Space-Time Correlations in Exchange-Coupled Paramagnets at Elevated Temperatures

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(Received 22 January 1969)

The frequency-dependent correlations $\Phi_n(\omega)\equiv\langle S_0{}^z(t)S_n{}^z(t')\rangle_{(\omega)}$, $n=0$, 1, 2, 3, and 4, and their inverse-lattice Fourier transforms $\Phi(\mathbf{K},\omega)$, have recently been exactly calculated by Carboni and Richards for finite linear chains consisting of N ($N=6$, 7, 8, 9, and 10) spins ($S=\frac{1}{2}$) in contact with a heat bath at temperature $T\rightarrow\infty$ and interacting via a nearest-neighbor isotropic Heisenberg exchange interaction. Carboni and Richards have used plausible extrapolation procedures to predict the corresponding correlations in the thermodynamic limit $N\rightarrow\infty$. As the extension of these exact calculations to two and three dimensions is of prohibitive difficulty, we have constructed an alternative theory, based upon a simple two-parameter Gaussian representation of the generalized diffusivity, for calculating these correlations for general spin and the dimensionality. To test the accuracy of this phenomenological theory (which is, however, free of any arbitrariness in the sense that the diffusivity is exactly specified by the second and the fourth frequency moments of the Fourier transform of the frequency-dependent correlation function, which are known at infinite temperature), we first compare our results for $\Phi_n(\omega)$ with those given by Carboni and Richards for the one-dimensional spin-$\frac{1}{2}$ system and find the agreement to be excellent for $n=0$, good for $n=1,2$ and adequate for $n=3,4$. The comparison of the corresponding results for $\Phi(\mathbf{K},\omega)$ reveals the agreement to be quite satisfactory for $|\mathbf{K}|\lesssim\frac{1}{2}\pi$ but only *adequate* for the higher-$\mathbf{K}$ range, i.e., $\frac{1}{2}\pi\lesssim\mathbf{K}\lesssim\pi$. Further support in favor of our phenomenological theory is obtained from a comparison with another set of available exact "computer experiment" results, whereby $\Phi_0(\omega)$, $\Phi_0(t)$, and $\Phi_1(t)$ are accurately known for a three-dimensional (simple-cubic) lattice of infinite spins, i.e., $S\rightarrow\infty$. (This is the limit in which the classical spin system studied by Windsor corresponds to the quantum spin system of interest to us.) The agreement of our result with Windsor's is excellent. The salient features of our results are as follows: (i) In two dimensions, the divergence of $\Phi_0(\omega)$ for $\omega\rightarrow 0$ becomes less sharp and disappears completely in three dimensions. (ii) The cutoff frequency, beyond which $\Phi_0(\omega)$ is effectively zero, increases with the dimensionality, being in three dimensions about twice what it is in one dimension. (iii) There exists a system of reduced units, i.e., $\Phi_n'(\omega)\rightarrow\Phi_n(\omega)[S(S+1)]^{-1}$, $I'\rightarrow[S(S+1)]^{1/2}I$, and $\omega'\rightarrow(\omega/I')$, in which the function $I'\Phi_n'(\omega')$ is approximately the same for all spins, and the accuracy of this law of corresponding states seems to increase with the increase in the dimensionality.

I. INTRODUCTION

RECENTLY, Carboni and Richards[1] (CR) have extended their *ab initio* numerical calculations of the frequency Fourier transform of the self-correlation[2] function for a one-dimensional quantum-mechanical spin system with $S=\frac{1}{2}$ (we shall use Dirac's units, where $\hbar=1$). Available now is also the time-dependent self-correlation function (i.e., rather than only a histogram version of its frequency Fourier transform), the frequency Fourier transforms of the nearest-neighbor and further-neighbor correlation functions, as well as the frequency and wave-vector–dependent Fourier transforms for a few selected values of $\mathbf{K}$. In view of the fact that these results are exact,[3] they provide an excellent testing ground of the accuracy and the validity of the various phenomenological approximations which in our opinion form the basis of much of the existing

* Supported by the Office of Naval Research, under Contract No. NOOO14-68-A-0264.

† Supported by an N.S.F. Predoctoral Fellowship.

‡ This work will form a part of a dissertation to be submitted to the Temple University in partial fulfillment of the requirements for the Ph.D. degree.

[1] F. Carboni and P. M. Richards, Phys. Rev. **177**, 889 (1969).

[2] F. Carboni and P. M. Richards, J. Appl. Phys. **39**, 967 (1968).

[3] In this regard, a few words of explanation are perhaps in order. The CR calculations in Refs. 1 and 2 are carried out exactly for a finite linear chain with periodic boundary conditions with the implicit stipulation that the finite system be in contact with a heat and angular momentum bath at temperature $T\rightarrow\infty$. Plausible extrapolation procedures are then used to predict the corresponding correlation functions in the thermodynamic limit, i.e., $N\rightarrow\infty$, in which the system possesses its own temperature T (T is then assumed to be infinite). Because our present interests are restricted to the consideration of the thermodynamic limit only, it is desirable to clarify the limitations of the extrapolated results, and in particular to have a feeling for how "exact" the various results are. In the opinion of CR, at the elevated temperature of interest, a meaningful thermodynamic description of the spin correlation functions can be achieved as long as clusters extending to at least three spins or more (away from the two spins under consideration) are considered exactly. With this criterion, the calculation of the self-correlation function begins to be meaningful when the total number of spins N in the array is equal to about six and larger. Therefore, from among the computations of CR, which utilize arrays of up to $N=10$, the results for the self-correlation may be considered to be the most exact, while by the same token the relative exactness of the results for the correlations of separated spins may be anticipated to be the smaller the larger the separation.

To make the above discussion more quantitative, it should be noted that CR used extrapolation procedures similar to those used by J. C. Bonner and M. E. Fisher, Phys. Rev. **135**, A640 (1964), who found that the ground-state energy as a function of N extrapolated to the limit $N=\infty$ to about 0.1% of the true thermodynamic limit. CR found that the extrapolation worked exceedingly well for the *self-correlation function* for $N=6$ to the highest N considered, i.e., $N=10$, except for the very small frequencies, i.e., $\omega\lesssim(0.1)I$. For correlation functions of separated spins, they do not give details of the adequacy of the extrapolation except for the implication that the special attention paid to the extrapolation of the self-correlation results was not used in the case of the separated correlations.

theoretical work on the in-equilibrium[4,5] phenomena in many-body systems. Specifically, these results offer a unique opportunity for verifying the adequacy of one of the most popular phenomenological approximations used for discussing the in-equilibrium effects in many-spin systems. This approximation consists of the introduction of a two-parameter Gaussian representation for the frequency-wave vector-dependent diffusivity, and it has been used to make statements about the nature of the dynamic effects obtaining both in the extreme paramagnetic regime and in the neighborhood of the second-order phase transition in Heisenberg spin systems.[6–8]

In this regard, it should be mentioned that in an earlier calculation we carried out a limited test of the validity of the two-parameter Gaussian representation of the generalized diffusivity by comparing its predictions for the self-correlation function of a one-dimensional $S=\frac{1}{2}$ Heisenberg spin system against the, then available, exact *ab initio* results of CR.[2] The good agreement of these results encourages us to enlarge the scope of the earlier calculation and to further test the adequacy of the Gaussian representation of the diffusivity by comparing its predictions against the more expanded program of *ab initio* calculations recently performed by CR.[1]

In addition to the above, there is also an important byproduct of this study, namely, the results for the dynamical correlations themselves. The exact computer studies of the type carried out by CR are expected to be painstaking and costly as the magnitude of the spin increases beyond $\frac{1}{2}$. Moreover, in two and three dimensions, the execution of the procedure even for spin $\frac{1}{2}$ would be prohibitive, except, of course, for the case of the so-called "classical spins" which we discuss below.

