ is the time-ordered two-spin correlation function introduced earlier in Eq. (2.11). Consequently,

$$\delta \langle S^\alpha(\vec{R}_1, t_1) \rangle / \delta h^\beta(\vec{R}_2, t_2) \equiv \chi_L^{\alpha\beta}(\vec{R}_1 t_1; \vec{R}_2 t_2) = i \langle T_L S^\alpha(\vec{R}_1 t_1) S^\beta(\vec{R}_2 t_2) \rangle_c. \quad (3.10)$$

Similarly, we generate the time-ordered higher correlation functions by differentiating $\langle S^\alpha(\vec{R}_1 t_1) \rangle$ with respect to the external field the required number of times.

The formal trick of introducing the double-time path above was used previously in deriving general transport equations for phonons,[42] and it is actually closely related to the procedure suggested earlier.[43] For further details and clarification, we refer to the above authors.

It is now obvious how to proceed from Eq. (3.4). By differentiating this equation with respect to the external field, we generate the equation for the time-ordered two-spin correlation function. By simply differentiating several times we would get the corresponding equations for the higher-order correlation functions. This yields

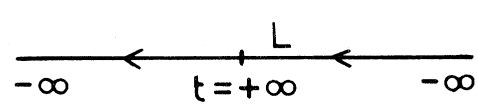

FIG. 1. Time-path L used in the definition of time ordering.

$$\frac{\partial}{\partial t_1}\chi_L^{\alpha\beta}(1,2)+\int_L d(1')d(1'')\,J(1;1'1'')\frac{\delta\langle S^{\alpha'}(1')S^{\alpha''}(1'')\rangle}{\delta h^{\beta}(2)}$$
$$=\delta(1,2)[-\delta_{\alpha'\beta}\langle S^{\alpha''}(1)\rangle+\delta_{\alpha''\beta}\langle S^{\alpha'}(1)\rangle]$$
$$-h^{\alpha'}(1)\chi_L^{\alpha''\beta}(1,2)+h^{\alpha''}(1)\chi_L^{\alpha'\beta}(1,2), \qquad (3.11)$$

with

$$\delta(1,2)=\delta(t_1-t_2)\delta_{\vec{R}_1\vec{R}_2}. \qquad (3.12)$$

Let us now consider the equilibrium situation with no external field present. Then $\chi_L^{\alpha\beta}$ is diagonal in the Cartesian components and all its diagonal elements are equal. Equation (3.11) goes over to

$$\frac{\partial}{\partial t_1}\chi_L(1,2)+\int_L d(1')d(1'')\,J(1;1'1'')\times\left(\frac{\delta\langle S^{\alpha'}(1')S^{\alpha''}(1'')\rangle}{\delta h^{\alpha}(2)}\right)_{\vec{h}=0}=0. \qquad (3.13)$$

Here and in the following we shall drop the Cartesian indices whenever we are considering the equilibrium situation with $\vec{h}=0$ and if no ambiguities result. As we see above, we are left with the problem of calculating how a certain equal-time correlation function is changed under the influence of the external field.

In addition to the direct effect when the external field is varied, a particular spin will also feel (as a result of interactions) the rearrangements of the surrounding spins. We shall take into account some of this through an effective magnetic field, which we assume to be of the form

$$h_{\text{eff}}^{\alpha}(1)=h^{\alpha}(1)+\int_L d(2)_{\beta}\,\Gamma^{\alpha\beta}(1,2)\langle S^{\beta}(2)\rangle, \qquad (3.14)$$

where the integration symbol includes summation over the Cartesian components and over the lattice points and further integration over the time path L. Also

$$\Gamma^{\alpha\beta}(1,2)=\delta_{\alpha\beta}\delta(t_1-t_2)\Gamma(\vec{R}_1-\vec{R}_2), \qquad (3.15)$$

where $\Gamma(\vec{R}_1-\vec{R}_2)$ is independent of the magnetic fields.[44]

In the following discussion we shall often make use of the two relations

$$\frac{\delta\langle\;\rangle}{\delta h^{\alpha}(1)}=\int_L d(2)_{\beta}\frac{\delta\langle\;\rangle}{\delta h_{\text{eff}}^{\beta}(2)}\frac{\delta h_{\text{eff}}^{\beta}(2)}{\delta h^{\alpha}(1)}, \qquad (3.16)$$

and

$$\frac{\delta h_{\text{eff}}^{\alpha}(1)}{\delta h^{\beta}(2)}=\delta_{\alpha\beta}\delta(1,2)+\int_L d(3)_{\gamma}\,\Gamma^{\alpha\gamma}(1,3)\chi_L^{\gamma\beta}(3,2), \qquad (3.17)$$

with the latter being a direct consequence of the definition in Eq. (3.14). Using these relations, Eq. (3.13) can be transformed into

$$\frac{\partial}{\partial t_1}\chi_L(1,2)+\int_L d(3)M_L(1,3)\chi_L(3,2)=-\Lambda_L(1,2), \qquad (3.18)$$

where

$$\Lambda_L(1,2)=\int_L d(1')d(1'')\,J(1;1'1'')\times\left(\frac{\delta\langle S^{\alpha'}(1')S^{\alpha''}(1'')\rangle}{\delta h_{\text{eff}}^{\alpha}(2)}\right)_{\vec{h}=0} \qquad (3.19)$$

and

$$M_L(1,2)=\int_L d(3)\,\Gamma(1,3)\Lambda_L(3,2). \qquad (3.20)$$

Now we can easily go over to the retarded response function (see Appendix A) and arrive at

$$\frac{\partial}{\partial t}\chi_r(\vec{q},t)+\int_0^t dt'\,M_r(\vec{q},t-t')\chi_r(\vec{q},t')=-\Lambda_r(\vec{q},t), \qquad (3.21)$$

where $\Lambda_r(\vec{q},t)$ and $M_r(\vec{q},t)$ are the retarded functions instead of the time-ordered ones above and

$$M_r(\vec{q},t)=\Gamma(\vec{q})\Lambda_r(\vec{q},t), \qquad (3.22)$$

$\Gamma(\vec{q})$ is the Fourier transform of $\Gamma(\vec{R})$.

