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SPACE-TIME MAGNETIC SPIN CORRELATIONS OF ANISOTROPIC HEISENBERG SYSTEMS BY THE METHOD OF MOMENTS OF A DIFFUSIVITY-SPECTRAL REPRESENTATION

Frederick A. Malinoski

Franford Arsenal
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The longitudinal and transverse time-dependent spin correlations of Heisenberg spin systems with cylindrical symmetry and with both uniaxial and exchange anisotropy are evaluated in the limit of elevated temperature, by the method of frequency moments. The zeroth, second, and fourth frequency moments are determined by thermal expansions of commutators of equal-time spin operators and their time derivatives. The frequency wave-vector spectral functions are established by these moments via the hydrodynamical representation, based on a two-parameter Gaussian diffusivity. The moment equations in a generic form apply for all spins, for arbitrary range of the exchange interactions, and for arbitrary dimensionality and lattice structures and are particularized with next nearest neighbor interactions for a linear chain, a two-dimensional net and a simple cubic lattice. The effects on the correlation functions of varying the axial anisotropy, the exchange anisotropy, and the interaction range are investigated. The resulting time-dependent correlations and their Fourier transforms are calculated, exhibiting features predicted by theoretical considerations and pointing to several conclusions; i.e., (1) The effect of increasing the axial anisotropy upon the longitudinal correlations is similar to that of decreasing the exchange anisotropy; (2) In contrast, the behavior of the transverse correlations is similar for increasing axial anisotropy or for increasing exchange anisotropy; (3) The anisotropic effects on the correlations are decreased as the strength of the exchange interactions is increased or its range is extended to the next nearest neighbors.

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Exchange Anisotropy Uniaxial Anisotropy Spin Diffusion Spectral Functions Frequency Moments Gaussian Diffusivity High Temperature Expansions Density Matrix Spin Hamiltonian of Heisenberg Model Lattice Structures and Dynamics Linear Chain XY Model Windsor Classical Model Statistical Correlation Functions Traces of Spin Operator Products Thermal Averages Hydrodynamical Representation Many-Particle Systems Green Functions Longitudinal and Transverse Moments Fourier Transforms Phenomenological Theory Next Nearest Neighbor Interactions Wave Vectors Time Derivatives of Dynamical Operators						

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SPACE-TIME MAGNETIC SPIN CORRELATIONS OF ANISOTROPIC HEISENBERG SYSTEMS BY THE METHOD OF MOMENTS OF A DIFFUSIVITY - SPECTRAL REPRESENTATION

by

FREDERICK A. MALINOSKI*

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ABSTRACT

The longitudinal and transverse time-dependent spin correlations of Heisenberg spin systems with cylindrical symmetry and with both uniaxial and exchange anisotropy are evaluated in the limit of elevated temperature, by the method of frequency moments. The zeroth, second, and fourth frequency moments are determined by thermal expansions of commutators of equal-time spin operators and their time derivatives. The frequency wave-vector spectral functions are established by these moments via the hydrodynamical representation, based on a two-parameter Gaussian diffusivity. The moment equations in a generic form apply for all spins, for arbitrary range of the exchange interactions, and for arbitrary dimensionality and lattice structures and are particularized with next nearest neighbor interactions for a linear chain, a two-dimensional net and a simple cubic lattice. The effects on the correlation functions of varying the axial anisotropy, the exchange anisotropy, and the interaction range are investigated. The resulting time-dependent correlations and their Fourier transforms are calculated, exhibiting features predicted by theoretical considerations and pointing to several conclusions; i.e., (1) The effect of increasing the axial anisotropy upon the longitudinal correlations is similar to that of decreasing the exchange anisotropy; (2) In contrast, the behavior of the transverse correlations is similar for increasing axial anisotropy or for increasing exchange anisotropy; (3) The anisotropic effects on the correlations are decreased as the strength of the exchange interaction is increased or its range is extended to the next nearest neighbors.

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INTRODUCTION

The thermodynamics of interacting spin systems is, in general, too complicated to be exactly soluble. Of the many approximate solutions which are known, only those relating to low temperature and to high temperature are reliable, and the dynamics of these systems are even more complicated. Nonetheless, the dynamical properties of Heisenberg spin systems have been the subject of much theoretical study in recent years,[1-9] stimulated by the appearance of inelastic neutron scattering data at high temperatures[10-12] and by the computer simulation calculations of precisely defined models.[13-17] The various methods used include diagrammatic perturbation theory,[5] interpolation schemes,[6,7] frequency moment calculations,[1,4,7] and microscopic theories of spin diffusion.[2,3] A useful review of much of this research is given by Marshall and Lowde.[8]

In exchange-coupled paramagnetic spin systems, the time-independent spin correlation functions have been accurately determined with the pertinent high temperature series expansion of the equilibrium density matrix. However, the dynamical time-dependent correlation functions have not been rigorously treated, even in the region of high temperatures, although several approaches[5,9] attack the problem from first principles, utilizing self-consistent Green function analysis.

A highly successful phenomenological approximation of the dynamics of Heisenberg spin systems has been the frequency wave-vector-dependent diffusivity represented by a two-parameter Gaussian.[3,7] This method circumvents much of the calculational difficulty in the extensive resummations of

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Feynman diagrams, used in many-body physics. This approximation has been seen to yield adequate to excellent agreement with several restrictive cases where the dynamics have been solved exactly and with experimental results of neutron scattering of exchange-coupled systems. In this present study the emphasis will be on the dynamical correlation functions of Heisenberg spin systems with uniaxial and exchange anisotropy, a problem area which has never before been treated adequately in the literature. This study will involve a kind of many-body physics technique arising from the statistical mechanics calculation of thermal averages of products of spin-paired exchange sums, and will treat specific solid-state magnetic structures.

An example of a uniaxial structure with a large anisotropy energy is FeF_2. Several papers[18, 19] deal with FeF_2 by the method of Green functions. It has been pointed out by Lines[19] that this anisotropy presents a new difficulty in the Green function treatment, namely, how to decouple the higher-order Green functions which appear in conjunction with the anisotropy. Tanaka et al.[20] applied a cluster-variation method to FeF_2 in the region of the Néel temperature by considering spin correlations between nearest and next nearest neighbors.

Terbium and other rare earths, also examples of axially anisotropic spin structures, have recently been the subjects of experimental investigation,[21] particularly neutron scattering studies[22-24] and theoretical studies based on scattering data and utilizing the method of moments.[25-27] These scattering studies attempt to formulate various parameters for exponential time decay and truncated Lorentzians, based on measured values of the exchange interaction and axial anisotropic constants, with some anomalous results. Several frequency moments for the axially anisotropic Hamiltonian are given by Sears,[25] Collins,[26] and Lindgård.[27] In addition, Sears has calculated the first-order temperature correction term of the zeroth and second moments. It must be emphasized that all of these moment calculations were performed with isotropic exchange, a less general case than that treated in this study.

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Although several cases exist in which spin correlations have been calculated exactly, they are highly restrictive in their system assumptions and do not permit generalizations to more realistic spin systems without excountering prohibitive computational difficulties. Still, these results are valuable for checking on the limiting cases of approximate phenomenological schemes. Carboni and Richards[15] exactly calculated the frequency-dependent spin correlations, the spectral functions, and the time-dependent correlations for a linear chain consisting of a finite number N of spins with $S=1/2$. These spins were considered to be in contact with a heat bath and to interact by a nearest neighbor isotropic exchange. The eigenfunctions and eigenvalues were evaluated and the results extrapolated to the thermodynamic limit, $N\rightarrow\infty$. This method presented the frequency Fourier transform of the temporal spin correlations in the form of histograms.