The essential usefulness of the concept of classical spins lies in the fact that when the magnitude of the quantum-spin vector becomes extremely large, the role of the spin quantization becomes diminished, and the spins begin to behave "classically." Thus the results for classical spins, and those for quantum spins, are expected to approach each other as S (the magnitude of the spins) approaches infinity. Therefore, the availability of the exceedingly accurate numerical results for the classical-spin case in three dimensions[9] offers yet another limiting behavior that we can use for testing our phenomenological construct of the generalized diffusivity and, in turn, to get a feel for the reliability of the results (i.e., the time-dependent correlation functions for finite-spin magnitudes) that we have said are the byproduct of the current analysis.

An outline of the salient details of the formulation are given in Sec. II. The results for an infinite linear chain (i.e., the one-dimensional case) are described in Sec. III and those for two- and three-dimensional systems are given in Secs. IV and V. The salient features of these results are briefly recapitulated in Sec. VI. The Appendix contains a mathematical detail regarding the calculation of the principal-parts integral needed in carrying out the calculations described in Secs. II–V.

II. FORMULATION

We shall adopt the Heisenberg model of magnetism which ascribes a localized spin to each lattice point of the d ($d=1, 2, 3$) dimensional crystal, interacting only with its nearest-neighbor spins through an isotropic exchange interaction. We assume that the spacing between the spins is uniform (for convenience, the distances will be measured in the units of this spacing) and that periodic boundary conditions apply. Furthermore, we assume that the spins are somehow not coupled to the lattice, or, alternatively, that it is sufficient to consider the strength of the exchange integral I to be some suitable scalar function of the temperature to describe any spin-lattice coupling.[10,11] The relevant Hamiltonian therefore is

$$\mathcal{H}=-\sum_{f_1,f_2} I(f_1f_2)\mathbf{S}_{f_1}\cdot\mathbf{S}_{f_2}, \tag{2.1}$$

where $\mathbf{S}_f$ is the spin vector associated with the lattice point f, and $I(f_1f_2)$ is the strength of the exchange interaction (being equal to I, when f_1 and f_2 are nearest neighbors and zero otherwise), which is a known implicit function of the temperature.

Let us define the spectral function $F_{\mathbf{K}}(\omega)$ which is related to the frequency–wave-vector Fourier transform of the correlation function:

$$F_K(\omega)=-F_K(-\omega)=-F_{-K}(-\omega)=F_{-K}(\omega)$$
$$=\frac{1}{2\pi}\sum_{f_1-f_2} e^{-i\mathbf{K}\cdot(\mathbf{f}_1-\mathbf{f}_2)}\int_{-\infty}^{+\infty}\langle[S_{f_1}^z(t),S_{f_2}^z(t')]_-\rangle \times e^{i\omega(t-t')}d(t-t'), \tag{2.2}$$

[4] The term "in-equilibrium" in the present context refers only to those situations where the departures from the equilibrium are exceedingly small, e.g., situations created by the application of infinitesimally small space-time-dependent external fields on systems in thermal equilibrium or, alternatively, to the regime of linear transport phenomena where gradients of the local thermodynamic potentials are infinitesimal (see, for example, Ref. 5 and references cited therein).

[5] P. C. Martin, in *1967 Les Houches Lectures*, edited by C. DeWitt and R. Balian (Gordon and Breach, Science Publishers, Inc., New York, 1968).

[6] H. S. Bennett and P. C. Martin, Phys. Rev. **138**, A608 (1965).

[7] H. S. Bennett, Phys. Rev. **174**, 629 (1968); R. Tahir-Kheli, J. Appl. Phys. **40**, 1550 (1969).

[8] R. A. Tahir-Kheli and D. G. McFadden, Phys. Rev. **178**, 800 (1969).

[9] C. G. Windsor, Proc. Phys. Soc. (London) **91**, 353 (1967).

[10] For describing dynamic effects, this assumption is likely to be an important restriction. The reason is that not only does the average interspin separation vary with the temperature, but the agitation of the lattice causes dynamic coupling between the spins and the lattice vibrations. It is hoped that the investigation of some of these effects will be the subject of a future study.

[11] This assumption is, however, often made in the literature. See, e.g., D. C. Mattis, *The Theory of Magnetism* (Harper and Row Publishers, Inc., New York, 1965); S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Plenum Press, Inc., New York, 1967), and references therein.

where $\sum_{f_1-f_2}$ indicates a single sum over all the relative position vectors f_1-f_2. The time dependence of the spin operators is in the Heisenberg representation with respect to the Hamiltonian $\mathcal{H}$, and the angular brackets, as usual, denote the statistical average over a canonical ensemble.

The retarded and advanced double-time Green's functions, i.e.,

$$M_{f_1,f_2}{}^{\mathrm{ret}}(t,t')=-i\Theta(t-t')\langle[S_{f_1}{}^z(t),S_{f_2}{}^z(t')]_-\rangle, \quad (2.3a)$$

$$M_{f_1,f_2}{}^{\mathrm{adv}}(t,t')=+i\Theta(t'-t)\langle[S_{f_1}{}^z(t),S_{f_2}{}^z(t')]_-\rangle, \quad (2.3b)$$

where Θ is the Heaviside unit step function, have the well-known spectral representation[12]

$$M_{\mathbf{K}}(Z)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\frac{F_{\mathbf{K}}(\omega)}{Z-\omega}d\omega, \quad (2.4)$$

where $M_{\mathbf{K}}(Z)$ is the analytic extension of the Fourier transforms $M_{\mathbf{K}}{}^{\mathrm{ret}}(E)$, and $M_{\mathbf{K}}{}^{\mathrm{adv}}(E)$;

$$M_{f_1,f_2}{}^{\alpha}(t,t')=\frac{1}{N}\sum_K e^{i\mathbf{K}\cdot(f_1-f_2)}\times\int_{-\infty}^{+\infty}M_{\mathbf{K}}{}^{\alpha}(E)e^{-iE(t-t')}dE, \quad \alpha\equiv\mathrm{ret,\ adv}, \quad (2.5)$$

into the upper and the lower half of the complex energy plane, respectively. The K sum in Eq. (2.5) is over the first Brillouin zone. Note that as long as we work with lattice structures which have inversion symmetry, we have the result $M_{\mathbf{K}}(Z)=M_{-\mathbf{K}}(+Z)$, and, because of the left-hand side of Eq. (2.2), we have the additional important result that $M_{\mathbf{K}}(Z)=M_{\mathbf{K}}(-Z)$.

A convenient spectral representation[6,13] of $M_{\mathbf{K}}(Z)$ is in terms of the generalized diffusivity $D_{\mathbf{K}}(\omega)$, i.e.,

$$M_{\mathbf{K}}(Z)=M_{\mathbf{K}}(0)\left[1-\left(1-\frac{1}{\pi}\int_{-\infty}^{+\infty}\frac{D_{\mathbf{K}}(\omega)}{Z^2-\omega^2}d\omega\right)^{-1}\right], \quad (2.6)$$

where $D_{\mathbf{K}}(\omega)=D_{-\mathbf{K}}(\omega)=D_{\pm\mathbf{K}}(-\omega)$. Equations (2.4) and (2.6) specify an exact relationship between the spectral function $F_K(\omega)$ and the diffusivity $D_K(\omega)$, i.e.,

$$\frac{F_{\mathbf{K}}(\omega)}{\omega}\bigg/\int_{-\infty}^{+\infty}\left(\frac{F_{\mathbf{K}}(\omega)}{\omega}\right)d\omega =\frac{D_{\mathbf{K}}(\omega)}{\pi}\bigg/\left[\omega^2\left(1-\frac{\mathcal{P}}{\pi}\int_{-\infty}^{+\infty}\frac{D_{\mathbf{K}}(\omega')}{\omega^2-\omega'^2}d\omega'\right)^2+[D_{\mathbf{K}}(\omega)]^2\right]. \quad (2.7)$$

The usefulness of the above representation, however, depends critically on whether the function $D_{\mathbf{K}}(\omega)$ turns out to be a simple function or not.