If we now compare Eq. (3.21) with Eq. (2.14), we see that they become identical by choosing

$$\Gamma(\vec{q})=-1/\chi_{is}(\vec{q}). \qquad (3.23)$$

This choice gives an effective field of the form

$$h_{\text{eff}}^{\alpha}(\vec{q},t)=h^{\alpha}(\vec{q},t)-\langle S^{\alpha}(\vec{q},t)\rangle/\chi_{is}(\vec{q}), \qquad (3.24)$$

which we are going to use in the following.

Our main task is now to calculate more explicitly the memory function defined in Eq. (3.19). Once $M_r(\vec{q},t)$ is known, Eq. (3.21) is easily solved by going over to the Fourier transform in time, $\chi_r(\vec{q},\omega)$, and we have then

$$\chi_r(\vec{q},\omega)=\frac{-\Lambda_r(\vec{q},\omega)}{-i\omega+M_r(\vec{q},\omega)}=\chi_{is}(\vec{q})\frac{M_r(\vec{q},\omega)}{-i\omega+M_r(\vec{q},\omega)}. \qquad (3.25)$$

We close this section by making some remarks on our procedure above. The first one concerns the choice of effective field. We recall that the relaxation function and the Mori equation are closely connected with the following situation. A magnetic field $\vec{h}(\vec{q})$, constant in time, is applied for negative times and is suddenly switched off at $t=0$. The relaxation function describes how the induced magnetization $\langle\vec{S}(\vec{q},t)\rangle$ decays to zero for positive times. When the field is applied, two counteracting processes occur. The external field tries to align the spins in a certain direction, whereas "spin-diffusion" processes try to misalign them. In the stationary case, these two processes keep each other in balance. Here we have represented the effect of "spin diffusion" by a certain depolarizing field to be added to the external field. Only when we have deviation from *local equilibrium*, and thus some unbalance between the two processes, will the effective field differ from zero.

Equation (3.25) goes over to the simple spin-diffusion form for $\vec{q}, \omega \to 0$ if $M_r(\vec{q}, \omega) \to Dq^2$ (D is the spin-diffusion constant). From the definition in Eq. (3.19), one immediately extracts a factor q^2 and the result is therefore very plausible. If we had chosen a different effective field, the corresponding memory function would become zero for $\omega = 0$ in order to have $\chi_r(\vec{q}, \omega=0) = \chi_{is}(\vec{q})$. One can further show that $M_r(\vec{q}, \omega)$ must then contain a spin-diffusion pole if $\chi_r(\vec{q}, \omega)$ does. This means that by choosing the effective field as we have done, we have removed an expected spin-diffusion pole from the memory function and possibly gotten this function to behave more regularly for $\vec{q}$ and ω tending to zero.

The microscopic processes connected with spin polarization are necessarily rather complicated. If we suddenly apply a magnetic field, the immediate effect is not a polarization of the spins along the magnetic field. Instead, the spins start to precess in a plane perpendicular to the magnetic field. This is a direct consequence of the commutation relations. Only after some later time would we note how the spins start to align along the magnetic field, and it is an effect of the interactions and relaxation processes. For these particular reasons, we would get into some difficulties if, in Eq. (3.13), we introduce functional differentiation with respect to $\langle \vec{S}(1) \rangle$, in analogy to what was done for phonons.[42] In the spin case, the corresponding functional derivatives are singular for zero time. The initial spin precession is contained in the quantity $\Lambda_L(1,2)$ and the same is true also for the "sloppy spin waves."

IV. CALCULATION OF THE MEMORY FUNCTION

In this section we shall proceed to find a somewhat more explicit expression for the memory function, based on the definition in Eq. (3.19) and on Eq. (3.11). In order to do this, we first introduce a set of irreducible correlation functions through

$$\chi_{0,L}^{\alpha\beta}(1,2) = \delta\langle S^\alpha(1)\rangle / \delta h_{\text{eff}}^\beta(2), \tag{4.1}$$

$$\chi_{0,L}^{\alpha\beta\gamma}(1,2,3) = \delta^2\langle S^\alpha(1)\rangle / \delta h_{\text{eff}}^\beta(2)\delta h_{\text{eff}}^\gamma(3), \tag{4.2}$$

etc. Using Eq. (3.17), we can write

$$\begin{aligned}\chi_L^{\alpha\beta}(1,2) &= \chi_{0,L}^{\alpha\beta}(1,2) + \int_L d(3)_\gamma\, d(4)_\delta\, \chi_{0,L}^{\alpha\gamma}(1,3)\,\Gamma^{\gamma\delta}(3,4)\,\chi_L^{\delta\beta}(4,2)\\ &= \chi_{0,L}^{\alpha\beta}(1,2) + \int_L d(3)_\gamma\, d(4)_\delta\, \chi_{0,L}^{\alpha\gamma}(1,3)\,\Gamma^{\gamma\delta}(3,4)\,\chi_{0,L}^{\delta\beta}(4,2) + \cdots .\end{aligned} \tag{4.3}$$