Another model with exact results, in the sense that no theoretical approximations are introduced except those related to the numerical calculation, is the Windsor formulation[11, 14] of the classical Heisenberg paramagnet. This model was constructed with a thousand classical spins situated on a simple cubic lattice and interacting with isotropic, nearest-neighbor exchange, at infinite temperature. The time development of the classical spin correlations were then calculated in the thermodynamic limit, i.e., with $N\rightarrow\infty$. These results can then be compared to the infinite spin, $S\rightarrow\infty$, correlations of approximate, phenomenological schemes, applied to the same lattice structure.

In addition, the so-called XY model [28] affords another exact computation, giving considerable insights into the temporal correlation functions. In this model no exchange coupling exists between the z components of the spins, while the x and y components are assumed to be bilinearly coupled among themselves by pairwise exchange potentials. For the particular one-dimensional case with $S=1/2$, the analysis of this type of spin coupling can be reduced to that of a system of non-interacting fermions so that the dynamics of the XY model can be solved exactly.[29, 30]

Many of the approximate, phenomenological approaches to the study of spin correlations have been related to the hydrodynamic limit of the spectral

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function. The usefulness of the two-parameter Gaussian representation of the generalized diffusivity, in the construction of spectral functions, has been demonstrated by Tahir-Kheli and McFadden,[7] particularly in those cases for which excellent comparisons have been shown with the exact calculations of Carboni and Richards in the one-dimensional, isotropic array with $S=1/2$ spins and with the exact classical spin results of Windsor for infinite spin. Therefore this phenomenological construct is adopted in this study, since its basic simplicity and its proven ability to give adequate-to-excellent meaningful results permit its application to more realistic spin systems, which would involve, at present, vast difficulties in exact calculations.

The general plan of this study is as follows: The section "Formulation" introduces the pertinent Hamiltonian with both exchange and axial anisotropy. Then the analytical relationships between temporal spin correlations, frequency wave-vector-dependent spectral functions, and frequency moments in a high-temperature expansion are discussed. Physical arguments are then presented to justify the phenomenological representation of the spectral function for low frequency and long wave length, as a generalized diffusivity in the hydrodynamic limit. A two-parameter Gaussian representation of the diffusivity is then related to the diffusivity moments and frequency moments. In the section "General Form of the Frequency Moments", the frequency moments are derived in a generic form, and some intermediate calculational details are shown. Next, high-temperature spin correlations based on the two-parameter generalized diffusivity Gaussian are particularized in the three sections that follow for one, two, and three dimensions with the inclusion of the effects of a larger range of the exchange interactions (extended to next nearest neighbors), axial anisotropy, and exchange anisotropy. The last section, "Concluding Remarks", summarizes results. Appendix A presents a listing of repeatedly applied traces of products of spin operators. In Appendix B some details of the application of the frequency moments to various lattice structures are presented.

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FORMULATION

The Heisenberg Model

We shall study, as the most general case, the anisotropic Heisenberg spin systems, where the Hamiltonian is in the form

$$H = -\sum_{g,p}\left[I_+(gp)\left(S_g^x S_p^x + S_g^y S_p^y\right) + I_0(gp) S_g^z S_p^z\right] - A\sum_k \left(S_k^z\right)^2 \tag{2.1}$$

or, alternatively

$$H = -\sum_{g,p}\left[I_+(gp)\ S_g^+ S_p^- + I_0(gp) S_g^z S_p^z\right] - A\sum_k \left(S_k^z\right)^2 \tag{2.2}$$

by the use of the raising and lowering operators

$$S_g^{\pm} = S_g^x \pm iS_g^y \tag{2.3}$$

This Heisenberg model assumes both exchange anisotropy, i.e., $I_+ \neq I_0$, and axial anisotropy through the A term. Here g and p are the spin sites, I(gp) is the exchange interaction integral between the two spins at sites g and p, S_g^{α} is the α component of the spin vector associated with the lattice point g, and A is the axial anisotropy constant (energy) characterizing the strength of the crystalline field. This latter type of anisotropy is often referred to as uniaxial, crystal-field or single-ion. We assume that the exchange integrals depend only upon the spatial separation of the positions and that

$$I_0(gg) = I_+(gg) = 0$$

The integrals I_0 and I_+ are, in general, allowed to be different. In the limiting case when $I_+ = A = 0$, this magnetic spin system reduces to a nondynamical Ising model, whereas the other limiting case of $I_0 = A = 0$ corresponds to the XY model.

In the calculations of the spin correlations, the Heisenberg system is considered to be at infinite temperature. Moreover, the range of the exchange interactions includes next nearest neighbors. The spacing between the spins is assumed to be uniform for the one, two, and three (simple cubic) dimensional lattices, and distances will be measured in the units of these spacings. Periodic boundary conditions will apply. Furthermore, we assume that the spins are not vibrationally coupled to the lattice so that it is sufficient to consider the strength of the exchange integral to be a suitable scalar function of the temperature in order to describe any spin-lattice coupling. The Dirac system of units with $\hbar=1$ is used throughout.

Correlations, Spectral Functions, and Frequency Moments

The space-time dependent spin correlation function is defined as

$$F^{\alpha\alpha'}(g-w,t-t') \equiv \left\langle \left[S^{\alpha}_{g}(t), S^{\alpha'}_{w}(t') \right] \right\rangle \tag{2.4}$$

where α and $\alpha' = x, y, z$. Correlations with $\alpha \neq \alpha'$ vanish because of the assumed cylindrical symmetry. For the same reason, correlations with $\alpha=x$ and $\alpha=y$ are equivalent and are called the transverse correlations. In the general anisotropic cases, the correlations with $\alpha=z$ differ from the transverse correlations and are called the longitudinal correlations. The time dependence of the spin operators is in the Heisenberg representation with respect to the Hamiltonian. The angular brackets denote a statistical thermal average over a canonical ensemble and the straight brackets denote a commutator.

The translational invariance of the system Hamiltonian determines the dependence of these correlation functions upon space and time. This fact is most conveniently incorporated by introducing the spectral function, $F^{\alpha\alpha}(K,\omega)$, which is the Fourier representation of the spin correlation function

$$F^{\alpha\alpha}(\vec{K},\omega) = \sum_{\vec{g}-\vec{w}} \frac{e^{-i\vec{K}\cdot(\vec{g}-\vec{w})}}{2\pi} \int_{-\infty}^{\infty} d(t-t')\, e^{i\omega(t-t')}\, F^{\alpha\alpha}(g-w,t-t') \tag{2.5}$$

where $\vec{K}$ is the inverse lattice vector, and ω is the frequency. This summation consists of N allowed $\vec{K}$-vectors falling within the first Brillouin zone and becomes an appropriate integral for large N.

