

Kinetic theory of the anisotropic Heisenberg Hamiltonian

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A set of kinetic equations for the correlation functions describing the simultaneous propagation of two-spin fluctuations in an anisotropic Heisenberg paramagnet [$H = -(1/2) \sum_{ij} a_{ij} \vec{S}_i \cdot \vec{S}_j - (1/2) \sum_{ij} b_{ij} S_i^z S_j^z$] is obtained that reduces to earlier results in the case of vanishing anisotropy. The equations conserve the total spin and energy, and have for their equilibrium solution the spherical-model static correlation functions. A prescription for obtaining a diagrammatic expansion of the moments of the spectral density of a single-spin fluctuation mode, $\langle \omega^n \rangle_q$, for any n , to lowest order in $1/C$, where C is the number of spins in the range of the interaction is given. The kinetic equation can be used to calculate this spectral density by providing a partial summation of terms in the diagrammatic representation of the moment expansion. The spectral density obtained by solving the kinetic equation will have the correct second and fourth moments to lowest order in $1/C$. An approximate solution for the response of the $q=0$ mode in a dipole lattice in a strong magnetic field is obtained using a constant-relaxation-time approximation, and shown to be in good agreement with the measurements in CaF_2 . A comparison of the theory with other attempts to calculate the free induction decay is given.

INTRODUCTION

In this paper we present a kinetic theory for the anisotropic Heisenberg Hamiltonian,

$$H = -\frac{1}{2} \hbar \sum_{ij} (a_{ij} \vec{S}_i \cdot \vec{S}_j + b_{ij} S_i^z S_j^z)$$

which is capable of describing most of the measurable dynamical properties of the system. The Hamiltonian describes, with particular choices of coefficients, the Heisenberg, Ising, x - y models as well as a system of interacting magnetic dipoles in a strong magnetic field.

The explicit calculation will be restricted to the truncated dipolar Hamiltonian, where the existence of precise data on the free induction decay, $[\sum_i \langle S_i^z(t) S_i^z(0) \rangle]$ in an essentially ideal dipole lattice makes possible a strong test of the accuracy of the theory.

The theory to be presented is an extension of earlier work done on the isotropic system¹ ($b_{ij} = 0$). The principal results obtained for the isotropic case can all be extended to the anisotropic system, and the difficulties connected with the lack of any suitable expansion parameter for the calculation of the dynamical properties of the system can be overcome in the same way.

Two distinct but complementary approaches are used: (i) diagrammatic expansion of the matrix elements of the resolvent of the Liouville operator for the system; (ii) a kinetic theory for the time evolution of the spin correlation functions.

The first method provides a diagrammatic way to calculate the moments of the spectral density. The second and fourth moments are calculated

explicitly and it is shown that $\langle \omega^{2n} \rangle_q \propto [\frac{1}{3} S(S+1)]^n$ to lowest order in $1/C$, where C is the number of spins in the range of the interaction. The diagrams for the moments may be summed to obtain the time-dependent correlation function, and the usual resummation procedure employed to express the series more compactly in terms of renormalized propagators. (The lowest-order diagram obtained in this way corresponds to the mode-mode coupling equations.) However the nonexistence of a small parameter prevents the selection of a dominant set of diagrams. To overcome this difficulty a set of kinetic equations for the two-spin $[\langle S^\alpha(q_1, t) S^\beta(q_2, t) \rangle]$ correlation function is obtained. The derivation is based on a cluster expansion of the equations of motion and on an intuitively plausible renormalization. The kinetic equations so obtained are shown to conserve total spin and energy and provide a self-consistent nonlinear equation for the time-dependent vertex function governing the decay of a fluctuation mode of wave vector \vec{q} into two modes of wave vectors \vec{q}_1 and \vec{q}_2 . The equation, when iterated, generates a series expansion whose individual terms can be put into correspondence with a subset of graphs appearing in the moment expansion of the spectral density. Thus its solution provides a partial resummation of the diagrams.

To obtain concrete results, an approximate solution for the vertex function at infinite temperatures is obtained using a constant-relaxation-time approximation. Improvements can be obtained by iteration. The kinetic equations are valid at any temperature $T \geq T_c$.

Underlying the diagrammatic expansion is a physical picture in which the states of the system are taken to be one-, two-, ..., many-spin-fluctuations modes superimposed on a background determined by thermal equilibrium. The dynamics is then determined by the decay rates of these modes into one another. This picture is consistent in an operational sense, with the exact equations of motion, in the lowest order of the expansion of the dynamical matrix elements in $1/C$. In this approximation the only decay process possible is the decay of one mode into two others.

I. MATHEMATICAL PRELIMINARIES

The system under study is composed of N identical particles with spin \vec{S} localized at lattice sites, denoted by i . The Hamiltonian describing the system is

$$\mathcal{H} = -\frac{1}{2}\hbar \sum_{(i,j)} (a_{ij}\vec{S}_i \cdot \vec{S}_j + b_{ij}S_i^z S_j^z). \quad (1.1)$$

The coefficients a_{ij} and b_{ij} are arbitrary and they shall be kept as such throughout the discussion. \vec{S} is the spin operator, of magnitude S , associated with lattice site i , whose components satisfy the equal-time commutation relations

$$[S_i^+, S_j^-] = 2\delta_{ij}S_i^z, \quad (1.2)$$

$$[S_i^z, S_j^\pm] = \pm \delta_{ij}S_i^\pm. \quad (1.3)$$

Defining the operator $\vec{S}(q)$ as $N^{-1/2}\sum_i e^{-iq\cdot r_i}\vec{S}_i$ one has

$$\mathcal{H} = -\frac{1}{2}\hbar \sum_q [a(q)\vec{S}(q) \cdot \vec{S}(-q) + b(q)S^z(q)S^z(-q)], \quad (1.4)$$

where

$$a(q) = N^{-1/2} \sum_i e^{-iq \cdot (\vec{r}_i - \vec{r}_j)} a_{ij}, \quad (1.5)$$

$$b(q) = N^{-1/2} \sum_j e^{-iq \cdot (\vec{r}_i - \vec{r}_j)} b_{ij}, \quad (1.6)$$

and the equal-time commutation relation for the components of $\vec{S}(q)$ are

$$[S^+(q), S^-(q')] = 2N^{-1/2}S^z(q+q'), \quad (1.7)$$

$$[S^z(q), S^\pm(q')] = \pm N^{-1/2}S^\pm(q+q'). \quad (1.8)$$

The equation of motion for the spin components are

$$i \frac{\partial}{\partial t} S^z(q) = \frac{1}{2}N^{-1/2} \sum_{q'} [a(q-q') - a(q')] S^-(q') S^+(q-q') \quad (1.9)$$

$$i \frac{\partial}{\partial t} S^\pm(q) = \pm N^{-1/2} \sum_{q'} [a(q') - a(q-q')] + b(q') S^\pm(q') S^\pm(q-q'). \quad (1.10)$$

The response of the system to an external perturbation² is given by

$$\Sigma^\alpha(q, z) = \int_0^\infty e^{izt} \langle S^\alpha(q) | S^\alpha(q, t) \rangle dt \quad (1.11)$$

or

$$\Sigma^\alpha(q, z) = i \langle S^\alpha(q) | [z - \mathcal{L}]^{-1} | S^\alpha(q) \rangle, \quad (1.11a)$$

where

$$\langle A | B \rangle = \langle \int_0^\beta e^{\tau\mathcal{H}} A^\dagger e^{-\tau\mathcal{H}} B d\tau \rangle,$$

$$\langle \langle O \rangle \rangle = \text{Tr} \rho_{\text{eq}} O,$$

$$\rho_{\text{eq}} = e^{-\beta\mathcal{H}} / \text{Tr} e^{-\beta\mathcal{H}},$$

and $\alpha = z, \pm$. \mathcal{L} is the Liouville operator for the system defined as $\mathcal{L}O = (1/\hbar)[O, \mathcal{H}]$, and $\langle \mathcal{L}A | B \rangle = \langle A | \mathcal{L}B \rangle$, $\langle \mathcal{L}A | B \rangle = \langle [B, A^\dagger] \rangle$.

It can be shown³ that

$$\Sigma^\alpha(\vec{q}, z) = i\chi^\alpha(\vec{q}, 0)[z - \phi^\alpha(\vec{q}, z)]^{-1}, \quad (1.12)$$

where

$$\phi^\alpha(\vec{q}, z) = \langle (I - P^\alpha)\mathcal{L}S^\alpha(\vec{q}) | [z - (I - P^\alpha)\mathcal{L}(I - P^\alpha)]^{-1} \times [(I - P^\alpha)\mathcal{L}S^\alpha(\vec{q})] / \chi^\alpha(\vec{q}, 0) \rangle, \quad (1.13)$$

$$\chi^\alpha(\vec{q}, 0) = \langle S^\alpha(\vec{q}) | S^\alpha(\vec{q}) \rangle.$$

\mathcal{L} is an operator on the linear vector space consisting of all bounded operators on the Hilbert space of the spins which we will call V . P^α is the projection operator $\sum_q |S^\alpha(q)\rangle \langle S^\alpha(q)| / \chi^\alpha(q)$ and I is the identity operator on V .

We define the vertex functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ with $\vec{q}_1 + \vec{q}_2 = \vec{q}$ as

$$\Gamma^+(\vec{q}_1, \vec{q}_2; z) = \langle S^-(\vec{q}_1) S^+(\vec{q}_2) | \times [z - (I - P^+) \mathcal{L} (I - P^+)]^{-1} | \mathcal{L} S^+(\vec{q}) \rangle / \chi^+(q, 0), \quad (1.14)$$

$$\Gamma^-(\vec{q}_1, \vec{q}_2; z) = \langle S^+(\vec{q}_1) S^-(\vec{q}_2) | \times [z - (I - P^-) \mathcal{L} (I - P^-)]^{-1} | \mathcal{L} S^-(q) \rangle / \chi^-(q, 0). \quad (1.15)$$

It follows then that

$$\phi^+(\vec{q}, z) = \frac{1}{2}N^{-1/2} \sum_{q'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^+(\vec{q}', \vec{q} - \vec{q}'; z), \quad (1.16)$$

$$\phi^-(\vec{q}, z) = -N^{-1/2} \sum_{q'} [a(\vec{q}') - a(\vec{q} - \vec{q}')] + b(\vec{q}') \Gamma^-(\vec{q}', \vec{q} - \vec{q}'; z). \quad (1.17)$$

We will derive a set of equations for the vertex functions, from which the ϕ functions can be determined. The vertex functions describe the time behavior of the longitudinal and transverse two-spin correlation functions for particular initial conditions.

II. MOMENT EXPANSION AND DIAGRAMMATIC METHOD

The standard method to calculate the moments of the spectral line, used in NMR and EPR, usually for $T = \infty$, is to evaluate the multiple commutation relation and then evaluate the equilibrium averages of the obtained result. The procedure

used here consists of representing the moments as products of the matrix elements of \mathcal{L} . Closely related procedures have been developed by Resibois and De Leener⁴ and Wegner.⁵ In this section, the second and fourth moments are calculated. There is no attempt to give details on the basic theory of the diagrams. This can be found in Reiter's¹ work. However, some basic formalism is given in order to make the results obtained understandable.

The resolvent operator can be written as

$$[z - \mathcal{L}]^{-1} = z^{-1} \sum_0^{\infty} (z^{-1} \mathcal{L})^n. \quad (2.1)$$

Using this result in (1.11a)

$$\Sigma^{\alpha}(\vec{q}, z) = i \sum_0^{\infty} z^{-(n+1)} \langle S^{\alpha}(\vec{q}) | \mathcal{L}^n | S^{\alpha}(\vec{q}) \rangle. \quad (2.2)$$

This result is the Laplace transform of the expansion in the time domain given by

$$\Sigma^{\alpha}(\vec{q}, t) = \sum_0^{\infty} \frac{(-it)^n}{n!} \langle S^{\alpha}(\vec{q}) | \mathcal{L}^n | S^{\alpha}(\vec{q}) \rangle. \quad (2.3)$$

The spectral density can be obtained from the response function

$$\frac{\chi''^{\alpha}(\vec{q}, \omega)}{\omega} = \text{Re} \Sigma^{\alpha}(\vec{q}, \omega + i\epsilon).$$

The Kramers-Kronig relation together with (2.4) gives the following result:

$$\text{Im} \Sigma^{\alpha}(\vec{q}, \omega + i\epsilon) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} \frac{\chi''^{\alpha}(\vec{q}, \omega')}{\omega'(\omega - \omega')} d\omega'. \quad (2.5)$$

In the last expression expand $(\omega - \omega')^{-1}$ in powers of ω'/ω and compare the result obtained with expression (2.2). The moments of the spectral density are found to be

$$\begin{aligned} \langle \omega^n \rangle_{\vec{q}}^{\alpha} &= \frac{1}{\pi} \int_{-\infty}^{\infty} \omega^{n-1} \frac{\chi''^{\alpha}(\vec{q}, \omega)}{\chi^{\alpha}(\vec{q}, 0)} d\omega \\ &= \langle S^{\alpha}(\vec{q}) | \mathcal{L}^n | S^{\alpha}(\vec{q}) \rangle / \chi^{\alpha}(\vec{q}, 0). \end{aligned} \quad (2.6)$$

Due to translation and inversion symmetry of the Hamiltonian the odd moments are zero.

The same expansion for $(z - \mathcal{L}')^{-1}$, $\mathcal{L}' = (1 - P^{\alpha}) \times \mathcal{L}(1 - P^{\alpha})$ can be used for $\phi^{\alpha}(\vec{q}, z)$ defined by (1.13).

From (1.13) one gets

$$\begin{aligned} \phi^{\alpha}(\vec{q}, z) &= \sum_0^{\infty} \langle \mathcal{L} S^{\alpha}(\vec{q}) | (\mathcal{L}')^{2n} \\ &\quad \times | S^{\alpha}(\vec{q}) \rangle z^{-(2n+1)} / \chi^{\alpha}(\vec{q}, 0) \\ &= \sum_0^{\infty} \langle \Omega^{2n+2} \rangle_{\vec{q}}^{\alpha} z^{-(2n+1)}. \end{aligned} \quad (2.7)$$

The last relation defines $\langle \Omega^{2n+2} \rangle_{\vec{q}}^{\alpha}$. Therefore for large $|z|$,

$$\phi^{\alpha}(\vec{q}, z) = \frac{\langle \omega^2 \rangle_{\vec{q}}^{\alpha}}{z} + \frac{\langle \Omega^4 \rangle_{\vec{q}}^{\alpha}}{z^3} + O(1/z^5). \quad (2.8)$$

This last result will become important when one tries to obtain the moments from the kinetic equa-

tions for the spin correlation functions.