The last line is obtained by iterating the previous one and expresses χ_L as an infinite series in $\chi_{0,L}$. We further note that

$$\langle T_L S^{\alpha'}(1')S^{\alpha''}(1'')\rangle = \langle S^{\alpha'}(1')\rangle\langle S^{\alpha''}(1'')\rangle - i\chi_L^{\alpha'\alpha''}(1',1''). \tag{4.4}$$

Differentiating with respect to $\vec{h}_{\text{eff}}$, using the last line of Eq. (4.3), gives

$$\begin{aligned}\frac{\delta\langle T_L S^{\alpha'}(1')S^{\alpha''}(1'')\rangle}{\delta h_{\text{eff}}^\beta(2)} &= \langle S^{\alpha'}(1')\rangle\chi_{0,L}^{\alpha''\beta}(1'',2) + \langle S^{\alpha''}(1'')\rangle\chi_{0,L}^{\alpha'\beta}(1',2)\\ &\quad - i\int_L d(3)_\gamma\, d(4)_\delta\, P_L^{\alpha'\gamma}(1',3)\,\chi_{0,L}^{\gamma\beta\delta}(3,2,4)\,P_R^{\delta\alpha''}(4,1''),\end{aligned} \tag{4.5}$$

where[45]

$$P_L^{\alpha\beta}(1,2) = \delta_{\alpha\beta}\delta(1,2) + \int_L d(3)_\gamma\, \chi_{0,L}^{\alpha\gamma}(1,3)\,\Gamma^{\gamma\beta}(3,2) + \cdots \tag{4.6}$$

and

$$P_R^{\alpha\beta}(1,2) = \delta_{\alpha\beta}\delta(1,2) + \int_L d(3)_\gamma\, \Gamma^{\alpha\gamma}(1,3)\,\chi_{0,L}^{\gamma\beta}(3,2) + \cdots . \tag{4.7}$$

If we now insert Eq. (4.5) into the definition for $\Lambda_L(1,2)$, we obtain

$$\Lambda_L(1,2) = -i\int_L d(1')\cdots d(3'')\, J(1;1'1'')P(1',3')P(1'',3'')\,\chi_{0,L}^{\alpha''\alpha'\alpha}(3'',3',2). \tag{4.8}$$

Thus we are faced with the problem of calculating the irreducible three-spin correlation function.

Let us therefore first write Eq. (3.4) in a slightly modified form,

$$\frac{\partial}{\partial t_2}\langle S^{\alpha}(2)\rangle + \int_L d(2')d(2'')\, J(2;2'2'')\langle S^{\alpha'}(2')S^{\alpha''}(2'')\rangle$$
$$= -h_{\text{eff}}^{\alpha'}(2)\langle S^{\alpha''}(2)\rangle + h_{\text{eff}}^{\alpha''}(2)\langle S^{\alpha'}(2)\rangle + \int_L d(4)\,\Gamma(2,4)[\langle S^{\alpha'}(4)\rangle\langle S^{\alpha''}(2)\rangle - \langle S^{\alpha''}(4)\rangle\langle S^{\alpha'}(2)\rangle]. \tag{4.9}$$

Here $\Gamma(2,4)=\delta(t_2-t_4)\Gamma(\vec{R}_2-\vec{R}_4)$. We differentiate the equation twice with respect to $\vec{h}_{\text{eff}}$ and consider the equilibrium situation again. This yields

$$\frac{\partial}{\partial t_2}\chi_{0,L}^{\alpha\alpha'\alpha''}(2,3',3'') + \int_L d(2')d(2'')\, J(2;2'2'')\left(\frac{\delta^2\langle S^{\alpha'}(2')S^{\alpha''}(2'')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')}\right)_{\vec{h}=0}$$
$$= -\delta(2,3')\chi_{0,L}(2,3'') + \delta(2,3'')\chi_{0,L}(2,3') + \int_L d(4)\,\Gamma(2,4)[\chi_{0,L}(4,3')\chi_{0,L}(2,3'') - \chi_{0,L}(4,3'')\chi_{0,L}(2,3')]. \tag{4.10}$$

If we now take the time derivative of $\Lambda_L(1,2)$ in Eq. (4.8), use the fact that it depends only on the difference (t_1-t_2), and insert Eq. (4.10) on the right-hand side, we arrive at the following equation:

$$\frac{\partial}{\partial t_1}\Lambda_L(1,2) = i\int_L d(1')d(1'')\, J(1;1'1'')[\delta(1'',2)\chi_L(1',2) - \delta(1',2)\chi_L(1'',2)]$$
$$- i\int_L d(1')\cdots d(3'')\, J(1;1'1'')P_L(1',3')P_L(1'',3'')\left(\frac{\delta^2\langle S^{\alpha'}(2')S^{\alpha''}(2'')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')}\right)_{\vec{h}=0} J(2;2'2''). \tag{4.11}$$

The right-hand side of Eq. (4.10) has gone into the first term, using the identities

$$P_R^{-1}(1,2) = \delta(1,2) - \int_L d(3)\,\Gamma(1,3)\chi_{0,L}(3,2),\quad P_L^{-1}(1,2) = \delta(1,2) - \int_L d(3)\,\chi_{0,L}(1,3)\Gamma(3,2) \tag{4.12}$$

and

$$\chi_L(1,2) = \int_L d(3)P_L(1,3)\chi_{0,L}(3,2) = \int_L d(3)\chi_{0,L}(1,3)P_R(3,2). \tag{4.13}$$