It should be remarked that in complete analogy with the above formulation, a variety of existing phenomenological theories of in-equilibrium phenomena can also be conveniently formulated in terms of the generalized diffusivity.[5,14] In these analyses, the usual choices for the functional form of the diffusivity are the two-parameter exponential[15] or the two-parameter Gaussian.[5–8,14,16] In view of our recent observation,[17] whereby a two-parameter Gaussian representation of the diffusivity led to very satisfactory results for the frequency Fourier transform of the self-correlation function for a one-dimensional spin-$\frac{1}{2}$ system, we shall in the present paper work only with the latter representation, i.e.,

$$D_{\mathbf{K}}(\omega)=\pi\Delta(\mathbf{K})\Gamma(\mathbf{K})e^{-\omega^2\Gamma^2(\mathbf{K})}, \quad (2.8)$$

where the choice of the coefficients $\Delta(k)$ and $\Gamma(k)$ is not arbitrary but rather is given by the requirement that it reproduce as many of the frequency moments of the spectral function $F_K(\omega)$ as possible. In addition to the simple sum rule $\langle\omega^0\rangle_{\mathbf{K}}=1$, which is exactly satisfied for all nontrivial choices of Δ and Γ in Eq. (2.8), the moments $\langle\omega^2\rangle_{\mathbf{K}}$ and $\langle\omega^4\rangle_{\mathbf{K}}$, where

$$\langle\omega^{2n}\rangle_{\mathbf{K}}=\int_{-\infty}^{+\infty}\left(\frac{F_{\mathbf{K}}(\omega)}{\omega}\right)\omega^{2n}d\omega\bigg/\left(\int_{-\infty}^{+\infty}[F_{\mathbf{K}}(\omega)/\omega]d\omega\right), \quad (2.9)$$

uniquely fix the coefficients $\Delta(\mathbf{K})$ and $\Gamma(\mathbf{K})$ through the following relations [which are readily derived from Eqs. (2.4) and (2.6)]:

$$D_{\mathbf{K}}{}^{(0)}=(\sqrt{\pi})\Delta(\mathbf{K})=\langle\omega^2\rangle_{\mathbf{K}}, \quad (2.10)$$

$$D_{\mathbf{K}}{}^{(2)}=(\sqrt{\pi})(\Delta(\mathbf{K})/2\Gamma^2(\mathbf{K}))=\langle\omega^4\rangle_{\mathbf{K}}-[\langle\omega^2\rangle_{\mathbf{K}}]^2, \quad (2.11)$$

where

$$D_{\mathbf{K}}{}^{(2n)}=\frac{1}{\pi}\int_{-\infty}^{+\infty}D_{\mathbf{K}}(\omega)\omega^{2n}d\omega. \quad (2.12)$$

It should be emphasized that insofar as the representation (2.8) is approximate, it is not expected to preserve the higher frequency moments exactly. Indeed, a useful measure of the accuracy of this representation would be to compare the exact result[18] for the next

[12] D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

[13] L. P. Kadanoff and P. C. Martin, Ann. Phys. (N. Y.) **24**, 419 (1963); R. A. Tahir-Kheli, Phys. Rev. **159**, 439 (1967).

[14] P. C. Martin and S. Yip, Phys. Rev. **170**. 151 (1968).

[15] B. J. Berne, J. P. Boon, and S. A. Rice, J. Chem. Phys. **45**, 1086 (1966).

[16] K. S. Singwi and M. P. Tosi, Phys. Rev. **157**, 153 (1967).

[17] Note that the remarks made in Ref. 8 concerning the structure of the results for the correlation functions referring to large spins, i.e., $S\gg1$, are irrelevant in view of the discovery of the reduced scale in which these correlations obey an approximate law of corresponding states. See also Ref. 20.

[18] The computation of the moment $\langle\omega^6\rangle_{\mathbf{K}}$ is a very tedious business and, as far as we are aware, has not so far been carried out. The present authors have recently undertaken its computation.

higher moment, $\langle\omega^6\rangle_{\mathbf{K}}$ with the approximate one, $(\langle\omega^6\rangle_{\mathbf{K}})_{\text{app}}$ computed from Eqs. (2.6) and (2.8), i.e.,

$$(\langle\omega^6\rangle_{\mathbf{K}})_{\text{app}}=\left(\frac{3\Delta(\mathbf{K})\sqrt{\pi}}{4\Gamma^4(\mathbf{K})}\right)+\left(\frac{\pi\Delta^2(\mathbf{K})}{\Gamma^2(\mathbf{K})}\right)+\pi^{3/2}\Delta^3(\mathbf{K}). \quad (2.13)$$

Finally, in this section it remains to be recorded that once, through the artifice of the phenomenological construct for the diffusivity, the spectral function $F_K(\omega)$ is calculated [see Eq. (2.7)], the correlation function is readily obtained,[12] i.e.,

$$\langle S_0^z(t)S_n^z(0)\rangle\equiv\Phi_n(t)=\int_{-\infty}^{+\infty}\Phi_n(\omega)e^{-i\omega t}d\omega=\frac{1}{N}\sum_{\mathbf{K}}\int_{-\infty}^{+\infty}\frac{F_{\mathbf{K}}(\omega)e^{-i(\mathbf{K}\cdot\mathbf{R}_n+\omega t)}}{1-e^{-\beta\omega}}d\omega. \quad (2.14)$$

(For convenience, the subscript n will denote both the spatial location $\mathbf{R}_n$ and the order of the neighbor; e.g., $n=1$ denotes nearest-neighbor correlation.)

At the elevated temperatures of interest, an adequate approximation is to replace the denominator of the integrand in Eq. (2.14) by the first term in the expansion, i.e., $(1-e^{-\beta\omega})\sim\beta\omega$. Moreover, since the wave-vector-dependent susceptibility $\int_{-\infty}^{+\infty}[F_{\mathbf{K}}(\omega)/\omega]d\omega$ is readily evaluated to have the form

$$\int_{-\infty}^{+\infty}\left(\frac{F_{\mathbf{K}}(\omega)}{\omega}\right)d\omega=\tfrac{1}{3}\beta S(S+1)[1+o(\beta J)]. \quad (2.15)$$

Therefore, at infinite temperatures, i.e., $\beta\rightarrow 0$, the correlation function

$$\Phi_n(\omega)=\frac{1}{N}\sum_{\mathbf{K}}\cos(\mathbf{K}\cdot\mathbf{R}_n)\Phi(\mathbf{K},\omega) \quad (2.16)$$

is given by the relation [see Eq. (2.7)]

$$\lim_{T\rightarrow\infty}[\Phi(\mathbf{K},\omega)]=\frac{F_{\mathbf{K}}(\omega)}{\beta\omega}=\frac{[S(S+1)/3\pi]D_{\mathbf{K}}(\omega)}{\omega^2[1+Q(\omega)]^2+[D_{\mathbf{K}}(\omega)]^2}, \quad (2.17)$$

where

$$Q(\omega)=+\left(\frac{1}{\pi}\right)\mathcal{P}\int_{-\infty}^{+\infty}\frac{D_{\mathbf{K}}(\omega')}{\omega'^2-\omega^2}d\omega'. \quad (2.18)$$

III. RESULTS IN ONE DIMENSION

The exact calculation of the moments $\langle\omega^2\rangle_{\mathbf{K}}$ and $\langle\omega^4\rangle_{\mathbf{K}}$ at infinite temperatures has been performed by Marshall and by others,[19] and for the one-dimensional case the results are

$$\langle\omega^2\rangle_K=(2I)^2\tfrac{4}{3}S(S+1)(1-\cos K_x), \quad (3.1)$$

$$\langle\omega^4\rangle_K=(2I)^4(8/9)[S(S+1)]^2(1-\cos K_x)\times\{5-3\cos K_x-\tfrac{3}{4}[S(S+1)]^{-1}\}. \quad (3.2)$$

Inserting these values for $\langle\omega^2\rangle_K$ and $\langle\omega^4\rangle_K$ into Eqs. (2.10) and (2.11) and using Eqs. (2.8), (2.17), and (A6), the Fourier transforms $\Phi_n(\omega)$ for $n=0$, 1, 2, 3, and $\Phi(\mathbf{K},\omega)$ are determined by numerical integration for various values of the spin S in the range $S=\frac{1}{2}$ and $S=\infty$. It turns out that in the reduced unit scale

$$I'\rightarrow I[S(S+1)]^{1/2},\quad \Phi_n'(\omega)\rightarrow[S(S+1)]^{-1}\Phi_n(\omega),$$

the plots of $I'\Phi_n'(\omega)$ versus $\omega'=\omega/I'$ have only a small relative spread as S varies between the extreme limits $S=\frac{1}{2}$ and $S=\infty$.