There is a relation between the quantity $\langle \Omega^{2n+2} \rangle_{\vec{q}}^{\alpha}$ defined as

$$\langle \Omega^{2n+2} \rangle_{\vec{q}}^{\alpha} \equiv \langle \mathcal{L} S^{\alpha}(\vec{q}) | (\mathcal{L}')^{2n} | \mathcal{L} S^{\alpha}(\vec{q}) \rangle / \chi^{\alpha}(\vec{q}, 0)$$

and the moments $\langle \omega^{2n} \rangle_{\vec{q}}^{\alpha}$ defined by (2.6).

In particular,

$$\begin{aligned} \langle \Omega^2 \rangle_{\vec{q}}^{\alpha} &= \langle \omega^2 \rangle_{\vec{q}}^{\alpha}, \\ \langle \Omega^4 \rangle_{\vec{q}}^{\alpha} &= \langle \omega^4 - \langle \omega^2 \rangle_{\vec{q}}^{\alpha} \rangle_{\vec{q}}^{\alpha}. \end{aligned}$$

In order to calculate the moments, insert a complete set of states in V between the factors of \mathcal{L} in expression (2.6). This complete set can be obtained from the $(2S+1)^2$ tensor operators Y_i^{nm} defined by

$$\sum_{m=-n}^{+n} Y_i^{nm} t^m = (-t S_i^+ + 2S_i^z + S_i^- / t)^n, \quad (2.9)$$

where $n = 0, \dots, 2S$, and t is a parameter.

The set obtained by taking all possible products of the Y_i^{nm} for all i, n, m is a complete set, because any operator in the Hilbert space V_i can be expressed by linear combinations of the tensor operators.

In the following the case $T = \infty$, $S = \frac{1}{2}$ is described. For $T = \infty$ the inner product is given by

$$\begin{aligned} \langle A | B \rangle &= (A^\dagger, B) \xrightarrow{T \rightarrow \infty} \beta [\text{Tr} A^\dagger B / (2S+1)^N], \\ \beta &= 1/kT. \end{aligned}$$

Define the new inner product

$$\langle A | B \rangle_{\infty} \equiv (1/\beta) \langle A | B \rangle = \text{Tr} A^\dagger B / (2S+1)^N. \quad (2.10)$$

The operator A_i^{nm} defined by (2.11) are orthonormal,

$$A_i^{nm} \equiv Y_i^{nm} / c_{nm}^{1/2}, \quad (2.11)$$

where

$$c_{nm} = \frac{(2S+1+n)!(n!)^2}{(2n+1)^2(2S-n)!(n-m)!(n+m)!} \frac{2n!}{(2S+1)}$$

and

$$\langle A_i^{nm} | A_j^{n'm'} \rangle_{\infty} = \delta_{ij} \delta_{n'm} \delta_{mm'}.$$

In the $(2S+1)$ dimensional space there are $(2S+1)^2$ independent operators. For $S = \frac{1}{2}$ those operators are I, S^+, S^-, S^z .

Take for the complete normalized set the operators given by:

$$\begin{aligned} A_i^{00} &= I, \\ A_i^{1,-1} &= \frac{S_i^-}{[\frac{2}{3}S(S+1)]^{1/2}}, \\ A_i^{1,1} &= -\frac{S_i^+}{[\frac{2}{3}S(S+1)]^{1/2}}, \\ A_i^{1,0} &= \frac{S_i^z}{[\frac{1}{3}S(S+1)]^{1/2}}. \end{aligned} \quad (2.12)$$

The identify operator on the space V is

$$I = |A^{00}\rangle\langle A^{00}| + \sum_i \sum_{n=1}^{2S} \sum_{m=-n}^n |A_i^{nm}\rangle\langle A_i^{nm}| + \sum_{\{i,j\}} \sum_{n=1}^{2S} \sum_{n'=1}^{2S} \sum_{m=-n}^n \sum_{m'=-n'}^{n'} |A_i^{nm}A_j^{n'm'}\rangle\langle A_i^{nm}A_j^{n'm'}| + \dots, \quad (2.13)$$

where $\{i, j\}$ is summed over all distinct pairs with $i \neq j$.

The moments are obtained by inserting the identify operator I between the powers of \mathcal{L} . The second moment is given by

$$\langle \omega^2 \rangle_{\vec{q}}^\alpha = N^{-1} \sum_j e^{-i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)} \times \sum_{\{A\}} (A_i^{1\alpha} | \mathcal{L} | \{A\} \rangle_\infty \langle \{A\} | \mathcal{L} | A_i^{1\alpha} \rangle_\infty), \quad (2.14)$$

where $\{A\}$ denotes all intermediate states. From (2.14) one sees that the moments are given by products of the matrix elements of \mathcal{L} . In the graphical representation, those matrix elements are the vertices.

In the momentum representation denote the operators $A_i^{1\alpha}$ by $A^\alpha(\vec{q})$, where $\alpha = z, \pm$. From the equations of motion for $S^\alpha(\vec{q})$ the following results are obtained:

$$\mathcal{L}A^\alpha(\vec{q}) = -[\frac{1}{3}S(S+1)]^{1/2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \times A^{-1}(\vec{q}') A^{+1}(\vec{q} - \vec{q}'), \quad (2.15a)$$

$$\mathcal{L}A^{+1}(\vec{q}) = [\frac{1}{3}S(S+1)]^{1/2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q} - \vec{q}')] + b(\vec{q}') A^0(\vec{q}') A^{+1}(\vec{q} - \vec{q}'). \quad (2.15b)$$

Using Eqs. (2.15) and the orthogonality condi-

tion for the states the matrix elements can be obtained. An example of a nonzero matrix element is

$$\begin{aligned} \langle A(\vec{q}) | \mathcal{L} | A^{+1}(\vec{q}_1) A^{-1}(\vec{q}_2) \rangle \\ = \langle \mathcal{L}A^\alpha(\vec{q}) | A^{+1}(\vec{q}_1) A^{-1}(\vec{q}_2) \rangle \\ = -N^{-1/2} [\frac{1}{3}S(S+1)]^{1/2} [a(\vec{q}_1) - a(\vec{q}_2)] \\ \times \delta(\vec{q}_1 + \vec{q}_2 - \vec{q}). \end{aligned}$$

Table I gives all possible vertices from which the graphs representing the moments are composed.

The total contribution to the moments is obtained by drawing all distinct graphs using the basic vertices. The rules for calculating the moments $\langle \omega^{2n} \rangle_{\vec{q}}^\alpha$ are the following: (i) Draw all distinct graphs beginning with a dotted line (left-to-right arrow) and ending with a dotted line (left-to-right arrow). (ii) Label initial and final lines with momentum index \vec{q} and all internal lines with indices \vec{q}_i . (iii) Associate with each vertex the analytic expression taken from the table above. Take their products. (iv) Sum over all indices \vec{q}_i . (v) Add results of all graphs. Using those rules the graphs representing the second and fourth moments, shown in Fig. 1 and their analytic expression, Eqs. (2.16) are obtained:

$$\begin{aligned} \langle \omega^2 \rangle_{\vec{q}}^\alpha &= N^{-1\frac{1}{3}} S(S+1) \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')]^2, \\ \langle \Omega^4 \rangle_{\vec{q}}^\alpha &= 2[N^{-1\frac{1}{3}} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')]^2 [a(\vec{q}'') - a(\vec{q}' - \vec{q}'') + b(\vec{q}'')]^2 \\ &\quad + 2[N^{-1\frac{1}{3}} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [a(\vec{q} - \vec{q}' - \vec{q}'') - a(\vec{q}' + \vec{q}'')] [a(\vec{q}'') - a(\vec{q}') + b(\vec{q}'')] \\ &\quad \times [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')], \end{aligned} \quad (2.16a)$$

$$\langle \omega^2 \rangle_{\vec{q}}^\alpha = N^{-1\frac{1}{3}} S(S+1) \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2,$$

$$\begin{aligned} \langle \Omega^4 \rangle_{\vec{q}}^\alpha &= [N^{-1\frac{1}{3}} S(S+1)]^2 \left(\sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2 [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')]^2 \right. \\ &\quad + \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2 [a(\vec{q}' - \vec{q}'') - a(\vec{q}'')]^2 + \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')] \\ &\quad \times [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')] [a(\vec{q}'') - a(\vec{q} - \vec{q}'') + b(\vec{q}'')] [a(\vec{q}') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}')] \\ &\quad \left. + \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')] [a(\vec{q}' - \vec{q}'') - a(\vec{q} - \vec{q}')] [a(\vec{q}' - \vec{q}'') - a(\vec{q}'')] \right. \\ &\quad \left. \times [a(\vec{q} - \vec{q}'') - a(\vec{q}'') + b(\vec{q} - \vec{q}'')] \right). \end{aligned} \quad (2.16b)$$

TABLE I. Matrix elements, equivalent diagrams, and associated analytic expressions at infinite temperature, for calculation of moments to lowest order in $1/C$.

$\langle A^0(\vec{q}) \mathcal{L} A^1(\vec{q}_1) A^{-1}(\vec{q}_2) \rangle$		$= N^{-1/2} \sqrt{1/3 S(S+1)} [a(\vec{q}_1) - a(\vec{q}_2)] \times \delta(\vec{q}_1 + \vec{q}_2 - \vec{q})$
$\langle A^1(\vec{q}_1) A^1(\vec{q}_2) \mathcal{L} A^0(\vec{q}) \rangle$		
$\langle A^1(\vec{q}) \mathcal{L} A^0(\vec{q}_1) A^{-1}(\vec{q}_2) \rangle$		$= N^{-1/2} \sqrt{1/3 S(S+1)} [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \delta(\vec{q}_1 + \vec{q}_2 - \vec{q})$
$\langle A^1(\vec{q}_2) A^1(\vec{q}_1) \mathcal{L} A^1(\vec{q}) \rangle$		
$\langle A^1(\vec{q}) \mathcal{L} A^0(\vec{q}_1) A^1(\vec{q}_2) \rangle$		$= N^{-1/2} \sqrt{1/3 S(S+1)} [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \delta(\vec{q}_1 + \vec{q}_2 - \vec{q})$
$\langle A^1(\vec{q}_1) A^1(\vec{q}_2) \mathcal{L} A^1(\vec{q}) \rangle$		

Graphs of type shown in Fig. 2 also exist but have been omitted because they do not enter in the formalism since they are eliminated by the projection operators. Due to the omission of the restriction on the sums in the intermediate states, these rules give the moments correct to lowest order in $1/C$, where C is the number of spins in the range of the interaction (a_{ij} and b_{ij} are assumed to have the same range).

For the finite temperature case the set of operators A_i^{nm} are not orthogonal, that is

$$\beta^{-1} \langle A_i^{nm} | A_j^{n'm'} \rangle \neq \delta_{ij} \delta_{nm} \delta_{m'm'}$$

However, the rules outlined before to calculate the moments are still valid if instead of the operators $A^\alpha(q)$ one uses the operators $\tilde{A}^\alpha(q)$ defined

$$\tilde{A}^\alpha(q) = A^\alpha(q) \rho(q)^{-1/2},$$

where the functions $\rho(q)$ are the equilibrium two-spin correlation functions defined in Eq. (2.19). The set of operators $\tilde{A}^\alpha(q)$ can be treated as an orthonormal set.

The prescription then is to insert the identity operator I between the powers of \mathcal{L}^{2n} in expression for $\langle \omega^{2n} \rangle$, where I is

$$\begin{aligned} I = & |A^{00}\rangle \langle A^{00}| + \sum_{\vec{q}} \sum_{i=1}^1 |A^{\alpha i}(\vec{q})\rangle \langle A^{\alpha j}(\vec{q})| \rho^{-1}(\vec{q}) \\ & + \frac{1}{2} \sum_{q_1} \sum_{q_2} \sum_{i=-1}^1 \sum_{j=-1}^1 |A^{\alpha i}(\vec{q}_1) A^{\alpha j}(\vec{q}_2)\rangle \\ & \times \langle A^{\alpha i}(\vec{q}_1) A^{\alpha j}(\vec{q}_2) | \rho^{-1}(\vec{q}_1) \rho^{-1}(\vec{q}_2) + \dots \end{aligned} \quad (2.17)$$

It is also "true" that

$$\beta^{-1} \prod_{\vec{q}_i} \rho^{-1}(\vec{q}_i) \left\langle A \left[\begin{array}{c} \alpha_i \\ \vec{q}_i \end{array} \right] \middle| A \left[\begin{array}{c} \alpha_j \\ \vec{q}_j \end{array} \right] \right\rangle = \delta \left(\left[\begin{array}{c} \alpha_i \\ \vec{q}_i \end{array} \right], \left[\begin{array}{c} \alpha_j \\ \vec{q}_j \end{array} \right] \right). \quad (2.18)$$

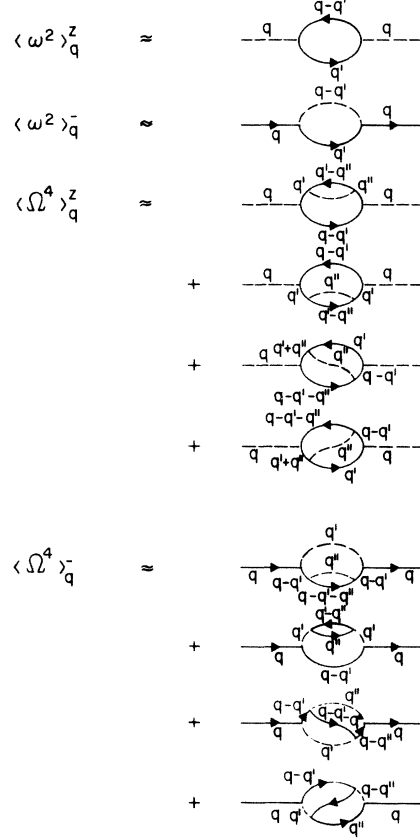


FIG. 1. Diagrammatic expansion of second and fourth moments to lowest order in $1/C$.

It should be pointed out that the relations (2.17) and (2.18) are not strictly correct even for order $1/C$. What was shown¹ is that in calculating the moments the terms coming from the nonorthogonal part of (2.18) cancel in order $1/C$. The two relations are correct only in an operative sense. This may be shown by the same method in the present case.

The equilibrium spin correlation functions are

$$\rho_n(i-j) = \langle\langle A_i^{-1} A_j^{+1} \rangle\rangle = \frac{\langle\langle S_i^- S_j^+ \rangle\rangle}{\frac{2}{3} S(S+1)} = \frac{n(i-j)}{\frac{2}{3} S(S+1)}, \quad i \neq j \quad (2.19a)$$

$$\rho_m(i-j) = \langle\langle A_i^0 A_j^0 \rangle\rangle = \frac{\langle\langle S_i^z S_j^z \rangle\rangle}{\frac{1}{3} S(S+1)} = \frac{m(i-j)}{\frac{1}{3} S(S+1)},$$

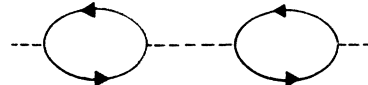


FIG. 2. Diagrams omitted in expansion of Ω^4 due to projection operator in propagator.

$$\rho_n(0) = \rho_m(0) = 1. \quad (2.19b)$$

In Sec. IV more will be said about those functions. An explicit form will be obtained in terms of the coefficients $a(q)$ and $b(q)$ and the static susceptibility.