From the definition of the spin correlation function $F^{aa}(g-w, t-t')$ in Equation (2.4) and from the invariance of the spin system under spatial inversion, the relevant symmetry properties are obtained

$$\begin{aligned} F^{aa}(g-w, t-t') &= -F^{aa}(w-g, t'-t) \\ &= F^{aa}(w-g, t-t') = -F^{aa}(g-w, t'-t) \end{aligned} \tag{2.6}$$

If these properties are used in Equation (2.5), the symmetry properties of the spectral function result

$$F^{aa}(\vec{K}, \omega) = F^{aa}(-\vec{K}, \omega) = -F^{aa}(\vec{K}, -\omega) \tag{2.7}$$

showing that the spectral function is even in $\vec{K}$ and odd in ω, similar to the spin correlation function's relation to space and time.

The commutator correlation $F^{aa}(g-w, t-t')$ is closely related to the statistical correlation function $\Phi^{aa}(g-w, t-t')$, defined as

$$\Phi^{aa}(g-w, t-t') \equiv \left\langle S_g^a(t)\, S_w^a(t') \right\rangle \tag{2.8}$$

If two general time-dependent operators, say A(t) and B(t), develop in the Heisenberg picture, then the well-known statistical mechanical identity

$$\left\langle A(t)B(t') \right\rangle = \left\langle B(t'-i\beta)\, A(t) \right\rangle \tag{2.9}$$

results, by noting the invariance of the trace under cyclic permutation. Using this result with Equations (2.8), (2.5), and (2.4), we find that

$$\Phi^{aa}(g-w, t-t') = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega(t-t')} \left\langle S_g^a(t) S_w^a(t') \right\rangle_{\omega} \tag{2.10a}$$

where

$$\left\langle S^a_g(t)\, S^a_w(t')\right\rangle_\omega = \frac{1}{N}\sum_{\vec{K}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\ \frac{F^{aa}(\vec{K},\omega)}{1-e^{-\beta\omega}} \tag{2.10b}$$

Here the quantity $\beta = (k_B T)^{-1}$, where k_B= Boltzmann constant and T= temperature.

The definition of the frequency moments of the spectral function is, for integer n

$$\left\langle \omega^n \right\rangle^{aa}_{\vec{K}} \equiv \int_{-\infty}^{\infty} F^{aa}(\vec{K},\omega)\,\omega^{n-1}\,d\omega \tag{2.11}$$

Since the spectral function is odd in ω, the odd moments vanish.

Time-independent statistical averages determine the frequency moments of the spectral function. If the time derivatives of a corresponding Fourier representation of the spin correlation function in the following form are determined

$$F^{aa}(g-w,t-t') = \frac{1}{N}\sum_{\vec{K}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\int_{-\infty}^{\infty} F^{aa}(\vec{K},\omega)\,e^{-i\omega(t-t')}\,d\omega \tag{2.12}$$

then it can be shown, from Equations (2.4), (2.5), and (2.11), that one obtains, by proceeding to the limit t'=t

$$\sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})}\left[\left(i\frac{d}{dt}\right)^r\left(-i\frac{d}{dt'}\right)^{n-r} F^{aa}(g-w,t-t')\right]_{t=t'}$$

$$= \int_{-\infty}^{\infty} F^{aa}(\vec{K},\omega)\,\omega^n\,d\omega = \left\langle \omega^{n+1}\right\rangle^{aa}_{\vec{K}} \tag{2.13}$$

Here the indices n and r are non-negative integers such that n ≥ r, and the sum over all position vectors $\vec{g}-\vec{w}$ includes the origin. The significance of this result is that to compute, for example, the second moment, it is necessary to consider only the first time derivatives of only one of the spin operators in Equation (2.4).

Therefore, when the temperature is finite, the moments should be calculated from Equation (2.13), where, it is possible, in principle, to calculate the moments to any desired order in β (inverse temperature), although the amount of calculations grows rapidly with the expansion in β. However, if the high-temperature limit is of primary interest, then a simplification in the moment calculation can be effected. Equation (2.10b) becomes, for large temperatures

$$\lim_{\beta \to 0} \left\langle S_g^a(t)\, S_w^a(t') \right\rangle_\omega = \frac{1}{N} \sum_{\vec{K}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})} \frac{F^{aa}(\vec{K},\omega)}{\beta\omega} \qquad (2.14)$$

and, with Equation (2.10a), one obtains

$$\lim_{\beta \to 0} \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\, \Phi^{aa}(g-w, t-t') = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega(t-t')} \frac{F^{aa}(\vec{K},\omega)}{\beta\omega} \qquad (2.15)$$

The time derivations of this expression lead to

$$\lim_{\beta \to 0} \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})} \left[\left(i \frac{d}{dt} \right)^r \left(-i \frac{d}{dt'} \right)^{n-r} \Phi^{aa}(g-w, t-t') \right]_{t=t'}$$

$$= \frac{1}{\beta} \int_{-\infty}^{\infty} F^{aa}(\vec{K},\omega)\, \omega^{n-1}\, d\omega = \frac{1}{\beta} \left\langle \omega^n \right\rangle_{\vec{K}}^{aa} \qquad (2.16)$$

with the same restrictions on n and r, as before. The appearance of $1/\beta$ as a factor of the moment can be explained by the fact that the statistical correlation function Φ^{aa} (g-w, t-t') contains products of spin operators, for which the trace is finite, as contrasted to the spin correlation function F^{aa} (g-w, t-t') which contains commutators, for which the trace vanishes but the first contribution to the thermal average arises from the next term, $-\beta H$, in the expansion. Equation (2.16) therefore avoids considerable commutator algebra but is valid only at elevated temperatures.

The method for the computation of the frequency moments first involves the repeated application of the von Neumann, quantum mechanical equation of motion to the spin operators

$$i \frac{d}{dt} S_g^a(t) = \left[S_g^a(t), H \right] \qquad (2.17a)$$

$$\left(i\,\frac{d}{dt}\right)^2 S_g^a(t) = \left[\left[S_g^a(t), H\right], H\right] \qquad (2.17b)$$

Next the time-independent statistical thermal averages $\langle Q \rangle$, where Q is a generalized spin operator product (or commutator), are computed by the usual high temperature expansion procedure, with the density matrix

$$\rho = e^{-\beta H} \qquad (2.18)$$

Therefore

$$\langle Q \rangle = \mathrm{Tr}\left\{e^{-\beta H} Q\right\} / \mathrm{Tr}\left\{e^{-\beta H}\right\}$$

$$= \frac{\mathrm{Tr}\left\{\left[1 - \beta H + \frac{1}{2!}\beta^2 H^2 - \ldots\right] Q\right\}}{\mathrm{Tr}\left\{1 - \beta H + \frac{1}{2!}\beta^2 H^2 - \ldots\right\}}$$

$$= \frac{\mathrm{Tr}\left\{Q - \beta H Q + \frac{1}{2}\beta^2 H^2 Q - \ldots\right\}}{\mathrm{Tr}\{1\}}\left(1 + \frac{\mathrm{Tr}\{\beta H\}}{\mathrm{Tr}\{1\}}\right.$$

$$\left. + \left[\left(\mathrm{Tr}\{\beta H\}\right)^2 - \frac{1}{2}\mathrm{Tr}\{\beta H\}^2\right] / \,\mathrm{Tr}\{1\} + \ldots\right) \qquad (2.19)$$

The computation of the zeroth moment

$$\left\langle \omega^0 \right\rangle_{\vec{K}}^{aa} = \int_{-\infty}^{\infty} \frac{F^{aa}(\vec{K}, \omega)}{\omega}\, d\omega \qquad (2.20)$$

or wave-dependent susceptibility, involves a special case. From Equations (2.4, 2.5, and 2.20), one finds

$$\left\langle \omega^0 \right\rangle_{\vec{K}}^{aa} = \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega\, \frac{e^{i\omega t}}{2\pi\omega} \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[S_g^a(t), S_w^a(0)\right] \right\rangle \qquad (2.21)$$