The above behavior is, of course, not entirely unexpected. The structure of the zeroth, second, and fourth moments [see Eqs. (2.15), (3.1), and (3.2)] is clearly indicative of a law of "corresponding states" in terms of the reduced quantities I', $\Phi_n'(\omega)$, and ω', except for a slight additional spin dependence of the fourth moment through the occurence of the factor $\frac{3}{4}[S(S+1)]^{-1}$ in the last term on the right-hand side of Eq. (3.2). We believe that this reduced scale correspondence offers an explanation for the observation made by CR,[1] namely, that the classical-spin results of Windsor,[20] which are traditionally plotted on a reduced scale of the sort $I''\rightarrow JS$, $\Phi_n''(\omega)\rightarrow\Phi_n(\omega)(S)^{-2}$, and $\omega''\rightarrow\omega/I''$, can be made to yield results very close to the CR results for spin $\frac{1}{2}$ by the transformation[20] $S\rightarrow\frac{1}{2}(3)^{1/2}$. [Note that the classical spins have the kinematic restriction $\mathbf{S}\cdot\mathbf{S}=S^2$ in contrast to $\mathbf{S}\cdot\mathbf{S}=S(S+1)$ for the quantum spins.]

The results for spin $\frac{1}{2}$ are compared with the exact results given by CR (see Figs. 1–11). In our opinion, the agreement of our results for $\Phi_n(\omega)$ with the exact results is excellent for the self-correlation $\Phi_0(\omega)$, good for the nearest-neighbor $\Phi_1(\omega)$ and the next nearest-neighbor correlation $\Phi_2(\omega)$, and barely adequate for $\Phi_3(\omega)$ and $\Phi_4(\omega)$. There can, of course, be only three possible reasons for this, namely, that either our formulation generates less accurate results for larger spin separations, or the CR results are becoming rapidly less exact with the increase in the spatial separation of the spins (for example, compare Ref. 3), or, alternatively, both these results are inaccurate for $n\gtrsim 3$.

To investigate further this interesting question, we compare our results for transforms $\Phi(\mathbf{K},\omega)$ with the cor-

[19] P. G. de Gennes, J. Phys. Chem. Solids 4, 223 (1958). The errors in this calculation were corrected and the results extended to close-packed lattices by W. Marshall, in *Critical Phenomena*, edited by M. S. Green and J. V. Sengers (National Bureau of Standards Misc. Publ. 273, 1966). See also M. F. Collins and W. Marshall, Proc. Phys. Soc. (London) **92**, 390 (1967).

[20] Note that since the classical-spin results are appropriate to the limiting case $S\rightarrow\infty$, one might have expected that the mere use of the transformation $S^2\rightarrow\frac{3}{4}$ would not be sufficient to yield reasonable results appropriate to the spin-$\frac{1}{2}$ case. However, our observation that the results for the infinite-spin case are not too different (the differences becomes even smaller in two and three dimensions) when plotted in the reduced scale seems to explain the good fit obtained by CR with the results of M. Windsor (unpublished).

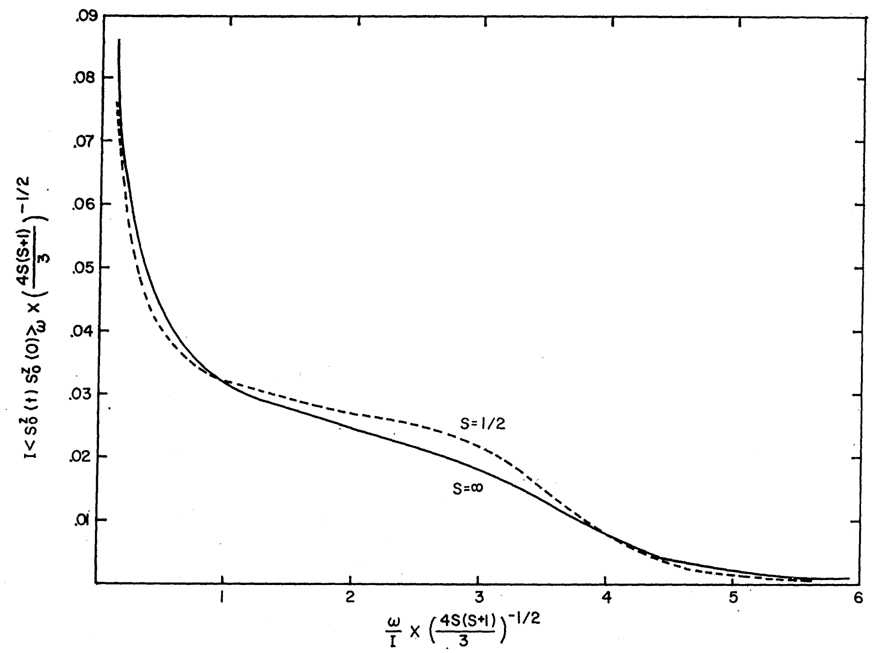

FIG. 1. Plot of the frequency Fourier transform of the self-correlation in one dimension versus the frequency in the reduced units. Note the similarity of the results for $S=\frac{1}{2}$ and $S=\infty$. The results for intermediate values of S lie in between the two curves.

responding ones supplied by CR, i.e., for $K=[(2/9)\pi p]$, $p=1$, 2, 3, and 4 (see Figs. 6–9). To our surprise, we find that the agreement between our $\Phi(\mathbf{K},\omega)$ and the results of CR is excellent for the smaller $|\mathbf{K}|$'s given by CR, i.e., $|\mathbf{K}|=(2/9)\pi$ and $(4/9)\pi$, whereas it is barely adequate for $|\mathbf{K}|=(6/9)\pi$ and $(8/9)\pi$. The reason why this result is unexpected in the light of our previous observation is, of course, the following: The correlations with large spatial separation were expected to contribute dominantly to small K Fourier transforms $\Phi(\mathbf{K},\omega)$, and therefore the agreement of the CR results with ours should be relatively less satisfactory for small K's. Similarly, since the correlations for smaller spatial separations are expected to dominantly determine the transform $\Phi(\mathbf{K},\omega)$ for large K values the opposite should be the case for these.

It should be added that so far the present authors have not found a satisfactory resolution of this dilemma.