For finite temperature the vertices can be ob-

tained in the same way as for the $T = \infty$ case, where the only difference is the presence of the function ρ . Table II gives all possible vertices from which the graphs representing the moments are composed.

Using the rules obtained before and the orthogonality condition the moments obtained are

$$\langle \omega^2 \rangle_{\vec{q}}^{\pm} = N^{-1} \frac{1}{3} S(S+1) \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \rho_n(\vec{q}') \rho_n(\vec{q} - \vec{q}') \rho_m^{-1}(\vec{q}), \quad (2.20a)$$

$$\begin{aligned} \langle \omega^4 \rangle_{\vec{q}}^{\pm} &= 2 [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}' - \vec{q}'') - a(\vec{q}' + \vec{q}'')] [a(\vec{q}'') - a(\vec{q}') + b(\vec{q}'')] \\ &\quad \times [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')] [a(\vec{q} - \vec{q}') - a(\vec{q}')] \rho_n(\vec{q} - \vec{q}' - \vec{q}'') \rho_m(\vec{q}'') \rho_n(\vec{q}') \rho_m^{-1}(\vec{q}) \\ &\quad + 2 [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')]^2 [a(\vec{q}'') - a(\vec{q}' - \vec{q}'') + b(\vec{q}'')]^2 \\ &\quad \times \rho_n(\vec{q} - \vec{q}') \rho_m(\vec{q}'') \rho_n(\vec{q}') \rho_m^{-1}(\vec{q}), \end{aligned} \quad (2.20b)$$

$$\langle \omega^2 \rangle_{\vec{q}}^{-} = N^{-1} \frac{1}{3} S(S+1) \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2 \rho_n(\vec{q} - \vec{q}') \rho_m(\vec{q}') \rho_n^{-1}(\vec{q}), \quad (2.21a)$$

$$\begin{aligned} \langle \Omega^4 \rangle_{\vec{q}}^{-} &= [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2 [a(\vec{q}' - \vec{q}'') - a(\vec{q}'')]^2 \\ &\quad \times \rho_n(\vec{q} - \vec{q}') \rho_n(\vec{q}' - \vec{q}'') \rho_n(\vec{q}'') \rho_n^{-1}(\vec{q}) + [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')]^2 \\ &\quad \times [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')]^2 \rho_m(\vec{q}') \rho_m(\vec{q}'') \rho_n(\vec{q} - \vec{q}' - \vec{q}'') \rho_n^{-1}(\vec{q}) \\ &\quad + [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}' - \vec{q}'') - a(\vec{q}'')] [a(\vec{q}' - \vec{q}'') - a(\vec{q} - \vec{q}')] \\ &\quad \times [a(\vec{q} - \vec{q}'') - a(\vec{q}'') + b(\vec{q} - \vec{q}'')] [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')] \rho_n(\vec{q}'') \rho_n(\vec{q} - \vec{q}') \rho_n(\vec{q}' - \vec{q}'') \rho_n^{-1}(\vec{q}) \\ &\quad + [N^{-1} \frac{1}{3} S(S+1)]^2 \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q}') - a(\vec{q} - \vec{q}') + b(\vec{q}')] [a(\vec{q}'') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}'')] \\ &\quad \times [a(\vec{q}') - a(\vec{q} - \vec{q}' - \vec{q}'') + b(\vec{q}')] [a(\vec{q}'') - a(\vec{q} - \vec{q}'') + b(\vec{q}'')] \rho_m(\vec{q}'') \rho_n(\vec{q} - \vec{q}' - \vec{q}'') \rho_m(\vec{q}') \rho_n(\vec{q}). \end{aligned} \quad (2.21b)$$

III. KINETIC EQUATIONS

The study developed in this section is intended to provide the complementary information to the diagrammatic method. A set of kinetic equations for the two-spin correlation function will be derived and they will give a set of nonlinear equations for the vertex functions. The derivation is based on the decoupling of the equations of motion for the spin operators. The decoupling is obtained from the cluster expansion of the density matrix and will be such that the second and fourth moments of the two-spin spectra density are correct to order $1/C$. The procedure for decoupling a correlation function is to replace the higher-order function by the sum of all possible lower-order functions and neglect the cumulant part. In lowest order in $1/C$ the cumulant part has no contribution. In the course of the derivation the vertex functions,

TABLE II. Matrix elements, equivalent diagrams, and associated analytic expressions at finite temperatures, $\rho_n(q) = \langle\langle S^+(q) \cdot S^+(-q) \rangle\rangle / \frac{2}{3} S(S+1)$; $\rho_m(q) = \langle\langle S^x(q) \times S^x(-q) \rangle\rangle / \frac{1}{3} S(S+1)$.

	} = $-N^{-1/2} \sqrt{\frac{1}{3} S(S+1)} [a(\vec{q}_1) - a(\vec{q}_2)] \rho_n^{1/2}(\vec{q}_1) \rho_n^{1/2}(\vec{q}_2) \rho_m^{1/2}(\vec{q}) \delta(\vec{q}_1, \vec{q}_2 - \vec{q})$
	} = $N^{-1/2} \sqrt{\frac{1}{3} S(S+1)} [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \rho_m^{1/2}(\vec{q}_1) \rho_m^{1/2}(\vec{q}_2) \rho_n^{1/2}(\vec{q}) \delta(\vec{q}_1, \vec{q}_2 - \vec{q})$

to be called $\Gamma^\alpha(q_1, q_2; z)$, are renormalized. The renormalization procedure is not exact in any mathematical or physical sense. This renormalization although intuitive is nevertheless suggested by the diagrammatic method. The information

about the physical systems obtained from the kinetic equations will be the *a posteriori* justification for the renormalization.

The equation of motion for $\langle S^-(q_1)S^+(q_2); t \rangle$ can be obtained from (1.10) and it is

$$i \frac{\partial}{\partial t} \langle S^-(\vec{q}_1)S^+(\vec{q}_2); t \rangle = N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_1 - \vec{q}_3) - a(\vec{q}_3) - b(\vec{q}_3)] \langle S^+(\vec{q}_3)S^-(\vec{q}_1 - \vec{q}_3)S^+(\vec{q}_2); t \rangle \\ - N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_2 - \vec{q}_3) - a(\vec{q}_3) - b(\vec{q}_3)] \langle S^-(\vec{q}_1)S^+(\vec{q}_3)S^+(\vec{q}_2 - \vec{q}_3); t \rangle. \quad (3.1)$$

The brackets $\langle \rangle$ denote an average over an arbitrary nonequilibrium density matrix. The averages are time dependent although the time t is omitted; if necessary the notation $\langle AB; t \rangle$ will be used and $\langle AB; z \rangle$ for its Laplace transform.

The equation can be closed by means of the approximation

$$\langle S_i^- S_j^+ S_k^+ \rangle = \langle S_i^- \rangle \langle S_j^+ S_k^+ \rangle + \langle S_j^+ \rangle \langle S_i^- S_k^+ \rangle \\ + \langle S_k^+ \rangle \langle S_i^- S_j^+ \rangle + 2 \langle S_i^- \rangle \langle S_j^+ \rangle \langle S_k^+ \rangle, \\ i \neq j \neq k. \quad (3.2)$$

Using the equations for $\langle S^+ \rangle$ and $\langle S^- \rangle$ and a similar decoupling, together with (3.1) a closed set of coupled nonlinear equations can be obtained. Those equations have a stationary solution described by arbitrary parameters that can be chosen to have

values appropriate to thermal equilibrium. The linearization of the decoupled equation (3.1) yields

$$\langle S^+(\vec{q}, 0) \rangle = 0, \\ \langle S(\vec{q}, 0) \rangle = \langle\langle S(\vec{q}) \rangle\rangle + \delta \langle S(\vec{q}) \rangle, \\ \langle S^-(\vec{q}_1)S^+(\vec{q}_2) \rangle = \langle\langle S^-(\vec{q}_1)S^+(\vec{q}_2) \rangle\rangle \\ + \delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2) \rangle.$$

In other words the function consists of the equilibrium part plus its deviation from equilibrium (δ indicates this deviation). Under those conditions only the second term of (3.2) contributes. A set of linear equations (order of δ) describing the behavior in time of the small disturbance from equilibrium is obtained for the paramagnetic regime:

$$i \frac{\partial}{\partial t} \delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2) \rangle = N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_1 - \vec{q}_3) - a(\vec{q}_3) - b(\vec{q}_3)] \langle\langle S^-(\vec{q}_1 - \vec{q}_3)S^+(\vec{q}_2) \rangle\rangle \delta \langle S^+(\vec{q}_3) \rangle \\ - N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_2 - \vec{q}_3) - a(\vec{q}_3) - b(\vec{q}_3)] \langle\langle S^-(\vec{q}_2 - \vec{q}_3)S^+(\vec{q}_1) \rangle\rangle \delta \langle S^+(\vec{q}_3) \rangle;$$

but

$$\langle\langle S^-(\vec{q})S^+(\vec{q}') \rangle\rangle = \delta_{\vec{q}-\vec{q}'} \langle\langle S^-(\vec{q})S^+(-\vec{q}) \rangle\rangle = \delta_{\vec{q}-\vec{q}'} n(\vec{q}).$$

It follows that

$$i \frac{\partial}{\partial t} \delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2) \rangle \\ = N^{-1/2} \gamma^\alpha(\vec{q}_1, \vec{q}_2) \delta \langle S^+(\vec{q}_1, \vec{q}_2) \rangle, \quad (3.3)$$

where

$$\gamma^\alpha(\vec{q}_1, \vec{q}_2) = [a(\vec{q}_1 + \vec{q}_2) + b(\vec{q}_1 + \vec{q}_2)] [n(\vec{q}_1) - n(\vec{q}_2)] \\ - a(\vec{q}_1)n(\vec{q}_1) + a(\vec{q}_2)n(\vec{q}_2). \quad (3.4)$$

An approximation for $\Sigma^\alpha(\vec{q}, t)$ can be obtained from (3.3) and from the equation

$$i \frac{\partial}{\partial t} \delta \langle S^+(\vec{q}_1 t) \rangle = \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \times \delta \langle S^-(\vec{q}')S^+(\vec{q} - \vec{q}'); t \rangle, \quad (3.5)$$

which was obtained from (1.9).

By definition $\delta \langle S^+(\vec{q}, t) \rangle = h^\alpha(\vec{q}) \Sigma^\alpha(\vec{q}, t)$ when $h^\alpha(\vec{q})$ is a field applied up until $t=0$, and then removed. Since the equations of motion are linear, $h^\alpha(q)$ can be taken equal to one.

The Laplace transforms of (3.5) and (3.3) are

$$= \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \\ \times \delta \langle S^-(\vec{q}')S^+(\vec{q} - \vec{q}'); z \rangle, \quad (3.6a)$$

$$i \langle S^-(\vec{q}')S^+(\vec{q} - \vec{q}') | S^+(\vec{q}) \rangle + z \delta \langle S^-(\vec{q}')S^+(\vec{q} - \vec{q}') \rangle; z \\ = N^{-1/2} \gamma^\alpha(\vec{q}', \vec{q} - \vec{q}') \Sigma^\alpha(\vec{q}, z), \quad (3.6b)$$

where the initial values are obtained from the initial density matrix

$$\rho_{\text{initial}} = \rho_{\text{eq}} - \int_0^\beta e^{\tau H} S^z(-\vec{q}) e^{-\tau H} d\tau \rho_{\text{eq}} .$$

Solving (3.6) for $\Sigma^z(\vec{q}, z)$ we obtain

$$\Sigma^z(\vec{q}, z) = i\chi^z(\vec{q}, 0)[z - \omega^2(\vec{q})/z]^{-1} , \quad (3.7a)$$

with

$$\begin{aligned} \frac{\omega^2(\vec{q})}{z} &\equiv \phi^z(\vec{q}, z) \\ &= \frac{1}{z} \frac{1}{2} N^{-1} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \\ &\quad \times \gamma^z(\vec{q}', \vec{q} - \vec{q}') . \end{aligned} \quad (3.7b)$$

We have used in the preceding step the fact that $\langle \mathcal{L}A|B \rangle = \langle \langle [B, A^\dagger] \rangle \rangle$, that is, $\langle \mathcal{L}S^z(\vec{q}) | S^z(\vec{q}) \rangle = 0$.

The results predicted by Eq. (3.3), i. e., (3.7a) and (3.7b), show that this equation is inadequate. The function $\Phi^z(\vec{q}, z)$ is not analytic at $z=0$ as it should be, and the equation gives also a wrong result for $\vec{q}=0$. In this case it follows that

$$i \frac{\partial}{\partial t} \delta \langle S^-(\vec{q}_1) S^+(\vec{q}_2) \rangle = 0 .$$

This implies that the disturbance of the two-spin correlation functions would persist in time, when it is expected that they should decay until they have reached their asymptotic value. The decay processes can be obtained by adding to Eq. (3.3) a term of the type $-\nu \langle S^-(\vec{q}') S^+(\vec{q} - \vec{q}') \rangle$. The response function obtained from this new equation would be

$$\Sigma^z(\vec{q}, z) = i\chi^z(\vec{q}, 0)[z - \omega^2(\vec{q})/(z + i\nu)]^{-1} . \quad (3.8)$$

Equation (3.8) has the proper behavior for $z=0$.

This phenomenological approach, although giving at this stage a correct qualitative description, will not be pursued. Instead a more systematic derivation will be given.

Note that if result (3.7) is compared with (1.12) and (1.13) one sees that the approximation leading to (3.7) is

$$\begin{aligned} \Gamma^z(\vec{q}_1, \vec{q}_2; z) &\cong N^{-1/2} [\gamma^z(\vec{q}_1, \vec{q}_2)/z] \\ &\equiv \Gamma_0^z(\vec{q}_1, \vec{q}_2; z) . \end{aligned} \quad (3.9)$$

It has been shown above that this approximation for the function $\Gamma^z(\vec{q}_1, \vec{q}_2; z)$ neglects the decay of the two-spin correlation. So, the decay processes are contained in the exact $\Gamma^z(\vec{q}_1, \vec{q}_2; t)$ which gives the rate at which the function $\langle S^-(\vec{q}_1) S^+(\vec{q}_2); t \rangle$

changes due to the unit disturbance $h(q)$ introduced in the system until $t=0$.

