Spin Correlation Functions at High Temperatures*

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A method for the self-consistent calculation of correlation functions is described and applied to the Heisenberg model at high temperatures. The technique is based on a straightforward physical picture. It is used (i) to derive a simple analytic approximation for the autocorrelation function valid at times $t \lesssim 2JS/\hbar$; (ii) to derive an equation given by Résibois and DeLeener which is shown to be valid at short times also; (iii) to derive a set of integrodifferential equations for the general correlation functions $\langle \mathbf{S}_{-\mathbf{q}} \cdot \mathbf{S}_{\mathbf{q}}(t) \rangle$. The latter equations are solved numerically for the case of a simple cubic lattice with nearest-neighbor interactions, and are shown to give results in excellent agreement with computer simulation calculations for the same model. A discussion is given of the physical motivation of the approximations employed and the special mathematical aspects of the problem.

1. INTRODUCTION

The dynamical properties of Heisenberg spin systems above the Curie temperature have been the subject of much theoretical study in recent years[1–11] stimulated by the appearance of inelastic neutron scattering data at high temperatures[12–16] and by the data becoming available from computer simulation calculations for precisely defined models.[17,18] The methods used include moment calculations,[1,6] microscopic theories of spin diffusion,[2,5] diagrammatic perturbation theory,[7,8] and interpolation schemes.[9,10] A useful review of much of this work is contained in Ref. 11.

The theoretical treatment of high-temperature spin systems has conceptual and mathematical difficulties typical of a certain class of many-body problems in which the "self-energy" effects due to the interactions are large compared to the "unperturbed single-particle energies." Other examples are the theories of classical liquids, of homogeneous turbulence,[19] of low density electron gases, or, indeed, of any many-body problem with very strong interactions. The situation can perhaps be made clearer by comparing the low- and high-temperature regimes of a spin system. At low temperatures in (say) a ferromagnet, each spin is acted on by a mean field $\sim JSz_1$ (J= exchange energy, S= spin magnitude, z_1= number of interacting neighbors), which represents to a first approximation the effect of the spin interactions. The higher-order effects of the interaction then appear as small fluctuations of the effective field (acting on a spin) about this mean field. One can think of the mean field as providing a set of "unperturbed" energy levels for a spin and of the higher-order effects of the interactions as a perturbation, a picture which is good, provided that the fluctuations of the effective field are small compared to the mean field. One then has a strong analogy with a system of particles with weak interactions, an analogy which is exploited in spin-wave theory. If, however, one now raises the temperature, the mean field decreases and the fluctuations increase, until at the Curie temperature the mean field and with it the unperturbed energies disappear altogether—one only has the perturbation left.

To overcome this difficulty, there have been two main lines of attack. One is the adoption of "fitting" schemes,[2,5,6,9,10] in which one postulates a certain plausible shape (e.g., Gaussian) for some suitable function (e.g., the spin correlation function, its Fourier transform, or the generalized diffusion function) and adjusts parameters to fit certain known moments, a procedure which can be very successful in practice. The other[3,4,7,8] (of which the theory to be described here is an example) is to attack the microscopic problem from first principles in spite of the difficulty alluded to above. All such calculations have been based upon the notion that although the mean field disappears, each spin is still acted on by an effective field $\sim JS\sqrt{z_1}$ which, albeit fluctuating in magnitude and direction, can play the role of generating unperturbed energy levels. Essentially, in the absence

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19 The methods used in this paper are similar to those first used by Kraichnan in his treatment of turbulence; cf. R. H. Kraichnan, Phys. Rev. **109**, 1407 (1958).

of a real mean field, or unperturbed Hamiltonian, one is asking the self-energy to serve the same purpose. Since the strength of interaction $\sim JS^2$ and the self-energy $\sim JS^2\sqrt{z_1}$, such a theory will have an expansion parameter $\sim 1/\sqrt{z_1}$ (or $1/z_1$ for some quantities), which in practice is small enough to make the approach useful. An essential feature of such an approach is that the self-energy has to be calculated self-consistently, since its expansion in powers of $1/z_1$ expresses it in terms of itself.

The first approach of this kind was that of Résibois and DeLeener,[7] who used a perturbation technique to derive integral equations for the high-temperature correlation functions. These equations are of the self-consistent kind required and are physically plausible. They later extended their approach to discuss the critical region[8] which has also been studied by Kawasaki[4] who, using a different (but again self-consistent) technique, obtained identical equations. In this paper we present another treatment of this kind, motivated by certain simple physical considerations, and derive several approximations of varying degrees of elaboration for the high-temperature case. One of these coincides with Résibois and DeLeener's original result[7] which we show is only valid at short times $t<t_c\sim 2JS/\hbar$. We also derive a simpler analytic solution which is equally accurate in the same time interval, and a more elaborate self-consistent equation which turns out to be the high-temperature form of the equations derived by DeLeener and Résibois[8] and Kawasaki[4] for the critical region. This equation has been solved numerically for the case of a simple cubic lattice with nearest-neighbor interactions and is found to give spin correlation functions $\langle S_{-q}^z S_q^z(t)\rangle$ in excellent agreement with the computer simulation calculations of Windsor[18] and a spin diffusion coefficient in agreement with that calculated by Mori and Kawasaki[2] and also found by Windsor.

The physical idea behind our mathematical calculation is that each spin moves in a randomly varying effective magnetic field produced by its neighbors. The correlation function for the motion of a spin in a randomly varying field can be calculated approximately in terms of the correlation function of the random field, as was done by Kubo and Toyabe,[20] among others. Indeed, by making assumptions about the nature of the random field, the latter authors derived numerical expressions for the spin-spin correlation function. In our treatment, however, we make no such assumption about the correlation functions for the field. Instead, we recognize that the fluctuating field is itself produced by neighboring spins with the same correlation function. This enables us to express the correlation function of the random field in terms of the correlation function of the spins. Combining this result with the calculation of the spin correlation function in terms of that of the random field leads to a self-consistent equation for the spin correlation function which is in some sense an analog of the self-consistent treatment of the magnetization at low temperatures. Needless to say, the mathematical implementation of these physical ideas requires a number of approximations, some of which are straightforward and easily understood, while others are less directly accessible physically. We have made an effort to indicate clearly the approximations made and to discuss, as far as is possible, their physical significance. The principal approximation made is the neglect of the detailed correlations between three or more spins, an approximation which we argue should only lead to errors $O(1/z_1)$ at high temperatures. From the point of view of stochastic theory, this assumption is equivalent to saying that the spin motion is approximately a Gaussian random process.[21]

In Sec. 2 we collect some general theory which is unfamiliar but necessary for the mathematical development. In Sec. 3 we apply our technique to derive a simple approximate equation for the self-correlation function. This equation, which is valid for short times, $t<2JS/\hbar$, has the analytical solution $\langle \mathbf{S}_i(0)\cdot\mathbf{S}_i(t)\rangle = S(S+1)\times\operatorname{sech}^2(\tau/\sqrt{2})$, where $\tau=[\frac{2}{3}S(S+1)\sum_{\mathbf{r}} J^2(\mathbf{r})]^{1/2}t$. In Sec. 4 we show that the use of a more accurate approximation for this problem yields the equation of DeLeener and Résibois, which, however, is still only valid for $t<2JS/\hbar$ and has a solution not much different from the above solution. In Sec. 5 we consider the general wave-vector–dependent correlation functions $\langle \mathbf{S}_{-\mathbf{q}}(0)\cdot\mathbf{S}_{\mathbf{q}}(t)\rangle$ and use our technique to derive a set of approximate nonlinear integrodifferential equations for them. These equations have been integrated numerically, yielding both the correlation functions and their temporal Fourier transforms discussed in Sec. 6. The generalized diffusivity has also been calculated and a brief discussion is given of the Gaussian interpolation schemes[9,10] for this quantity.

The second moment is given exactly in our approximation, while the fourth moment differs from the exact value by 10–20%. Finally, the long-time behavior of the small-q correlation functions yields the spin diffusion coefficient. In several appendixes we give a somewhat more detailed discussion of the approximations involved.

2. GENERAL THEORY

It is our purpose to show how one may calculate approximately the correlation functions $C(t)=\langle S_i^z S_i^z(t)\rangle$ and $C_{\mathbf{q}}(t)=\langle S_{-\mathbf{q}}^z S_{\mathbf{q}}^z(t)\rangle$ of the Heisenberg model in the high-temperature limit $T\to\infty$. Here $\mathbf{S}_i(t)=e^{iH_0t}\mathbf{S}_i e^{-iH_0t}$ is the quantum-mechanical spin operator of length $[S(S+1)]^{1/2}$ at lattice site i with Cartesian components $S_i^x(t)$, $S_i^y(t)$, $S_i^z(t)$; we adopt the notational convention

[20] R. Kubo and T. Toyabe, in *Magnetic Resonance and Relaxation*, edited by R. Blinc (North-Holland, Amsterdam, 1967), p. 810.

[21] Of course, the assertion that this motion is a *Gaussian random process* does not imply that the spin correlation function is itself Gaussian. Indeed, as our calculations show, it is not; cf. P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. 25, 269 (1953).

of labeling lattice sites $i, j, k, \ldots$, and denote the position vector of site i by $\mathbf{R}_i$. $\mathbf{S}_\mathbf{q}(t)$ is the Fourier transformed spin

$$\mathbf{S}_\mathbf{q}(t)=N^{-1}\sum_i \mathbf{S}_i(t)e^{i\mathbf{q}\cdot\mathbf{R}_i}, \quad (1)$$

where the sum runs over the N lattice sites and $\mathbf{q}$ may be any wave vector; we will use the notation $\mathbf{S}_\mathbf{q}$, $\mathbf{S}_{\mathbf{q}'}$, $\mathbf{S}_{\mathbf{q}''}, \ldots$, for such operators. The average $\langle A\rangle$ of any operator is defined according to

$$\langle A\rangle=\mathrm{Tr}(Ae^{-\beta H_0})/\mathrm{Tr}(e^{-\beta H_0}), \quad (2)$$

where Tr denotes the trace over the Hilbert space of the spin system and $\beta=1/\kappa T$, $\kappa=$Boltzmann's constant. Throughout the remainder of the paper we take $\hbar=1$. The Hamiltonian of the Heisenberg model will be written

$$H_0=-\tfrac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j, \quad (3)$$

where J_{ij} is the exchange interaction of spins at sites i, j. We assume that $J_{ii}=0$, that $J_{ij}=J_{ji}$ depends only upon the difference $\mathbf{R}_i-\mathbf{R}_j$, and that $J_{ij}\to 0$ sufficiently quickly (e.g., exponentially) as $|\mathbf{R}_i-\mathbf{R}_j|\to\infty$. The Hamiltonian (3) and the spin commutation relations lead to the usual equation of motion

$$\dot{\mathbf{S}}_i=-\sum_j J_{ij}\mathbf{S}_j\times\mathbf{S}_i \quad (4)$$

for the $\mathbf{S}_i$.

Let the symbols $A, B, C, \ldots$, stand for either S_i^z or $S_\mathbf{q}^z$. We shall calculate the correlation functions $\langle AB(t)\rangle$ indirectly via the relaxation functions[22] defined by

$$\{A,B(t)\}=\int_0^\beta \langle e^{\lambda H_0}Ae^{-\lambda H_0}B(t)\rangle d\lambda, \quad (5)$$

where we have omitted the term $\langle A_0B_0\rangle$ (in Kubo's notation) which will vanish in the present application. In the limit $T\to\infty$, $\beta\to 0$, we deduce at once from (5) that

$$\beta\langle AB(t)\rangle=\{A,B(t)\}. \quad (6)$$

Dividing (6) by the corresponding equation taken at $t=0$ gives

$$\frac{\langle AB(t)\rangle}{\langle AB\rangle}=\frac{\{A,B(t)\}}{\{A,B\}}\equiv F_{AB}(t), \quad (7)$$

where $F_{AB}(t)$ is the "relaxation shape function." It is this function F_{AB} that we aim to calculate. The correlation functions may then be deduced from (7), since the averages $\langle AB\rangle$ are given in the high-temperature limit by $\langle S_i^zS_j^z\rangle=\frac{1}{3}S(S+1)\delta_{ij}$ and [using (1)] $\langle S_{-q}^zS_q^z\rangle=\frac{1}{3}N^{-1}S(S+1)$.

Thus we find

$$C(t)=\tfrac{1}{3}S(S+1)\frac{\{S_i^z,S_i^z(t)\}}{\{S_i^z,S_j^z\}}\equiv\tfrac{1}{3}S(S+1)F(t) \quad (8)$$

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

and

$$C_\mathbf{q}(t)=\tfrac{1}{3}N^{-1}S(S+1)\frac{\{S_{-\mathbf{q}}^z,S_\mathbf{q}^z(t)\}}{\{S_{-\mathbf{q}}^z,S_\mathbf{q}^z\}}\equiv\tfrac{1}{3}N^{-1}S(S+1)F_\mathbf{q}(t), \quad (9)$$

defining the functions $F(t)$, $F_\mathbf{q}(t)$.