A contour integration yields

$$\lim_{\epsilon\to 0}\int_{-\infty}^{\infty}\frac{d\omega e^{i\omega t}}{2\pi(\omega+i\epsilon)} = \frac{1}{i}\,\theta(-t) \tag{2.22}$$

where the step function $\theta(t)=1$, for $t>0$ and $=0$, for $t<0$. Now using the interchangeability of the symmetric spatial variables g and w in the correlation function and the identity of Equation (2.9), one obtains

$$\left\langle \omega^{0}\right\rangle_{\vec{K}}^{aa} = \frac{1}{i}\sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\int_{-\infty}^{0} dt\left\langle S_{g}^{a}(0)\left[e^{iH(t+i\beta)}\,S_{w}^{a}(0)\,e^{-iH(t+i\beta)}\right.\right.$$

$$\left.\left.-e^{iHt}\,S_{w}^{a}(0)\,e^{-iHt}\right]\right\rangle \tag{2.23}$$

and by a change of variable, $t+i\beta\to i\mu$, the following relation results

$$\left\langle \omega^{0}\right\rangle_{\vec{K}}^{aa} = \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\int_{0}^{\beta} d\mu\left\langle S_{g}^{a}(0)\,e^{-\mu H}\,S_{w}^{a}(0)\,e^{\mu H}\right\rangle \tag{2.24}$$

which is valid for all temperatures. In a second-order temperature expansion, Equation (2.24) becomes (omitting the unnecessary equal time specification of the spin operators)

$$\left\langle \omega^{0}\right\rangle_{\vec{K}}^{aa} = \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})}\left[\beta\left\langle S_{g}^{a}\,S_{w}^{a}\right\rangle + \frac{\beta^{2}}{2}\left\langle S_{g}^{a}\left[S_{w}^{a},H\right]\right\rangle\right] \tag{2.25}$$

The knowledge of the short time behavior of the correlation functions is of limited usefulness. Low-frequency phenomena, such as the magnetic scattering of slow, long-wavelength neutrons, the electromagnetic field resonance experiments, and various transport phenomena in fluid mechanics applications depend upon the long-time behavior of correlations.

The frequency moments actually yield no information about the behavior of the spectral function, as shown by Grant,[31] but do point out unreasonable representations of the spectral function. The moments also provide only limited information about the time dependence of the correlations F^{aa} (g-w, t-t') and Φ^{aa} (g-w, t-t'). As shown by Tahir-Kheli and McFadden,[7] the expansion of the correlations in terms of the first few frequency moments adequately determine only their short time behavior.

In some respects, the spin diffusion process would be a difficult part of the long-time, large-distance behavior of spin correlations, if conventional perturbation theory requiring integral equations or extensive resummation of diagrams were applied. The long-time description of an interacting many-particle system is dominated by many inter-particle collisions.

The possible analogy of such a collision in the spin system is a mutual exchange of a single spin-flip. However, such a collision dominated behavior can be described in terms of hydrodynamics, which refers to a system in local thermodynamic equilibrium, produced and maintained by frequent collisions between particles. Therefore, in the limit of long wavelengths (small K vectors), the behavior of the spin correlations relates to the appropriate hydrodynamic description of the diffusion of the magnetization density in equilibrium.

The required formulation, therefore, should couple the rather limited information available in the first three non-vanishing frequency moments, $\langle \omega^{2n} \rangle^{aa}_{\vec{K}}$, n=0, 1, 2, with the limiting hydrodynamic behavior of the spin diffusion. Kadanoff and Martin[32] have described the prediction of the spin

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[7] R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969); B-1, No. 7, 3178 (1970); B-1, No. 9, 3649 (1970); B-1, No. 9, 3671 (1970).

[31] W.C. Grant, Physica 30, 1433 (1964).

[32] L.P. Kadanoff and P.C. Martin, Ann. of Phys. 24, 419 (1963).

diffusion process by hydrodynamic equations. The theory begins with the conservation of the total z component (S_T^z) of the spin, or

$$\left[S_T^z, H\right] = 0 = i \frac{d}{dt} S_T^z \tag{2.26}$$

In the direction of quantization z, the magnetization density $m^z(\vec{r}, t)$ and the total magnetization $M^z(t)$ are related by

$$M^z(t) = \int d^3r \; m^z(\vec{r}, t) = \left\langle S_T^z \right\rangle \tag{2.27}$$

With this, Equation (2.26) can be expressed in the differential form, a continuity equation

$$\frac{\partial}{\partial t} m^z(\vec{r}, t) + \nabla \cdot \vec{j}(\vec{r}, t) = 0 \tag{2.28}$$

where $\vec{j}(\vec{r}, t)$ is the magnetization spin current. When the properties of the system vary slowly in space and time, local equilibrium occurs, and the spin current is proportional to the gradient of the magnetization density, that is

$$\vec{j}(\vec{r}, t) = - D^z \nabla m^z(\vec{r}, t) \tag{2.29}$$

where the analogous transport coefficient D^z is now called the spin diffusion coefficient. Combining Equations (2.28) and (2.29), one finds the diffusion equation for the magnetization density

$$\frac{\partial}{\partial t} m^z(\vec{r}, t) = D^z \nabla^2 m^z(\vec{r}, t) \tag{2.30}$$

If we now define a combined wave-vector Fourier transform and frequency Laplace transform of the magnetization density as

$$m^z(\vec{K}, \theta) = \int d^3r \; e^{-i\vec{K}\cdot\vec{r}} \int_0^\infty dt \, e^{i\theta t} \, m^z(\vec{r}, t) \tag{2.31}$$

and then substitute Equation (2.30) and integrate by parts, we find

$$m^z(\vec{K},\theta) = \frac{m^z(\vec{K},0)}{D^z K^2 - i\theta} \tag{2.32}$$

To connect this result to the spectral function, suppose that a weak space-time varying field $h^z(\vec{r},t)$ has been turned on at time t, such that

$$\begin{aligned} h^z(r,t) &= h^z(\vec{r})\, e^{-\epsilon t};\, t > 0 \\ &= 0;\, t < 0 \end{aligned} \tag{2.33}$$

where ϵ is an infinitesimal positive quantity. First-order perturbation theory yields the response of the system

$$m^z(\vec{r},t) = i\int dr' \int_0^t dt' \left\langle \left[S^z_r(t), S^z_{r'}(t') \right] \right\rangle h^z(\vec{r},t) \tag{2.34}$$

If the spectral function from Equation (2.5), along with Equation (2.4) is used, one obtains

$$m^z(\vec{K},\theta) = \int_{-\infty}^{\infty} d\omega\, h^z(\vec{K}) \frac{F^{zz}(\vec{K},\omega)}{i\omega(\omega-\theta)} \tag{2.35a}$$

where

$$h^z(\vec{K}) = \int dr\, e^{-i\vec{K}\cdot\vec{r}}\, h^z(\vec{r}) \tag{2.35b}$$