A similar study for the function $\langle S^z(\vec{q}_1) S^-(\vec{q}_2); t \rangle$ gives

$$\begin{aligned} i \frac{\partial}{\partial t} \langle S^z(\vec{q}_1) S^-(\vec{q}_2) \rangle &= \frac{1}{2} N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_1 - \vec{q}_3) - a(\vec{q}_3)] \\ &\quad \times \langle S^-(\vec{q}_3) S^+(\vec{q}_1 - \vec{q}_3) S^-(\vec{q}_2) \rangle \\ &\quad - N^{-1/2} \sum_{\vec{q}_3} [a(\vec{q}_3) - a(\vec{q}_2 - \vec{q}_3) + b(\vec{q}_3)] \\ &\quad \times \langle S^z(\vec{q}_1) S^z(\vec{q}_3) S^-(\vec{q}_2 - \vec{q}_3) \rangle , \end{aligned} \quad (3.10)$$

$$i \frac{\partial}{\partial t} \delta \langle S^z(\vec{q}_1) S^-(\vec{q}_2) \rangle$$

$$= N^{-1/2} \gamma^-(\vec{q}_1, \vec{q}_2) \delta \langle S^-(\vec{q}_1 + \vec{q}_2) \rangle , \quad (3.11)$$

where

$$\begin{aligned} \gamma^-(\vec{q}_1, \vec{q}_2) &= a(\vec{q}_1 + \vec{q}_2) [m(\vec{q}_1) - \frac{1}{2} n(\vec{q}_2)] \\ &\quad - [a(\vec{q}_1) + b(\vec{q}_1)] m(\vec{q}_1) + \frac{1}{2} a(\vec{q}_2) n(\vec{q}_2) \end{aligned} \quad (3.12)$$

and

$$m(\vec{q}) = \langle \langle S^z(\vec{q}) S^z(-\vec{q}) \rangle \rangle .$$

The approximation leading to (3.11) is

$$\Gamma^-(\vec{q}_1, \vec{q}_2; z) \cong N^{-1/2} \frac{\gamma^-(\vec{q}_1, \vec{q}_2)}{z} \equiv \Gamma_0^-(\vec{q}_1, \vec{q}_2; z) . \quad (3.13)$$

A kinetic theory must be able to describe internal relaxation processes and at this level of approximation this has not been accomplished. To obtain a description of these processes, we will consider the evolution of the correlation function $\langle S^\alpha(q_1) S^\alpha(\vec{q}_2); t \rangle$, for the case that there is no change in the external variables of the system, i. e., $\langle S^\alpha(\vec{q}, t) \rangle = 0$. We will see that this suffices to calculate $\Phi(\vec{q}, z)$, so that we will obtain at the same time an expression for $\Sigma(\vec{q}, z)$ that includes the effect of the internal relaxation on the dynamics of the external fluctuation. The time dependence of the two-spin correlation functions now depends on the three-spin correlation function and this implies the need to study these functions. The three-spin functions entering in the equations for the two-spin functions are $\langle S^-(\vec{q}_1) S^z(\vec{q}_2) S^+(\vec{q}_3); t \rangle$, $\langle S^-(\vec{q}_1) S^+(\vec{q}_2) S^-(\vec{q}_3); t \rangle$, and $\langle S^z(\vec{q}_1) S^z(\vec{q}_2) S^-(\vec{q}_3); t \rangle$.

The equation of motion for the first of those functions is

$$\begin{aligned} i \frac{\partial}{\partial t} \langle S^-(\vec{q}_1) S^z(\vec{q}_2) S^+(\vec{q}_3) \rangle &= -N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_1 - \vec{q}_4) + b(\vec{q}_4)] \langle S^z(\vec{q}_4) S^-(\vec{q}_1 - \vec{q}_4) S^z(\vec{q}_2) S^+(\vec{q}_3) \rangle \\ &\quad + \frac{1}{2} N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_2 - \vec{q}_4) - a(\vec{q}_4)] \langle S^-(\vec{q}_1) S^-(\vec{q}_4) S^+(\vec{q}_2 - \vec{q}_4) S^+(\vec{q}_3) \rangle \end{aligned}$$

$$+ N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_3 - \vec{q}_4) + b(\vec{q}_4)] \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^s(\vec{q}_4) S^+(\vec{q}_3 - \vec{q}_4) \rangle. \quad (3.14)$$

Making the cluster expansion for the four-spin functions we get

$$\langle S^s(\vec{q}_1) S^-(\vec{q}_2) S^s(\vec{q}_3) S^+(\vec{q}_4) \rangle = \langle S^s(\vec{q}_1) S^s(\vec{q}_3) \rangle \langle S^-(\vec{q}_2) S^+(\vec{q}_4) \rangle, \quad (3.15a)$$

$$\langle S^-(\vec{q}_1) S^-(\vec{q}_2) S^+(\vec{q}_3) S^+(\vec{q}_4) \rangle = \langle S^-(\vec{q}_1) S^+(\vec{q}_3) \rangle \langle S^-(\vec{q}_2) S^+(\vec{q}_4) \rangle + \langle S^-(\vec{q}_1) S^+(\vec{q}_4) \rangle \langle S^-(\vec{q}_2) S^+(\vec{q}_3) \rangle. \quad (3.15b)$$

The initial density matrix will be taken rotationally invariant about the z axis, and therefore $\langle S_i^+ S_j^+ \rangle = \langle S_i^- S_j^- \rangle = 0$. Insert (3.15) in (3.14) to get

$$\begin{aligned} i \frac{\partial}{\partial t} \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3) \rangle &= -N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_1 - \vec{q}_4) + b(\vec{q}_4)] \langle S^s(\vec{q}_4) S^s(\vec{q}_2) \rangle \langle S^-(\vec{q}_1 - \vec{q}_4) S^+(\vec{q}_3) \rangle \\ &+ \frac{1}{2} N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_2 - \vec{q}_4) - a(\vec{q}_4)] \langle S^-(\vec{q}_1) S^+(\vec{q}_2 - \vec{q}_4) \rangle \langle S^-(\vec{q}_4) S^+(\vec{q}_3) \rangle \\ &+ N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_3 - \vec{q}_4) + b(\vec{q}_4)] \langle S^-(\vec{q}_1) S^+(\vec{q}_3 - \vec{q}_4) \rangle \langle S^s(\vec{q}_2) S^s(\vec{q}_3) \rangle. \end{aligned} \quad (3.16)$$

In obtaining (3.16) we used the fact $\langle S^s(\vec{q}) \rangle = 0$. Equation (3.16) now has to be linearized about equilibrium (restrict to linear response). Therefore, the equilibrium values for $\langle S^-(\vec{q}_1) S^+(\vec{q}_2) \rangle$ and $\langle S^s(\vec{q}_1) S^s(\vec{q}_2) \rangle$ must be such that the time derivative of the three-spin function vanishes. The condition on the equilibrium values $n(\vec{q})$ and $m(\vec{q})$, when $(\partial/\partial t) \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3) \rangle = 0$, is

$$[a(\vec{q}_3) - a(\vec{q}_2)] n(\vec{q}_3) m(\vec{q}_2) + [a(\vec{q}_1) - a(\vec{q}_3)] n(\vec{q}_1) \frac{1}{2} [n(\vec{q}_3)] + [a(\vec{q}_2) - a(\vec{q}_1)] n(\vec{q}_1) m(\vec{q}_2) + b(\vec{q}_2) m(\vec{q}_2) [n(\vec{q}_1) - n(\vec{q}_3)] = 0. \quad (3.17)$$

Equation (3.16) is now linearized about correlation functions which satisfy (3.17), with the result

$$\begin{aligned} i \frac{\partial}{\partial t} \delta \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3) \rangle &= -N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_1 - \vec{q}_4) + b(\vec{q}_4)] \\ &\times [\langle S^s(\vec{q}_4) S^s(\vec{q}_2) \rangle \delta \langle S^-(\vec{q}_1 - \vec{q}_4) S^+(\vec{q}_3) \rangle + \langle S^-(\vec{q}_1 - \vec{q}_4) S^+(\vec{q}_3) \rangle \delta \langle S^s(\vec{q}_4) S^s(\vec{q}_2) \rangle] \\ &+ \frac{1}{2} N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_2 - \vec{q}_4) - a(\vec{q}_4)] [\langle S^-(\vec{q}_1) S^+(\vec{q}_2 - \vec{q}_4) \rangle \delta \langle S^-(\vec{q}_4) S^+(\vec{q}_3) \rangle \\ &+ \langle S^-(\vec{q}_4) S^+(\vec{q}_3) \rangle \delta \langle S^-(\vec{q}_1) S^+(\vec{q}_2 - \vec{q}_4) \rangle] + N^{-1/2} \sum_{\vec{q}_4} [a(\vec{q}_4) - a(\vec{q}_3 - \vec{q}_4) + b(\vec{q}_4)] \\ &\times [\langle S^-(\vec{q}_1) S^+(\vec{q}_3 - \vec{q}_4) \rangle \delta \langle S^s(\vec{q}_2) S^s(\vec{q}_4) \rangle + \langle S^s(\vec{q}_2) S^s(\vec{q}_4) \rangle \delta \langle S^-(\vec{q}_1) S^+(\vec{q}_3 - \vec{q}_4) \rangle]. \end{aligned} \quad (3.18)$$

Using the fact that $\langle S^s(\vec{q}) S^s(\vec{q}') \rangle = \delta_{\vec{q}, -\vec{q}'} m(\vec{q})$, $\langle S^-(\vec{q}) S^+(\vec{q}') \rangle = \delta_{\vec{q}, -\vec{q}'} n(\vec{q})$, and defining

$$n^0(\vec{q}_1, \vec{q}_2) \equiv \delta \langle S^-(\vec{q}_1) S^+(\vec{q}_2) \rangle,$$

$$m(\vec{q}_1, \vec{q}_2) \equiv \delta \langle S^s(\vec{q}_1) S^s(\vec{q}_2) \rangle,$$

the result (3.18) is obtained after substitution and rearrangement of the terms:

$$\begin{aligned} i \frac{\partial}{\partial t} \delta \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3) \rangle &= N^{-1/2} \gamma^-(\vec{q}_2, \vec{q}_1) n^0(\vec{q}_1 + \vec{q}_2, \vec{q}_3; t) \\ &- N^{-1/2} \gamma^-(\vec{q}_2, \vec{q}_3) n^0(\vec{q}_1, \vec{q}_2 + \vec{q}_3; t) \\ &+ N^{-1/2} \gamma^s(\vec{q}_1, \vec{q}_3) m(\vec{q}_1 + \vec{q}_3, \vec{q}_2; t), \end{aligned} \quad (3.19)$$

where $\gamma^s(\vec{q}_1, \vec{q}_2)$ and $\gamma^-(\vec{q}_1, \vec{q}_2)$ are given by (3.4) and (3.12).

This equation shows that the rate at which three-spin fluctuations are produced is equal to the sums of rates at which one of the fluctuations in the excited pair decays into two. This rate for fluctuations of wave vector $\vec{q}_1 + \vec{q}_2$ is just γ^- and γ^s in this approximation. The Laplace transform of (3.19) with the initial value $\delta \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3); t=0 \rangle = 0$ is

$$\begin{aligned} \delta \langle S^-(\vec{q}_1) S^s(\vec{q}_2) S^+(\vec{q}_3); z \rangle &= \Gamma_0^-(\vec{q}_2, \vec{q}_1; z) n^0(\vec{q}_1 + \vec{q}_2, \vec{q}_3; z) \\ &- \Gamma_0^-(\vec{q}_2, \vec{q}_3; z) n^0(\vec{q}_1, \vec{q}_2 + \vec{q}_3; z) \\ &+ \Gamma_0^s(\vec{q}_1, \vec{q}_3; z) m(\vec{q}_1 + \vec{q}_3, \vec{q}_2; z), \end{aligned} \quad (3.20)$$

where (3.9) and (3.13) were used.

Taking the initial value equal to zero implies neglecting the possibility of a fluctuation decaying

directly into three fluctuations. The initial value enters in the equation of motion as $\langle S^-(\vec{q}_1)S^+(\vec{q}_2)S^+(\vec{q}_3) | \mathcal{L} | S^+(\vec{q}) \rangle$ and this is a matrix element of order $1/C$ and hence can be neglected.

The presence of $\Gamma_0^\alpha(\vec{q}_1, \vec{q}_2; z)$ in Eq. (3.20) indicates that the equation will not describe the relaxation process.

One way of including those processes is to substitute in (3.20) the exact function $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ for $\Gamma_0^\alpha(\vec{q}_1, \vec{q}_2; z)$. This modification will include the internal relaxation processes in the response of the three-spin function to a change in the two-spin function:

$$\begin{aligned} \delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2)S^+(\vec{q}_3); z \rangle \\ = \Gamma^-(\vec{q}_2, \vec{q}_1; z)n^0(\vec{q}_1 + \vec{q}_2, \vec{q}_3; z) \\ - \Gamma^-(\vec{q}_2, \vec{q}_3; z)n^0(\vec{q}_1, \vec{q}_2 + \vec{q}_3; z) \end{aligned}$$

$$+ \Gamma^+(\vec{q}_1, \vec{q}_3; z)m(\vec{q}_1 + \vec{q}_3, \vec{q}_2; z). \quad (3.21a)$$

Similar study for the other three-spin functions gives

$$\begin{aligned} \delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2)S^-(\vec{q}_3); z \rangle \\ = \Gamma^+(\vec{q}_1, \vec{q}_2; z)n^-(\vec{q}_1 + \vec{q}_2, \vec{q}_3; z) \\ + \Gamma^+(\vec{q}_3, \vec{q}_2; z)n^-(\vec{q}_2 + \vec{q}_3, \vec{q}_1; z), \quad (3.21b) \end{aligned}$$

$$\begin{aligned} \delta \langle S^+(\vec{q}_1)S^+(\vec{q}_2)S^-(\vec{q}_3); z \rangle \\ = \Gamma^-(\vec{q}_2, \vec{q}_3; z)n^-(\vec{q}_1, \vec{q}_2 + \vec{q}_3; z) \\ + \Gamma^-(\vec{q}_1, \vec{q}_3; z)n^-(\vec{q}_2, \vec{q}_1 + \vec{q}_3; z), \quad (3.21c) \end{aligned}$$

where $n^-(q_1, q_2; z) = \delta \langle S^+(\vec{q}_1)S^-(\vec{q}_2); z \rangle$.