We shall use an unfamiliar technique to calculate the functions $F_{AB}(t)$. From Eqs. (2.24b) and (3.3) of Ref. 22, one may deduce an alternative expression for the relaxation function, namely,

$$\{A,B(t)\}=\lim -i\int_{-\infty}^0 \langle[A,B(t-t')]\rangle e^{\epsilon t'}dt', \quad \text{as } \epsilon\to 0_+ \quad (10)$$

which can be rearranged into the form

$$\begin{aligned}\{A,B(t)\}&=\lim -i\int_{-\infty}^0 \langle[A(t'),B(t)]\rangle e^{\epsilon t'}dt', \quad \text{as } \epsilon\to 0_+\\ &=\langle[X,B(t)]\rangle\\ &=\langle\delta B(t)\rangle,\end{aligned} \quad (11)$$

where

$$X=\lim \frac{-}{i}\int_{-\infty}^0 A(t')e^{\epsilon t'}dt' \text{ as } \epsilon\to 0_+$$

and we define

$$\delta B(t)\equiv[X,B(t)]. \quad (12)$$

The operator $\delta B(t)$ has a simple physical interpretation. The relaxation function $\{A,B(t)\}$ describes how the variable B relaxes for $t>0$ when a perturbation proportional to A is switched on adiabatically between $t=-\infty$ and $t=0$ and switched off abruptly at $t=0$. In the absence of the perturbation A the operator B evolves in time in the usual way into $B(t)=e^{iH_0t}Be^{-iH_0t}$. However, the perturbation A disturbs this evolution and, as is shown in Appendix A, causes it to evolve instead into the operator $\hat{B}(t)=B(t)+\delta B(t)$ (for $t>0$). Thus $\delta B(t)$ represents the change in $B(t)$ (at positive t) due to the application of the perturbation A, and is the exact analog of the change of a classical dynamical variable due to such a perturbation.

From the definition (12) and the operator equations of motion, one may deduce (using the identity $[[A,B],C]+[[C,A],B]+[[B,C],A]=0$) that $\hat{B}(t)$ satisfies the equation of motion

$$i\frac{d}{dt}\hat{B}(t)=[\hat{B}(t),\hat{H}_0], \quad (13)$$

where $\hat{H}_0=H_0+\delta H_0\equiv H_0+[X,H_0]$ is the perturbed Hamiltonian operator. Equation (13) is simply the ordinary equation of motion for B with all operators $A(t)$ replaced by the corresponding circumflexed operators

$\hat{A}(t)$. For example, $\hat{\mathbf{S}}_i(t)$ obeys the equation of motion

$$\frac{d}{dt}\hat{\mathbf{S}}_i(t) = -\sum_j J_{ij}\hat{\mathbf{S}}_j(t)\times\hat{\mathbf{S}}_i(t). \tag{14}$$

One may now combine (8) and (11) to write

$$F(t) = \langle\delta S_i^z(t)\rangle/\langle\delta S_i^z\rangle, \tag{15}$$

where δS_i^z is due to a perturbation proportional to S_i^z acting up to $t=0$, and, similarly,

$$F_{\mathbf{q}}(t) = \langle\delta S_{\mathbf{q}}^z(t)\rangle/\langle\delta S_{\mathbf{q}}^z\rangle, \tag{16}$$

where $\delta S_{\mathbf{q}}^z(t)$ is due to a perturbation proportional to $S_{-\mathbf{q}}^z$ acting up to $t=0$. Our program of calculation is to estimate the ratios in (15) and (16), and therefore the functions F and $F_{\mathbf{q}}$, and thence C and $C_{\mathbf{q}}$ via (8) and (9).

3. APPROXIMATION FOR $\langle S_i^z(0)S_i^z(t)\rangle$

We begin by developing an analytic approximation for $C(t)$. This approximation has the disadvantage of being valid only for short times, but the mathematical expression of the physical picture described in Sec. 1 is most clearly seen in this treatment. Following our program, we imagine a magnetic field in the z direction to have been applied to a single spin (say the ith) up to $t=0$ and ask how $\langle\delta S_i^z(t)\rangle$ evolves for $t>0$. To do this, we use the equation of motion

$$\delta\dot{\mathbf{S}}_i(t) = -\sum_j J_{ij}\mathbf{S}_j(t)\times\delta\mathbf{S}_i(t) - \sum_j J_{ij}\delta\mathbf{S}_j(t)\times\mathbf{S}_i(t) \tag{17}$$

for $\delta\mathbf{S}_i$ derived from (4) and (14). Since the perturbing field was applied to the ith spin alone, one knows that in the $T\to\infty$ limit $\langle\delta\mathbf{S}_j\rangle=0$ for $j\neq i$, that is, $\delta\mathbf{S}_j(j\neq i)$ vanishes on the average at $t=0$. This suggests that at short times one might neglect the term in $\delta\mathbf{S}_j(t)$ on the right-hand side of (17) compared to the other; this neglect will be referred to as approximation (A). One would expect this approximation to be good at times t for which $\langle\delta S_j^z(t)\rangle \ll \langle\delta S_i^z(t)\rangle$. However, from the symmetry of the situation and the conservation of spin, one has $\langle\delta S_j^z(t)\rangle \lesssim \langle\delta S_i^z\rangle/z_1$, where z_1 is the number of nearest neighbors. Thus the approximation (A) should be good for times t for which $\langle\delta S_i^z(t)\rangle \gg \langle\delta S_i^z\rangle/z_1$, i.e., for which $F(t)\gg 1/z_1$, i.e., for $t\lesssim t_c$ where $F(t_c)\simeq 1/z_1$. We estimate below $t_c\sim 2/S\hat{J}$, where

$$\hat{J}^2 = \sum_{ij} J_{ij}^2. \tag{18}$$

[For nearest-neighbor interactions of strength J, one has $\hat{J}=(\sqrt{z_1})J$.] This expectation is, in fact, borne out by comparison of the results obtained here with the more elaborate calculations of Secs. 5 and 6, which do not involve an approximation of the kind (A). We therefore make the approximation (A) and expect the theory to be good for $t<t_c$; t_c will be estimated below. Later (in Secs. 5 and 6) we shall show how it is possible to dispense with this kind of approximation.

With the use of approximation (A), Eq. (17) can be written

$$\delta\dot{\mathbf{S}}_i(t) = \mathbf{h}(t)\times\delta\mathbf{S}_i(t), \tag{19}$$

where $\mathbf{h}(t)$ may be thought of as an "effective magnetic field" acting on the ith spin given by

$$\mathbf{h}(t) = -\sum_j J_{ij}\mathbf{S}_j(t). \tag{20}$$

To solve (20), we formally rewrite (19) (following Kubo[23])

$$\delta\dot{\mathbf{S}}_i = ih^\times(t)\mathbf{S}_i(t), \tag{21}$$

where $ih^\times(t)$ is a linear "superoperator" (by which we mean an entity which, acting on a quantum-mechanical operator, turns it into another operator) defined by

$$ih^\times(t)A \equiv \mathbf{h}(t)\times\mathbf{A}, \tag{22}$$

where A is any vector operator. The factor i has been included in the definition so that $h^\times$ will be Hermitian. Written in the form (21) the equation may be solved in the usual way to give

$$\delta\mathbf{S}_i(t) = \exp_+\left\{i\int_0^t h^\times(t')dt'\right\}\delta\mathbf{S}_i(0), \tag{23}$$

where $\exp_+$ means the time-ordered exponential. Using this expression in (15) gives

$$F(t) = \left\langle\left[\exp_+\left\{i\int_0^t h^\times(t')dt'\right\}\delta\mathbf{S}_i\right]_z\right\rangle \Big/ \langle\delta S_i^z\rangle, \tag{24}$$

where $[\ ]_z$ means the z component of the operator in $[\ ]$. This formula may be thought of as describing how the perturbation $\delta\mathbf{S}_i$ is relaxed away by the action on $\mathbf{S}_i$ of the effective fluctuating field $\mathbf{h}(t)$ due to the other spins.

The right-hand side of (24) cannot be calculated exactly and further approximations are necessary. In this and the next section, two different lines of approximation will be described. Here we adopt the simpler of the two which will lead to an analytic approximation for $F(t)$.

Let us define an averaging operation $\langle\langle A\rangle\rangle$ for superoperators according to

$$\langle\langle A\rangle\rangle = \langle[A\,\delta\mathbf{S}_i]_z\rangle/\langle\delta S_i^z\rangle. \tag{25}$$

We note that $\langle\langle 1\rangle\rangle=1$ and that $\langle\langle\cdots\rangle\rangle$ is linear, confirming that $\langle\langle\cdots\rangle\rangle$ is a proper averaging operation and that (24) can then be written

$$F(t) = \left\langle\left\langle\exp_+\left\{i\int_0^t h^\times(t')dt'\right\}\right\rangle\right\rangle. \tag{26}$$

[23] R. Kubo, in *Fluctuation, Relaxation and Resonance in Magnetic Systems*, edited by D. ter Haar (Oliver and Boyd, Edinburgh, 1962), p. 23.

To evaluate (approximately) the right-hand side, we will use the cumulant expansion theory of Kubo,[24] transforming it to

$$F(t)=\exp\left\{i\int_0^t \langle\langle h^\times(t')\rangle\rangle dt' + \tfrac{1}{2}i^2\int_0^t dt_1\int_0^t dt_2\langle\langle(h^\times(t_1)h^\times(t_2))_+\rangle\rangle_c+\cdots\right\}, \quad (27)$$

where $(A(t_1)A(t_2)\cdots)_+$ means the time-ordered product of the operators (or superoperators), the cumulant averages $\langle\langle\cdots\rangle\rangle_c$ are defined in the usual way,[24] e.g.,

$$\langle\langle(h^\times(t_1)h^\times(t_2))_+\rangle\rangle_c = \langle\langle(h^\times(t_1)h^\times(t_2))_+\rangle\rangle - \langle\langle h^\times(t_1)\rangle\rangle\langle\langle h^\times(t_2)\rangle\rangle, \quad (28)$$

and the exponent is the usual cumulant expansion.

The derivation of (27) has involved only the approximation (A). To evaluate the exponent, however, further approximations are needed, since [using (20) and (25)] we see that the terms of the cumulant series involve spin averages of all orders. The kind of approximation we use is motivated by noticing that according to (20) the effective field $\mathbf{h}(t)$ is a superposition of contributions from the z_1 neighboring spins. If these z_1 contributions were statistically independent then each (nonvanishing) term in the cumulant series would be smaller by a factor $1/z_1$ than its predecessor (in the limit $z_1\to\infty$ this would be equivalent to the central limit theorem). In practice we do not expect the z_1 neighboring spins to be very strongly correlated at high temperatures, so the z_1 contributions to $\mathbf{h}$ are nearly independent. Furthermore z_1 is a moderately large number ($z_1\sim 6$–12). Thus we may expect the cumulant series to be fairly rapidly convergent and will therefore approximate it by its first two terms [those written explicitly in (27)], neglecting the third- and higher-order cumulants; we shall refer to this as approximation (B′).

We are still faced with the problem of evaluating the averages $\langle\langle h^\times(t')\rangle\rangle$ and $\langle\langle h^\times(t_1)h^\times(t_2)\rangle\rangle_c$. To do this we shall make use of a rather general approximation principle [approximation (B)], namely we neglect the *detailed* correlations of three or more spin operators; or, to be more precise, we assume that the cumulant averages $\langle S_1S_2S_3\cdots\rangle_c$ of three or more spins are negligible. This is the basic approximation used in this paper. All other approximations can be either avoided or are readily refined. It is really a more general statement of the considerations underlying approximation (B′) discussed above. To the extent that the operators S_1, S_2, ..., in $\langle S_1S_2\cdots\rangle_c$ refer to different sites, it reflects the notion that correlations between spins at different sites are not too strong. On the other hand, examination of the structure of the cumulant series shows that it only involves the neglect of correlations of spins at the same site in terms which are already small by a factor $1/z_1$ (relative to the leading terms of the cumulant expansion).

The approximation (B) is very powerful. Approximation (B′) above is to a considerable extent a corollary of (B). In fact, in the theory of Secs. 4 and 5, where use is made of approximation (B′) [in the context of a definition of average that is different from (25)], one can show (see Appendix B) that (B′) is a consequence of (B). In the context of the average definition (25) used here, however, (B′) involves not only (B), but also an additional kind of approximation, and so must be introduced as a separate assumption. This point is discussed briefly in Appendix C. Approximation (B) also has two other consequences which are of use here. The way in which they follow from (B) is discussed in Appendix B. These consequences are as follows. Approximation (B″): Any average $\langle S_1S_2\cdots S_n\delta S\rangle$ may be approximated as $\langle S_1S_2\cdots S_n\rangle\langle\delta S\rangle$. Approximation (B‴): Any average of an odd number of factors $\langle\langle h_1h_2\cdots h_n\rangle\rangle$ may be neglected.