Equation (4.11) can be approximated in various ways and we shall find that the simplest approximation leads to the conventional mode-mode theory. The first term on the right-hand side contains a factor $\delta(t_1-t_2)$ and it gives the discontinuity of $\Lambda_L(1,2)$ for $t_1=t_2$. For the retarded function, it gives the initial value which depends on the equal-time two-spin correlation function. The second term contains a four-spin correlation function and by differentiating Eq. (4.3) twice we can express it in terms of the irreducible correlation functions. For completeness, we give the full expression

$$\left(\frac{\delta^2\langle S^{\alpha'}(2')S^{\alpha''}(2'')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')}\right)_{\vec{h}=0} = \chi_{0,L}(2',3')\chi_{0,L}(2'',3'')$$
$$- i\int_L d(4')\cdots d(6)\,\chi_{0,L}^{\alpha'\alpha\alpha''}(3',5'',4'')\chi_{0,L}^{\alpha''\alpha\alpha'}(3'',5',4')\{P(5',6)\Gamma(6,5'')\}P(4',2')P(4'',2'')$$
$$- i\int_L d(4')d(4'')\,\chi_{0,L}^{\alpha'\alpha''\alpha'\alpha''}(3',3'',4',4'')P(4',2')P(4'',2''). \tag{4.14}$$

The first term is obtained simply by factorizing the two-spin correlation function on the left. Keeping only this term we will recover the mode-mode theory after making a high-temperature approximation. This means that all the higher-order irreducible correlation functions are ignored and gives

$$\frac{\partial}{\partial t_1}\Lambda_L(1,2) = i\delta(t_1-t_2)\int_L d(1')d(1'')\, J(1;1'1'')[\delta_{\vec{R}_1''\vec{R}_2} - \delta_{\vec{R}_1'\vec{R}_2}]\chi_L(1',1'')$$
$$- i\int_L d(1')\cdots d(2'')\, J(1;1'1'')\chi_L(1',2')\chi_L(1'',2'')J(2;2'2'') \tag{4.15}$$

for the memory function. Together with Eq. (3.18), this gives a closed system of equations for the two-spin correlation function. We could actually go one step further. Ignoring only the four-spin correlation function in Eq. (4.14) and combining this with Eqs. (3.18) and (4.10), we obtain a closed set of equations for the two- and three-spin correlation functions. However, we do not expect this to give any major improvement. The mechanism for the sloppy spin waves seems to be contained in the four-spin correlation function. We will comment on this in Sec. VI.

We note that Eq. (4.11) does not depend explicitly on our choice of effective field. This enters only in the meaning of the functional derivatives and in the relation between $M_L(1,2)$ and $\Lambda_L(1,2)$.

In the treatment of Kawasaki, only the long-wavelength and low-frequency motions were considered, and he argued for a certain renormalization of the exchange integrals in Eq. (4.15). In Appendix B, we show how the same kind of renormalization can be extracted in our formalism.

V. CONVENTIONAL MODE-MODE COUPLING THEORY

We shall here show how we recover the ordinary mode-mode theory by accepting the approximation in Eq. (4.15) and further making a high-temperature approximation. Written in Fourier space Eq. (4.15) reads

$$\frac{\partial}{\partial t}\Lambda_L(\vec{q},t)=\delta(t)\Lambda_r(\vec{q},t=0) - i\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2\chi_L(\vec{q}',t)\chi_L(\vec{q}-\vec{q}',t), \tag{5.1}$$

where

$$\Lambda_r(\vec{q},t=0)=-2\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]C(\vec{q}',t=0) \tag{5.2}$$

and $C(\vec{q},t=0)$ is the equal-time correlation function. We can now easily go over to the retarded function by noting that (see Appendix A)

$$\Lambda_r(\vec{q},t)=\Lambda(\vec{q},t)-\Lambda^{<}(\vec{q},t), \tag{5.3}$$

and that

$$\chi(\vec{q}',t)\chi(\vec{q}-\vec{q}',t)-\chi^{<}(\vec{q}',t)\chi^{<}(\vec{q}-\vec{q}',t) = i[C(\vec{q}',t)\chi_r(\vec{q}-\vec{q}',t)+\chi_r(\vec{q}',t)C(\vec{q}-\vec{q}',t)]. \tag{5.4}$$

We obtain

$$\frac{\partial}{\partial t}\Lambda_r(\vec{q},t)=\delta(t)\Lambda_r(\vec{q},t=0)+\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2 \times[C(\vec{q}',t)\chi_r(\vec{q}-\vec{q}',t)+\chi_r(\vec{q}',t)C(\vec{q}-\vec{q}',t)]. \tag{5.5}$$

In order to have $\Lambda_r(\vec{q},t)\to 0$ for $t\to\infty$, we must impose the condition

$$\Lambda_r(\vec{q},t=0)=-2\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2 \times\int_0^\infty dt\,C(\vec{q}',t)\chi_r(\vec{q}-\vec{q}',t), \tag{5.6}$$

and this provides certain restrictions on the wave-vector-dependent static susceptibility.

In the various derivations of the mode-mode theory, the high-temperature form of Eq. (2.4) is often used, so that [see also Eq. (2.9)]

$$C(\vec{q},t)=(1/\beta)F(\vec{q},t). \tag{5.7}$$

We further recall that for $t\geq 0$ [Eq. (2.8)]

$$\chi_r(\vec{q},t)=-\frac{\partial}{\partial t}F(\vec{q},t), \tag{5.8}$$

and we then find that Eq. (5.5) leads to

$$\Lambda_r(\vec{q},t)=\frac{-1}{\beta}\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2 \times F(\vec{q}',t)F(\vec{q}-\vec{q}',t). \tag{5.9}$$

Together with the relation

$$M_r(\vec{q},t)=-\Lambda_r(\vec{q},t)/\chi_{is}(\vec{q}) \tag{5.10}$$

Eq. (5.9) gives the same expression for $M_r(\vec{q},t)$ as in Eq. (2.13).