The Laplace transform of the equations of motion for the two-spin correlation functions $\delta \langle S^-(\vec{q}_1)S^+(\vec{q}_2); t \rangle$, $\delta \langle S^+(\vec{q}_1)S^-(\vec{q}_2); t \rangle$, and $\delta \langle S^+(\vec{q}_1)S^+(\vec{q}_2); t \rangle$ are

$$\begin{aligned} zn^0(\vec{q}_1, \vec{q}_2; z) = in^0(\vec{q}_1, \vec{q}_2; t=0) + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \delta \langle S^+(\vec{q}')S^-(\vec{q}_1 - \vec{q}')S^+(\vec{q}_2); z \rangle \\ - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \delta \langle S^-(\vec{q}_1)S^+(\vec{q}')S^+(\vec{q}_2 - \vec{q}'); z \rangle, \quad (3.22a) \end{aligned}$$

$$\begin{aligned} zm(\vec{q}_1, \vec{q}_2; z) = im(\vec{q}_1, \vec{q}_2; t=0) + \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}')] \delta \langle S^-(\vec{q}')S^+(\vec{q}_1 - \vec{q}')S^+(\vec{q}_2); z \rangle \\ + \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}')] \delta \langle S^+(\vec{q}_1)S^-(\vec{q}')S^+(\vec{q}_2 - \vec{q}'); z \rangle, \quad (3.22b) \end{aligned}$$

$$\begin{aligned} zn^-(\vec{q}_1, \vec{q}_2; z) = in^-(\vec{q}_1, \vec{q}_2; t=0) + \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}')] \delta \langle S^-(\vec{q}')S^+(\vec{q}_1 - \vec{q}')S^-(\vec{q}_2); z \rangle \\ - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q}_2 - \vec{q}') + b(\vec{q}')] \delta \langle S^+(\vec{q}_1)S^+(\vec{q}')S^-(\vec{q}_2 - \vec{q}'); z \rangle. \quad (3.22c) \end{aligned}$$

Substituting (3.21) into (3.22) and making use of results (1.16) and (1.17), the kinetic equations are obtained:

$$\begin{aligned} zn^0(\vec{q}_1, \vec{q}_2; z) = in^0(\vec{q}_1, \vec{q}_2; t=0) + [\phi^-(\vec{q}_1, z) + \phi^-(\vec{q}_2, z)]n^0(\vec{q}_1, \vec{q}_2; z) \\ - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q}_1 - \vec{q}') - b(\vec{q}_1 - \vec{q}')] \Gamma^-(\vec{q}_1 - \vec{q}', \vec{q}_2; z)n^0(\vec{q}', \vec{q} - \vec{q}'; z) \\ - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q}_2 - \vec{q}') - b(\vec{q}_2 - \vec{q}')] \Gamma^-(\vec{q}_2 - \vec{q}', \vec{q}_1; z)m^0(\vec{q} - \vec{q}', \vec{q}'; z) \\ + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^+(\vec{q}_1 - \vec{q}', \vec{q}_2; z)m(\vec{q} - \vec{q}', \vec{q}'; z) \\ + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^+(\vec{q}_2 - \vec{q}', \vec{q}_1; z)m(\vec{q} - \vec{q}', \vec{q}'; z), \quad (3.23a) \end{aligned}$$

$$\begin{aligned} zm(\vec{q}_1, \vec{q}_2; z) = im(\vec{q}_1, \vec{q}_2; t=0) + [\phi^+(\vec{q}_1, z) + \phi^+(\vec{q}_2, z)]m(\vec{q}_1, \vec{q}_2; z) \\ - \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}_2, \vec{q}_1 - \vec{q}'; z) [n^0(\vec{q} - \vec{q}', \vec{q}'; z) + n^0(\vec{q}', \vec{q} - \vec{q}'; z)] \\ - \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}_1, \vec{q}_2 - \vec{q}'; z) \{n^0(\vec{q} - \vec{q}', \vec{q}'; z) + n^0(\vec{q}', \vec{q} - \vec{q}'; z)\}, \quad (3.23b) \end{aligned}$$

$$\begin{aligned} zn^-(\vec{q}_1, \vec{q}_2; z) = in^-(\vec{q}_1, \vec{q}_2; t=0) + [\phi^-(\vec{q}_1, z) + \phi^-(\vec{q}_2, z)]n^-(\vec{q}_1, \vec{q}_2; z) \\ + \frac{1}{2}N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}')] \Gamma^+(\vec{q}_2, \vec{q}_1 - \vec{q}'; z)n^-(\vec{q} - \vec{q}', \vec{q}'; z) \end{aligned}$$

$$-N^{-1/2} \sum_{\vec{q}} [a(\vec{q}') - a(\vec{q}_2 - \vec{q}') + b(\vec{q}')] \Gamma^-(\vec{q}_1, \vec{q}_2 - \vec{q}'; z) n^-(\vec{q}', \vec{q} - \vec{q}'; z). \quad (3.23c)$$

Equations (3.23) form a set of linear equations for the two-spin correlation functions with initial values still arbitrary. Because the functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ are still unknown the set is not closed. It will be shown next that for a particular initial condition $\Gamma^+(\vec{q}_1, \vec{q}_2; t)$ is simply $\langle S^-(\vec{q}_1) S^+(\vec{q}_2); t \rangle$ and $\Gamma^-(\vec{q}_1, \vec{q}_2; t)$ is $\langle S^+(\vec{q}_1) S^-(\vec{q}_2); t \rangle$. From the definition of $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ given by (1.14) follows

$$\begin{aligned} \Gamma^+(\vec{q}_1, \vec{q}_2; t) &= -i \frac{1}{2} N^{-1/2} \sum_{\vec{q}} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \\ &\quad \times \langle S^-(\vec{q}_1) S^+(\vec{q}_2) | e^{i\mathcal{L}'t} | S^-(\vec{q}') \\ &\quad \times S^+(\vec{q} - \vec{q}') \rangle / \chi^+(\vec{q}, 0), \end{aligned}$$

and for $t=0$

$$\Gamma^+(\vec{q}_1, \vec{q}_2; t=0) = -i \langle S^-(\vec{q}_1) S^+(\vec{q}_2) | \mathcal{L} S^+(\vec{q}) \rangle / \chi^+(\vec{q}, 0).$$

Because the set (3.23) is still valid for any initial value we choose the initial condition to be such that

$$i \langle S^-(\vec{q}_1) S^+(\vec{q}_2); t=0 \rangle = i \Gamma^+(\vec{q}_1, \vec{q}_2; t=0)$$

$$\begin{aligned} &= \langle S^-(\vec{q}_1) S^+(\vec{q}_2) | \mathcal{L} S^+(\vec{q}) \rangle / \chi^+(\vec{q}, 0) \\ &= \langle \langle [S^+(\vec{q}_1 + \vec{q}_2), S^-(\vec{q}_1) S^+(\vec{q}_2)] \rangle \rangle / \\ &\quad \chi^+(\vec{q}, 0) \\ &= [N^{-1/2} / \chi^+(\vec{q}, 0)] [n(\vec{q}_2) - n(\vec{q}_1)], \end{aligned} \quad (3.24a)$$

$$\begin{aligned} i \langle S^+(\vec{q}_1) S^-(\vec{q}_2); t=0 \rangle \\ = [N^{-1/2} / \chi^-(\vec{q}, 0)] [n(\vec{q}_2) - 2m(\vec{q}_1)]. \end{aligned} \quad (3.24b)$$

Because the initial value

$$m(\vec{q}_1, \vec{q}_2; t=0) = \langle S^+(\vec{q}_1) S^+(\vec{q}_2) | \mathcal{L} S^+(\vec{q}) \rangle = 0$$

and using the fact that $\Gamma^+(\vec{q}_1, \vec{q}_2; z) = -\Gamma^+(\vec{q}_2, \vec{q}_1; z)$ it follows from (3.23) that

$$m(\vec{q}_1, \vec{q}_2; z) = 0$$

and

$$\begin{aligned} z \Gamma^+(\vec{q}_1, \vec{q}_2; z) &= N^{-1/2} \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^+(\vec{q}_1 + \vec{q}_2, 0)} + [\phi^-(\vec{q}_1, z) + \phi^-(\vec{q}_2, z)] \Gamma^+(\vec{q}_1, \vec{q}_2; z) \\ &\quad + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}') + b(\vec{q}_1 - \vec{q}')] \Gamma^-(\vec{q}_1 - \vec{q}', \vec{q}_2; z) \Gamma^+(\vec{q}', \vec{q} - \vec{q}'; z) \\ &\quad + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}') + b(\vec{q}_2 - \vec{q}')] \Gamma^-(\vec{q}_2 - \vec{q}', \vec{q}_1; z) \Gamma^+(\vec{q} - \vec{q}', \vec{q}'; z), \end{aligned} \quad (3.25a)$$

$$\begin{aligned} z \Gamma^-(\vec{q}_1, \vec{q}_2; z) &= N^{-1/2} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^-(\vec{q}_1 + \vec{q}_2, 0)} + [\phi^+(\vec{q}_1, z) + \phi^-(\vec{q}_2, z)] \Gamma^-(\vec{q}_1, \vec{q}_2; z) \\ &\quad + \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}')] \Gamma^+(\vec{q}_2, \vec{q}_1 - \vec{q}'; z) \Gamma^-(\vec{q} - \vec{q}', \vec{q}'; z) \\ &\quad - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q}_2 - \vec{q}') + b(\vec{q}')] \Gamma^-(\vec{q}_1, \vec{q}_2 - \vec{q}'; z) \Gamma^-(\vec{q}', \vec{q} - \vec{q}'; z). \end{aligned} \quad (3.25b)$$

It should be noted that the initial perturbation for which $\delta \langle S^-(\vec{q}_1) S^+(\vec{q}_2); t=0 \rangle = \Gamma^+(\vec{q}_1, \vec{q}_2; t=0)$ has the property $\delta \langle S^+(\vec{q}); t=0 \rangle$, and that furthermore since Γ^+ evolves in accordance with \mathcal{L}' , not \mathcal{L} , this property will be preserved in time. Hence, the kinetic equations derived assuming $\delta \langle S^+(q); t \rangle = 0$ are the appropriate equations to use. A similar statement holds for $\Gamma^-(\vec{q}_1, \vec{q}_2; t)$. This is a coupled system of nonlinear equations satisfied by the $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ functions. Those functions contain most of the dynamical properties of systems described by Hamiltonian (1.1). The equations are valid for any spin and any temperature above T_c , and give the correct second and fourth moments.

IV. APPROXIMATE HIGH-TEMPERATURE SOLUTION

If the solution to the kinetic equations are known the time-dependent spin correlation function

$$\langle S^{\alpha_1}(\vec{q}_1, t) S^{\alpha_2}(\vec{q}_2, t) | S^{\alpha_3}(\vec{q}_3, 0) S^{\alpha_4}(\vec{q}_4, 0) \rangle$$

can be calculated and with the aid of this function a wide range of experiments can be discussed. The proposed solution will be based on a physically motivated approximation for the functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ at high temperature. By an iterative procedure the equation can be solved.

In the preceding sections the equilibrium correlation functions $n(\vec{q})$ and $m(\vec{q})$ have appeared con-

stantly throughout the discussion. In the following an explicit formula will be obtained for those functions and together with the initial values for the vertex functions the approximate solution is presented.

From the definition of $\Gamma^s(\vec{q}_1, \vec{q}_2; z)$ the following result is obtained for large values of z :

$$\begin{aligned} \Gamma^s(q_1, q_2; z) &= \frac{\langle S^-(q_1)S^+(q_2) | \mathcal{L} S^s(q) \rangle}{z\chi^s(q, 0)} + O(1/z^3) \\ &= \frac{1}{z} N^{-1/2} \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^s(\vec{q}_1 + \vec{q}_2, 0)}, \end{aligned}$$

where we have the identity $\langle A | \mathcal{L} | B \rangle = \langle [B, A^+] \rangle$. It was shown that the first approximation for $\Gamma^s(\vec{q}_1, \vec{q}_2; z)$ is

$$\Gamma^s(\vec{q}_1, \vec{q}_2; z) \cong N^{-1/2} [\gamma^s(q_1, q_2)/z].$$

The last two relations combine to give

$$\gamma^s(\vec{q}_1, \vec{q}_2) = \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^s(\vec{q}_1 + \vec{q}_2, 0)}, \quad (4.1)$$

where, according to (3.4), $\gamma^s(q_1, q_2)$ is

$$\begin{aligned} \gamma^s(\vec{q}_1, \vec{q}_2) &= a[(\vec{q}_1 + \vec{q}_2) + b(\vec{q}_1 + \vec{q}_2)] \\ &\quad \times (n(\vec{q}_1) - n(\vec{q}_2)) - a(\vec{q}_1)n(\vec{q}_2)n(\vec{q}_2). \end{aligned}$$

Taking $\vec{q}_2 = \vec{q} - \vec{q}'$ and $\vec{q}_1 = \vec{q}'$ in (4.1),

$$\begin{aligned} n(\vec{q} - \vec{q}') - n(\vec{q}') &= \{ [a(\vec{q}) + b(\vec{q})] (n(\vec{q}') - n(\vec{q} - \vec{q}')) \\ &\quad - a(\vec{q}')n(\vec{q}') + a(\vec{q} - \vec{q}')n(\vec{q} - \vec{q}') \} \\ &\quad \times \chi^s(\vec{q}, 0), \end{aligned}$$

$$\begin{aligned} n(\vec{q} - \vec{q}') \{ 1 + \chi^s(\vec{q}, 0) [a(\vec{q}) + b(\vec{q}) - a(\vec{q} - \vec{q}')] \} \\ - n(\vec{q}') \{ 1 + \chi^s(\vec{q}, 0) [a(\vec{q}) + b(\vec{q}) - a(\vec{q})] \} = 0. \end{aligned}$$

Taking $\vec{q} \rightarrow 0$,

$$\vec{q} \cdot \nabla_{\vec{q}'} \{ 1 + \chi^s(0, 0) [a(0) + b(0) - a(\vec{q}')] \} n(\vec{q}') = 0.$$

Therefore

$$n(\vec{q}) = K \{ 1 + \chi^s [a(0) + b(0) - a(q)] \}^{-1}, \quad (4.2)$$

where $\chi^s \equiv \chi^s(0, 0)$.