The use of approximations (B′) and (B‴) in (27) now leads to

$$F(t)=\exp\left\{i^2\int_0^t dt_1\int_0^{t_1} dt_2\langle\langle h^\times(t_1)h^\times(t_2)\rangle\rangle\right\}, \quad (29)$$

expressing the relaxation F approximately in terms of the correlation function of the random field $\mathbf{h}$. We now evaluate the correlation function $\langle\langle h^x(t_1)h^x(t_2)\rangle\rangle$ in terms of F returning to the definitions (22) and (25) which give

$$\begin{aligned} i^2\langle\langle h^\times(t_1)h^\times(t_2)\rangle\rangle &= -\langle[h^\times(t_1)h^\times(t_2)\delta\mathbf{S}_i]_z\rangle/\langle\delta S_i^z\rangle\\ &= \langle[\mathbf{h}(t_1)\times(\mathbf{h}(t_2)\times\delta\mathbf{S}_i)]_z\rangle/\langle\delta S_i^z\rangle\\ &= \epsilon^{z\mu\nu}\epsilon^{\nu\sigma\tau}\langle h^\mu(t_1)h^\sigma(t_2)\delta S_i^\tau\rangle/\langle\delta S_i^z\rangle\\ &= (\delta_{z\sigma}\delta_{\mu\tau}-\delta_{z\tau}\delta_{\mu\sigma})\langle h^\mu(t_1)h^\sigma(t_2)\rangle\langle\delta S_i^\tau\rangle/\langle\delta S_i^z\rangle, \end{aligned} \quad (30)$$

where the last line follows by the application of approximation (B″). The $\epsilon^{\nu\sigma\tau}$ are the usual permutation symbols and we have adopted the summation convention that any repeated Greek superscript is to be summed over x, y, and z. From consideration of rotation symmetry in spin space [and the definition (20)], it follows that $\langle h^\mu(t_1)h^\sigma(t_2)\rangle=\delta_{\mu\sigma}\langle h^z(t_1)h^z(t_2)\rangle$. Using this result and (20) one obtains

$$\begin{aligned} i^2\langle\langle h^x(t_1)h^x(t_2)\rangle\rangle &= -2\langle h^z(t_1)h^z(t_2)\rangle\\ &= -2\sum_{jk} J_{ij}J_{ik}\langle S_j^z(t_1)S_k^z(t_2)\rangle. \end{aligned} \quad (31)$$

We now introduce our final approximation (A′), closely related to (A). The present theory is only useful for times $t<t_c$, for which $z_1\langle\delta S_j^z(t)\rangle\ll\langle\delta S_i^z(t)\rangle$, which through (6) and (11) implies $z_1\langle S_j^z(t)S_i^z\rangle\ll\langle S_i^z(t)S_i^z\rangle$. In this time range, therefore, the $j\neq k$ terms in the sum in (31) are small compared to the $j=k$ terms and may

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

be neglected. This gives [using (8) and (18)]

$$\langle h^z(t_1)h^z(t_2)\rangle = \sum_j J_{ij}^2 C(t_1-t_2)$$
$$= \tfrac{1}{3}S(S+1)\hat{J}^2F(t_1-t_2)\,, \qquad (32)$$

and, from (31),

$$i^2\langle\langle h^\times(t_1)h^\times(t)\rangle\rangle = -\tfrac{2}{3}S(S+1)\hat{J}^2F(t_1-t_2),$$

so that (29) reduces to the equation

$$F(t)=\exp\left\{-\tfrac{2}{3}S(S+1)\hat{J}^2\int_0^t dt_1\int_0^{t_1} dt_2 F(t_1-t_2)\right\}, \qquad (33)$$

or, introducing the reduced time variable

$$\tau = [\tfrac{2}{3}S(S+1)]^{1/2}\hat{J}t, \qquad (34)$$

and rearranging a little,

$$F(\tau)=\exp\left\{-\int_0^\tau (\tau-\tau')F(\tau')d\tau'\right\}, \qquad (35)$$

which is the self-consistent equation for $F(\tau)$ that was sought.

The self-consistency has entered because F was determined by the relaxation of $\delta\mathbf{S}_i$, which was in turn determined by the fluctuating field $\mathbf{h}(t)$, the statistics of which depend upon $C(t_1-t_2)$, and these on F again via (8). The self-consistent equation (35) can be quite easily solved by differentiating twice. This leads to the differential equation

$$\ddot{F}=\dot{F}^2/F-F^2, \qquad (36)$$

with the solution

$$C(\tau)/\tfrac{1}{3}S(S+1)=F(\tau)=\operatorname{sech}^2(\tau/\sqrt{2})\,, \qquad (37)$$

satisfying the boundary condition $F(0)=1$, $\dot{F}(0)=0$. This is, in fact, our simple analytic solution for $C(\tau)$ valid for $\tau<\tau_c\approx[\tfrac{2}{3}S(S+1)]^{1/2}\hat{J}t_c$.

For small τ our solution (37) has the expansion

$$F(\tau)=1-\tfrac{1}{2}\tau^2+\tfrac{1}{6}\tau^4+O(\tau^6)\,, \qquad (38)$$

which should be compared with the exact result (derived from Collins and Marshall[6]) for the nearest-neighbor interaction case

$$F(\tau)=1-\tfrac{1}{2}\tau^2 + \frac{7}{48}\tau^4\left[1-\frac{1}{7z_1}\left\{4+2z_2+\frac{3}{2S(S+1)}\right\}\right], \qquad (39)$$

where z_1 is the number of nearest neighbors and z_2 is the number of nearest neighbors of a site i that are also nearest neighbors of a given nearest neighbor j of i ($z_1=6$, $z_2=0$ for simple cubic (sc), $z_1=8$, $z_2=0$ for bcc, $z_1=12$, $z_2=4$ for fcc). In practice, the expression in square brackets is always close to 1, so our approximation (38) has the correct second derivative at $\tau=0$ and a fourth derivative accurate to within $\sim 5\%$.

For large τ the solution (37) is proportional to $\exp(-\tau\sqrt{2})$, but this lies outside its region of validity $\tau<\tau_c$. In fact, we expect that for large τ the correlation function obeys a diffusion law $\langle S_i^zS_j^z(\tau)\rangle \propto \tau^{-3/2}\times\exp[-(\mathbf{R}_i-\mathbf{R}_j)^2/4D\tau]$, so $F(\tau)\propto\tau^{-3/2}$. We may estimate the range of validity $\tau<\tau_c$ from the equation $F(\tau_c)\simeq 1/z_1$, which gives $\tau_c\simeq 2$ using (37). In fact, in Fig. 1 the solution (37) is compared with other approximations derived in this paper and with the computer simulation experiments of Windsor[18] for the sc case and is seen to be tolerably good out to $\tau\sim 3$.

We finally note that in the limit $T\to\infty$, $C(t)$ is an even function of t given by $C(t)=\tfrac{1}{3}S(S+1)F(|t|)$. Its Fourier transform is therefore given by

$$\hat{C}(\Omega)=\tfrac{2}{3}S(S+1)\int_0^\infty F(t)\cos\Omega t\,dt$$
$$=[\tfrac{2}{3}S(S+1)]^{1/2}\hat{F}(\omega)/\hat{J}\,, \qquad (40)$$

where ω is the scaled frequency

$$\omega=\frac{\Omega}{[\tfrac{2}{3}S(S+1)]^{1/2}\hat{J}} \qquad (41)$$

and

$$\hat{F}(\omega)=\int_0^\infty F(\tau)\cos\omega\tau\,d\tau \qquad (42)$$

$$=\frac{\pi\omega}{\sinh(\pi\omega/\sqrt{2})} \qquad (43)$$

using (37). This result is compared in Fig. 2 with the more accurate calculations of Sec. 6 and with the corre-

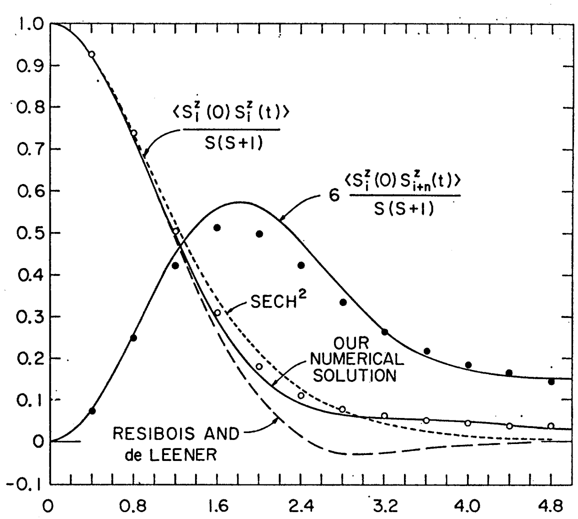

FIG. 1. Autocorrelation function and the nearest-neighbor correlation function for a simple cubic lattice with nearest-neighbor exchange. The circles represent Windsor's computer simulation data. The analytical approximation $\operatorname{sech}^2\tau/\sqrt{2}$ and the Résibois and DeLeener result are seen to be reasonable approximations at short times. The scaled time τ is defined in Eq. (34).

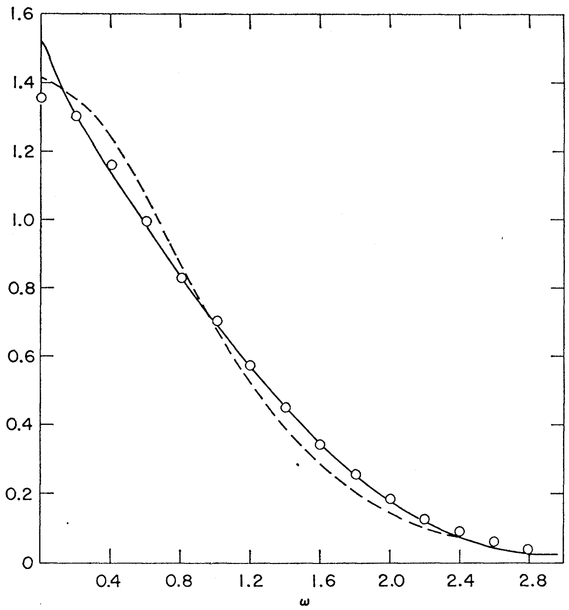

FIG. 2. Fourier transform $\hat{F}(\omega)$ of the autocorrelation function. The solid line is our numerical result, while the dashed line is the analytical approximation Eq. (37). The open circles are Windsor's computer results. The scaled frequency ω is defined in Eq. (41).

sponding result obtained in the computer simulation calculation of Windsor,[18] and is seen to be in fair agreement with both. It is of interest to note that for large ω, (43) becomes proportional to $\omega\exp(-\pi\omega/\sqrt{2})$, rather than a Gaussian form. In fact $\hat{C}(\Omega)$ is a quantity which is accessible to experiment, since it gives the shape (as a function of energy loss) of the inelastic neutron scattering cross-section curve for polycrystalline materials at large momentum transfer, and Windsor has shown that his calculation of $\hat{S}(\omega)$ [and therefore our approximation (43) which agrees with it] is in fair agreement with his experiments[13] on $RbMnF_3$.

4. RÉSIBOIS-DELEENER EQUATION FOR $C(t)$

Résibois and DeLeener,[7,8] using a special technique, derived an integrodifferential equation for $C(t)$ [the $\Gamma(t)$ of their notation is our $F(t)$]. We will next show how their equation may be derived by a modification of the approximation procedure of Sec. 3. It will be found that its derivation also involves approximations (A) and (A′), so their theory is also valid only for $\tau<\tau_c$. Furthermore, although the approximation procedure used in this section is more sophisticated in principle than that of Sec. 3, in practice the solution does not differ very much from that of Sec. 3 in the range $\tau<\tau_c$.