IV. RESULTS IN TWO DIMENSIONS

The motivation for studying the nature of the space- and time-dependent correlation function in two dimensions is twofold: Firstly, it is of interest to know whether or not the structure of the results, especially the non-Gaussian nature of the spectral line-shapes [i.e., the Fourier transforms $\Phi(\mathbf{K},\omega)$ for very small and large $|\mathbf{K}|$'s and $\Phi_n(\omega)$] is a critical function of the lattice dimensionality. Secondly, by analogy with the case of

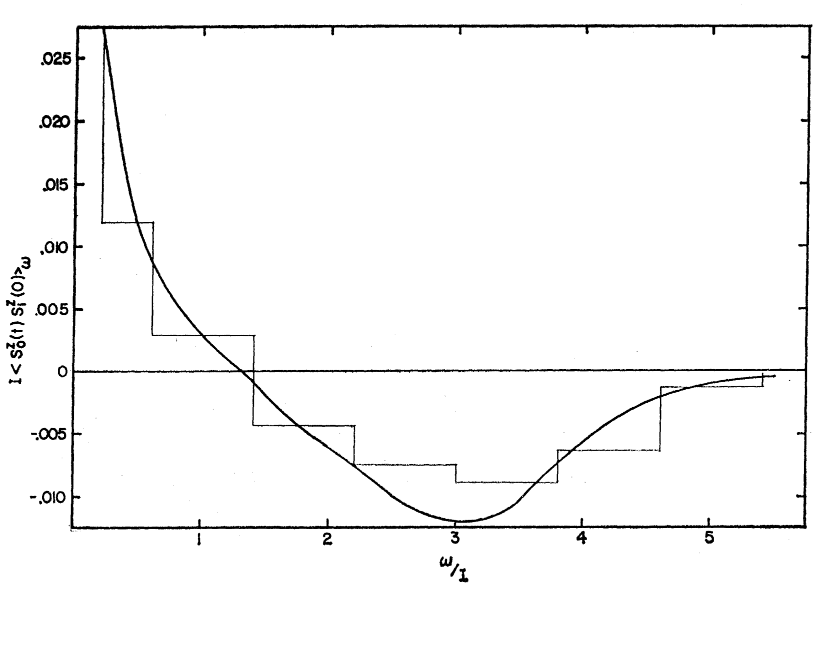

FIG. 2. Comparison of our results (given as the continuous curve) for the frequency Fourier transform of the nearest-neighbor correlation in one dimension for spin $\frac{1}{2}$ with the corresponding exact results of CR (given as the histogram).

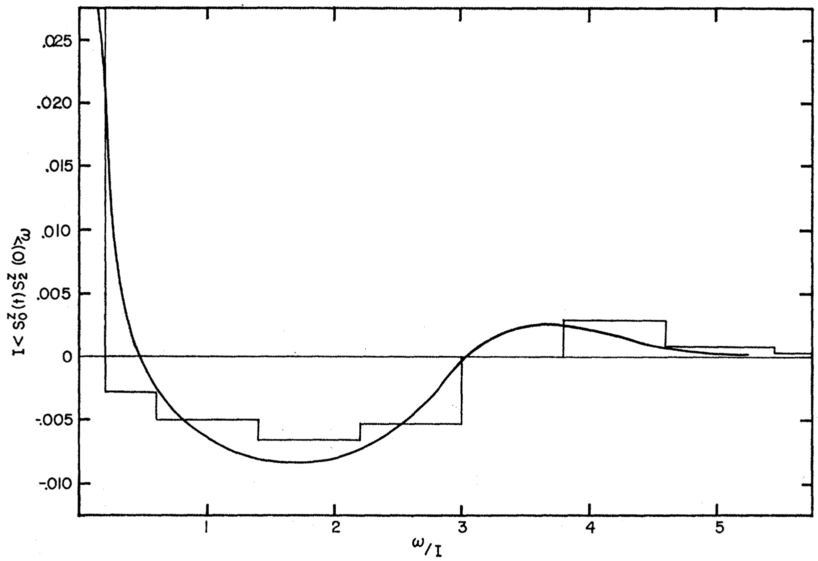

FIG. 3. Continuous curve gives our results for the frequency Fourier transform of the second-neighbor correlation for $S=\frac{1}{2}$ in one dimension. The corresponding exact results of CR are plotted as a histogram.

one dimension, where a reasonably good experimental realization of a Heisenberg linear chain of exchange-coupled spins exists in the form[21] of $Cu(NH_3)_4SO_4\cdot H_2O$, and which therefore makes the theoretical study of the spectral line shapes physically relevant,[1,22] the recent efforts to find satisfactory experimental systems corresponding to the two-dimensional model of the exchange-coupled Heisenberg spin system have to a certain extent succeeded.[23] To this extent, the present study may also be considered to be physically motivated.

For simplicity, we shall restrict our consideration to the simplest two-dimensional lattice, the square net. If desired, these results may be extended to other lattice structures with only slight modification in the algebra.

The frequency moments for this system at infinite temperature are

$$\langle\omega^2\rangle_K=(2I)^2\tfrac{4}{3}S(S+1)[2-\cos K_x-\cos K_y], \tag{4.1}$$

$$\langle\omega^4\rangle_K=(2I)^4(8/3)[S(S+1)]^2[2-\cos K_x-\cos K_y]\times\{4-\cos K_x-\cos K_y-[4S(S+1)]^{-1}\}. \tag{4.2}$$

Using Eqs. (4.1) and (4.2) and following the procedure described in the previous sections, we determine the parameters $\Delta(\mathbf{K})$ and $\Gamma(\mathbf{K})$; consequently, through the use of Eqs. (2.8) and (A6), we write down the

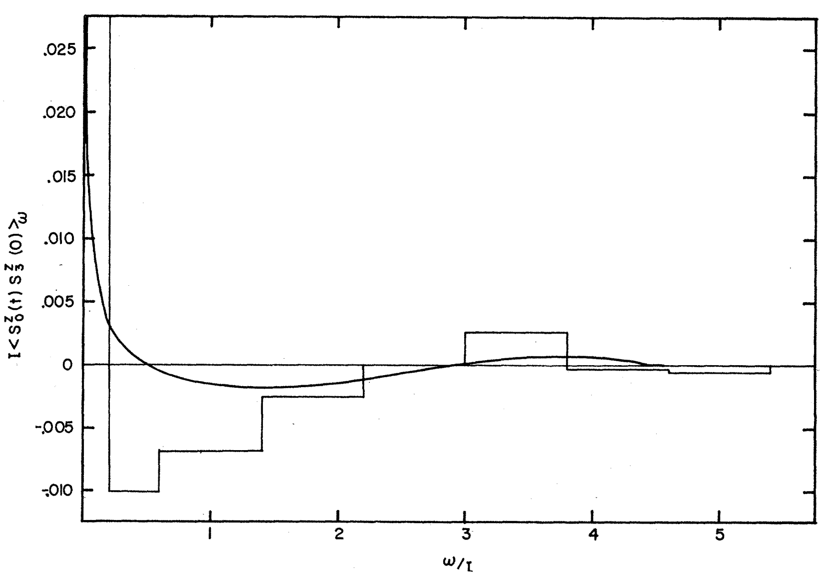

FIG. 4. As in Figs. 2 and 3, our results (continuous curve) for the third-neighbor correlation for $S=\frac{1}{2}$ in one dimension are compared with the CR results. The quantitative agreement is unsatisfactory in the range of small ω, i.e., $\omega\lesssim I$.

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[22] See, e.g., R. N. Rogers, F. Carboni, and P. M. Richards, Phys. Rev. Letters **19**, 1016 (1964).
[23] M. E. Lines, J. Appl. Phys. **40**, 1352 (1969).

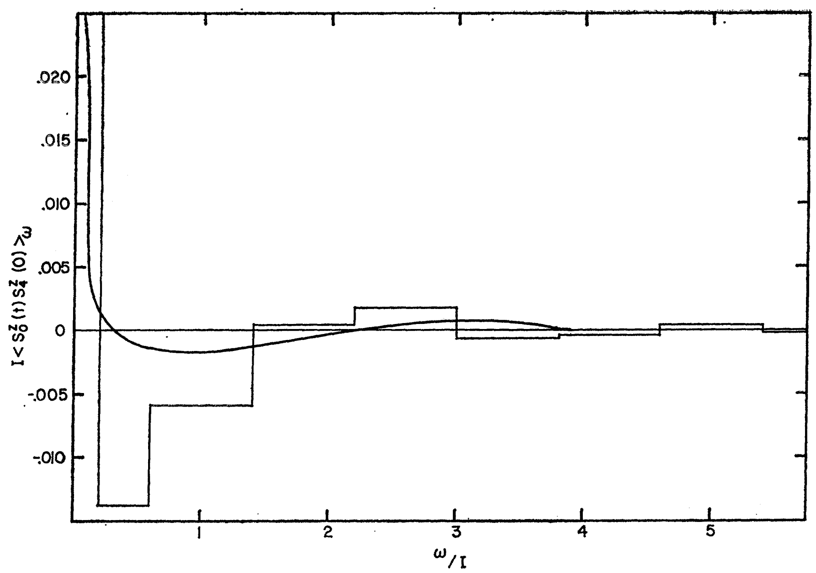

FIG. 5. Our results (continuous curve) for the frequency Fourier transform of the fourth-neighbor correlation in one dimension for spin $\frac{1}{2}$ are compared with the CR results. The quantitative agreement of these results is poor in the range of small-small ω, i.e., $\omega \lesssim I$ and is probably somewhat worse than it was in the case of third-neighbor correlation (see Fig. 4). As mentioned in the text, there are three possible explanations for this, namely, that *either* our theory is inadequate in describing correlations of larger spatial separation *or* the corresponding results of CR (the reliability of which crucially depends upon the accuracy of the extrapolation procedure used for predicting the results in the thermodynamic limit $N=\infty$ from their exact finite linear-chain results) are less reliable for the third and the fourth nearest-neighbor correlations, or a bit of both these statements is true.