The expansion in z gives for $\Gamma^-(\vec{q}_1, \vec{q}_2; z)$ the value

$$\begin{aligned} \Gamma^-(\vec{q}_1, \vec{q}_2; z) &= \frac{\langle S^s(\vec{q}_1)S^-(\vec{q}_2) | \mathcal{L} S^-(\vec{q}) \rangle}{z\chi^-(\vec{q}_1, 0)} + O(1/z^3) \\ &= 1/z N^{-1/2} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^-(\vec{q}_1 + \vec{q}_2, 0)}, \end{aligned}$$

and the first approximation for $\Gamma^-(\vec{q}_1, \vec{q}_2; z)$ is

$$\Gamma^-(q_1, q_2; z) \cong N^{-1/2} \frac{\gamma^-(q_1, q_2)}{z},$$

and therefore

$$\gamma^-(\vec{q}_1, \vec{q}_2) = \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^-(\vec{q}_1 + \vec{q}_2, 0)}. \quad (4.3)$$

The condition for the stationarity of the three-spin correlation function $\langle S^-(\vec{q}_1)S^s(\vec{q}_2)S^-(\vec{q}_3) \rangle$ gives a relation for $n(q)$ and $m(q)$. Take $\vec{q}_3 = \vec{q}_1 + \vec{q}_2$ in (3.17):

$$\begin{aligned} [a(\vec{q}_1 + \vec{q}_2) - a(\vec{q}_2)] n(\vec{q}_1 + \vec{q}_2) m(\vec{q}_2) \\ + [a(\vec{q}_1) - a(\vec{q}_1 + \vec{q}_2)] n(\vec{q}_1) \frac{1}{2} [n(\vec{q}_1 + \vec{q}_2)] \\ + [a(\vec{q}_2) - a(\vec{q}_1) + b(\vec{q}_2)] n(\vec{q}_1) m(\vec{q}_2) \\ - b(\vec{q}_2) m(\vec{q}_2) m(\vec{q}_1 + \vec{q}_2) = 0. \end{aligned}$$

Divide by $n(\vec{q}_1 + \vec{q}_2)$ to get

$$\begin{aligned} a(\vec{q}_1 + \vec{q}_2) [m(\vec{q}_2) - \frac{1}{2} n(\vec{q}_1)] - [a(\vec{q}_2) + b(\vec{q}_2)] \\ \times m(\vec{q}_2) + \frac{1}{2} a(\vec{q}_1) n(\vec{q}_1) \\ = - [a(\vec{q}_2) - a(\vec{q}_1) + b(\vec{q}_2)] \frac{n(\vec{q}_1) m(\vec{q}_2)}{n(\vec{q}_1 + \vec{q}_2)}. \quad (4.4) \end{aligned}$$

By interchanging \vec{q}_1 and \vec{q}_2 one sees that the left-hand side of (4.4) is just $\gamma^-(\vec{q}_1, \vec{q}_2)$ [see Eq. (3.11)]. Combining (4.3) and (4.4)

$$\begin{aligned} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^-(\vec{q}_1 + \vec{q}_2, 0)} = \gamma^-(\vec{q}_1, \vec{q}_2) \\ = - [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \frac{n(\vec{q}_2) m(\vec{q}_1)}{n(\vec{q}_1 + \vec{q}_2)}. \quad (4.5) \end{aligned}$$

From the fact that matrix elements of \mathcal{L} are $O(1/C)$,

$$\langle A \begin{bmatrix} \alpha \\ i \end{bmatrix}_n | A \begin{bmatrix} \alpha' \\ j \end{bmatrix}_n \rangle = \beta \langle A \begin{bmatrix} \alpha'' \\ k \end{bmatrix}_n \rangle (1 + O(1/C))$$

and therefore

$$\chi^s(\vec{q}, 0) = \langle S^s(\vec{q}) | S^s(\vec{q}) \rangle = \beta \langle S^s(-\vec{q}) S^s(\vec{q}) \rangle = \beta m(\vec{q}), \quad (4.6)$$

$$\chi^-(\vec{q}, 0) = \langle S^-(\vec{q}) | S^-(\vec{q}) \rangle = \beta \langle S^+(-\vec{q}) S^-(\vec{q}) \rangle = \beta n(\vec{q}). \quad (4.7)$$

Using result (4.7) in (4.5),

$$\begin{aligned} n(\vec{q}_2) - 2m(\vec{q}_1) &= - [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \beta n(\vec{q}_2) m(\vec{q}_1), \\ m(q_1) &= \{ 2/n(q_2) - [a(q_1) - a(q_2) + b(q_1)] \beta \}^{-1}. \end{aligned}$$

Substitute $n(\vec{q}_2)$ given by (4.2) in the last result to get

$$\begin{aligned} m(q_1) &= ((2/K) \{ 1 + \chi^s [a(0) + b(0) - a(\vec{q}_2)] \} \\ &\quad - [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \beta)^{-1} \end{aligned}$$

For $q_1 = 0$, $m(0) = \chi^s/\beta$ and from this obtain $K = 2\chi^s/\beta$. The equilibrium spin correlation functions are given by

$$n(\vec{q}) = (2/\beta) \{ 1/\chi^s + [a(0) + b(0) - a(\vec{q})] \}^{-1}, \quad (4.8)$$

$$m(\vec{q}) = (1/\beta) \{ 1/\chi^s + [a(0) + b(0) - a(\vec{q}) - b(\vec{q})] \}^{-1}. \quad (4.9)$$

These are anisotropic spherical model values, which could also have been obtained directly by making the usual spherical-model approximation on the Hamiltonian (1.1). The equivalence of the spherical model and the stationary solution is a consequence of evaluating the decay rates correct-

ly to lowest order in $1/C$. The relationship between χ and β , that completes the description of the model, is obtained by requiring that

$$\sum_{\vec{q}} [m(\vec{q}) + n(\vec{q})] = S(S+1).$$

Using results (4.6)–(4.9) the initial values for $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ can be written as

$$N^{-1/2} \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^\alpha(\vec{q}_1 + \vec{q}_2, 0)} = N^{-1/2} [a(\vec{q}_2) - a(\vec{q}_1)] \frac{n(\vec{q}_1)n(\vec{q}_2)}{m(\vec{q}_1 + \vec{q}_2)}, \quad (4.10)$$

$$N^{-1/2} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^-(\vec{q}_1 + \vec{q}_2, 0)} = -N^{-1/2} [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \times \frac{n(\vec{q}_2)m(\vec{q}_1)}{n(\vec{q}_1 + \vec{q}_2)}. \quad (4.11)$$

We have derived the expressions (4.8) and (4.9) from the equations of motion, but they can as readily be derived from the diagrammatic expansion, by equating the matrix elements given on the right-hand side of Table II with the expression calculated exactly using the identity $\langle A | \mathcal{L} | B \rangle = \langle\langle [B, A^\dagger] \rangle\rangle$. The result would be that

$$\rho_m(q) = [\frac{1}{3}S(S+1)]^{-1}m(q), \\ \rho_n(q) = [\frac{2}{3}S(S+1)]^{-1}n(q).$$

However, in order to derive the expression for the vertices, and to verify the rules for calculating the diagrams at finite temperature, we required the condition $\rho_n(i-i)=1$, $\rho_m(i-i)=1$, which is not satisfied, since $m(q) \neq \frac{1}{2}n(q)$ in the presence of anisotropy. From (4.11) we have, however, that

$$\sum_q [n(q) - 2m(q)] = \frac{1}{2}\beta \sum_q b(q)n(q)m(q) = O(1/C).$$

Thus $\rho_n(i-i) = 1 + O(1/C)$, $\rho_m(i-i) = 1 + O(1/C)$, and the corrections to a diagram arising from the deviation from 1 will be of the same order as the terms that we have been neglecting. The inconsistency is only apparent, therefore. We note also that this correction vanishes at $T = \infty$.

In order to get an estimate of the accuracy of the kinetic equations, we will introduce a simple physically motivated approximation that will allow us to obtain an approximate solution of the equations that will have the correct second and fourth moments. We will assume that the solution of the kinetic equation for the vertex function can be written

$$\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z) = \{N^{-1/2} \frac{2}{3}S(S+1)[a(\vec{q}_2) - a(\vec{q}_1)] \times \rho_n(\vec{q}_1)\rho_n(\vec{q}_2)\rho_m^{-1}(\vec{q})\} / \nu(z), \quad (4.12a)$$

$$\Gamma^-(\vec{q}_1, \vec{q}_2; z) = \{-N^{-1/2} \frac{1}{3}S(S+1)[a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)]$$

$$\times \rho_m(\vec{q}_1)\rho_n(\vec{q}_2)\rho_n^{-1}(\vec{q})\} / \mu(z). \quad (4.12b)$$

This is the constant relaxation-time approximation for the functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$. The functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; t)$ determine the rate of decay of one spin fluctuation of wave vector $\vec{q}_1 + \vec{q}_2$ into two fluctuations of wave vectors \vec{q}_1 and \vec{q}_2 and the quantities in the brackets of (4.12) are the values of $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; t)$ for $t=0$. Therefore the approximation states that the relaxation of those functions is independent of \vec{q}_1 and \vec{q}_2 . The approximations are valid and will lead to correct descriptions of the experiments, because at high temperatures fluctuations of any wave vector will decay primarily into short-wavelength fluctuations, since the phase space for such decays is large. Moreover, the relaxation time for short-wavelength fluctuations is approximately constant, and therefore the relaxation of the functions $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; t)$ are nearly independent of wave vector. The functions $\nu(z)$, $\mu(z)$ will be determined by using the assumption (4.12a) and (4.12b) to calculate these functions self-consistently from the kinetic equation at $T = \infty$.

Insert (4.12a) and (4.12b) in the right-hand side of (3.25a) and (3.25b). Divide by z and substitute the resultant expression for $\Gamma^\alpha(\vec{q}_1, \vec{q}_2; z)$ and $\Gamma^-(\vec{q}_1, \vec{q}_2; z)$ in (1.16) and (1.17). After some algebra we obtain the result shown in Fig. 3(a).

If (4.12a) and (4.12b) are substituted directly in (1.16) and (1.17), we obtain the result shown in Fig. 3(b),

$$\phi^+(q, z) = \langle \omega^2 \rangle_q^+ / \nu(z), \quad (4.13)$$

$$\phi^-(q, z) = \langle \omega^2 \rangle_q^- / \mu(z). \quad (4.14)$$

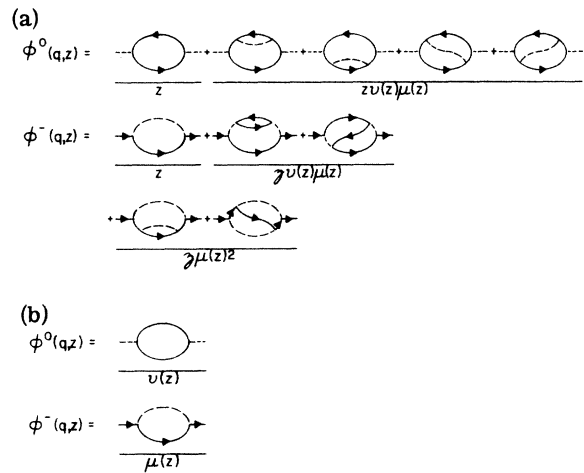


FIG. 3. (a) Approximate expression for Φ obtained by using constant relaxation-time approximation in equation of motion for Γ . (b) Approximate expression for Φ using constant relaxation-time approximation directly in equation for Φ in terms of Γ .

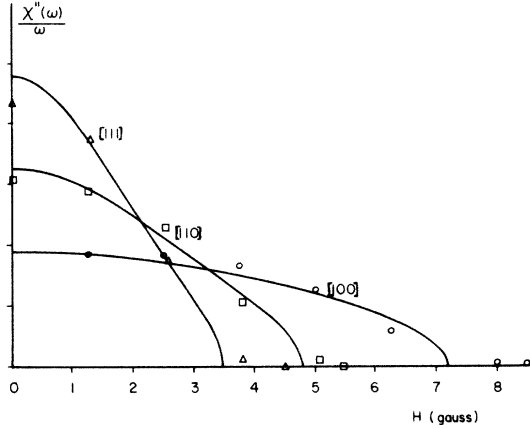


FIG. 4. Comparison of spectral densities predicted by constant relaxation-time approximation with data of Bruce on CaF_2 . Intensity axis normalized by fitting experimental and theoretical results at $\omega=0$ for the [1, 0, 0] direction.

These results when combined give the following equations from which $\nu(z)$ and $\mu(z)$ can be calculated:

$$\frac{\langle \omega^2 \rangle_q^2}{\nu(z)} = \frac{\langle \omega^2 \rangle_q^2}{z} + \frac{\langle \Omega^4 \rangle_q^2}{z\nu(z)\mu(z)}, \quad (4.15)$$

$$\frac{\langle \omega^2 \rangle_q^-}{\mu(z)} = \frac{\langle \omega^2 \rangle_q^-}{z} + \frac{A\langle \Omega^4 \rangle_q^-}{z\nu(z)\mu(z)} + \frac{B\langle \Omega^4 \rangle_q^-}{z\mu^2(z)}. \quad (4.16)$$

The solutions must satisfy the condition $\nu(z), \mu(z) \rightarrow z$ for large z . That the solution will have the correct second and fourth moment is then evident by comparing the expansion of the right-hand sides of (4.15) and (4.16) for large z with (2.8). The values of $\nu(z), \mu(z)$ obtained by solving these equations will have $\langle \omega^2 \rangle_q^2, \langle \Omega^4 \rangle_q^2$ as parameters, and when substituted in (4.13) and (4.14) yield a solution for the spectral density. Strictly speaking, $\nu(z)$ and $\mu(z)$ depend also upon q , and since this dependence has been neglected in deriving (4.15) and (4.16), the approximation is only a good one if the variation with q is small. In order to compare with experiment, we have used the values of a_{ij} and b_{ij} appropriate for the truncated dipole Hamiltonian on a simple cubic lattice:

$$a_{ij} = [(\gamma\hbar)^2/2r_{ij}^3](1 - 3\cos^2\theta_{ij}),$$

$$b_{ij} = -3a_{ij},$$

which allows us to calculate the NMR line shape and free-induction decay in a system of coupled nuclear dipoles,⁶ CaF_2 , in a strong magnetic field. All that is required to interpret these experiments is the value of the spectral density at $q=0$, and since $A\langle \Omega \rangle_q = \frac{1}{3}B\langle \Omega \rangle_q$ for $q=0$, there is little loss of accuracy in making $\mu(z) = \nu(z)$ in (4.16). We get

$$\mu(\omega + i\epsilon) = \frac{1}{2}\omega + i(\langle \Omega^4 \rangle_{q=0}^- / \langle \omega^2 \rangle_{q=0}^- - \frac{1}{4}\omega^2)^{1/2}, \quad (4.17)$$

when

$$\omega < 2(\langle \Omega^4 \rangle_{q=0}^- / \langle \omega^2 \rangle_{q=0}^-)^{1/2}.$$

Using this result in (4.14) we get from (1.12) and (2.4) the line shape

$$f(\omega) = \frac{1}{\langle \omega^2 \rangle_q^-} \left(\frac{\langle \Omega^4 \rangle_q^-}{\langle \omega^2 \rangle_q^-} \right)^{1/2} \left(1 - \frac{\omega^2}{4} \frac{\langle \omega^2 \rangle_q^-}{\langle \Omega^4 \rangle_q^-} \right)^{1/2} \times \left[1 + \frac{\omega^2}{\langle \omega^2 \rangle_q^-} \left(\frac{\langle \Omega^4 \rangle_q^-}{\langle \omega^2 \rangle_q^-} - 1 \right) \right]^{-1}, \quad (4.18)$$

where $f(\omega) = \text{Re}\Sigma(0, \omega)/\chi^-(0, 0)$.