Our starting point will be Eq. (24), whose derivation has already made use of approximation (A). It will be convenient to rewrite (22) in the form

$$ih^{\times}(t)\mathbf{A}=h^{\mu}(t)\epsilon^{\mu}\mathbf{A}\quad\text{or}\quad[ih^{\times}(t)\mathbf{A}]_{\alpha}=h^{\mu}(t)\epsilon^{\alpha\mu\beta}A^{\beta}(t)\,,\tag{44}$$

where ϵ^{μ} is a tensor with elements $(\epsilon^{\mu})^{\alpha\beta}=\epsilon^{\alpha\mu\beta}$ acting on vector operators. Thus one can think of $h^{\times}(t)$ as a tensor operator in spin space [it is also, of course, an operator in Hilbert space, since $h^{\mu}(t)$ depends upon the S_j according to the definition (20)]. Similarly $\exp_{+}\{i\int_0^t h^{\times}(t')dt'\}$ can be thought of as a tensor in spin space (and an operator in Hilbert space), and (24) can be written out in component form

$$F(t)=\left\langle\left(\exp_{+}\left\{i\int_0^t h^{\times}(t')dt'\right\}\right)^{z\alpha}\delta S_i^{\alpha}\right\rangle\Big/\langle\delta S_i^{z}\rangle\,,\tag{45}$$

where $(\cdots)^{\alpha\beta}$ means the α,β component of the tensor $(\cdots)$. If we now apply approximation (B″) (see Sec. 3, and Appendix B) directly to this expression, we get

$$F(t)=\left\langle\left(\exp_{+}\left\{i\int_0^t h^{\times}(t')dt'\right\}\right)^{z\alpha}\right\rangle\langle\delta S_i^{\alpha}\rangle/\langle\delta S_i^{z}\rangle\,.\tag{46}$$

However, from rotational symmetry in spin space, one has $\langle\delta S_i^{\alpha}\rangle=0$ unless $z=\alpha$ (since the applied field leading to $\delta\mathbf{S}_i$ was proportional to S_i^z), so that (46) gives

$$F(t)=\left\langle\left(\exp_{+}\left\{i\int_0^t h^{\times}(t')dt'\right\}\right)^{zz}\right\rangle.\tag{47}$$

To evaluate the right-hand side of (47), we now regard $\langle\cdots\rangle$ as an averaging operation and apply the cumulant expansion to obtain

$$F(t)=\left(\exp_{+}\left\{i\int_0^t\langle h^{\times}(t')\rangle dt'+\tfrac{1}{2}i^2\int_0^t dt_1\int_0^t dt_2\langle h^{\times}(t_1)h^{\times}(t_2)\rangle_c+\cdots\right\}\right)^{zz},\tag{48}$$

where the exponential is still time-ordered since the terms in the exponent are (noncommuting) tensors in spin space; for example, according to (44),

$$\langle h^{\times}(t_1)h^{\times}(t_2)\rangle=\langle h^{\mu}(t_1)h^{\nu}(t_2)\rangle\epsilon^{\mu}\epsilon^{\nu}\tag{49}$$

involves the noncommuting ϵ^{μ}.

We next use approximation (B′) to neglect all but the first two terms in (48). In this case (see Appendix B), as opposed to that discussed in Sec. 3, approximation (B′) follows solely from the principle of neglecting the detailed correlation between three or more spins. One again finds $\langle h^{\times}(t)\rangle=0$, so that

$$\begin{aligned}F(t)&=\left(\exp_{+}\left\{\tfrac{1}{2}i^2\int_0^t dt_1\int_0^t dt_2\langle h^{\times}(t_1)h^{\times}(t_2)\rangle\right\}\right)^{zz}\\&=\left(\exp_{+}\left\{\int_0^t dt_1\int_0^{t_1}dt_2\langle h^{z}(t_1)h^{z}(t_2)\rangle\epsilon_{t_1}{}^{\mu}\epsilon_{t_2}{}^{\mu}\right\}\right)^{zz},\end{aligned}\tag{50}$$

where the second line follows by the use of (44) and observing that $\langle h^{\mu}(t_1)h^{\nu}(t_2)\rangle=\delta_{\mu\nu}\langle h^{z}(t_1)h^{z}(t_2)\rangle$;

$\epsilon_t{}^\mu = \epsilon^\mu$, defined after (44), the subscript t only serving to indicate where the tensor is to be inserted in the time-ordered product. Since the present theory, like that of Sec. 3, is supposed valid only for $\tau < \tau_c$, we can again use approximation (A′) which yields the expression (32) for $\langle h^z(t_1)h^z(t_2)\rangle$. Using this in (50), and changing to the reduced time variable τ according to (34), gives

$$F(\tau)=\left(\exp_+\left\{\frac{1}{2}\int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2 \times F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\mu\epsilon_{\tau_2}{}^\mu\right\}\right)^{zz}. \quad (51)$$

If in this expression one neglected the time ordering of the $\epsilon_\tau{}^\mu$ operators and simply wrote $\epsilon_{\tau_1}{}^\mu\epsilon_{\tau_2}{}^\mu = \epsilon^\mu\epsilon^\mu = -2$, then (51) would reduce to (35) and we would reproduce the theory of Sec. 3; the relation between the theory of Sec. 3 and (51) is discussed briefly in Appendix C. To the extent that the time ordering in (51) is taken seriously in this section, we may regard it as more accurate than the theory of Sec. 3. On the other hand, as shown in Appendix C, at short times $\tau<\tau_c$ the time ordering in (51) is not a very important effect, so we would not expect to obtain much difference between the solution of (51) and (35) for $\tau<\tau_c$. For large τ, the method of this section would be superior, but the theory developed here is invalid for large τ because of approximations (A) and (A′).

We cannot evaluate the time-ordered exponential in (51) exactly, but will derive an approximate integro-differential equation for it. To this end we differentiate (51) to obtain

$$\dot F(\tau)=\frac{1}{2}\int_0^\tau d\tau' F(\tau-\tau')\epsilon^\mu\left(\epsilon_{\tau'}{}^\mu \exp\left\{\frac{1}{2}\int_0^\tau d\tau_1 \times\int_0^{\tau_1} d\tau_2 F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\nu\epsilon_{\tau_2}{}^\nu\right\}\right)_+^{zz}, \quad (52)$$

where $(\cdots)_+{}^{zz}$ means the zz component of the tensor given by the time-ordered product $(\;)_+$, and introduce a new approximation (C) to evaluate the time-ordered product, namely,

$$\begin{aligned}\left(\epsilon_{\tau'}{}^\mu \exp\left\{\frac{1}{2}\int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2 F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\nu\epsilon_{\tau_2}{}^\nu\right\}\right)_+ &\simeq \exp_+\left\{\frac{1}{2}\int_{\tau'}^\tau d\tau_1\int_{\tau'}^{\tau_1} d\tau_2 F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\nu\epsilon_{\tau_2}{}^\nu\right\}\epsilon^\mu \\ &\quad\times \exp_+\left\{\frac{1}{2}\int_0^{\tau'} d\tau_1\int_0^{\tau_1} d\tau_2 F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\sigma\epsilon_{\tau_2}{}^\sigma\right\} \\ &= F(\tau-\tau')\epsilon^\mu F(\tau'), \end{aligned} \quad (53)$$

where the last line follows by using the rotational symmetry in spin space to infer that

$$\begin{aligned}\left(\exp_+\left\{\frac{1}{2}\int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2 F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\nu\epsilon_{\tau_2}{}^\nu\right\}\right)^{\alpha\beta} &= \delta_{\alpha\beta}\left(\exp_+\left\{\frac{1}{2}\int^\tau d\tau_1\int^{\tau_1} F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^\nu\epsilon_{\tau_2}{}^\nu\right\}\right)^{zz} \\ &= \delta_{\alpha\beta}F(\tau).\end{aligned}$$

The approximation leading the second line of (53) is discussed in Appendix D, where it is argued that it should not lead to very large errors. Using (53) in (52) now gives

$$\dot F(\tau) = -\int_0^\tau F^2(\tau-\tau')F(\tau')d\tau', \quad (54)$$

which is the equation derived by Résibois and DeLeener.

The solution of (54) has been evaluated numerically by its originators and is plotted in Fig. 1 for comparison with that of Sec. 6. We see that they all agree fairly well for $\tau<\tau_c\simeq 2$. Thus one finds that (54) is only valid in the region $\tau<\tau_c$ and does not differ too much from the solution $\mathrm{sech}^2(\tau/\sqrt{2})$ of Sec. 3 in this range. For small τ, Eq. (54) gives the expansion

$$F(\tau) = 1-\tfrac{1}{2}\tau^2+\tfrac{1}{8}\tau^4+O(\tau^6),$$

which should be compared with (38) and (39).

5. APPROXIMATE EQUATION FOR $C_{\mathbf{q}}(t)$

We now apply the methods used in the Sec. 4 for the study of $C(t)$ to calculate the $C_{\mathbf{q}}(t)$. This application is more general than that discussed above, since a knowledge of the $C_{\mathbf{q}}(t)$ determines not only $C(t)$, but also the correlation function $\langle S_i{}^z S_j{}^z(t)\rangle$ for $i\neq j$. Furthermore, within this more general framework one can avoid the use of the approximations (A) and (A′), which limited the validity of the above calculations to the region $\tau<\tau_c$, so one may hope to obtain a theory valid for all τ.

We proceed as was outlined in Sec. 2, aiming to calculate $F_{\mathbf{q}}(t)$ as the ratio in (16) and using (9) to get $C_{\mathbf{q}}(\tau)$. From (1) and (4) one finds that $\mathbf{S}_{\mathbf{q}}(t)$ obeys the equation of motion

$$\dot{\mathbf{S}}_{\mathbf{q}}(t) = -\sum_{\mathbf{q}'} J_{\mathbf{q}'}\mathbf{S}_{\mathbf{q}'}\times\mathbf{S}_{\mathbf{q}-\mathbf{q}'}, \quad (55)$$

where the $\mathbf{q}$ sum is over all the N wave vectors in the first Brillouin zone (as are all wave-vector sums in this paper) and

$$J_{\mathbf{q}} = \sum_j J_{ij} e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_i)}. \quad (56)$$

We suppose (see Sec. 2) that the system has been prepared by the application of the time-independent but spatially varying magnetic field $h_{\mathbf{q}}{}^z e^{i\mathbf{q}\cdot\mathbf{r}}$ of wave vector $\mathbf{q}$ for $t<0$. The perturbed operator $\hat{\mathbf{S}}_{\mathbf{q}'}(t)$ obeys an equation similar to (55), and the difference $\delta\mathbf{S}_{\mathbf{q}'}$ is found, after a little rearrangement, to obey the equation

$$\delta\dot{\mathbf{S}}_{\mathbf{q}'}=\sum_{\mathbf{q}''} J_{\mathbf{q}'\mathbf{q}''}\mathbf{S}_{\mathbf{q}'-\mathbf{q}''}\times\delta\mathbf{S}_{\mathbf{q}''}\,, \tag{57}$$

where

$$J_{\mathbf{q}'\mathbf{q}''}=-J_{\mathbf{q}'-\mathbf{q}''}+J_{\mathbf{q}''}.$$

$\delta\mathbf{S}_{\mathbf{q}'}$ is the change in the operator $\mathbf{S}_{\mathbf{q}'}$ due to the application of the perturbation of wave vector $\mathbf{q}$. We do not indicate the special wave vector $\mathbf{q}$ in the notation, but it should be kept in mind.

We now introduce the column vector δS whose components are the spin operators $\delta S_{\mathbf{q}'}{}^{\alpha'}$, where $\alpha'=x$, y, and z and $\mathbf{q}'$ is any wave vector in the first Brillouin zone. Written as a row vector, the $3N$ components are $\delta S\equiv(\delta S_{\mathbf{q}_1}{}^x,\delta S_{\mathbf{q}_1}{}^y,\ldots,\delta S_{\mathbf{q}N}{}^y\delta S_{\mathbf{q}N}{}^z)$. With this notation the equations of motion (58) can be written as

$$\delta\dot{S}(t)=i\mathfrak{h}(t)\delta S(t) \tag{58}$$

or

$$\delta S_{\mathbf{q}'}{}^{\alpha'}(t)=\sum_{\mathbf{q}''} i\mathfrak{h}_{\mathbf{q}'\mathbf{q}''}{}^{\alpha'\alpha''}(t)\delta S_{\mathbf{q}''}{}^{\alpha''}(t)\,, \tag{59}$$

where $\mathfrak{h}(t)$ is a linear superoperator with matrix elements

$$i\mathfrak{h}_{\mathbf{q}'\mathbf{q}''}{}^{\alpha'\alpha''}(t)=J_{\mathbf{q}'\mathbf{q}''}S_{\mathbf{q}'-\mathbf{q}''}{}^{\mu}(t)\epsilon^{\alpha'\mu\alpha''}. \tag{60}$$

$\mathfrak{h}(t)$ is both a linear operator in spin and $\mathbf{q}$ space, as implied by (59), and an operator in Hilbert space, as implied by (60). Comparison of (59) with (21) shows that $\mathfrak{h}(t)$ may be thought of as a "generalized magnetic field" acting on the "generalized spin" δS.

We wish to calculate the average $\langle\delta S_{\mathbf{q}}{}^z(t)\rangle$. Proceeding as in Sec. 3, we obtain from (59) the analog of (24), i.e.,

$$F_{\mathbf{q}}(t)=\frac{\langle\delta S_{\mathbf{q}}{}^z(t)\rangle}{\langle\delta S_{\mathbf{q}}{}^z(0)\rangle}=\left\langle\left(\exp_+\left(i\int_0^t\mathfrak{h}(t')dt'\right)\delta S(0)\right)_{\mathbf{q}}^{z}\right\rangle\Big/\langle\delta S_{\mathbf{q}}{}^z(0)\rangle, \tag{61}$$

when $(\cdots)_{\mathbf{q}}{}^z$ means the $z,\mathbf{q}$ component of the generalized spin vector $(\cdots)$.