The condition in Eq. (5.6) imposes the relation

$$\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]^2\chi_{is}(\vec{q}')\chi_{is}(\vec{q}-\vec{q}') =\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')][\chi_{is}(\vec{q}')-\chi_{is}(\vec{q}-\vec{q}')], \tag{5.11}$$

and it is easily seen that this is satisfied by[27]

$$\chi_{is}(\vec{q})=1/[\lambda-J(\vec{q})]. \tag{5.12}$$

The parameter λ is arbitrary but can be specified by the condition

$$\langle S^\alpha(\vec{R})S^\alpha(\vec{R})\rangle=\int\frac{d\vec{q}}{v}C(\vec{q},t=0)=\frac{1}{\beta}\int\frac{d\vec{q}}{v}\chi_{is}(\vec{q}). \tag{5.13}$$

The last equality follows from Eq. (5.7) at $t=0$. In the long-wavelength limit, Eq. (5.12) leads to the Ornstein-Zernike form

$$\chi_{is}(\vec{q})\sim 1/(\kappa^2+q^2), \tag{5.14}$$

with $\kappa\propto T-T_c$ near the critical point (see Appendix C). This is the same result as obtained in the spherical model,[41] and it deviates significantly from the generally accepted one, which is close to $\kappa\propto(T-T_c)^{2/3}$. A more detailed discussion of the mode-mode theory and, in particular, of the explicit time dependence of $M_r(\vec{q},t)$ is given in Ref. 28.

VI. CONCLUDING REMARKS

In this paper, we have presented a procedure for obtaining equations of motion for the two-spin correlation function and also for the higher-order ones. As expected, the various correlation functions are coupled to each other and we cannot get a closed set of equations without making some approximations. The conventional mode-mode theory was obtained by truncating this hierarchy of equations at the very first stage, ignoring three-spin and higher-order irreducible correlation functions in Eq. (4.14). As mentioned in the Introduction, this theory has been quite successful in explaining various experimental results. It was also mentioned that in other respects it is in strong disagreement with some observations. We refer particularly to the observation of damped spin waves in the paramagnetic region. We may also refer to the poor result obtained for the critical exponent of the static susceptibility.

Concerning the "sloppy spin waves," something is certainly missing in the ordinary mode-mode theory. For smaller times, at least, it is by no means obvious that the factorization made in Eq. (4.15) is a proper approximation. If, instead, we first differentiate $\Lambda_L(1,2)$ in Eq. (4.11) with respect to t_2 we obtain for $t \neq 0$

$$\frac{\partial^2}{\partial t_1^2}\Lambda_L(1,2) = -i\int_L d(1')\cdots d(4'')\, J(1;1'1'')P_L(1',3')P_L(1'',3'') \times\left(\frac{\delta^2\langle S^{\alpha''}(4')S^{\alpha}(4'')S^{\alpha''}(2'')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')}J(2';4'4'') + \frac{\delta^2\langle S^{\alpha'}(2')S^{\alpha}(4'')S^{\alpha'}(4')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')}J(2'';4''4')\right)J(2;2'2''). \tag{6.1}$$

A natural factorization here would be

$$\frac{\delta^2\langle S^{\alpha''}(4')S^{\alpha}(4'')S^{\alpha''}(2'')\rangle}{\delta h_{\text{eff}}^{\alpha'}(3')\delta h_{\text{eff}}^{\alpha''}(3'')} = \langle S^{\alpha''}(4')S^{\alpha''}(2'')\rangle\chi_{0,L}^{\alpha\alpha'\alpha''}(4'',3',3''), \tag{6.2}$$

and this leads to

$$\frac{\partial^2}{\partial t^2}\Lambda_r(\vec{q},t)+\lambda(\vec{q})\Lambda_r(\vec{q},t)=\delta'(t)\Lambda_r(\vec{q},t=0), \tag{6.3}$$

where

$$\lambda(\vec{q}) = 2\int\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')] \times[J(\vec{q}')-J(\vec{q})]C(\vec{q}',t=0). \tag{6.4}$$

Using this in the expression for $\chi_r(\vec{q},t)$ yields

$$\chi_r(\vec{q},\omega)=\frac{\Lambda_r(\vec{q},t=0)}{\omega^2-\lambda(\vec{q})+\Lambda_r(\vec{q},t=0)/\chi_{is}(\vec{q})}, \tag{6.5}$$

which leads to sharp spin waves for $[\lambda(\vec{q})-\Lambda_r(\vec{q},t=0)/\chi_{is}(\vec{q})]>0$. This inequality certainly holds if Eqs. (5.7) and (5.12) are valid.[46] A closer inspection shows that the kind of terms we are here including in $\Lambda_L(1,2)$ are not doubly counted by also including the ordinary mode-mode theory terms. The proper theory should, in our opinion, contain effects of both kinds of terms. This is actually in line with what was done by McLean and Blume[13] in their analysis of the experiments on TMMC. From Eq. (4.11) we can formally extract an equation of the form

$$\frac{\partial^2}{\partial t^2}\Lambda_r(\vec{q},t)+\lambda(\vec{q})\Lambda_r(\vec{q},t)-\int_0^t dt'\,R(\vec{q},t-t')\Lambda_r(\vec{q},t') =\delta'(t)[\Lambda_r(\vec{q},t=0)-\Lambda_r^M(\vec{q},t=0)]+\frac{\partial^2}{\partial t^2}\Lambda_r^M(\vec{q},t), \tag{6.6}$$