summand given in the right-hand side of Eq. (2.17). Now we exploit the symmetry of the square Brillouin zone, noting that its boundary is square, and K_x and K_y vary between the limits $-\pi$ to $+\pi$. In this manner, we calculate the correlations $\Phi_n(\omega)$. The self-correlation, i.e., $n=0$ and the nearest-neighbor correlation, i.e., $n=1$, are the only ones we study here, but, if desired, further neighbor correlations can also be calculated readily in the same fashion.

In Fig. 12 we give a plot of $I'\Phi'_0(\omega')$ versus ω' (primes indicate the reduced units) for the minimum, i.e., $\frac{1}{2}$, and the maximum, i.e., ∞, values of the spin S. Three points are worth noting. Firstly, the divergence of $\Phi_0(\omega')$ as $\omega' \to 0$ is much less pronounced in two dimensions than is the case in one dimension (compare Fig. 1). Secondly, the curves for $S=\frac{1}{2}$ and $S=\infty$ are closer together in two dimensions than in one. Thirdly, the cutoff is not as sharp and dramatic in two dimensions as was observed to be the case in one dimension and occurs at a higher value.

Figure 13 depicts the nearest-neighbor correlation $\Phi_1(\omega)$ for spin $\frac{1}{2}$ in two dimensions. The results are mildly similar to the corresponding ones in one dimension (see Fig. 2) except for the general features of the

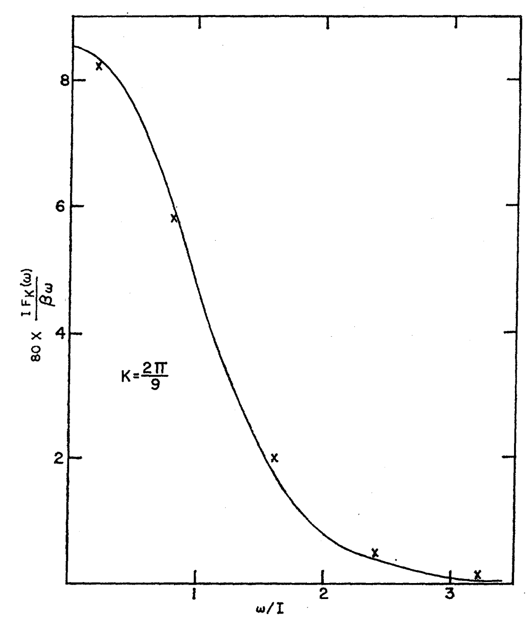

FIG. 6. Continuous curve depicts our one-dimensional spin-$\frac{1}{2}$ results for $\Phi(\mathbf{K},\omega)=F_{\mathbf{K}}(\omega)/\beta\omega$ with $K=2\pi/9$. [The ordinate is $80\Phi(\mathbf{K},\omega)I$ and the abscissa is ω/I.] The crosses indicate the corresponding results obtained by CR.

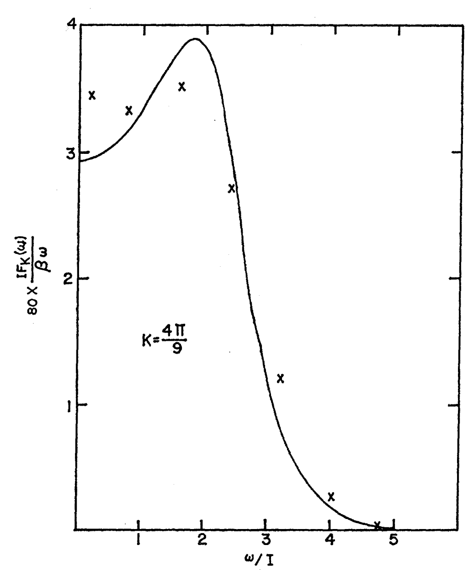

FIG. 7. Plot of our one-dimensional spin-$\frac{1}{2}$ results for (80 times) $I\Phi(\mathbf{K},\omega)$, with $K=4\pi/9$ versus ω/I. The crosses indicate the CR results. Note that while the quantitative agreement of these results for small ω, i.e., $\omega \lesssim I$, is no longer as good as was the case in Fig. 6, they do demonstrate (as already noted in Ref. 1) that $\Phi(K,\omega)$ is not a monotonically decreasing function of ω when K becomes bigger than roughly about a third of the zone boundary.

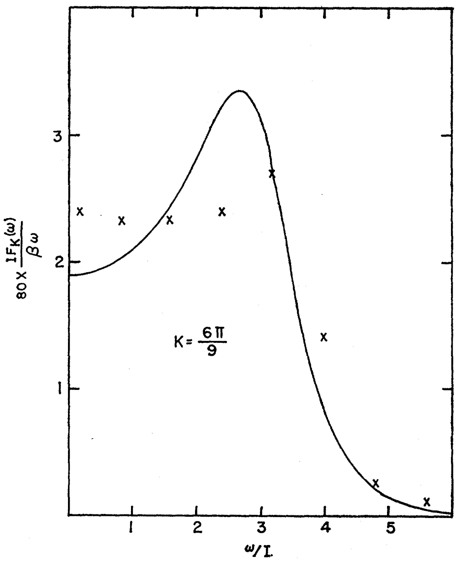

FIG. 8. Plot of our one-dimensional spin-½ results for $80I\Phi(\mathbf{K},\omega)$ versus (ω/I) for $|\mathbf{K}|=6\pi/9$. The crosses indicate the CR results. The trend noted in Fig. 7 persists, namely, that the quantitative agreement of these results is deteriorating for small ω values.

differences noted in the first and third observations made in the preceding paragraph and the fact that these differences are somewhat less pronounced for the neighboring correlations than is the case for the self-correlations.

V. THREE DIMENSIONS

For convenience, we shall once again treat only the simplest lattice, i.e., the simple-cubic (sc) lattice. The

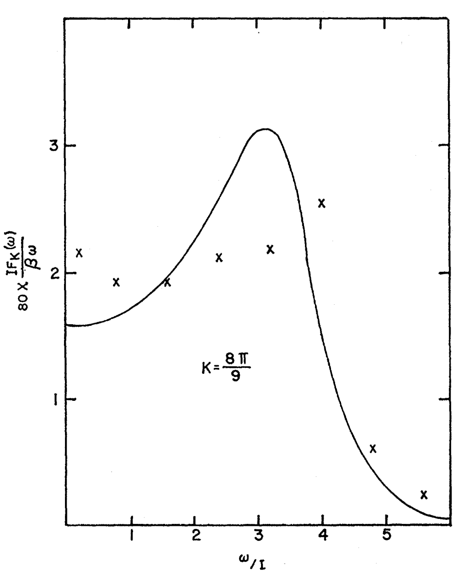

FIG. 9. Analogously to Figs. 7 and 8, the plot shows results for $80I\Phi(\mathbf{K},\omega)$ versus ω/I for a one-dimensional spin-½ system for $|\mathbf{K}|=8\pi/9$. The quantitative agreement with the corresponding results of CR (shown as crosses) is no longer adequate—except at large frequencies. Notice that all $\Phi(\mathbf{K},\omega)$ (compare Figs. 6–9) seem to cut off before ω gets to about $6I$. Moreover, the non-Gaussian characteristics of $\Phi(\mathbf{K},\omega)$ should be noted.