If the following identity, implied for integrations, is used

$$\lim_{\epsilon \to 0} \frac{1}{\omega' - \omega \pm i\epsilon} = P\frac{1}{\omega' - \omega} \mp i\pi\delta(\omega - \omega') \tag{2.36}$$

where P denotes the principal value, we find from Equation (2.35) that

$$\mathrm{Re}\left\{ m^z(K, \omega + i\epsilon) \right\} = \pi \int d\omega' \, h^z(\vec{K}) \, \frac{F^{zz}(\vec{K}, \omega')}{\omega'} \, \delta(\omega - \omega')$$

$$= \pi h^z(\vec{K}) \, \frac{F^{zz}(\vec{K}, \omega)}{\omega} \tag{2.37}$$

Comparing Equation (2.32) with this result, then we obtain

$$\frac{F^{zz}(\vec{K}, \omega)}{\omega} = \frac{m^z(\vec{K}, 0)}{h^z(\vec{K})} \; \frac{D^z K^2/\pi}{\omega^2 + (D^z K^2)^2} \tag{2.38}$$

Or, if we denote the static wave-vector dependent susceptibility by $\chi_{\vec{K}}$ in the relation

$$m^z(\vec{K}, 0) = \chi_{\vec{K}} \, h^z(\vec{K}) \tag{2.39}$$

then Equation (2.38) becomes

$$\frac{F^{zz}(\vec{K}, \omega)}{\omega} = \frac{\chi_{\vec{K}} \, D^z K^2/\pi}{\omega^2 + (D^z K^2)^2} \tag{2.40}$$

It is noted here that the zeroth frequency moment is equivalent to this susceptibility, i.e., $\left\langle \omega^0 \right\rangle_{\vec{K}} = \chi_{\vec{K}}$. The quantity on each side of Equation (2.40) is often referred to as the spectral line shape function

$$S(\vec{K}, \omega) = F^{zz}(K, \omega) / \omega \tag{2.40a}$$

The hydrodynamic limit expressed in Equation (2.40) is valid for slowly varying disturbances, i.e., for small ω and small $\vec{K}$. Under these conditions, the value of the spectral function is sensitive to the relative magnitude of ω and $\vec{K}$. If $\omega << D^z K^2$

$$\frac{F^{zz}(\vec{K}, \omega)}{\omega} \cong \frac{\chi_{\vec{K}}}{\pi D^z K^2} \tag{2.41}$$

and when $D^z K^2 << \omega$

$$\frac{F^{zz}(\vec{K},\omega)}{\omega} \cong \frac{\chi_{\vec{K}}\, D^z K^2}{\pi \omega^2} \tag{2.42}$$

Although only the longitudinal spectral function is shown in Equation (2.40), the transverse spectral function, in the isotropic case, will possess the same hydrodynamic limit.

For computational purposes, the zero frequency behavior of the frequency Fourier transform of the longitudinal correlations given in Equation (2.14) can be roughly described in their variation with lattice dimensionality. Since in this case, Equation (2.41) shows that $F^{zz}(\vec{K},\omega)/\omega \sim K^{-2}$, then

$$\left\langle S^z_g(t)\, S^z_w(0)\right\rangle_\omega \Big|_{\omega=0} \sim \int \frac{d^n K}{K^2} \sim \int K^{n-3}\, dK$$

where n is the dimensionality. Therefore, in one and two dimensions these transforms will diverge as $\omega \to 0$, less rapidly so in two dimensions. For three dimensions no divergence will be exhibited.

The Gaussian Diffusivity Method

The generalization of the spin diffusion constant so that it could be a function of $\vec{K}$ and ω was first suggested by Martin and Bennett,[3] with a postulation of a two-parameter Gaussian form.

The objective here is to develop a representation of the spin and spectral correlation functions, in terms of a generalized frequency wave-vector dependent diffusivity, $D^{\alpha\alpha}(\vec{K},\omega)$, which reduces to the hydrodynamic limit of Equation (2.40) for small ω and $\vec{K}$. This phenomenological representation of the well-behaved function $D^{\alpha\alpha}(\vec{K},\omega)$ might then pose fewer hazards in its application than the rather singular spectral function $F^{\alpha\alpha}(\vec{K},\omega)$.

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

If a retarded double-time Green function, utilizing the spin correlation function, is defined

$$M^{aa}_{gw}(t-t') = -i\theta(t-t') \left\langle \left[S^{a}_{g}(t),\ S^{a}_{w}(t') \right] \right\rangle \tag{2.43}$$

where θ is the Heaviside unit step function, then its Fourier transform is

$$M^{aa}(\vec{K}, \omega) = \frac{1}{2\pi} \sum_{g-w} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \int_{-\infty}^{\infty} e^{i\omega(t-t')} M^{aa}_{gw}(t-t')\, d(t-t') \tag{2.44}$$

Now, if Equations (2.4), (2.12), and (2.43) are substituted in Equation (2.44), and if this resulting Fourier transform is extended into the upper half of the complex plane by means of the complex quantity

$$z = \omega + i\epsilon$$

where ϵ is a positive infinitesimal, one obtains the following spectral representation

$$M^{aa}(\vec{K}, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{F^{aa}(\vec{K}, \omega)}{z-\omega}\, d\omega \tag{2.45}$$

The generalized diffusivity $D^{aa}(\vec{K}, \omega)$ can now be defined as

$$M^{aa}(\vec{K}, z) \equiv M^{aa}(\vec{K}, 0) \left[1 - \left(1 - \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{D^{aa}(\vec{K}, \omega)}{z^{2} - \omega^{2}}\, d\omega \right)^{-1} \right] \tag{2.46}$$

Now Equations (2.45) and (2.46) relate the diffusivity and the spectral function, through the frequency moments in Equation (2.11) and the now defined diffusion moments

$$\left\langle D^{2n} \right\rangle^{aa}_{\vec{K}} \equiv \frac{1}{\pi} \int_{-\infty}^{\infty} D^{aa}(\vec{K}, \omega)\, \omega^{2n}\, d\omega \tag{2.47}$$

It should be noted that the functions $F^{aa}(\vec{K}, \omega)/\omega$ and $D^{aa}(\vec{K}, \omega)$ are both even in ω.

If large z expansions are performed on Equations (2.45) and (2.46), and if we note that

$$M^{aa}(\vec{K}, 0) = -\frac{1}{2\pi}\left\langle \omega^{0} \right\rangle_{\vec{K}}^{aa} \tag{2.48}$$

then we find, for Equation (2.46)

$$\frac{M(\vec{K}, z)}{M(\vec{K}, 0)} = 1 - \left[1 - \frac{1}{\pi z^{2}} \int_{-\infty}^{\infty} D^{aa}(\vec{K}, \omega) \sum_{n=1}^{\infty} \left(\frac{\omega}{z}\right)^{2n} d\omega\right]^{-1}$$

$$\cong -\sum_{m=1}^{\infty} \left[\frac{1}{z^{2}} \sum_{n=0}^{\infty} \frac{\left\langle D^{2n} \right\rangle_{\vec{K}}^{aa}}{z^{2n}}\right]^{m} \tag{2.49}$$