where $\Lambda_r^M(\vec{q},t)$ is the memory function in the mode-mode theory and $R(\vec{q},t)$ is a certain functional derivative. It can be shown that

$$R(\vec{q},\omega)\to 0, \tag{6.7}$$

for $\omega\to\infty$, and that

$$R(\vec{q},\omega)\to\lambda(\vec{q}), \tag{6.8}$$

for $\omega\to 0$. This implies that, for very high frequencies, we have a certain "harmonic" restoring force which disappears at lower frequencies. In this sense, the situation is similar to what happens for transverse modes in liquids. In order to draw any useful conclusions, we have to know at least an approximate form for $R(\vec{q},t)$. In particular, we must know how it approaches $\lambda(\vec{q})$ for $\omega\to 0$ in order to determine whether or not the mode-mode theory is correct for low-frequency dynamics. Our remarks above indicate that we should not apply the mode-mode theory for short times, at least not for temperatures where "sloppy spin waves" appear. Our procedure in Sec. V to extract an equation for $\chi_{is}(\vec{q})$ would then be incorrect.

It is clear that much more work has to be done on this problem. At present, we are trying to find an appropriate transport equation for $\chi^{\alpha\beta}(1,2)$ and thereby to get some more insight into the whole problem.

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APPENDIX A

In the way time ordering has been introduced, we can split $\chi_L(1,2)$ into four different parts. Let us denote the time interval $(-\infty, +\infty)$ by F (forward) and the interval $(+\infty, -\infty)$ by B (backward).[42] We have then, for

$t_1, t_2 \in F$,

$$\chi_L^{\alpha\beta}(1,2) = \chi^{\alpha\beta}(1,2) = i\begin{cases}\langle S^\alpha(1)S^\beta(2)\rangle_c, & t_1 > t_2\\ \langle S^\beta(2)S^\alpha(1)\rangle_c, & t_1 < t_2\end{cases} \tag{A1}$$

$t_1 \in F, t_2 \in B$,

$$\chi_L^{\alpha\beta}(1,2) = \chi^{\alpha\beta<}(1,2) = i\langle S^\beta(2)S^\alpha(1)\rangle_c, \tag{A2}$$

$t_1 \in B, t_2 \in F$,

$$\chi_L^{\alpha\beta}(1,2) = \chi^{\alpha\beta>}(1,2) = i\langle S^\alpha(1)S^\beta(2)\rangle_c, \tag{A3}$$

$t_1, t_2 \in B$,

$$\chi_L^{\alpha\beta}(1,2) = \tilde{\chi}^{\alpha\beta}(1,2) = i\begin{cases}\langle S^\beta(2)S^\alpha(1)\rangle_c, & t_1 > t_2\\ \langle S^\alpha(1)S^\beta(2)\rangle_c, & t_1 < t_2.\end{cases} \tag{A4}$$

In a similar way, any other time-ordered function can be split into four parts. From Eq. (3.18), we now get, for $t_1, t_2 \in F$,

$$\frac{\partial}{\partial t_1}\chi(1,2) + \int_{-\infty}^{\infty} d(3)\,[M(1,3)\chi(3,2) - M^<(1,3)\chi^>(3,2)] = -\Lambda(1,2), \tag{A5}$$

and similarly for $t_1 \in F, t_2 \in B$,

$$\frac{\partial}{\partial t_1}\chi^<(1,2) + \int_{-\infty}^{\infty} d(3)\,[M(1,3)\chi^<(3,2) - M^<(1,3)\tilde{\chi}(3,2)] = -\Lambda^<(1,2). \tag{A6}$$

The time integration extends here only over the interval $(-\infty, +\infty)$. Subtracting (A6) from (A5) and using

$$\chi_r(1,2) = \chi(1,2) - \chi^<(1,2) = \chi^>(1,2) - \tilde{\chi}(1,2), \tag{A7}$$

we obtain

$$\frac{\partial}{\partial t_1}\chi_r(1,2) + \int_{-\infty}^{\infty} d(3)\,[M(1,3)\chi_r(3,2) - M^<(1,3)\chi_r(3,2)] = -\Lambda_r(1,2). \tag{A8}$$

The two terms within the square brackets combine to give $M_r(1,3)\chi_r(3,2)$, and we finally arrive at

$$\frac{\partial}{\partial t_1}\chi_r(1,2) + \int_{-\infty}^{\infty} d(3)\,M_r(1,3)\chi_r(3,2) = -\Lambda_r(1,2). \tag{A9}$$

Here the time integration can actually be restricted to the interval (t_1, t_2).

APPENDIX B

We shall here consider only the long-wavelength (hydrodynamic) modes and follow the prescription of Kawasaki[35] to eliminate all the rapid short-wavelength modes. Therefore, we split the wave-vector space into an inner region, denoted by v_0, and a remaining outer region. The former is chosen very small compared with the whole Brillouin zone, but, near the critical point, it will contain all the relevant critical fluctuations. All modes of wave vectors inside v_0 are considered to be hydrodynamic and all the other modes are said to be nonhydrodynamic and will be averaged out in the final equation of motion.