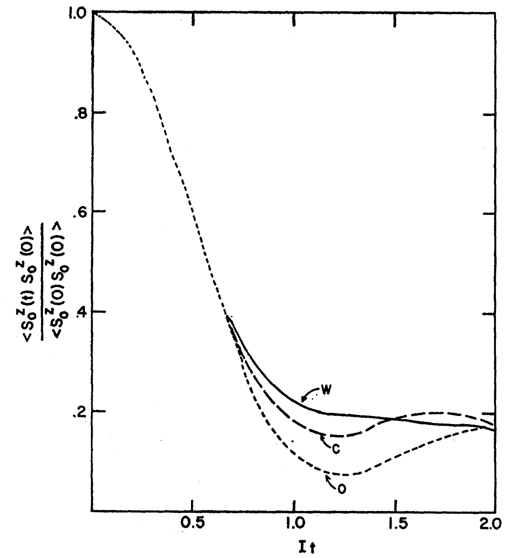

FIG. 10. Plot of $\Phi_0(t)/\Phi_0(0)$ versus It for one-dimensional spin-½ system. The broken curves marked C and O are the CR and our results, respectively. The full curve marked W has been taken from Ref. 1 and represents a transcription of Windsor's unpublished classical-spin results (which are appropriate to $S\rightarrow\infty$) to the spin-½ case through the transformation $S=\frac{1}{2}\sqrt{3}$.

results for other loose packed (three-dimensional) lattices are likely to be qualitatively similar to these.

The infinite temperature-frequency moments for the sc lattice with nearest-neighbor exchange are

$$\langle\omega^2\rangle_K=(2I)^2\tfrac{4}{3}S(S+1)\times(3-\cos K_x-\cos K_y-\cos K_z), \qquad (5.1)$$

$$\langle\omega^4\rangle_K=(2I)^4(8/3)[S(S+1)]^2\times(3-\cos K_x-\cos K_y-\cos K_z)\times\{19/3-[4S(S+1)]^{-1}-\cos K_x-\cos K_y-\cos K_z\}. \qquad (5.2)$$

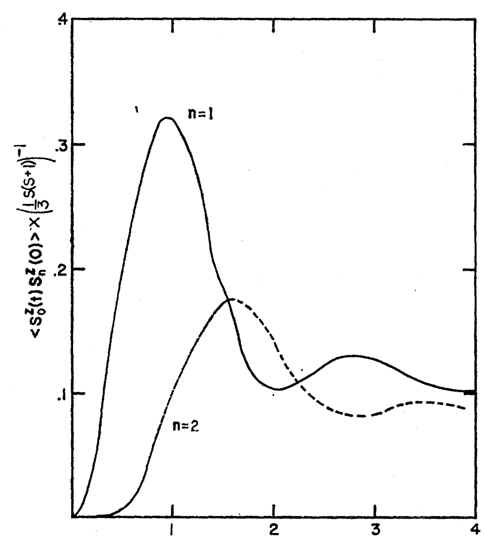

FIG. 11. Time-dependent nearest-neighbor and next-nearest-neighbor results for $S=\frac{1}{2}$ one-dimensional system.

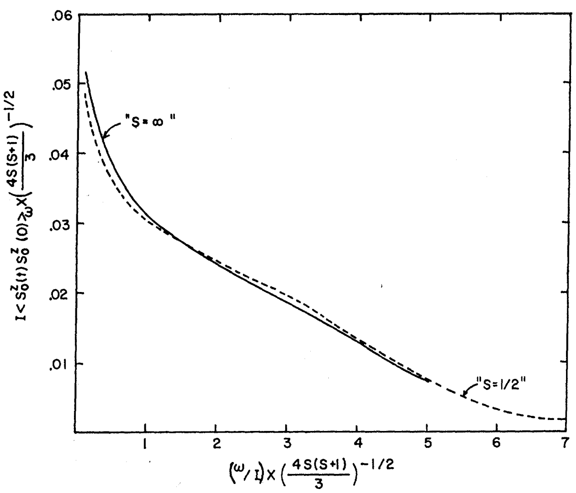

FIG. 12. Plot of $I\Phi_0'(\omega)$ versus ω' in a two-dimensional square lattice. The continuous curve corresponds to the infinite-spin case (this result should be compared with the corresponding classical-spin calculations of Windsor whenever they become available). The broken curve refers to spin $\frac{1}{2}$. Compare with the corresponding reduced unit plots for one dimension given in Fig. 1 and note the somewhat closer agreement of the two curves in two dimensions.

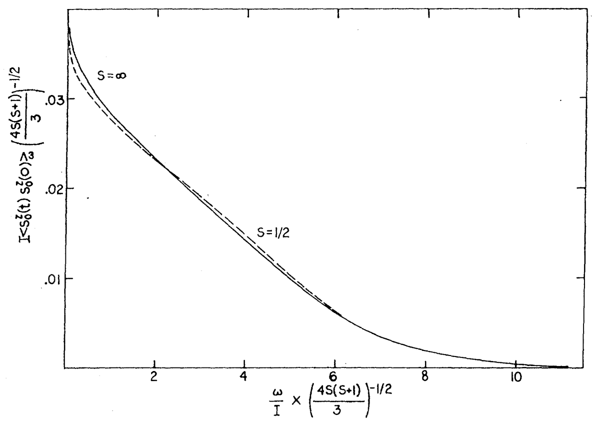

FIG. 14. Reduced unit plot of the self-correlation function in three dimensions (i.e., in a sc lattice). The infinite-spin results (full curve) are even closer to the $S=\frac{1}{2}$ results (dashed curve) in three dimensions than in two (compare Figs. 1 and 12). Note also that in striking contrast with the corresponding results in one and two dimensions, $\Phi_0(\omega)$ remains as $\omega\rightarrow 0$. In fact, for $\omega=0$, $[\frac{4}{3}S(S+1)]^{-1/2}I\langle S_0^z(t)S_0^z(0)\rangle_{(\omega)}=0.0367$ and 0.0383 for spin $\frac{1}{2}$ and ∞, respectively.

The structure of the Brillouin zone for this system is once again very simple, i.e., K_x, K_y, and K_z vary between $-\pi$ and $+\pi$, and the correlations of interest are computed by following the previously described procedure and exploiting the symmetry of the integrand and the Brillouin zone.

The self-correlation function is plotted in Fig. 14 for spins $S=\frac{1}{2}$ and $S=\infty$ in the reduced unit scale. These three-dimensional results differ strikingly from the corresponding results in one and two dimensions in the fact that the transform $\Phi_0(\omega)$ remains finite as $\omega\rightarrow 0$. Moreover, in keeping with the trend noticed earlier, the cutoff occurs at a higher frequency, and the results for $S=\frac{1}{2}$ and $S=\infty$ are closer together here than was the case in one and two dimensions. The results for the nearest-neighbor correlation $\Phi_1(\omega)$ are plotted in Fig. 15, and when compared and contrasted with the corresponding results in one and two dimensions (i.e., Figs. 2 and 13) they too confirm the general trend noted earlier.

In Figs. 16 and 17 we compare our spin-∞ results with the corresponding computer experiments results of Windsor.[9] (Note that it is essential to use the reduced scale for this purpose.) The agreement of these results is generally quite good.