In a similar manner, Equation (2.45) becomes

$$\frac{M(\vec{K}, z)}{M(\vec{K}, 0)} = -\frac{1}{\left\langle \omega^{0} \right\rangle_{\vec{K}}^{aa}} \int_{-\infty}^{\infty} \frac{F^{aa}(\vec{K}, \omega)}{\omega} \sum_{n=1}^{\infty} \left(\frac{\omega}{z}\right)^{n} d\omega$$

$$= -\frac{1}{\left\langle \omega^{0} \right\rangle_{\vec{K}}^{aa}} \sum_{n=1}^{\infty} \frac{\left\langle \omega^{2n} \right\rangle_{\vec{K}}^{aa}}{z^{2n}} \tag{2.50}$$

where the odd frequency moments vanish. Equating relations (2.49) and (2.50), we find for equivalent powers of $1/Z^{2n}$, out to n=4

$$\left\langle \omega^{2} \right\rangle_{\vec{K}}^{aa} \Big/ \left\langle \omega^{0} \right\rangle_{\vec{K}}^{aa} = \left\langle D^{0} \right\rangle_{\vec{K}}^{aa} \tag{2.51a}$$

$$\left\langle \omega^{4} \right\rangle_{\vec{K}}^{aa} \Big/ \left\langle \omega^{0} \right\rangle_{\vec{K}}^{aa} = \left\langle D^{2} \right\rangle_{\vec{K}}^{aa} + \left(\left\langle D^{0} \right\rangle_{\vec{K}}^{aa}\right)^{2} \tag{2.51b}$$

$$\left\langle \omega^6 \right\rangle^{aa}_{\vec{K}} \Big/ \left\langle \omega^0 \right\rangle^{aa}_{\vec{K}} = \left\langle D^4 \right\rangle^{aa}_{\vec{K}} + 2 \left\langle D^0 \right\rangle^{aa}_{\vec{K}} \left\langle D^2 \right\rangle^{aa}_{\vec{K}} + \left(\left\langle D^0 \right\rangle^{aa}_{\vec{K}} \right)^3 \tag{2.51c}$$

$$\begin{aligned} \left\langle \omega^8 \right\rangle^{aa}_{\vec{K}} \Big/ \left\langle \omega^0 \right\rangle^{aa}_{\vec{K}} = {} & \left\langle D^6 \right\rangle^{aa}_{\vec{K}} + 2 \left\langle D^0 \right\rangle^{aa}_{\vec{K}} \left\langle D^4 \right\rangle^{aa}_{\vec{K}} + \left(\left\langle D^2 \right\rangle^{aa}_{\vec{K}} \right)^2 \\ & + 3 \left(\left\langle D^0 \right\rangle^{aa}_{\vec{K}} \right)^2 \left\langle D^2 \right\rangle^{aa}_{\vec{K}} + \left(\left\langle D^0 \right\rangle^{aa}_{\vec{K}} \right)^4 \end{aligned} \tag{2.51d}$$

Therefore, with the frequency moments determined from the thermal expansion of the time-independent spin correlations, these equations yield the diffusivity moments. It can be seen that the higher order diffusivity moments are essentially given in terms of the lower order diffusivity moments, this fact leading to a significant computational convenience in the final calculation of the time-dependent spin correlations. Solving Equation (2.51) for the second and fourth diffusivity moments, one obtains

$$\left\langle D^2 \right\rangle^{aa}_{\vec{K}} = \frac{\left\langle \omega^4 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} - \left(\frac{\left\langle \omega^2 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} \right)^2 \tag{2.52a}$$

$$\left\langle D^4 \right\rangle^{aa}_{\vec{K}} = \frac{\left\langle \omega^6 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} - 2 \frac{\left\langle \omega^2 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} \left[\frac{\left\langle \omega^4 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} - \left(\frac{\left\langle \omega^2 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} \right)^2 \right] - \left(\frac{\left\langle \omega^2 \right\rangle^{aa}_{\vec{K}}}{\left\langle \omega^0 \right\rangle^{aa}_{\vec{K}}} \right)^3 \tag{2.52b}$$

If the identity in Equation (2.36) is applied to the integral in Equation (2.46), then

$$\frac{1}{\pi} \int_{-\infty}^{\infty} d\phi \, \frac{D^{aa}(\vec{K}, \phi)}{\phi^2 - z^2} = Q(\omega) + \frac{i}{\omega} D^{aa}(\vec{K}, \omega) \tag{2.53a}$$

where

$$Q(\omega) = P \int_{-\infty}^{\infty} \frac{d\phi}{\pi} \, \frac{D^{aa}(\vec{K}, \phi)}{\phi^2 - \omega^2} \tag{2.53b}$$

From Equation (2.45), we find

$$F^{aa}(\vec{K}, \omega) = \lim_{\epsilon \to 0} i \left[M^{aa}(\vec{K}, \omega + i\epsilon) - M^{aa}(\vec{K}, \omega - i\epsilon) \right] \tag{2.54}$$

which, when used with Equations (2.46) and (2.53), yields the spectral function in terms of the generalized diffusivity

$$\frac{F^{aa}(\vec{K}, \omega)}{\omega} = \frac{-2M(K, 0)\, D^{aa}(\vec{K}, \omega)}{\omega^2 \left[1 + Q(\omega)\right]^2 + \left[D^{aa}(\vec{K}, \omega)\right]^2} \tag{2.55}$$

The usefulness of this representation depends critically upon whether the function $D^{aa}(\vec{K}, \omega)$ turns out to be simple and well-behaved or not.

In complete analogy with the above formulation, a variety of existing phenomenological theories of equilibrium phenomena can also be conveniently cast in terms of the generalized diffusivity.[33, 34] In these analyses, the usual choices for the functional form of the diffusivity are the two-parameter exponential[35] or the two-parameter Gaussian.[3, 6, 7, 33, 34, 36] Since the latter has been used quite successfully, it will now be discussed. An adequate representation of generalized diffusivity by a two-parameter Gaussian is

$$D^{aa}(\vec{K}, \omega) = \Delta^{aa}(\vec{K})\, \Gamma^{aa}(\vec{K})\, \exp\left\{ - \left[\Gamma^{aa}(\vec{K})\, \omega \right]^2 \right\} \tag{2.56}$$

where the normalization and collision parameters, $\Delta^{aa}(\vec{K})$ and $\Gamma^{aa}(\vec{K})$ are functions only of $\vec{K}$, through their relation with the previously determined frequency moments and diffusivity moments. The parameter $\Gamma^{aa}(\vec{K})$ can be considered as a quasi-collision time.

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[3] H.S. Bennett & P.C. Martin, Phys. Rev. 138, A608 (1965).

[6] H.S. Bennett, Phys. Rev. 174, 629 (1968); 176, 650 (1968).

[7] R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969); B-1, No. 7, 3178 (1970); B-1, No. 9, 3649 (1970); B-1, No. 9, 3671 (1970).

[33] P.C. Martin, in 1967 *Les Houches Lectures*, ed. by C. DeWitt and R. Balian. New York: Gordon and Breach, Science Publishers, Inc., 1968.

[34] P.C. Martin & S. Yip, Phys. Rev. 170, 151 (1968).

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

[36] R.A. Tahir-Kheli, J. Appl. Phys. 40, 1550 (1969).