The second and fourth moments appearing in (4.18) are those predicted by Eqs. (3.25). The second moment is exact and the fourth moment differs by only a few percent from the exact value. In actually calculating the line shape, we will use the exact value of the fourth moment in (4.18).⁷ In Fig. 4 we show the line shape for CaF_2 for the three different directions of the external field. The points represent Bruce's⁸ data. In Fig. 5 we show the free-induction decay, which is the Laplace transform of the line shape, also for the three different directions of the external field. The data are due to Lowe and Barnaal,⁹ who measure the free-induction decay directly, and the Fourier transform of Bruce's line shape.

We can see that the kinetic equations together with the constant relaxation time approximation accounts not only for the shape of the free-induction decay but for the systematic variation of the shape with the direction of the magnetic field, i.e., with the parameters of the Hamiltonian, which are known precisely from the lattice constant. We emphasize that there are no adjustable parameters in the fit in Fig. 5, and that only the intensity at one point has been fit in Fig. 4, which is made necessary by the arbitrariness in the intensity scale for the experimental data. The differences between the theoretical and experimental curves are comparable to the correction to be expected upon solving (3.25a) and (3.25b) exactly. One can estimate these corrections to $f(\omega)$ by iterating the solution, which has been done for the isotropic case, $b_{ij} = 0$.¹ The corrections amount to about 20% at most (at $\omega=0$), in this case, and are expected to be smaller for the present situation, since the constant relaxation time approximation can be expected to hold for the entire zone, whereas it fails when $q_1, q_2 \cong 0$ in the isotropic case. We have not calculated the variation of the ratio $\langle \Omega^4 \rangle_q^- / \langle \omega^2 \rangle_q^-$ with q , but again we expect it to be smaller than in the isotropic case, where it is 20% over the whole zone. We point out that numerical solutions of the equations are readily ob-

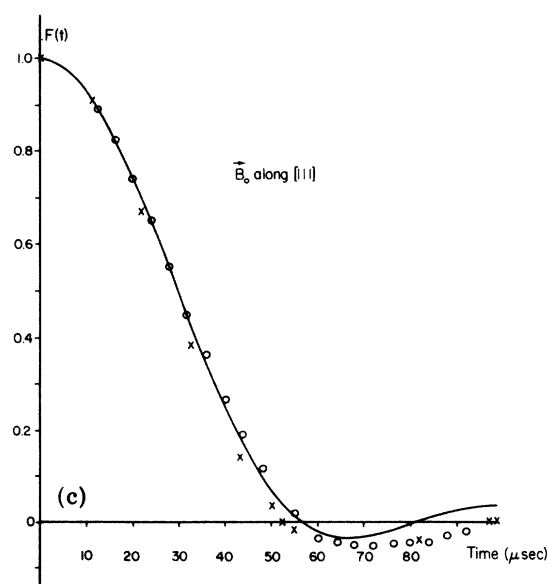
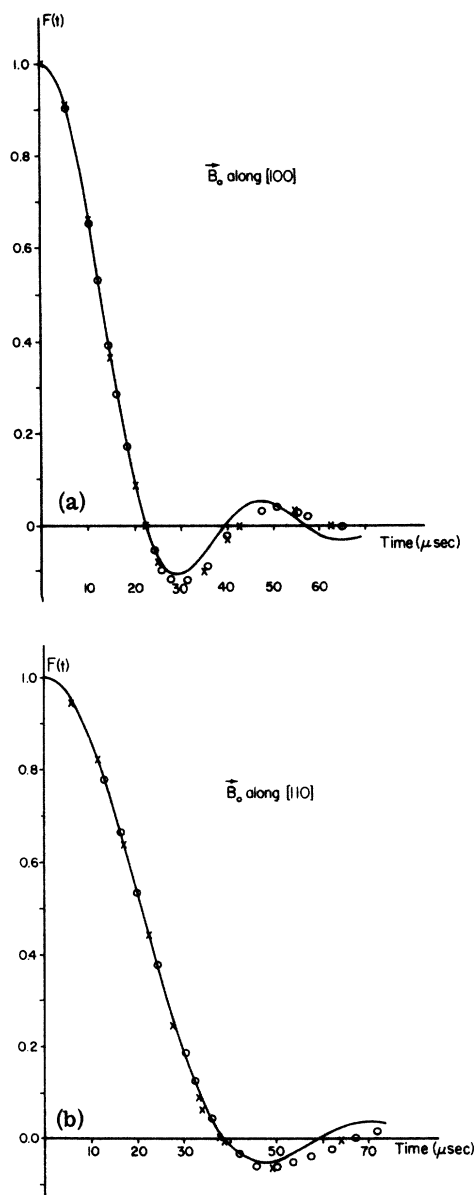


FIG. 5. Free-induction decay in CaF_2 compared with theoretical prediction using constant relaxation-time approximation. Circles are the data of Lowe and Bar-naal, the crosses the Fourier transform of the data of Bruce, shown in Fig. 4.

The spin dynamics of systems described by the Hamiltonian (1.1) has been the subject of work for a long time. However, only recently attempts towards microscopic theories were made. The early works¹¹⁻¹⁵ have been restricted to the study of particular problems, such as NMR or EPR line shape, valid only for $T = \infty$ for special values of the spin. Moreover, those theories have made use of *ad hoc* assumptions about either the spin correlation function or another related dynamical function. More recently, the thermodynamical Green's-function method was used to study the NMR line shape¹⁶ and the exchange narrowing problem.¹⁷ The first of those theories contains an adjustable parameter which is chosen to fit the experimental data and no attempt to derive the equation for the correlation functions is made. The second theory obtains the equations of motion for the two-spin function. However, the equations cannot be solved and the analysis of the problem proceeds only if one assumes *ad hoc* the correct form for the spectral function valid only in the hydrodynamic limit.

The first attempt towards a microscopic theory capable of describing the fluctuations of the dipole system was done by Borkmans and Walgraef.¹⁸ They have made a systematic analysis of a perturbation solution of the function $\langle S_a^\alpha(t) S_b^\alpha(0) \rangle$ by renormalizing the perturbation theory. This method does not yield accurate results if the kinetic equations are solved directly, and gives an incor-

tained in one dimension, and have been carried out for the isotropic case by Foster and Reiter.¹⁰

CONCLUDING REMARKS AND COMPARISON WITH OTHER WORK

We have derived the set of kinetic equations (3.25) based on first principles, which is able to describe the dynamics of a system of spins \vec{S} at any temperature $T \geq T_c$. As a result of the renormalization of higher-order spin correlation functions most of the internal relaxation processes are described by the kinetic equations. The renormalization procedure is the essential point of the theory.

rect value for the fourth moment. A better approximation on this method has been done,¹⁹ which consists in factorizing the four-spin correlation function $\langle S^{\alpha 1}(q_1, t) S^{\alpha 2}(q_2, t) S^{\alpha 3}(q_3, 0) S^{\alpha 4}(q_4, 0) \rangle$, but it does not lead to any significant improvement. The predicted spectral density and fourth moment are still incorrect. It is evident that this approximation does not allow for the scattering process. This is the reason why the fourth moment predicted by the equations^{4,5,18,20,21} is seriously in error, and also it leads to an exact solution of the equations which underestimates the damping of the oscillations appearing in the experiments. On the other hand, the derivation of the present work which is based on the renormalization of the function $\langle S^-(q_1, t) S^+(q_2, t) S^z(q_3, t) S^-(q_4, 0) S^+(q_5, 0) \rangle$ leading to the equations (3.25) for the functions $\Gamma^\alpha(q_1, q_2; t)$ explicitly shows both the decay of the single excitation and the scattering between excitations. The fourth moment calculated from the equations (3.25) is correct to the order of $1/C$ (see Appendix).

Although the solutions of the kinetic equations for the vertex functions have been used to describe dipole spin systems at $T = \infty$, the theory is not restricted in any sense. For a system composed of spin \vec{S} we can treat, using the derived formalism, the temperature dependence of the diffusion constant which can be obtained from $\phi^z(q, z) = -iDq^2$ as $q \rightarrow 0$ and $z \rightarrow 0$, the temperature dependence of the exchange narrowing problem, etc. Due to fact that the coefficients a_{ij} and b_{ij} are arbitrary, we could as well study the intermediate cases for dipolar-exchange systems, the x - y model, and the Ising model. The isotropic case⁴ ($b_{ij} = 0$) has been

shown to give good agreement with experiments,²² and we can expect the same order of accuracy for the calculations based on Eqs. (3.25) together with the constant relaxation-time approximation.

APPENDIX: CONSERVATION LAWS AND MOMENTS FROM KINETIC EQUATIONS

In this appendix it will be shown that the proposed kinetic equations guarantee the conservation of total spin and energy, i. e.,

$$\frac{\partial}{\partial t} N^{-1} \sum_{\vec{q}} \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}) \rangle = 0, \quad (\text{A1})$$

$$\begin{aligned} \frac{\partial}{\partial t} N^{-1} \sum_{\vec{q}} [a(\vec{q}) \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}) \rangle \\ + b(\vec{q}) \langle S^z(\vec{q}) S^z(-\vec{q}) \rangle] = 0, \end{aligned} \quad (\text{A2})$$

and that the moments derived from the kinetic equations agree with the values calculated from the diagrams in Sec. II.

From the commutation relations

$$\begin{aligned} \langle S^+(\vec{q}_1) S^-(\vec{q}_2); t \rangle \\ = \langle S^-(\vec{q}_2) S^+(\vec{q}_1); t \rangle + N^{1/2} \langle S^z(\vec{q}_1 + \vec{q}_2); t \rangle \\ = n^0(\vec{q}_2, \vec{q}_1; t) + N^{1/2} \langle S^z(\vec{q}_1 + \vec{q}_2); t \rangle. \end{aligned}$$

Using the fact that $\langle S^z(\vec{q}); t \rangle = 0$,

$$\begin{aligned} \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}); t \rangle \\ = \langle \frac{1}{2} [S^-(\vec{q}) S^+(-\vec{q}) + S^+(\vec{q}) S^-(\vec{q})] + S^z(\vec{q}) S^z(-\vec{q}) \rangle \\ = \frac{1}{2} [n^0(\vec{q}, -\vec{q}; t) + n^0(-\vec{q}, \vec{q}; t)] + m(\vec{q}, -\vec{q}; t). \end{aligned} \quad (\text{A3})$$

Substitute (A3) in Eqs. (3.23) to get

$$\begin{aligned} z \langle S(\vec{q}) \cdot S(-\vec{q}); z \rangle = & i \langle S(\vec{q}) \cdot S(-\vec{q}); t=0 \rangle + \phi^-(\vec{q}, z) \frac{1}{2} [n^0(\vec{q}, -\vec{q}; z) + n^0(-\vec{q}, \vec{q}; z)] \\ & + \phi^-(\vec{q}_1, z) \frac{1}{2} [n^0(\vec{q}, -\vec{q}; z) + n^0(-\vec{q}, \vec{q}; z)] + \phi^z(\vec{q}, z) m(\vec{q}, -\vec{q}; z) + \phi^z(-\vec{q}, z) m(\vec{q}, -\vec{q}; z) \\ & - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(\vec{q} - \vec{q}') - b(\vec{q} - \vec{q}')] \Gamma^-(\vec{q} - \vec{q}', -\vec{q}; z) \frac{1}{2} [n^0(\vec{q}', -\vec{q}'; z) + n^0(-\vec{q}', \vec{q}'; z)] \\ & - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}') - a(-\vec{q} - \vec{q}') - b(-\vec{q} - \vec{q}')] \Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) \frac{1}{2} [n^0(\vec{q}', -\vec{q}'; z) + n^0(-\vec{q}', \vec{q}'; z)] \\ & + N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^z(\vec{q} - \vec{q}', -\vec{q}; z) m(-\vec{q}', \vec{q}'; z) \\ & + N^{-1/2} \sum_{\vec{q}'} [a(-\vec{q} - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^z(-\vec{q} - \vec{q}', \vec{q}; z) m(-\vec{q}', \vec{q}'; z) \\ & - N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) \frac{1}{2} [n^0(-\vec{q}', \vec{q}'; z) + n^0(\vec{q}', -\vec{q}'; z)] \\ & - N^{-1/2} \sum_{\vec{q}'} [a(-\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) \frac{1}{2} [n^0(-\vec{q}', \vec{q}'; z) + n^0(\vec{q}', -\vec{q}'; z)]. \end{aligned} \quad (\text{A4})$$

Sum (A4) over \vec{q} and consider the seventh and eleventh terms:

$$\begin{aligned} N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} [a(-\vec{q} - \vec{q}') + b(-\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) f(\vec{q}') \\ - N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} [a(-\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) f(\vec{q}'), \end{aligned}$$

where

$$f(\vec{q}) = \frac{1}{2}[n^0(\vec{q}', -\vec{q}'; z) + n^0(-\vec{q}', \vec{q}'; z)].$$

Consider term by term

$$\begin{aligned} N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} f(\vec{q}') a(-\vec{q} - \vec{q}') [\Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z)] \\ = N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') \sum_{\vec{q}} [a(-\vec{q} - \vec{q}') - a(\vec{q})] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) \end{aligned}$$

by redefining the indices in the second term as $-\vec{q} - \vec{q}' = \vec{q}''$ and call $\vec{q}'' = \vec{q}$. The other term is

$$\begin{aligned} -N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} a(\vec{q}') f(\vec{q}') [\Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z)] \\ = -N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') a(\vec{q}') \left\{ \sum_{\vec{q}} [\Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z)] \right\}. \end{aligned}$$

The term in the curly brackets is zero because the sum is over all \vec{q} . So it contains

$$\Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) + \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z).$$

The first and last term add up to zero, as well as the second and third, by redefining the indices

$$\sum_{\vec{q}} [\Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z)] = \sum_{\vec{q}} \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) - \sum_{\vec{q}''} \Gamma^-(\vec{q}'' - \vec{q}', \vec{q}''; z) = 0.$$

Therefore the contribution from terms seven and eleven is

$$\begin{aligned} N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') \sum_{\vec{q}} [a(-\vec{q} - \vec{q}') - a(\vec{q}) + b(-\vec{q} - \vec{q}')] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) \\ = \sum_{\vec{q}'} f(\vec{q}') \left\{ N^{-1/2} \sum_{\vec{q}} [a(\vec{q}) - a(-\vec{q} - \vec{q}') + b(\vec{q})] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) \right\} \\ = - \sum_{\vec{q}'} f(\vec{q}') \phi^-(\vec{q}'; z), \end{aligned}$$

which cancels the result obtained from the third term when summed over \vec{q} . Similarly the sixth and tenth terms cancel the second term.