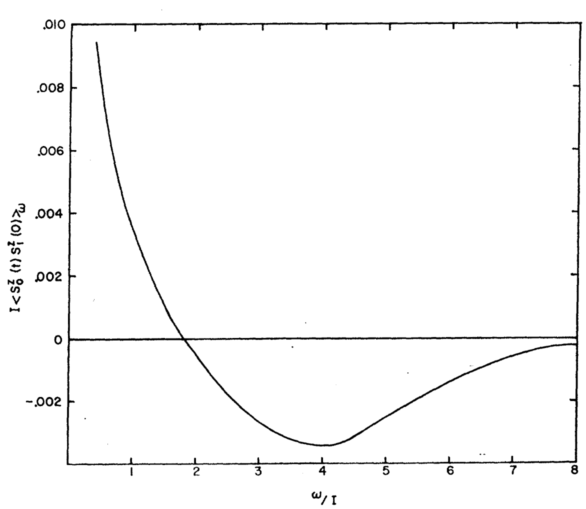

FIG. 13. Nearest-neighbor correlation $\Phi_1(\omega)$ in two dimensions for spin $\frac{1}{2}$. Compare this plot with the corresponding result in one dimension given in Fig. 2 and note their general similarity except for the fact that the relative scale of events in two dimensions is extended in frequency by about 30–50%. For instance, note the cutoff frequencies and also the frequency at which $\Phi_1(\omega)$ attains its minimum value.

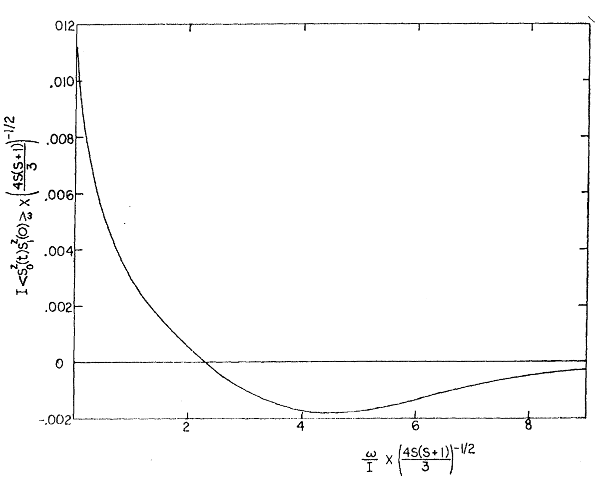

FIG. 15. Nearest-neighbor correlation $\Phi_1(\omega)$ for spin ∞ in the sc lattice.

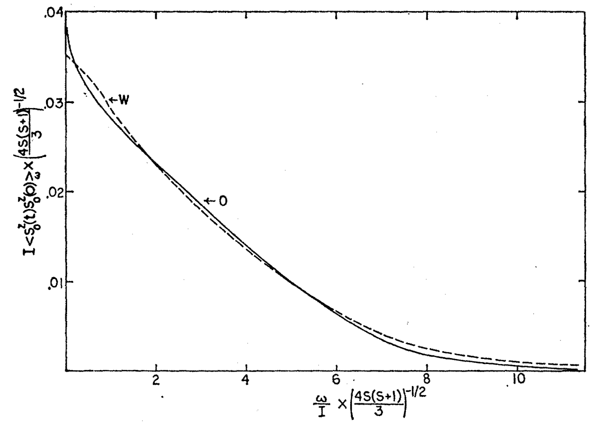

FIG. 16. Windsor's computer experiment results for the self-correlation $\Phi_0(\omega)$ for classical spins of magnitude $S=\infty$ in a sc lattice (dashed curve) are compared with our results in the corresponding limit (the plot is in the reduced units). The agreement is excellent.

VI. CONCLUSIONS

The phenomenological two-parameter Gaussian construct for the generalized diffusivity seems to be an adequate approximation for calculating the dynamic properties of Heisenberg spin systems at elevated temperatures. This conclusion is based on the following observations.

(a) In one dimension, the steep rise and the finite cutoff frequency of the Fourier transform $\Phi_n(\omega)$ is adequately reproduced by this procedure. Moreover, the agreement between the present results for spin $\frac{1}{2}$ and the exact corresponding calculations of CR is largely satisfactory. Nevertheless, it should be recorded that some quantitative discrepancies between the predicted Fourier transforms $\Phi(\mathbf{K},\omega)$ and those computed by CR *are* observed for large $|\mathbf{K}|$'s. Similarly, the agreement of the corresponding results for $\Phi_n(\omega)$, $n\gtrsim 3$, is mainly qualitative.

(b) In three dimensions, our predicted results for $S\rightarrow\infty$ are in very satisfactory agreement with the computer-experiment results of Windsor for classical spins.

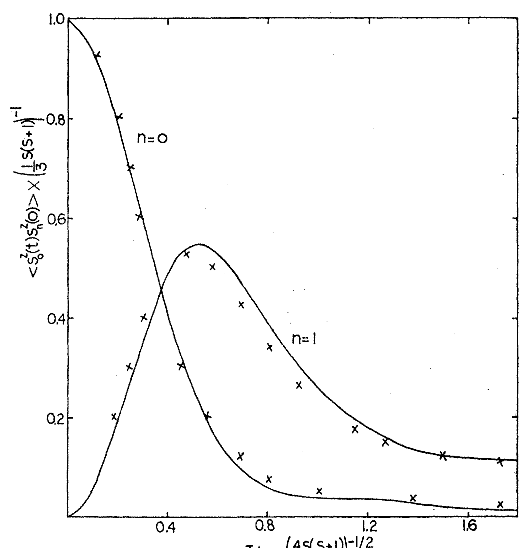

FIG. 17. Plots of the time-dependent self- and the nearest-neighbor correlation in a sc lattice in the reduced units for classical spins of magnitude ∞ (the crosses indicate Windsor's computer experiment results for classical spins). For convenience of display the magnitude of the nearest-neighbor correlation has been multiplied by a factor of 6. Our results for $S=\infty$ are given as continuous curves. The agreement is very good for the self-correlation. The relative scatter between the corresponding results for the nearest-neighbor correlation is somewhat greater.

It is therefore believed that the present calculations are a meaningful first estimate of the structure of the space-time-dependent correlation functions. In particular, our results for $S>\frac{1}{2}$ in one dimension, for $S\geq\frac{1}{2}$ in two dimensions, and those for quantum spins in three dimensions (i.e., all those cases for which no reliable *ab initio* or computer-experiment type of calculations are available) should be of value in analyzing the relevant experiments.

ACKNOWLEDGMENT

We are greatly indebted to the Temple University Computer Center for their generous allotment of computer time on their CDC 6400.

APPENDIX

The principal part integral $Q(\omega)$ [see Eq. (2.18)] is readily calculated when the diffusivity is represented by the two-parameter Gaussian given in Eq. (2.8). To see this, let us rewrite $Q(\omega)$ as

$$Q(\omega)=-\left(\frac{\Delta\Gamma}{2\pi\omega}\right)\mathcal{P}\int_{-\infty}^{+\infty}e^{-\omega'^2\Gamma^2}\left(\frac{1}{\omega-\omega'}+\frac{1}{\omega+\omega'}\right)d\omega'. \quad \text{(A1)}$$

The two terms in the right-hand side of (A1) are clearly equal, as can be ascertained by changing the variable from $\omega'\rightarrow-\omega'$. The substitution $\Gamma(\omega'-\omega)=x$ now transforms $Q(\omega)$ to the following:

$$Q(\omega)=(\Delta\Gamma/\pi\omega)U(y), \quad \text{(A2)}$$

$$U(y)=-U(-y)=\mathcal{P}\int_{-\infty}^{+\infty}\frac{e^{-(x+y)^2}}{x}dx. \quad \text{(A3)}$$

The function $U(y)$ satisfies the first-order differential equation

$$dU/dy+2yU=-2\sqrt{\pi}, \quad \text{(A4)}$$

which, consistent with the boundary condition derived from Eq. (A3), i.e., $U(0)=-U(0)=0$, has the solution

$$U(y)=-(2\sqrt{\pi})e^{-y^2}\int_0^y e^{x^2}dx. \quad \text{(A5)}$$

Therefore, $Q(\omega)$ has the result

$$Q(\omega)=-\left(\frac{2\Delta(\mathbf{K})\Gamma(\mathbf{K})}{\omega\sqrt{\pi}}\right)e^{-\omega^2\Gamma^2(\mathbf{K})}\int_0^{\omega\Gamma(\mathbf{K})}e^{x^2}dx. \quad \text{(A6)}$$