Writing out the components, this becomes

$$F_{\mathbf{q}}(t)=\sum_{\alpha'\mathbf{q}'}\left\langle\left(\exp_+\left(i\int_0^t\mathfrak{h}(t')dt'\right)\right)_{\mathbf{q}\mathbf{q}'}^{z\alpha'}\delta S_{\mathbf{q}'}{}^{\alpha'}(0)\right\rangle\Big/\langle\delta S_{\mathbf{q}}{}^z(0)\rangle, \tag{62}$$

where

$$\left(\exp_+\left(i\int_0^t\mathfrak{h}(t')dt'\right)\right)_{\mathbf{q}\mathbf{q}'}^{z\alpha'}$$

is the $\mathbf{q}z-\mathbf{q}'\alpha'$ matrix element of the superoperator.

At this point we have the choice of the alternative approximation procedures of Secs. 3 and 4. In fact, as discussed in Sec. 4, for small t it makes little difference which procedure we use. However, we are now seeking a theory valid for large t also, in which case the procedure of Sec. 4 is expected to be more accurate and is adopted here. [In fact, the application of the method of Sec. 3 to (62) was studied and was found to be much less accurate at large t.]

The first step is to use approximation (B″), which again follows from (B) (see Appendix B). This involves the decoupling of $\delta S_{\mathbf{q}'}{}^{\alpha'}(0)$ in (62), yielding the analog of (47),

$$F_{\mathbf{q}}(t)\approx\sum_{\alpha'\mathbf{q}'}\left\langle\left(\exp_+\left(i\int_0^t\mathfrak{h}(t')dt'\right)\right)_{\mathbf{q}\mathbf{q}'}^{z\alpha'}\right\rangle\langle\delta S_{\mathbf{q}'}{}^{\alpha'}(0)\rangle\Big/\langle\delta S_{\mathbf{q}}{}^z(0)\rangle. \tag{63}$$

The rotational and translational symmetry of the system now requires that $\langle\delta S_{\mathbf{q}'}{}^{\alpha'}(0)\rangle=0$, unless $\mathbf{q}'=\mathbf{q}$ and $\alpha'=z$, so that we have

$$F_{\mathbf{q}}(t)\approx\left\langle\left(\exp_+ i\int_0^t\mathfrak{h}(t')dt'\right)_{\mathbf{q}\mathbf{q}}^{zz}\right\rangle. \tag{64}$$

We now make the cumulant expansion, applying approximation (B′) to neglect all but the first two terms, and since $\langle\mathfrak{h}(t)\rangle=0$ again, we have

$$F_{\mathbf{q}}(t)\approx\left(\exp_+\left(-\int_0^t dt'\int_0^{t'}dt''\langle\mathfrak{h}(t)\mathfrak{h}(t')\rangle\right)\right)_{\mathbf{q}\mathbf{q}}^{zz}. \tag{65}$$

An approximate equation for $F_{\mathbf{q}}(t)$ analogous to (54) is obtained by differentiating (65) which, with due care to the time ordering, gives

$$\dot{F}_{\mathbf{q}}(t)=-\int_0^t dt'\left(\langle\mathfrak{h}(t)\mathfrak{h}(t')\rangle\times\exp\left(-\int_0^t dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\right)\right)_{+\mathbf{q}\mathbf{q}}^{zz}. \tag{66}$$

We now apply approximation (C) to the time-ordered product [cf. (53) and Appendix II], and write

$$\exp\left(-\int_0^t dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\right)\approx\exp\left(-\int_{t'}^t dt_1\int_{t'}^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\right)\times\exp\left(-\int_0^{t'}dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\right), \tag{67}$$

giving

$$\dot{F}_{\mathbf{q}}(t)=-\int_0^t dt'\times\left[\left\langle\mathfrak{h}(t)\exp_+\left(-\int_{t'}^t dt_1\int_{t'}^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\right)\mathfrak{h}(t')\right\rangle\times\exp_+\left(-\int_0^{t'}dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\right)\right]_{\mathbf{q}\mathbf{q}}^{zz}, \tag{68}$$

where, as a consequence of (67), we can disentangle the time ordering.

We now write the components of the matrix in (68):

$$\dot{F}_{\mathbf{q}}(t)=-\int_0^t dt'\sum_{\alpha\beta\gamma}\sum_{\mathbf{q}'\mathbf{q}''\mathbf{q}'''}\Bigg\langle \mathfrak{h}_{\mathbf{q}\mathbf{q}'}{}^{z\alpha}(t)\Bigg(\exp_+\Bigg(-\int_{t'}^{t}dt_1 \times\int_{t'}^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\Bigg)\Bigg)_{\mathbf{q}'\mathbf{q}''}^{\alpha\beta}\mathfrak{h}_{\mathbf{q}''\mathbf{q}'''}{}^{\beta\gamma}(t') \times\Bigg(\exp_+\Bigg(-\int_0^{t'}dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\Bigg)\Bigg)_{\mathbf{q}'''\mathbf{q}}^{\gamma z}\Bigg\rangle. \quad (69)$$

We again use the rotational and translational symmetry of the system to argue that

$$\Bigg(\exp_+\Bigg(-\int_0^{t}dt_1\int_0^{t_1}dt_2\langle\mathfrak{h}(t_1)\mathfrak{h}(t_2)\rangle\Bigg)\Bigg)_{\mathbf{q}\mathbf{q}'}^{\alpha\beta} =\delta_{\alpha\beta}\delta_{\mathbf{q}\mathbf{q}'}F_{\mathbf{q}}(t), \quad (70)$$

so that (69) becomes

$$\dot{F}_{\mathbf{q}}(t)=-\int_0^t dt'\sum_{\alpha\mathbf{q}'}\langle\mathfrak{h}_{\mathbf{q}\mathbf{q}'}{}^{z\alpha}(t)\mathfrak{h}_{\mathbf{q}'\mathbf{q}}{}^{\alpha z}(t')\rangle \times F_{\mathbf{q}'}(t-t')F_{\mathbf{q}}(t'). \quad (71)$$

The quantities $\mathfrak{h}_{\mathbf{q}\mathbf{q}'}{}^{z\alpha}$ are given in (60), so that

$$\sum_{\alpha\mathbf{q}'}\langle\mathfrak{h}_{\mathbf{q}\mathbf{q}'}{}^{z\alpha}(t)\mathfrak{h}_{\mathbf{q}'\mathbf{q}}{}^{\alpha z}(t')\rangle =-\sum_{\alpha\mathbf{q}'}J_{\mathbf{q}\mathbf{q}'}J_{\mathbf{q}'\mathbf{q}}\epsilon^{z\mu\alpha}\epsilon^{\alpha\nu z}\langle S_{\mathbf{q}-\mathbf{q}'}{}^{\mu}(t)S_{\mathbf{q}'-\mathbf{q}}{}^{\nu}(t')\rangle \quad (72)$$

$$=\tfrac{2}{3}S(S+1)N^{-1}\sum_{\mathbf{q}'}J_{\mathbf{q}\mathbf{q}'}J_{\mathbf{q}'\mathbf{q}}F_{\mathbf{q}-\mathbf{q}'}(t-t'), \quad (73)$$

where we have used (9) to obtain the last line. Hence

$$\dot{F}_{\mathbf{q}}(\tau)=-N^{-1}\sum_{\mathbf{q}'}K_{\mathbf{q}\mathbf{q}'}\int_0^{\tau}F_{\mathbf{q}'}(\tau-\tau') \times F_{\mathbf{q}-\mathbf{q}'}(\tau-\tau')F_{\mathbf{q}}(\tau')d\tau', \quad (74)$$

where

$$K_{\mathbf{q}\mathbf{q}'}=J_{\mathbf{q}\mathbf{q}'}J_{\mathbf{q}\mathbf{q}'}/\hat{J}^2=(J_{\mathbf{q}'}-J_{\mathbf{q}-\mathbf{q}'})(J_{\mathbf{q}}-J_{\mathbf{q}'-\mathbf{q}})/\hat{J}^2. \quad (75)$$

This form for $K_{\mathbf{q}\mathbf{q}'}$ follows directly from (73). It is one of several alternative ways of writing the $K_{\mathbf{q}\mathbf{q}'}$ in (74) which can be seen to be equivalent by making the variable change $\mathbf{q}'\rightleftharpoons\mathbf{q}-\mathbf{q}'$ and noting that

$$\sum_{\mathbf{q}'}(J_{\mathbf{q}'}-J_{\mathbf{q}-\mathbf{q}'})F_{\mathbf{q}'}F_{\mathbf{q}-\mathbf{q}'}=0$$

We have introduced the scaled time defined in (34).

Equation (74) is closely related to that derived by Résibois and DeLeener[7,8] and Kawasaki[4] to describe correlations near the critical point; in fact, it is the high-temperature limit of Kawasaki's Eq. (B4). The nature of its solution will be considered in some detail for a special case in Sec. 6. Here we review a few of its general properties.

First, we note that $K_{\mathbf{q}=0,\mathbf{q}'}=0$, so that $\dot{F}_{\mathbf{q}=0}(\tau)=0$ and $F_{\mathbf{q}=0}(\tau)=1$ for all τ, which is correct since $S_{\mathbf{q}=0}(t)=\sum_i S_i$ is a constant of the motion.

For small τ, (74) may be solved by iteration to obtain for nearest-neighbor interactions

$$F_{\mathbf{q}}(\tau)=1-\tfrac{1}{2}\tau^2\left(\frac{\gamma_0-\gamma_{\mathbf{q}}}{\gamma_0}\right) +\frac{1}{24}\tau^4\frac{(\gamma_0-\gamma_{\mathbf{q}})(3\gamma_0-\gamma_{\mathbf{q}}-z_2)}{\gamma_0{}^2}+O(\tau^6), \quad (76)$$

where $\gamma_0\equiv\gamma_{\mathbf{q}=0}=z_1$ and

$$\gamma_{\mathbf{q}}=\sum_{\mathbf{R}}^{\mathrm{nn}}e^{i\mathbf{q}\cdot\mathbf{R}}, \quad (77)$$

where the sum $\sum_{\mathbf{R}}{}^{\mathrm{nn}}$ is over nearest neighbors [z_1, z_2 have the meaning explained after (39)]. The coefficient of τ^2 is exactly correct, while that of τ^4 differs a little from the correct coefficient

$$(1/24)[(\gamma_0-\gamma_{\mathbf{q}})/\gamma_0{}^2](\tfrac{7}{2}\gamma_0-\tfrac{3}{2}\gamma_{\mathbf{q}}-2-3/4S(S+2)) \quad (78)$$

derived from the formulas of Ref. 6; the error is ~5–20%.

The factor

$$k_{\mathbf{q}}(\tau-\tau')=N^{-1}\sum_{\mathbf{q}'}K_{\mathbf{q}\mathbf{q}'}F_{\mathbf{q}'}(\tau-\tau')F_{\mathbf{q}-\mathbf{q}'}(\tau-\tau') \quad (79)$$

plays the role of the kernel of Eq. (74). (Its Fourier transform, the diffusivity, is discussed below.) This expression has been derived as a result of the approximations discussed in Appendixes C and D. It should be noted that small errors in the *theoretical* expression for the kernel are self-correcting in that a small increase in the kernel leads to a decrease in the correlation function, which in turn decreases the kernel. This is due to the cubic nature of Eq. (74).

For small $\mathbf{q}$ one may partially solve (74). Because $K_{\mathbf{q}\mathbf{q}'}$ is very small when $\mathbf{q}\to 0$, $F_{\mathbf{q}}(\tau)$ varies very slowly with τ. On the other hand, the factor (79) appearing in the integrand of (74) $\to 0$ fairly rapidly as $\tau-\tau'$ becomes large. Consequently one may replace the factor $F_{\mathbf{q}}(\tau')$ in the integral by $F_{\mathbf{q}}(\tau)$ for small $\mathbf{q}$ and the equation may be solved to give

$$F_{\mathbf{q}}(\tau)=\exp\left\{-N^{-1}\sum_{\mathbf{q}'}K_{\mathbf{q}\mathbf{q}'}\int_0^{\tau}d\tau' \times\int_0^{\tau'}F_{\mathbf{q}'}(\tau'-\tau'')F_{\mathbf{q}-\mathbf{q}'}(\tau'-\tau'')d\tau''\right\}$$

$$=\exp\left\{-N^{-1}\sum_{\mathbf{q}'}K_{\mathbf{q}\mathbf{q}'}\int_0^{\tau}(\tau-\tau') \times F_{\mathbf{q}'}(\tau')F_{\mathbf{q}-\mathbf{q}'}(\tau')d\tau'\right\}. \quad (80)$$

TABLE I. q-dependent correlation functions and the autocorrelation and nearest-neighbor correlation functions for the nearest-neighbor exchange simple cubic lattice.