Now take the eighth term and sum over \vec{q} :

$$N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^s(\vec{q} - \vec{q}', -\vec{q}; z) m(-\vec{q}', \vec{q}'; z).$$

The function $\Gamma^s(\vec{q}_1, \vec{q}_2; z)$ is antisymmetric in interchange of \vec{q}_1 and \vec{q}_2 . Therefore

$$\sum_{\vec{q}'} a(\vec{q}') m(-\vec{q}', \vec{q}'; z) \sum_{\vec{q}} \Gamma^s(\vec{q} - \vec{q}', -\vec{q}; z) = 0.$$

The only nonzero contribution comes from the first term, and using the antisymmetric property of $\Gamma^s(\vec{q}_1, \vec{q}_2; z)$ follows

$$\begin{aligned} N^{-1/2} \sum_{\vec{q}'} m(-\vec{q}', \vec{q}'; z) \left\{ \sum_{\vec{q}} a(\vec{q} - \vec{q}') \frac{1}{2} [\Gamma^s(\vec{q} - \vec{q}', -\vec{q}; z) - \Gamma^s(-\vec{q}, \vec{q} - \vec{q}'; z)] \right\} \\ = N^{-1/2} \sum_{\vec{q}'} m(-\vec{q}', \vec{q}'; z) \left(\frac{1}{2} \sum_{\vec{q}} [a(\vec{q} - \vec{q}') - a(-\vec{q})] \Gamma^s(\vec{q} - \vec{q}', -\vec{q}; z) \right) \\ = \sum_{\vec{q}'} m(-\vec{q}', \vec{q}'; z) \left(-\frac{1}{2} N^{-1/2} \sum_{\vec{q}} [a(\vec{q} - \vec{q}') - a(\vec{q})] \Gamma^s(\vec{q}, \vec{q}' - \vec{q}; z) \right) \\ = - \sum_{\vec{q}'} m(-\vec{q}', \vec{q}'; z) \phi^s(\vec{q}', z) \end{aligned}$$

which cancels the result obtained from the fourth term when summed over \vec{q} . Similarly, the ninth term cancels the fifth. Therefore,

$$z N^{-1} \sum_{\vec{q}} \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}); z \rangle = i N^{-1} \sum_{\vec{q}} \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}); t=0 \rangle = i S(S+1),$$

which is the Laplace transform of (A1). In order to prove relation (A2) multiply result (A4) by $a(q)$ and

(3.23b) with $\vec{q}_1 = \vec{q}$, $\vec{q}_2 = -\vec{q}$ by $b(\vec{q})$ to get

$$\begin{aligned}
& z[a(\vec{q}) \langle S(\vec{q}) \cdot S(-\vec{q}); z \rangle + b(\vec{q}) \langle S^e(\vec{q}) S^e(-\vec{q}); z \rangle] \\
&= i[a(\vec{q}) \langle S(\vec{q}) \cdot S(-\vec{q}); t=0 \rangle + b(\vec{q}) \langle S^e(\vec{q}) S^e(-\vec{q}); t=0 \rangle] + a(\vec{q}) \phi^-(\vec{q}, z) f(\vec{q}) + a(\vec{q}) \phi^-(\vec{q}, z) f(\vec{q}) \\
&+ [a(\vec{q}) + b(\vec{q})] \phi^e(\vec{q}, z) m(\vec{q}_1 - \vec{q}; z) + [a(\vec{q}) + b(\vec{q})] \phi^e(-\vec{q}, z) m(\vec{q}_1 - \vec{q}; z) \\
&- N^{-1/2} \sum_{\vec{q}'} a(\vec{q}) [a(\vec{q}') - a(\vec{q} - \vec{q}') - b(\vec{q} - \vec{q}')] \Gamma^-(\vec{q} - \vec{q}', -\vec{q}; z) f(\vec{q}') \\
&- N^{-1/2} \sum_{\vec{q}'} a(\vec{q}) [a(\vec{q}') - a(-\vec{q} - \vec{q}') - b(-\vec{q} - \vec{q}')] \Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) f(\vec{q}') \\
&+ N^{-1/2} \sum_{\vec{q}'} a(\vec{q}) [a(\vec{q} - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^e(\vec{q} - \vec{q}', -\vec{q}; z) m(\vec{q}', -\vec{q}; z) \\
&+ N^{-1/2} \sum_{\vec{q}'} a(\vec{q}) [a(-\vec{q} - \vec{q}') - a(\vec{q}') - b(\vec{q}')] \Gamma^e(-\vec{q} - \vec{q}', \vec{q}; z) m(\vec{q}', -\vec{q}; z) \\
&- N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}) + b(\vec{q})] [a(\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}, \vec{q} - \vec{q}'; z) f(\vec{q}') \\
&- N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}) + b(\vec{q})] [a(-\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) f(\vec{q}') , \tag{A5}
\end{aligned}$$

where $f(\vec{q}) = \frac{1}{2}[n^0(\vec{q}, -\vec{q}; z) + n^0(-\vec{q}, \vec{q}; z)]$. Consider terms seven and eleven and sum over \vec{q}

$$\begin{aligned}
& - N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} [a(\vec{q}) a(\vec{q}') - a(\vec{q}) a(-\vec{q} - \vec{q}') - a(\vec{q}) b(-\vec{q} - \vec{q}')] \Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) f(\vec{q}') \\
& - N^{-1/2} \sum_{\vec{q}} \sum_{\vec{q}'} [a(\vec{q}) a(-\vec{q} - \vec{q}') - a(\vec{q}) a(\vec{q}') + b(\vec{q}) a(-\vec{q} - \vec{q}') - b(\vec{q}) a(\vec{q}')] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) f(\vec{q}') . \tag{A6}
\end{aligned}$$

Take term by term

$$\begin{aligned}
& - N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') \sum_{\vec{q}} a(\vec{q}) a(\vec{q}') [\Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) - \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z)] \\
&= - N^{-1/2} \sum_{\vec{q}'} a(\vec{q}') f(\vec{q}') \sum_{\vec{q}} [a(\vec{q} + \vec{q}') - a(\vec{q})] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) .
\end{aligned}$$

This result, when combined with the contribution coming from $b(\vec{q}) a(\vec{q}')$, gives

$$N^{-1/2} \sum_{\vec{q}'} a(\vec{q}') f(\vec{q}') \sum_{\vec{q}} [a(\vec{q}) - a(\vec{q} + \vec{q}') + b(\vec{q})] \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z) = - \sum_{\vec{q}'} a(\vec{q}') f(\vec{q}') \phi^-(\vec{q}', z)$$

and this cancels the result obtained from the third term when summed over \vec{q} . It has to be shown also that the other terms in (A6) have zero contribution. Consider

$$\begin{aligned}
& N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') \sum_{\vec{q}} a(\vec{q}) a(-\vec{q} - \vec{q}') [\Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) - \Gamma^-(\vec{q}, -\vec{q} - \vec{q}'; z)] \\
&= N^{-1/2} \sum_{\vec{q}'} f(\vec{q}') \left(\sum_{\vec{q}} [a(\vec{q}) a(-\vec{q} - \vec{q}') - a(-\vec{q} - \vec{q}') a(\vec{q})] \Gamma^-(\vec{q} - \vec{q}', \vec{q}; z) \right) = 0 ,
\end{aligned}$$

where in second term $\vec{q} \rightarrow -\vec{q}'' - \vec{q}'$ and $\sum_{\vec{q}''} \rightarrow \sum_{\vec{q}}$.

The same procedure, when applied to the group of terms (2), (6), (10), (5), (9), and (4), (8) would lead to a cancellation.

It follows then

$$z N^{-1} \sum_{\vec{q}} [a(\vec{q}) \langle \vec{S}(\vec{q}) \cdot \vec{S}(-\vec{q}); z \rangle + b(\vec{q}) \langle S^e(\vec{q}) S^e(-\vec{q}); z \rangle] = i \langle H; t=0 \rangle ,$$

which is the Laplace transform of (A2).

The derivation of the kinetic equations was based on a cluster expansion of the correlation function and the terms kept in this expansion were correct to the order $1/C$. The diagrammatic expansion of the matrix elements of the Liouville operator was also correct to order $1/C$. Therefore the moments of the spectral function calculated from the kinetic equations should agree with those calculated from the diagrams as will be shown now.

Take Eq. (3.25a) and divide by z , and iterate it once using the first approximation $\Gamma^\alpha(\vec{q}_1 \vec{q}_2; z) = \Gamma_0^\alpha(\vec{q}_1 \vec{q}_2)$;

$$\begin{aligned}
\Gamma^{\epsilon}(\vec{q}_1, \vec{q}_2; z) &= \Gamma_0^{\epsilon}(\vec{q}_1, \vec{q}_2) + [\langle \omega^2 \rangle_{\vec{q}_1}^{-} + \langle \omega^2 \rangle_{\vec{q}_2}^{-}] \Gamma_0^{\epsilon}(\vec{q}_1, \vec{q}_2)(1/z^2) \\
&+ N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_1 - \vec{q}') - a(\vec{q}') + b(\vec{q}_1 - \vec{q}')] \Gamma_0^{-}(\vec{q}_1 - \vec{q}', \vec{q}_2) \Gamma_0^{\epsilon}(\vec{q}', \vec{q} - \vec{q}')(1/z) \\
&+ N^{-1/2} \sum_{\vec{q}'} [a(\vec{q}_2 - \vec{q}') - a(\vec{q}') + b(\vec{q}_2 - \vec{q}')] \Gamma_0^{-}(\vec{q}_2 - \vec{q}', \vec{q}_1) \Gamma_0^{\epsilon}(\vec{q} - \vec{q}', \vec{q}')(1/z), \tag{A7}
\end{aligned}$$

where $\phi^{-}(q, z) = \langle \omega^2 \rangle_{\vec{q}}^{-}/z$ in first approximation. Substitute (A7) in (1.16):

$$\begin{aligned}
\phi^{\epsilon}(\vec{q}, z) &= \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \Gamma_0^{\epsilon}(\vec{q}', \vec{q} - \vec{q}') \\
&+ \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [\langle \omega^2 \rangle_{\vec{q}'}^{-} + \langle \omega^2 \rangle_{\vec{q} - \vec{q}'}^{-}] \Gamma_0^{\epsilon}(\vec{q}', \vec{q} - \vec{q}')(1/z^2) \\
&+ \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [a(\vec{q}' - \vec{q}'') - a(\vec{q}'') + b(\vec{q}' - \vec{q}'')] \Gamma_0^{-}(\vec{q}' - \vec{q}'', \vec{q} - \vec{q}') \Gamma_0^{\epsilon}(\vec{q}'', \vec{q} - \vec{q}'')(1/z) \\
&+ \frac{1}{2} N^{-1/2} \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [a(\vec{q} - \vec{q}' - \vec{q}'') - a(\vec{q}'') + b(\vec{q} - \vec{q}' - \vec{q}'')] \Gamma_0^{-}(\vec{q} - \vec{q}' - \vec{q}'', \vec{q}') \Gamma_0^{\epsilon}(\vec{q} - \vec{q}'', \vec{q}'')(1/z).
\end{aligned}$$

Insert in this expression the values

$$\Gamma_0^{\epsilon}(\vec{q}_1, \vec{q}_2) = \frac{1}{z} N^{-1/2} \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^{\epsilon}(\vec{q}, 0)}, \quad \Gamma_0^{-}(\vec{q}_1, \vec{q}_2) = \frac{1}{z} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^{-}(\vec{q}, 0)},$$

and compare the result obtained with (2.8):

$$\phi^{\epsilon}(\vec{q}_1, z) = \frac{\langle \omega^2 \rangle_{\vec{q}_1}^{\epsilon}}{z} + \frac{\langle \Omega^4 \rangle_{\vec{q}_1}^{\epsilon}}{z^3} + O(1/z^5).$$

The second and fourth moments are given by

$$\langle \omega^2 \rangle_{\vec{q}}^{\epsilon} = \frac{1}{2} N^{-1} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] \frac{n(\vec{q} - \vec{q}') - n(\vec{q}')}{\chi^{\epsilon}(\vec{q}, 0)}, \tag{A8a}$$

$$\begin{aligned}
\langle \Omega^4 \rangle_{\vec{q}}^{\epsilon} &= \frac{1}{2} N^{-1} \sum_{\vec{q}'} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [\langle \omega^2 \rangle_{\vec{q}'}^{-} + \langle \omega^2 \rangle_{\vec{q} - \vec{q}'}^{-}] \frac{n(\vec{q} - \vec{q}') - n(\vec{q}')}{\chi^{\epsilon}(\vec{q}, 0)} \\
&+ \frac{1}{2} N^{-2} \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [a(\vec{q}' - \vec{q}'') - a(\vec{q}'') + b(\vec{q}' - \vec{q}'')] \frac{n(\vec{q}' - \vec{q}'') - 2m(\vec{q} - \vec{q}')}{\chi^{-}(q - q'', 0)} \frac{n(\vec{q} - \vec{q}'') - n(\vec{q}'')}{\chi^{\epsilon}(q, 0)} \\
&+ \frac{1}{2} N^{-2} \sum_{\vec{q}'} \sum_{\vec{q}''} [a(\vec{q} - \vec{q}') - a(\vec{q}')] [a(\vec{q} - \vec{q}' - \vec{q}'') - a(\vec{q}'') + b(\vec{q} - \vec{q}' - \vec{q}'')] \frac{n(\vec{q}') - 2m(\vec{q} - \vec{q}' - \vec{q}'')}{\chi^{-}(\vec{q} - \vec{q}'', 0)} \\
&\times \frac{n(\vec{q}'') - n(\vec{q} - \vec{q}'')}{\chi^{\epsilon}(\vec{q}, 0)}. \tag{A8b}
\end{aligned}$$

From results (4.10) and (4.11) of Sec. IV the following is obtained:

$$N^{-1/2} \frac{n(\vec{q}_2) - n(\vec{q}_1)}{\chi^{\epsilon}(\vec{q}, 0)} = N^{-1/2} \frac{2}{3} S(S+1) [a(\vec{q}_2) - a(\vec{q}_1)] \rho_n(\vec{q}_1) \rho_n(\vec{q}_2) \rho_m^{-1}(\vec{q}), \tag{A9}$$

$$N^{-1/2} \frac{n(\vec{q}_2) - 2m(\vec{q}_1)}{\chi^{-}(\vec{q}, 0)} = -N^{-1/2} \frac{1}{3} S(S+1) [a(\vec{q}_1) - a(\vec{q}_2) + b(\vec{q}_1)] \rho_m(\vec{q}_1) \rho_n(\vec{q}_2) \rho_n^{-1}(\vec{q}).$$

Substituting (A9) into (A8) yields the result (2.20). Equation (2.21) can be derived in the same manner, proving the equivalence of the second and fourth moments of the solution of the kinetic equations with the diagrammatic results.

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