Let us go back to Eq. (3.4) and write the second term in the Fourier space, i.e.,

$$\int \frac{d\vec{q}'}{v}[J(\vec{q}') - J(\vec{q}-\vec{q}')]\langle S^{\alpha'}(\vec{q}', t)S^{\alpha''}(\vec{q}-\vec{q}', t)\rangle. \tag{B1}$$

We consider $\vec{q}\in v_0$. The integral can then be split into two parts, with $\vec{q}'\in v_0$ and $\vec{q}'\notin v_0$, respectively. We concentrate on the latter part. This contains short-wavelength modes (relative to the other part), and these may rapidly attain local equilibrium in the presence of the slowly varying inhomogeneities, having wave vectors inside v_0. The correlation function $\langle S^{\alpha'}(\vec{q}', t)S^{\alpha''}(\vec{q}-\vec{q}', t)\rangle$ will, for $\vec{q}'\notin v_0$, become a certain functional of $\langle S^\alpha(\vec{q}, t)\rangle$, $\langle S^{\alpha'}(\vec{q}'', t)S^{\alpha''}(\vec{q}-\vec{q}'', t)\rangle_c$, etc., where $\vec{q}, \vec{q}''$, etc. all lie inside v_0. Following the procedure of Kawasaki, we write

$$\begin{aligned}\delta\langle S^{\alpha'}(\vec{q}', t)S^{\alpha''}(\vec{q}-\vec{q}', t)\rangle &= \frac{(S^{\alpha'}(\vec{q}')S^{\alpha''}(\vec{q}-\vec{q}'), S^\alpha(-\vec{q}))}{(S^\alpha(\vec{q}), S^\alpha(-\vec{q}))}\\ &\times \delta\langle S^\alpha(\vec{q}, t)\rangle + \int_{\vec{q}''\in v_0}\frac{d\vec{q}''}{v}\\ &\times \frac{(S^{\alpha'}(\vec{q}')S^{\alpha''}(\vec{q}-\vec{q}'), S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}{(S^{\alpha'}(\vec{q}'')S^{\alpha''}(\vec{q}-\vec{q}''), S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}\\ &\times \delta\langle S^{\alpha'}(\vec{q}'', t)S^{\alpha''}(\vec{q}-\vec{q}'', t)\rangle_c + \cdots,\end{aligned} \tag{B2}$$

where [cf. Eq. (2.7)]

$$(A, B) = \int_0^\beta d\lambda\,\langle e^{\lambda H}Ae^{-\lambda H}B\rangle, \tag{B3}$$

and hence the coefficients in the above expansion are the isothermal ones. This means that we should always consider times larger than the relaxation time for the nonhydrodynamic modes.

Inserting (B2) into (B1) and considering the equilibrium situation, we find that

$$\int \frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]\frac{\delta\langle S^{\alpha'}(\vec{q}',t)S^{\alpha''}(\vec{q}-\vec{q}',t)\rangle}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}$$

$$=-\frac{\delta\langle \dot{S}^{\alpha}(\vec{q},t)\rangle}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}$$

$$=-\frac{(\dot{S}^{\alpha}(\vec{q}),S^{\alpha}(-\vec{q}))}{(S^{\alpha}(\vec{q}),S^{\alpha}(-\vec{q}))}\chi_{0,L}(\vec{q},t)-\int_{\vec{q}''\in v_0}\frac{d\vec{q}''}{v}$$

$$\times\frac{(\dot{S}^{\alpha}(\vec{q}),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}{(S^{\alpha'}(\vec{q}'')S^{\alpha''}(\vec{q}-\vec{q}''),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}$$

$$\times\frac{\delta\langle S^{\alpha'}(\vec{q}'',t)S^{\alpha''}(\vec{q}-\vec{q}'',t)\rangle_c}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}. \tag{B4}$$

Direct evaluation shows that

$$(\dot{S}^{\alpha}(\vec{q}),S^{\alpha}(-\vec{q}))=0, \tag{B5}$$

and, following Kawasaki, we approximate

$$\frac{(\dot{S}^{\alpha}(\vec{q}),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}{(S^{\alpha'}(\vec{q}'')S^{\alpha''}(\vec{q}-\vec{q}''),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}$$

$$=\left(\frac{1}{\chi_{is}(\vec{q}'')}-\frac{1}{\chi_{is}(\vec{q}-\vec{q}'')}\right). \tag{B6}$$

In obtaining this relation, the numerator is calculated exactly. Also, the denominator is factored and the high-temperature relation between $C(\vec{q})$ and $\chi_{is}(\vec{q})$ is used, remembering that here only long-wavelength modes are involved.

This now means that in Eq. (3.19) we change the expression for $\Lambda_L(\vec{q},t)$ to

$$\Lambda_L(\vec{q},t)=-\int_{\vec{q}'\in v_0}\frac{d\vec{q}'}{v}\left(\frac{1}{\chi_{is}(\vec{q}')}-\frac{1}{\chi_{is}(\vec{q}-\vec{q}')}\right)$$

$$\times\frac{\delta\langle S^{\alpha'}(\vec{q}',t)S^{\alpha''}(\vec{q}-\vec{q}',t)\rangle}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}. \tag{B7}$$

The integration is now restricted to only wave vectors inside v_0, and the exchange integral has been renormalized as

$$J(\vec{q}')-J(\vec{q}-\vec{q}')\to-\left(\frac{1}{\chi_{is}(\vec{q}')}-\frac{1}{\chi_{is}(\vec{q}-\vec{q}')}\right). \tag{B8}$$

If we follow the same procedure as above for handling the second term in Eq. (4.9), we get the same renormalization of the last exchange integral in Eq. (4.11). In other words, the expression for the memory function is unchanged, except that only small wave vectors are involved and the exchange integral is renormalized as in (B8). Still, irreducible four-spin correlation functions are present and it is not obvious to us that they are only of minor importance.