	$F_{\mathbf{q}}(\tau)$									
τ	$\mathbf{q}=(\frac{1}{8},0,0)$	$(\frac{1}{4},0,0)$	$(\frac{3}{8},0,0)$	$(\frac{1}{2},0,0)$	$(\frac{1}{8},\frac{1}{8},\frac{1}{8})$	$(\frac{1}{4},\frac{1}{4},\frac{1}{4})$	$(\frac{3}{8},\frac{3}{8},\frac{3}{8})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$F(\tau)$	$F(\mathbf{R},\tau)$
0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	0
0.2	0.998	0.993	0.989	0.987	0.994	0.980	0.966	0.960	0.980	0.003
0.4	0.992	0.974	0.956	0.948	0.977	0.923	0.870	0.848	0.923	0.013
0.6	0.983	0.944	0.905	0.889	0.951	0.835	0.725	0.680	0.836	0.027
0.8	0.972	0.905	0.840	0.814	0.916	0.726	0.550	0.481	0.728	0.043
1.0	0.958	0.860	0.766	0.728	0.877	0.605	0.367	0.278	0.611	0.060
1.2	0.943	0.812	0.688	0.639	0.834	0.483	0.197	0.097	0.494	0.075
1.4	0.927	0.762	0.610	0.551	0.789	0.368	0.056	−0.046	0.386	0.086
1.6	0.911	0.712	0.534	0.467	0.745	0.266	−0.049	−0.139	0.292	0.093
1.8	0.894	0.663	0.463	0.389	0.701	0.180	−0.113	−0.181	0.216	0.095
2.0	0.877	0.616	0.398	0.320	0.658	0.111	−0.139	−0.179	0.158	0.093
2.2	0.861	0.571	0.339	0.258	0.618	0.059	−0.135	−0.144	0.117	0.088
2.4	0.844	0.528	0.286	0.206	0.579	0.022	−0.110	−0.091	0.089	0.079
2.6	0.828	0.488	0.240	0.161	0.542	−0.003	−0.073	−0.033	0.072	0.070
2.8	0.811	0.450	0.200	0.124	0.507	−0.017	−0.035	0.018	0.063	0.061
3.0	0.795	0.415	0.165	0.093	0.474	−0.024	0.000	0.054	0.058	0.052
3.2	0.780	0.382	0.135	0.068	0.442	−0.026	0.025	0.073	0.056	0.044
3.4	0.764	0.351	0.110	0.048	0.413	−0.024	0.040	0.075	0.054	0.038
3.6	0.749	0.323	0.089	0.033	0.386	−0.020	0.044	0.062	0.052	0.034
3.8	0.733	0.297	0.071	0.021	0.360	−0.016	0.040	0.041	0.050	0.031
4.0	0.719	0.272	0.056	0.012	0.336	−0.012	0.031	0.016	0.047	0.029
4.2	0.704	0.250	0.044	0.005	0.313	−0.008	0.019	−0.006	0.044	0.027
4.4	0.690	0.229	0.034	0.001	0.292	−0.005	0.006	−0.023	0.040	0.027
4.6	0.676	0.210	0.026	−0.003	0.272	−0.003	−0.004	−0.032	0.036	0.026
4.8	0.662	0.192	0.020	−0.005	0.253	−0.001	−0.011	−0.034	0.033	0.025
5.0	0.648	0.176	0.015	−0.006	0.236	−0.000	−0.015	−0.029	0.030	0.024

For small $\mathbf{q}$ the exponent is like $-q^2\phi(\tau)+O(q^4)$, so that

$$F_{\mathbf{q}}(\tau)=\exp\{-q^2\phi(\tau)\},\quad \mathbf{q}\to 0 \tag{81}$$

where after some algebra one finds

$$\phi(\tau)=\tfrac{1}{6}N^{-1}\sum_{\mathbf{q}'}\hat{J}^{-2}(\nabla_{\mathbf{q}'}J_{\mathbf{q}'})^2\int_0^\tau(\tau-\tau')F_{\mathbf{q}'}{}^2(\tau')d\tau'.$$

For large τ, $\phi(\tau)\sim\tau D_1$, so that

$$F_{\mathbf{q}}(\tau)=\exp\{-q^2\tau D_1\},\quad \mathbf{q}\to 0,\tau\to\infty \tag{82}$$

which is just the ordinary diffusion-law form for $F_{\mathbf{q}}(\tau)$. D_1 is given by

$$D_1=\tfrac{1}{6}N^{-1}\sum_{\mathbf{q}'}\hat{J}^{-2}(\nabla_{\mathbf{q}'}J_{\mathbf{q}'})^2\int_0^\infty F_{\mathbf{q}'}{}^2(\tau')d\tau' \tag{83}$$

and is related to the conventionally defined diffusion constant by

$$D=D_1[\tfrac{2}{3}S(S+1)]^{1/2}\hat{J} \tag{84}$$

using (34).

6. $C_{\mathbf{q}}(t)$ FOR SIMPLE CUBIC LATTICE WITH NEAREST-NEIGHBOR INTERACTIONS

In order to obtain some impression of the nature of the solutions of (74), we have solved it numerically for the special case of a simple cubic lattice with nearest-neighbor exchange interactions. This is a particularly suitable choice of model since it affords the opportunity of comparison with the computer simulation calculations of Windsor.[18]

Equation (74) may be integrated forward in τ starting with $F_{\mathbf{q}}(0)=1$, the integration being reasonably stable provided that a small enough time interval is used. The $\mathbf{q}'$ sum was approximated by a sum over 512 points (on a simple cubic mesh) in the Brillouin zone; because of cubic symmetry only a small proportion (35) of those points had to be considered explicitly. The $J_{\mathbf{q}}$ for this model are given by

$$J_{\mathbf{q}}=2(\cos q_xa+\cos q_ya+\cos q_za)J, \tag{85}$$

where a is the cube edge, and $\hat{J}=J\sqrt{6}$, where J is the exchange interaction.

The results of these numerical calculations of $C_{\mathbf{q}}(\tau)$ are given for a few $\mathbf{q}$ vectors in Table I. In Fig. 3 the $C_{\mathbf{q}}(\tau)$ are plotted against τ together with the corresponding results obtained by Windsor, with which they are seen to be in fairly good agreement [in making this com-

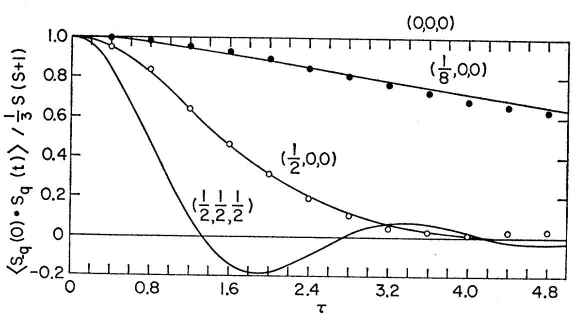

FIG. 3. q-dependent correlation functions for several different q vectors. These vectors are in units of $2\pi/a$ where a is the cube edge. $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ is thus the corner of the Brillouin zone, and $(\frac{1}{2},0,0)$ is the center of a square face. The circles again represent Windsor's computer simulation data.

parison, one must note that Windsor's calculations are for classical spins, corresponding to $S\rightarrow\infty$ in which case (34) becomes $\tau=2SJ$, and that his definition of J is half as large as that used here]. The characteristic behavior is a slow decrease for small $\mathbf{q}$ becoming more rapid for larger $\mathbf{q}$ with oscillations setting in for the largest $\mathbf{q}$.

From the $C_{\mathbf{q}}(\tau)$ one can also obtain the correlation function

$$C(\mathbf{R}_j-\mathbf{R}_i,\tau)=\langle S_i^zS_j^z(\tau)\rangle =N^{-1}\sum_{\mathbf{q}} C_{\mathbf{q}}(\tau)e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_i)}, \tag{86}$$

and, in particular,

$$C(\tau)=N^{-1}\sum_{\mathbf{q}} C_{\mathbf{q}}(\tau). \tag{87}$$

$C(\tau)$ and $C(\mathbf{R},\tau)$ for $\mathbf{R}=(a,0,0)$ are given in Table I and plotted in Fig. 1, where they are again compared with Windsor's corresponding results and with the approximations for $C(\tau)$ derived in Secs. 3 and 4. Again the agreement with Windsor is good [except for the height of the peak in $C(\mathbf{R},\tau)$].

The diffusion constants D and D_1 have been calculated by substituting the solution for $F_{\mathbf{q}}(\tau)$ in (83) and using (84). We find

$$D=0.33[S(S+1)]^{1/2}Ja^2, \tag{88}$$

which agrees with Mori and Kawasaki's result,[2] with the corrected value obtained by Bennett and Martin,[5] and with Windsor's[18] result, but is larger than that calculated by deGennes.[1]

The Fourier transforms $\hat{C}_{\mathbf{q}}(\Omega)$ and $\hat{C}(\Omega)$ are of interest since they determine the inelastic neutron scattering cross sections at high temperatures. For the large $\mathbf{q}$, $\hat{C}_{\mathbf{q}}(\Omega)$ could be determined by numerical transformation of the $C_{\mathbf{q}}(\tau)$. For small $\mathbf{q}$, however, this procedure is difficult because of the very slow decrease of $C_{\mathbf{q}}(\tau)$ at large τ. For this reason a different technique was used.

Using the definition (79), Eq. (74) may be written

$$\dot{F}_{\mathbf{q}}(\tau)=-\int_0^\tau k_{\mathbf{q}}(\tau-\tau')F_{\mathbf{q}}(\tau')d\tau'. \tag{89}$$

Hitherto the $F_{\mathbf{q}}(\tau)$ have only been considered for $\tau>0$. Suppose we now extend the definitions of $F_{\mathbf{q}}(\tau)$ and $k_{\mathbf{q}}(\tau)$ by requiring that $F_{\mathbf{q}}(\tau)=k_{\mathbf{q}}(\tau)=0$ for $\tau<0$. Then (79) will be satisfied for all τ, and $F_{\mathbf{q}}(\tau)$ will satisfy

$$\dot{F}_{\mathbf{q}}(\tau)=\delta(\tau)-\int_{-\infty}^{\infty} k_{\mathbf{q}}(\tau-\tau')F_{\mathbf{q}}(\tau')d\tau' \tag{90}$$

for all τ with the boundary condition $F_{\mathbf{q}}(\tau)=0$ for $\tau<0$; the δ function causes $F_{\mathbf{q}}(\tau)$ to jump from 0 to 1 at $\tau=0$. We may now Fourier-transform (90) to obtain

$$i\omega\hat{F}_{\mathbf{q}}(\omega)=1-\hat{k}_{\mathbf{q}}(\omega)\hat{F}_{\mathbf{q}}(\omega), \tag{91}$$

or

$$\hat{F}_{\mathbf{q}}(\omega)=\frac{1}{i\omega+\hat{k}_{\mathbf{q}}(\omega)}\equiv F_{\mathbf{q}}'(\omega)+iF_{\mathbf{q}}''(\omega). \tag{92}$$

We may remark in passing that this equation suggests that $\hat{k}_{\mathbf{q}}(\omega)$ is playing the role of a self-energy in our calculations. Combining Eqs. (80) and (92), we see that it is indeed determined by a self-consistent equation in the way discussed in the Introduction. Since $C_{\mathbf{q}}(t)=F_{\mathbf{q}}(|t|)$, one has from (92) [cf. (40)]

$$\frac{NJ\hat{C}_{\mathbf{q}}(\Omega)}{[\frac{2}{3}S(S+1)]^{1/2}}=\hat{F}_{\mathbf{q}}'(\omega)=\frac{k_{\mathbf{q}}'(\omega)}{[k_{\mathbf{q}}'(\omega)]^2+[\omega+k_{\mathbf{q}}''(\omega)]^2}, \tag{93}$$

where $\hat{k}_{\mathbf{q}}(\omega)=k_{\mathbf{q}}'(\omega)+ik_{\mathbf{q}}''(\omega)$. $\hat{k}_{\mathbf{q}}(\omega)$ is thus related to the generalized diffusivity by $\hat{k}_{\mathbf{q}}(\omega)=q^2D(\mathbf{q},\omega)$. The function $k_{\mathbf{q}}(\tau)$ always decreases quickly to 0 when τ becomes large, so its transform $\hat{k}_{\mathbf{q}}(\omega)$ may be readily evaluated numerically from $k_{\mathbf{q}}(\tau)$ which is in turn given by (80), and in this way $F_{\mathbf{q}}'(\omega)$ can be conveniently evaluated even for small $\mathbf{q}$. The transform $F(\omega)$ of $F(\tau)$ may be obtained from

$$\hat{F}(\omega)=N^{-1}\sum_{\mathbf{q}}\hat{F}_{\mathbf{q}}(\omega). \tag{94}$$

TABLE II. Fourier transforms of the $\mathbf{q}$-dependent correlation functions and of the autocorrelation function.