To determine these parameters, we use Equations (2.47) and (2.56) and find

$$\left\langle D^0 \right\rangle_{\vec{K}}^{aa} = \frac{1}{\sqrt{\pi}} \Delta^{aa}(\vec{K}) \tag{2.57a}$$

$$\left\langle D^2 \right\rangle_{\vec{K}}^{aa} = \frac{\Delta^{aa}(\vec{K})}{2\sqrt{\pi}\left[\Gamma^{aa}(\vec{K})\right]^2} \tag{2.57b}$$

$$\left\langle D^4 \right\rangle_{\vec{K}}^{aa} = \frac{3\Delta^{aa}(\vec{K})}{4\sqrt{\pi}\left[\Gamma^{aa}(\vec{K})\right]^4} \tag{2.57c}$$

and, solving for the parameters from Equations (2.51a) and (2.51b) or (2.52a), we obtain

$$\Delta^{aa}(\vec{K}) = \sqrt{\pi}\, \frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{aa}}{\left\langle \omega^0 \right\rangle_{\vec{K}}^{aa}} \tag{2.58a}$$

and

$$\Gamma^{aa}(\vec{K}) = \left[\frac{\left\langle \omega^0 \right\rangle_{\vec{K}}^{aa} \left\langle \omega^2 \right\rangle_{\vec{K}}^{aa}}{2\left(\left\langle \omega^4 \right\rangle_{\vec{K}}^{aa} \left\langle \omega^0 \right\rangle_{\vec{K}}^{aa} - \left[\left\langle \omega^2 \right\rangle_{\vec{K}}^{aa}\right]^2\right)}\right]^{1/2} \tag{2.58b}$$

Performing the principal parts integration in Equation (2.55), we obtain as the final form of the spectral function

$$\frac{F^{aa}(\vec{K},\omega)}{\omega} = \frac{-\,2M^{aa}(\vec{K},0)\,D^{aa}(\vec{K},\omega)}{\omega^2\left[1 - \frac{2D^{aa}(\vec{K},\omega)}{\sqrt{\pi}\,\omega}\int_0^{\omega\Gamma^{aa}(\vec{K})} \exp[x^2]\,dx\right]^2 + \left[D^{aa}(\vec{K},\omega)\right]^2} \tag{2.59}$$

This relation specifies the spectral function completely. The essential procedure is then to calculate in the following order:

1. The frequency moments by the time-independent Equation (2.13) or (2.16), which involve the thermal expansions embodied in Equations (2.17) and (2.19).
2. The Gaussian parameters by Equation (2.58).
3. The Gaussian diffusivity by Equation (2.56).
4. The spectral function (actually line shape function) by Equation (2.59).
5. The Fourier transform of the spin correlations by Equation (2.10b) or (2.14) and the time-dependent spin correlations by Equation (2.10a).

In this manner temporal spin correlations are constructed from time-independent spin correlations.

GENERAL FORM OF THE FREQUENCY MOMENTS

Although it is simpler to use the statistical correlation function of Equation (2.8) in the moment expression of Equation (2.16) in the infinite temperature computation of the frequency moments, the more complicated relation valid for any temperature, Equation (2.13), with the commutator spin correlation is used, in order to open the possibility of performing temperature expansions of the moments.

The procedure for calculating the moments by Equation (2.13) involves first finding the time derivatives by the equation of motion, Equation (2.17) and then computing the resulting time-independent statistical averages by the high temperature expansion of Equation (2.19). Considerable commutator algebra must be performed. With the notation now that S_1^α refers to the α - component of the spin at site 1, some of the more important basic commutator relations are

$$[S_1^x, S_2^y] = iS_1^z \delta_{12}$$

$$[S_1^+, S_2^-] = 2S_1^z \delta_{12}$$

$$[S_1^{\pm}, S_2^z] = \mp S_1^{\pm} \delta_{12}$$

$$[S_1^+ S_2^-, S_3^z] = S_1^+ S_2^- (\delta_{23} - \delta_{13})$$

$$[S_1^+, S_2^+] = [S_1^-, S_2^-] = [S_1^z, S_2^z] = 0$$

This procedure also requires the evaluation of traces of products of spin operators with highly variable permutations and combinations of site indices. For the lower order moments of present interest, the products are short to moderate in length, and fortunately, have been tabulated by Ambler et al.,[37] in a form, however, which requires some modification. Since the Heisenberg Hamiltonian in the form of Equation (2.2) is applied for overall computational convenience, the results of Reference 37, which give the traces of products of powers of S^x, S^y, and S^z, are reformulated in terms of S^+, S^-, and S^z by the defining Equation (2.3) for the raising and lowering spin operators, by the ancillary relations

$$S^+S^- = (S^x)^2 + (S^y)^2 + S^z \tag{3.1a}$$

$$S^-S^+ = (S^x)^2 + (S^y)^2 - S^z \tag{3.1b}$$

and by the various symmetry and permutation rules, regarding the spin exponents, given in Reference 37. A listing of repeatedly applied traces is given in Appendix A.

The special case of the zeroth frequency moment at infinite temperature follows, from the first term of Equation (2.25)

$$\left\langle \omega^0 \right\rangle_{\vec{K}}^{aa} = \beta \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})} \, \mathrm{Tr}(S_g^a S_w^a) / \mathrm{Tr}(1) = \beta a \tag{3.2}$$

where

$$a = \frac{1}{3} S(S+1) \tag{3.3}$$

and, of course,

$$\mathrm{Tr}(1) = 2S+1 \tag{3.4}$$

---

[37] E. Ambler, J.C. Eisenstein, and J.F. Schooley, J. Math. Phys. 3, 118 (1962).

If the distinct components $\alpha\alpha'$ in Equation (2.4) are extended through Equations (2.5) to (2.13) and are interpreted as "+ -", it can be shown that the transverse moments are

$$\left\langle \omega^n \right\rangle^{xx}_{\vec{K}} = \frac{1}{2} \left\langle \omega^n \right\rangle^{+-}_{\vec{K}} = \left\langle \omega^n \right\rangle^{yy}_{\vec{K}} \tag{3.5}$$

To calculate the non-vanishing second and fourth, longitudinal moments, the time derivatives of S^z_g (t) are found by Equation (2.17). Thus

$$i \frac{d}{dt} S^z_g(t) = \sum_f I_+(gf) \left[S^+_f S^-_g - S^+_g S^-_f \right] \tag{3.6}$$

and

$$\begin{aligned}
\left(i \frac{d}{dt} \right)^2 S^z_g(t) = & -2A \sum_f I_+(gf) \left[S^+_f S^-_g - S^+_g S^-_f \right] \\
& - 2A \sum_f I_+(gf) \left[S^z_g S^+_f S^-_g + S^z_g S^+_g S^-_f - S^z_f S^+_f S^-_g - S^z_f S^+_g S^-_f \right] \\
& - 2 \sum_{fm} I_+(gf) \left\{ I_+(mf) \left[S^z_f S^+_m S^-_g + S^z_f S^+_g S^-_m \right] \right. \\
& \left. - I_+(mg) \left[S^z_g S^+_f S^-_m + S^z_g S^+_m S^-_f \right] \right\} \\
& - 2 \sum_{fm} I_+(gf) \left[I_0(mg) - I_0(mf) \right] \left(S^z_m S^+_f S^-_g + S^z_m S^+_g S^-_f \right) \\
& - 2 \sum_f I_+(gf)\, I_0(gf) \left[S^+_g S^-_f - S^+_f S^-_g \right]
\end{aligned} \tag{3.7}$$

These equations omit the specification of time t in the spin operators on the right-hand side, since it is immaterial in the equal-time commutators of Equation (2.13).