In the derivation above, we did not exactly follow our original intention. When inserting (B2) into (B1) we should have excluded $\vec{q}'\in v_0$. We have no reason to believe that for these wave vectors the correlation function $\langle S^{\alpha'}(\vec{q}'t)S^{\alpha''}(\vec{q}-\vec{q}',t)\rangle$ takes its local equilibrium value. If we take this into account we get in (B7) extra terms

$$\int_{\vec{q}'\in v_0}\frac{d\vec{q}'}{v}[J(\vec{q}')-J(\vec{q}-\vec{q}')]\left(\frac{\delta\langle S^{\alpha'}(\vec{q}',t)S^{\alpha''}(\vec{q}-\vec{q}',t)\rangle_c}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}-\frac{(S^{\alpha'}(\vec{q}')S^{\alpha''}(\vec{q}-\vec{q}'),S^{\alpha}(-\vec{q}))}{\chi_{is}(\vec{q})}\chi_{0,L}(\vec{q},t)\right.$$

$$-\int_{\vec{q}''\in v_0}\frac{d\vec{q}''}{v}\frac{(S^{\alpha'}(\vec{q}')S^{\alpha''}(\vec{q}-\vec{q}'),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}{(S^{\alpha'}(\vec{q}'')S^{\alpha''}(\vec{q}-\vec{q}''),S^{\alpha'}(-\vec{q}'')S^{\alpha''}(-\vec{q}+\vec{q}''))}$$

$$\left.\times\frac{\delta\langle S^{\alpha'}(\vec{q}'',t)S^{\alpha''}(\vec{q}-\vec{q}'',t)\rangle_c}{\delta h^{\alpha}_{\rm eff}(\vec{q},0)}\right). \tag{B9}$$

These terms were ignored by Kawasaki without any comments. It seems to us that they should have no major effect on the results. The mode-mode equations (2.12) and (2.13) give precisely the dynamical scaling of Halperin and Hohenberg[23] after the above renormalization of the exchange integrals. Without the renormalization, one gets a discrepancy which is connected with the small critical exponent η.

The mode-mode equation now becomes applicable only for the long-wavelength modes and for long times. So, for instance, we cannot give any connection to the initial value $\Lambda_r(\vec{q},t=0)$, nor can we determine $\chi_{is}(\vec{q})$ self-consistently. The first term on the right-hand side of Eq. (4.11) is also modified according to the substitution in (B8). This follows directly from Eq. (B7). As a consequence of this, Eq. (5.11) is changed to

$$\int_{q'\in v_0}\frac{d\vec{q}'}{v}\left(\frac{1}{\chi_{is}(\vec{q}')}-\frac{1}{\chi_{is}(\vec{q}-\vec{q}')}\right)^2\chi_{is}(\vec{q}')\chi_{is}(\vec{q}-\vec{q}')$$

$$=-\int_{\vec{q}'\in v_0}\frac{d\vec{q}'}{v}\left(\frac{1}{\chi_{is}(\vec{q}')}-\frac{1}{\chi_{is}(\vec{q}-\vec{q}')}\right)$$

$$\times\,[\chi_{is}(\vec{q}\,')-\chi_{is}(\vec{q}-\vec{q}\,')], \qquad (B10)$$

and this relation is just an identity. Therefore it does not lead to any restriction on the form of $\chi_{is}(\vec{q})$. Actually, in evaluating the memory function we should use the exact values for $\chi_{is}(\vec{q})$, which may be obtained either from experiments or some other theoretical calculations. Whether the physics contained in $\chi_{is}(\vec{q})$ is then consistent with that retained in the mode-mode equation is, of course, a very basic question to answer. We can only conclude that the mode-mode theory and the prescription of using the exact $\chi_{is}(\vec{q})$ become consistent after making the renormalization of the exchange integral suggested by Kawasaki.

APPENDIX C

Accepting Eqs. (5.12) and (5.13) of the mode-mode theory, we will show that $\kappa \propto T-T_c$. We consider Eq. (5.13) for $T=T_c$ and for a slightly higher temperature and take the difference. This then leads to

$$\int \frac{d\vec{q}}{v}\,[\chi_{is}(\vec{q};\,T)-\chi_{is}(\vec{q};\,T_c)]=\tfrac{1}{3}S(S+1)(\beta-\beta_c). \qquad (C1)$$

The integral is dominated by the small-$\vec{q}$ region, and we can then use the form

$$\chi_{is}(\vec{q})=a/(\kappa^2+q^2), \qquad (C2)$$

where a is assumed to be a temperature-independent constant and where $\kappa\to 0$ for $T\to T_c$. Inserting this into the integral we immediately conclude that

$$a\int\frac{d\vec{q}}{v}\left(\frac{1}{\kappa^2+q^2}-\frac{1}{q^2}\right)\propto\kappa, \qquad (C3)$$

for $T\to T_c$, and thus that $\kappa\propto T-T_c$.

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[44]We could, without introducing any essential mathematical complications, let $\Gamma^{\alpha\beta}(1, 2)$ be a general time-ordered tensor, but we shall not do that in this paper. Other generalizations are also possible, but the above ansatz is sufficient for our purposes.

[45]At nonequilibrium we have to distinguish between P_L (left) and P_R (right), but this is not necessary at equilibrium, where we use the notation P.

[46]A similar decoupling was used by R. H. Knapp, Jr. and D. ter Haar [J. Stat. Phys. 1, 149 (1969)], and they also got sharp spin waves throughout the paramagnetic region. See also H. S. Bennet and P. C. Martin [Phys. Rev. 138, A608 (1965)] where a slightly different decoupling gives no evidence for spin waves.