	$\hat{F}_{\mathbf{q}}'(\omega)$								
ω	$\mathbf{q}=(\frac{1}{8},0,0)$	$(\frac{1}{4},0,0)$	$(\frac{3}{8},0,0)$	$(\frac{1}{2},0,0)$	$(\frac{1}{8},\frac{1}{8},\frac{1}{8})$	$(\frac{1}{4},\frac{1}{4},\frac{1}{4})$	$(\frac{3}{8},\frac{3}{8},\frac{3}{8})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$	$\hat{F}(\omega)$
0	10.348	3.143	1.899	1.640	3.617	1.181	0.747	0.654	1.518
0.2	2.173	2.554	1.802	1.588	2.704	1.167	0.746	0.655	1.309
0.4	0.596	1.562	1.537	1.435	1.464	1.126	0.745	0.656	1.131
0.6	0.247	0.877	1.181	1.198	0.770	1.053	0.743	0.657	0.978
0.8	0.125	0.498	0.833	0.919	0.428	0.948	0.740	0.660	0.838
1.0	0.071	0.293	0.554	0.653	0.251	0.813	0.735	0.664	0.707
1.2	0.043	0.178	0.358	0.439	0.153	0.658	0.724	0.669	0.584
1.4	0.027	0.112	0.228	0.286	0.096	0.500	0.703	0.675	0.468
1.6	0.017	0.071	0.146	0.184	0.062	0.357	0.660	0.677	0.361
1.8	0.012	0.046	0.094	0.118	0.040	0.240	0.578	0.661	0.264
2.0	0.008	0.031	0.061	0.076	0.027	0.155	0.447	0.593	0.181
2.2	0.005	0.020	0.039	0.049	0.108	0.096	0.294	0.441	0.115
2.4	0.004	0.013	0.026	0.032	0.012	0.059	0.169	0.257	0.069
2.6	0.002	0.009	0.017	0.021	0.008	0.036	0.090	0.129	0.040

TABLE III. Fourier cosine transform of the kernel $k_{\mathbf{q}}'(\omega)$ (the diffusivity).

	$k_{\mathbf{q}}'(\omega)$							
ω	$\mathbf{q}=(\frac{1}{8},0,0)$	$(\frac{1}{4},0,0)$	$(\frac{3}{8},0,0)$	$(\frac{1}{2},0,0)$	$(\frac{1}{8},\frac{1}{8},\frac{1}{8})$	$(\frac{1}{4},\frac{1}{4},\frac{1}{4})$	$(\frac{3}{8},\frac{3}{8},\frac{3}{8})$	$(\frac{1}{2},\frac{1}{2},\frac{1}{2})$
0	0.097	0.318	0.527	0.610	0.276	0.847	1.339	1.528
0.2	0.094	0.312	0.520	0.603	0.271	0.841	1.334	1.523
0.4	0.088	0.297	0.500	0.582	0.258	0.823	1.319	1.509
0.6	0.081	0.276	0.470	0.549	0.241	0.794	1.292	1.485
0.8	0.073	0.251	0.431	0.507	0.220	0.753	1.254	1.449
1.0	0.065	0.224	0.388	0.457	0.198	0.702	1.201	1.399
1.2	0.057	0.197	0.343	0.405	0.175	0.641	1.133	1.334
1.4	0.049	0.171	0.297	0.351	0.152	0.571	1.045	1.248
1.6	0.042	0.146	0.254	0.300	0.130	0.496	0.938	1.138
1.8	0.035	0.123	0.213	0.252	0.109	0.418	0.809	0.998
2.0	0.029	0.102	0.177	0.208	0.091	0.342	0.663	0.826
2.2	0.024	0.083	0.144	0.170	0.074	0.272	0.514	0.636
2.4	0.020	0.067	0.116	0.136	0.059	0.211	0.379	0.451
2.6	0.016	0.054	0.091	0.107	0.047	0.160	0.270	0.313

The transforms obtained in this way are given in Table II and are plotted in Figs. 2 and 4. $\hat{F}(\omega)$ is compared with the corresponding transform derived by Windsor and with the approximation of Sec. 3. Again the general agreement is good. For small $\mathbf{q}$, $F_{\mathbf{q}}'(\omega)$ is very narrow and Lorentzian. As q increases it broadens but the far wings of the line remain low. At still larger $\mathbf{q}$, $\hat{F}_{\mathbf{q}}'(\omega)$ "shoulders," reflecting the tendency of $C_{\mathbf{q}}(\tau)$ to oscillate, until at the $\mathbf{q}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ point it has a very flat plateau.

Bennett and Martin,[5] Bennett,[9] and Tahir-Kheli and McFadden[10] have given phenomenological treatments of the diffusion coefficient and the correlation functions

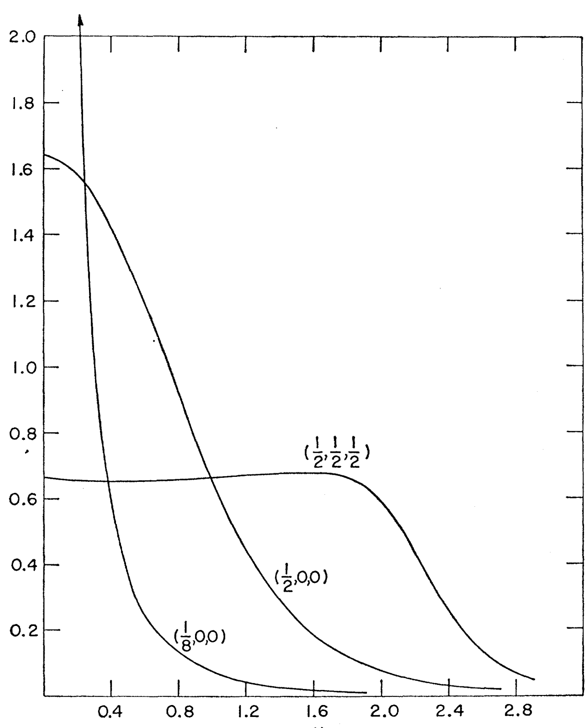

FIG. 4. Fourier transform $\hat{F}_{\mathbf{q}}'(\omega)$ of the $\mathbf{q}$-dependent correlation functions.

in which they assume a two-parameter Gaussian form for the diffusivity. In our notation they assume

$$\hat{k}_{\mathbf{q}}'(\omega)=(\sqrt{\pi})\frac{a_{\mathbf{q}}}{2b_{\mathbf{q}}}e^{-\omega^2/4b_{\mathbf{q}}^2}, \tag{95}$$

where $a_{\mathbf{q}}$ and $b_{\mathbf{q}}$ are parameters that are determined by fitting the exactly known second and fourth moments of the correlation functions. Since we have calculated $k_{\mathbf{q}}'(\omega)$ directly (these are given for several values of $\mathbf{q}$ in Table III), we are in a position to examine this hypothesis. In Fig. 5 we show $k_{\mathbf{q}}'(\omega)/k_{\mathbf{q}}'(0)$ for these different points in the Brillouin zone. The smallest and largest $\mathbf{q}$ vectors differ from the Gaussian shape. The intermediate value of $\mathbf{q}$ follows a curve which is quite closely Gaussian, however, and indeed fits closely the form

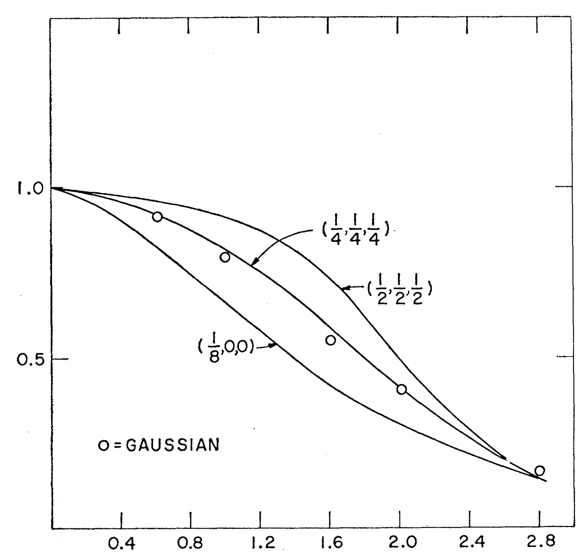

FIG. 5. Fourier cosine transform of the kernel $k_{\mathbf{q}}'(\omega)/k_{\mathbf{q}}'(0)$, which is equivalent to the diffusivity defined by Bennett and Martin (Ref. 5). The open circles represent the Gaussian approximation to the $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ curve, obtained by fitting (95) to the exact high-temperature moments (Ref. 12). The Gaussian approximation for the $(\frac{1}{8},0,0)$ and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ curves (not shown) is a much poorer fit.

derived from Eq. (95) with a_q and b_q determined by the exact values of the moments. Further, the amplitudes $k_q'(O)$ calculated by us agree very well with the values obtained from (95). The Gaussian approximation is thus apparently not very good for small or large **q** vectors, but in the intermediate **q** range the form (95) appears valid. Since a large number of **q** vectors fall in the range where the assumption is justified by our calculations, we expect and find good agreement between our correlation functions and those obtained by Tahir-Kheli and McFadden.[10]

7. CONCLUSION

It appears from the results discussed in the preceding sections that in spite of a number of approximations having been made, the theory still gives a good account of high-temperature correlation phenomena. Several points concerning the method should be noted. The theory involves a decoupling procedure as do the various Green's-function calculations. In our case, however, the decoupling is done in the exponential correlation functions rather than in the equations of motion, so that we take advantage of the property of cumulants—that they vanish if any of the operators are independent of the others in that term. Alternately we may compare our procedure with equation-of-motion methods by noting that in such methods a decoupling is performed directly on the equations of motion. In the present treatment the equations are formally solved and the decoupling is performed on the solution. This procedure should be applicable at finite temperatures and also in other many-body problems. The physical picture would be similar to that adopted here: A particle (in a general sense) is subject to a fluctuating potential. The correlation function for the particle is then expressed in terms of that of the fluctuating potential, which in turn is related to that for the particle—a self-consistent relation.

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APPENDIX A: INTERPRETATION OF $\delta B(t)$

It is our purpose here to confirm the assertion in Sec. 2 that $\hat{B}(t)$ is the operator which evolves in the presence of the perturbation from the operator which was $B(t)$ when $t\to-\infty$. To this end, we note that in the presence of the perturbation, $\hat{B}(t)$ will be given by

$$\hat{B}(t)=V^*(t)BV(t)\,, \tag{A1}$$

where $V(t)$ is an operator obeying the equation

$$i\frac{\partial V}{\partial t}=[H_0-\theta(-t)Ae^{\epsilon t}]V(t)\,, \tag{A2}$$

where $-\theta(-t)Ae^{\epsilon t}$ [$\theta(t)$ = step function, $\epsilon\to 0_+$] is the perturbation, and V satisfies a suitable boundary condition [i.e., such that $\hat{B}(t)\to B(t)$ as $t\to-\infty$]. One may easily verify that a suitable solution is given by

$$V(t)=e^{-iH_0t}\exp_+\left\{i\int_{-\infty}^{t}\theta(-t')A(t')e^{\epsilon t'}dt'\right\}. \tag{A3}$$

If we now expand the exponential to first order and substitute the result in (A1) we find, to first order in A for $t>0$,

$$\hat{B}(t)=(1+X)e^{iH_0t}Be^{-iH_0t}(1-X)=B(t)+\delta B(t)\,, \tag{A4}$$

since for $t>0$, one has $-i\int_{-\infty}^{t}\theta(-t')A(t')e^{\epsilon t'}dt'=X$.

APPENDIX B: SOME CONSEQUENCES OF APPROXIMATION (B)

We show here how the approximations (B′) (in the context of Secs. 4 and 5), (B″) and (B‴) follow from the approximation principle (B) enunciated in Sec. 3. We first note that any average $\langle S_1S_2\cdots S_n\rangle$ can be written as a sum of products of cumulant averages.[24] Since $\langle S_i\rangle=0$ at high temperatures, approximation (B) is equivalent to the assertion that any average of an even number of spins may be approximated by a sum of products of spin-pair averages

$$\langle S_1S_2\cdots S_{2n}\rangle\simeq\sum_{\text{pairings}}\langle S_{i_1}S_{j_1}\rangle\langle S_{i_2}S_{j_2}\rangle\cdots\langle S_{i_n}S_{j_n}\rangle\,, \tag{B1}$$

where the sum runs over all pairings of the spins in the product, while the average of an odd number of spins is negligible [in (B1) note that since $\langle S_i\rangle=0$, one has $\langle S_iS_j\rangle_c=\langle S_iS_j\rangle$].

We first use (B1) to establish (B″). Since [see Eq. (12)] $\delta S=[X,S]$, where in the application X is always proportional to $\int_{-\infty}^{0}S'(t')dt'$, we may write

$$\langle S_1S_2\cdots S_n\delta S\rangle=\int_{-\infty}^{0}[\langle S_1S_2\cdots S_nS'(t')S\rangle-\langle S_1\cdots S_nSS'(t')\rangle]dt.' \tag{B2}$$

We now apply (B1) to evaluate the averages in (B2). Consider the first term in the integrand expanded according to (B1). We distinguish two classes of pairings: (a) those in which S is paired with $S'(t')$; (b) those in which it is not. A typical term of class (b) will be

$$\int_{-\infty}^{0}\langle S_iS'(t')\rangle\langle S_jS\rangle\langle S_{p1}S_{q1}\rangle\langle S_{p2}S_{q2}\rangle\cdots dt'. \tag{B3}$$

If we now consider the second term of the integrand in (B2) and divide the pairings into two classes in the same way, then we find at once that in its expansion there occurs a term of class (b) which exactly cancels against (B3). In this way one easily finds that all pairings of class (b) cancel between the two terms. On the other hand, a pairing of class (a) from the first term of (B2) has the form

$$\int_{-\infty}^{0} \langle S'(t')S\rangle\langle S_{i_1}S_{j_1}\rangle\langle S_{i_2}S_{j_2}\rangle\cdots dt',$$

and is matched by

$$\int_{-\infty}^{0} \langle SS'(t')\rangle\langle S_{i_1}S_{j_1}\rangle\langle S_{i_2}S_{j_2}\rangle\cdots dt'$$

from the second term to give a total contribution

$$\langle S_{i_1}S_{j_1}\rangle\langle S_{i_2}S_{j_2}\rangle\cdots\int_{-\infty}^{0} \langle[S'(t'),S]\rangle dt' = \langle\delta S\rangle\langle S_{i_1}S_{j_1}\rangle\langle S_{i_2}S_{j_2}\rangle\cdots.$$

Combining all the pairing of class (a) and use of (B1), now leads to

$$\langle S_1S_2\cdots S_n\delta S\rangle = \langle S_1S_2\cdots S_n\rangle\langle\delta S\rangle,$$

i.e., (B″).

(B‴) follows rapidly from (B″). Any average of a product of an odd number of h superoperators ($h^\times$ or $\mathfrak{h}$) contains a factor of the form $\langle S_1\cdots S_n\delta S\rangle$, where n is odd. But by (B″) this is $\langle S_1\cdots S_n\rangle\langle\delta S\rangle = 0$, since $\langle S_1S_2\cdots S_n\rangle = 0$ if n is odd, establishing (B‴).

Within the context of Secs. 4 and 5, (B′) also follows from (B). This is because the cumulant expansions used there are based upon an averaging process which is just the usual $\langle\cdots\rangle$ average (as opposed to the $\langle\langle\cdots\rangle\rangle$ averaging process defined in Sec. 3, which is somewhat different). When the $\langle\cdots\rangle$ is used, we may easily show [using (B″)] that $\langle h_1h_2\cdots h_n\rangle_c$ is proportional to $\langle S_1S_2\cdots S_n\rangle_c\langle\delta S\rangle$ when $S_1\cdots S_n$ are certain spins. But for $n>3$, $\langle S_1S_2\cdots S_n\rangle_c$ has been assumed negligible; hence it follows that $\langle h_1h_2\cdots h_n\rangle$ is also for $n\geqslant 3$, which is just (B′).

APPENDIX C: RELATIONSHIP OF THEORIES OF SECS. 3 AND 4

We shall show here the nature of the approximation required to derive the theory of Sec. 3 from (51) of Sec. 4. We may regard the process $(O)^{zz}$ of taking the zz component of the tensor O as an averaging process. Then (51) has the form of such an average and we may apply the cumulant expansion to write

$$F(t) = \exp\Bigg\{\frac{1}{2}\int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2 F(\tau_1-\tau_2)(\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu})_+{}^{zz} + \tfrac{1}{2}(\tfrac{1}{2})^2\int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2\int_0^\tau d\tau_1'\int_0^{\tau_1'} d\tau_2' F(\tau_1-\tau_2) \times F(\tau_1'-\tau_2')(\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu}\epsilon_{\tau_1'}{}^{\nu}\epsilon_{\tau_2'}{}^{\nu})_{+c}{}^{zz}+\cdots\Bigg\}, \quad \text{(C1)}$$

where $(\cdots)_{+c}{}^{zz}$ means the zz component of the cumulant average of the time-ordered product of $(\cdots)$. What one means by the *cumulant* average here should be carefully noted; for the purpose of writing out the averages the units are the pairs of ϵ factors with the same superscript label, e.g., $\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu}$. For example,

$$(\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu}\epsilon_{\tau_1'}{}^{\nu}\epsilon_{\tau_2'}{}^{\nu})_{+c}{}^{zz} = (\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu}\epsilon_{\tau_1'}{}^{\nu}\epsilon_{\tau_2'}{}^{\nu})_+{}^{zz} - (\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu})_+{}^{zz}(\epsilon_{\tau_1'}{}^{\nu}\epsilon_{\tau_2'}{}^{\nu})_+{}^{zz}.$$

If we now neglect all but the first term in the cumulant expansion in (C1) and use $(\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu})^{zz} = -2$, then (C1) reduces to (35). Thus the theory of Sec. 3 involves the additional approximation of dropping the higher-order cumulants in (C1). At small τ this will be a good approximation.

The above discussion throws light on the different nature of approximation (B′) in the theory of Sec. 3 on the one hand, and Secs. 4 and 5 on the other. In the latter theories, (B′) follows from (B) as discussed in Sec. 2. But in Sec. 3 [because of the definition (25) for the average $\langle\langle\cdots\rangle\rangle$], approximation (B′) is equivalent to the application of both approximation (B) [which leads to (51)] *and* the approximation of dropping the higher-order cumulants in (C1), which is an additional approximation of a different nature, and thus (B′) of Sec. 3 does not follow solely from (B).

APPENDIX D: DISENTANGLING APPROXIMATION

At Eq. (53) of Sec. 4 and Eq. (71) of Sec. 5 we used approximations of the kind shown in (53) to estimate approximately certain time-ordered products. In this Appendix it will be shown that our approximation is in fact the leading term in a certain expansion for the time-ordered product, and the error introduced by the use of this approximation is estimated.

To obtain the expansion alluded to, we first expand part of the exponential in the first line of (53) to obtain

$$\begin{aligned}(\epsilon_{\tau'}{}^{\mu}U(\tau,0))_+ &= \Bigg(\epsilon_{\tau'}{}^{\mu}U(\tau,\tau')U(\tau',0) \times\exp\left\{\frac{1}{2}\int_{\tau'}^{\tau} d\tau_1\int_0^{\tau'} d\tau_1' F(\tau_1-\tau_1')\epsilon_{\tau_1}{}^{\nu}\epsilon_{\tau_1'}{}^{\nu}\right\}\Bigg)_+ \\ &= (U(\tau,\tau'))_+\epsilon_\mu(U(\tau',0))_+ + \frac{1}{2}\int_{\tau'}^{\tau} d\tau_1\int_0^{\tau'} d\tau_1' F(\tau_1-\tau_1')(\epsilon_{\tau_1}{}^{\nu}U(\tau,\tau'))_+\epsilon^{\mu} \\ &\quad\times(\epsilon_{\tau_1'}{}^{\nu}U(\tau',0))_+ + \frac{1}{2!}(\tfrac{1}{2})^2\int_{\tau'}^{\tau} d\tau_1\int_{\tau'}^{\tau} d\tau_2\int_0^{\tau'} d\tau_1' \\ &\quad\times\int_0^{\tau'} d\tau_2' F(\tau_1-\tau_1')F(\tau_2-\tau_2')(\epsilon_{\tau_1}{}^{\nu_1}\epsilon_{\tau_2}{}^{\nu_2}U(\tau,\tau'))_+ \\ &\quad\times\epsilon^{\mu}(\epsilon_{\tau_1'}{}^{\nu_1}\epsilon_{\tau_2'}{}^{\nu_2}U(\tau',0))_+ + \cdots, \quad \text{(D1)}\end{aligned}$$

where, for brevity, we have written

$$U(\tau,\tau')\equiv\exp\left\{\frac{1}{2}\int_{\tau'}^{\tau}d\tau_1\int_{\tau'}^{\tau}d\tau_2F(\tau_1-\tau_2)\epsilon_{\tau_1}{}^{\mu}\epsilon_{\tau_2}{}^{\mu}\right\}. \quad \text{(D2)}$$

The retention of just the first term, which is $\epsilon^{\mu}F(\tau_1-\tau')F(\tau')$, in (D1) gives the approximation (C) used in the text. If we retain the first two terms in (D1), we may iterate [since quantities of the form $(\epsilon^{\mu}U)_+$ appear on the right] to obtain an expansion of the form

$$(\epsilon_{\tau'}{}^{\mu}U(\tau,0))_+=F(\tau-\tau')F(\tau')\epsilon^{\mu}$$
$$-\tfrac{1}{2}\epsilon^{\mu}\int_{\tau'}^{\tau}d\tau_1\int_{0}^{\tau'}d\tau_2F(\tau_1-\tau_2)F(\tau-\tau_1)$$
$$\times F(\tau_1-\tau')F(\tau'-\tau_2)F(\tau_2)+\cdots. \quad \text{(D3)}$$

If we retain third- and higher-order terms in (D1), then we shall require expressions of the form

$$(\epsilon_{\tau_1}{}^{\nu_1}\epsilon_{\tau_2}{}^{\nu_2}\cdots U(\tau,\tau'))_+.$$

However we may expand these by exactly the same technique as led to (D1) to get a series beginning like

$$(\epsilon_{\tau_1}{}^{\nu_1}\epsilon_{\tau_2}{}^{\nu_2}\cdots U(\tau,\tau'))_+$$
$$=F(\tau-\tau_1)F(\tau_1-\tau_2)\cdots\epsilon^{\nu_1}\epsilon^{\nu_2}\cdots+\cdots.$$

In this way we may expand $(\epsilon_{\tau'}{}^{\mu}U(\tau,0))_+$ in a series, each of whose terms has the form of ϵ^{μ} times an integral with an integrand consisting of a product of F functions. The leading terms of this series are given in (D3).

If this series for $(\epsilon_{\tau'}{}^{\mu}U(\tau,0))_+$ is substituted into (52), we find after a little rearrangement that the resulting equation can be written

$$\dot{F}(t)=-\int_0^{\tau}k(\tau-\tau')F(\tau')d\tau', \quad \text{(D4)}$$

where

$$k(\tau)=F^2(\tau)-\frac{1}{2}\int_0^{\tau}d\tau_1\int_0^{\tau_1}d\tau_2F(\tau-\tau_1)F(\tau_1-\tau_2)$$
$$\times F(\tau_2)F(\tau_1)F(\tau-\tau_2)+\cdots. \quad \text{(D5)}$$

Our approximation (54) corresponds to keeping the first term is the kernel $k(\tau)$; evidently from (D5) this is a good approximation at small τ. If we write $k(\tau)=k_0(\tau)+k_1(\tau)$, where $k_0(\tau)=F^2(\tau)$, then we find from (D4) that

$$\begin{aligned}F(\tau)&=1-\tfrac{1}{2}\tau^2+(7/48)\tau^4+O(\tau^6),\\ k_0(\tau)&=1-\tau^2+O(\tau^4),\\ k_1(\tau)&=-\tfrac{1}{4}\tau^2+O(\tau^4).\end{aligned} \quad \text{(D6)}$$

Comparison with (54) and (38) shows that the leading correction to approximation (C) occurs in the τ^4 term where it changes the coefficient by 16%. In fact, (D6) gives $(d^4F/d\tau^4)_{\tau=0}=\frac{7}{2}$, which by comparison with (39) we see is good to $O(1/z_1)$. Thus the errors in τ^4 coefficients of (38) and (54) are due to approximation (C) rather than (B).

To estimate the error due to approximation (C) at large τ, we have estimated the ratio of $\int_0^{\infty}k_1(\tau)d\tau$ [with $k_1(\tau)$ given by the second term of (D5)] to $\int_0^{\infty}k_0(\tau)d\tau$, using the approximate representation $F(\tau)\simeq e^{-\lambda\tau}$ to give a rough estimate (a suitable choice is $\lambda\sim 1/\sqrt{2}$). We find $\int_0^{\infty}k_1(\tau)d\tau/\int_0^{\infty}k_0(\tau)d\tau\simeq-1/12\lambda^2\simeq-\frac{1}{6}$, so that the integrated error in the kernel $\sim$16%.

We expect the relative error in the solution of (D4) due to the use of approximation (C) to be rather less than the error in the kernel estimated above. The reason is that Eqs. (D4) and (D5) have a kind of "self-compensating" property. To see this, suppose $k_1<0$, so the addition of the correction k_1 to k_0 tends to decrease the kernel k. Then through (D5) we see that a decrease in k tends to cause an increase in F. However, since $k_0=F^2$, the increase in F causes an increase in k_0 which tends to cancel the decrease in k caused by k_1. The essential reason for this cancellation is the fact that (D4) with $k_0=F^2$ substituted for k, or (54), is cubic in F, and thus the addition of a term of relative magnitude η causes only a change of relative magnitude $\sim\frac{1}{3}\eta$ in F. We may comment that this latter argument applies not only to errors arising from the use of approximation (C), but also from other sources of error such as approximation (B). Because the fundamental equation is cubic, the errors in the solution are smaller than one might naively estimate.

The discussion above has all been concerned with the use of approximation (C) to derive (54). However, essentially the same arguments with the same consequences apply to its use in Sec. 5.