If these results are used in the following specific forms of Equation (2.13)

$$\left\langle \omega^{2} \right\rangle_{\vec{K}}^{zz} = \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[i \frac{d}{dt} S_{g}^{z}(t), S_{w}^{z}(t) \right] \right\rangle \tag{3.8}$$

and

$$\left\langle \omega^{4} \right\rangle_{\vec{K}}^{zz} = \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[i \frac{d}{dt} S_{g}^{z}(t), \left(i \frac{d}{dt} \right)^{2} S_{w}^{z}(t) \right] \right\rangle \tag{3.9}$$

and if the thermal expansion of Equation (2.19) is performed for infinite temperature, then the longitudinal frequency moments are found to be

$$\left\langle \omega^{2} \right\rangle_{\vec{K}}^{zz} = 8\beta a^{2} \sum_{\vec{R}} I_{+}^{2}(\vec{R}) \left(1 - e^{i\vec{K}\cdot\vec{R}} \right) \tag{3.10}$$

and

$$\begin{aligned} \frac{\left\langle \omega^{4} \right\rangle_{\vec{K}}^{zz}}{32\beta a^{3}} = {} & A \Big\{ \frac{12}{5} \left(1 - \frac{1}{4a} \right) \sum_{\vec{R}} \left(e^{i\vec{K}\cdot\vec{R}} - 1 \right) I_{+}^{2}(\vec{R})\, I_{0}(\vec{R}) \Big\} \\ & + A^{2} \Big\{ \frac{6}{5} \left(1 - \frac{1}{4a} \right) \sum_{\vec{R}} \left(1 - e^{i\vec{K}\cdot\vec{R}} \right) I_{+}^{2}(\vec{R}) \Big\} \\ & + \frac{1}{5} \sum_{\vec{R}} \left(e^{i\vec{K}\cdot\vec{R}} - 1 \right) I_{+}^{2}(\vec{R}) \Big\{ \left[4 + \frac{3}{2a} \right] I_{0}^{2}(\vec{R}) \\ & + \left(16 + \frac{1}{a} \right) I_{+}^{2}(\vec{R}) \Big\} \\ & + \sum_{\vec{R}\vec{U}} \Big\{ 4\, e^{i\vec{K}\cdot\vec{R}}\, I_{+}(\vec{R}+\vec{U})\, I_{+}(\vec{R})\, I_{+}(\vec{U}) \left[I_{0}(\vec{U}) - I_{0}(\vec{R}) \right] \\ & + 2 \left(1 - e^{i\vec{K}\cdot\vec{R}} \right) I_{+}^{2}(\vec{R})\, I_{0}(\vec{U}) \left[I_{0}(\vec{U}) - I_{0}(\vec{R}+\vec{U}) \right] \\ & + \left(1 - e^{i\vec{K}\cdot\vec{R}} \right) I_{+}^{2}(\vec{R}) \left[5 - 3e^{i\vec{K}\cdot\vec{U}} \right] I_{+}^{2}(\vec{U}) \Big\} \end{aligned} \tag{3.11}$$

where $\vec{R}$ is the lattice vector between sites g and w, and $\vec{U}$ is the lattice vector between site g and all other sites included within the range of the exchange interaction.

Following the same procedure for the second and fourth transverse moments, we obtain, in order to proceed in alternate forms, the time derivatives of both S_g^+ (t) and S_w^- (t). Thus, the first derivatives are

$$
\begin{aligned}
i \frac{d}{dt} S_g^+(t) &= 2A\, S_g^z S_g^+ - A\, S_g^+ \\
&\quad - 2 \sum_f I_+(fg)\, S_g^z S_f^+ + 2 \sum_f I_0(fg)\, S_f^z S_g^+
\end{aligned}
\tag{3.12}
$$

or

$$
\begin{aligned}
i \frac{d}{dt} S_w^-(t) &= -2A\, S_w^z S_w^- - A\, S_w^- \\
&\quad + 2 \sum_m I_+(wm)\, S_w^z S_m^- - 2 \sum_m I_0(mw)\, S_m^z S_w^-
\end{aligned}
\tag{3.13}
$$

The second derivatives are

$$
\begin{aligned}
\left(i \frac{d}{dt}\right)^2 S_g^+(t) &= -4A \sum_i I_+(ig)\, S_g^z S_i^z S_i^z + 8A \sum_i I_0(ig)\, S_i^z S_g^z S_g^+ \\
&\quad + 4A \sum_i I_+(ig)\, S_g^z S_i^+ - 4A \sum_i I_0(ig)\, S_i^z S_g^+ \\
&\quad - 4A \sum_i I_+(ig)\, S_g^z S_g^z S_i^+ + 2A \sum_i I_+(ig) \left[S_i^+ S_g^- - S_g^+ S_i^- \right] S_g^+ \\
&\quad + 4A^2 S_g^z S_g^z S_g^+ - 4A^2 S_g^z S_g^+ + A^2 S_g^+
\end{aligned}
$$

$$+\ 4 \sum_{ij} I_+(jg)\, I_+(ij)\, S^z_g S^z_j S^+_i$$

$$+\ 2 \sum_{ij} I_+(ig)\, I_+(jg) \left[S^+_g S^-_j - S^+_j S^-_g \right] S^+_i$$

$$-\ 4 \sum_{ij} I_+(jg)\, I_o(ij)\, S^z_g S^z_i S^+_j$$

$$-\ 4 \sum_{ij} I_o(jg)\, I_+(ig)\, S^z_j S^z_g S^+_i$$

$$+\ 2 \sum_{ij} I_o(jg)\, I_+(ij) \left[S^+_i S^-_j - S^+_j S^-_i \right] S^+_g$$

$$+\ 4 \sum_{ij} I_o(jg)\, I_o(ig)\, S^z_i S^z_j S^+_g \qquad (3.14)$$

or

$$\left(i \frac{d}{dt} \right)^2 S^-_w(t) = -\ 4A \sum_r I_+(rw)\, S^-_r S^z_r S^z_w + 8A \sum_r I_o(rw)\, S^-_w S^z_w S^z_r$$

$$+\ 4A \sum_r I_+(rw)\, S^-_r S^z_w - 4A \sum_r I_o(rw)\, S^-_w S^z_r$$

$$-\ 4A \sum_r I_+(rw)\, S^-_r S^z_w S^z_w$$

$$+\ 2A \sum_r I_+(rw) \left[S^-_w S^+_w S^-_r - S^-_w S^+_r S^-_w \right]$$

$$+\ 4A^2 S^-_w S^z_w S^z_w - 4A^2 S^-_w S^z_w + A^2 S^-_w$$

$$+ 4\sum_{rs} I_+(sw)\, I_+(rs)\, S_r^- S_w^z S_s^z$$

$$+ 2\sum_{rs} I_+(rw)\, I_+(sw) \left[S_r^- S_s^+ S_w^- - S_r^- S_w^+ S_s^- \right]$$

$$+ 4\sum_{rs} I_+(sw)\, I_0(rs)\, S_s^- S_r^z S_w^z - 4\sum_{rs} I_0(sw)\, I_+(rw)\, S_r^- S_w^z S_s^z$$

$$+ 2\sum_{rs} I_0(sw)\, I_+(rs) \left[S_w^- S_s^+ S_r^- - S_w^- S_r^+ S_s^- \right]$$

$$+ 4\sum_{rs} I_0(sw)\, I_0(rw)\, S_w^- S_s^z S_r^z \tag{3.15}$$

The transverse moments are, by Equation (2.13)

$$\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx} = \frac{1}{2} \sum_{\vec{g}-\vec{w}} e^{-iK\cdot(\vec{g}-\vec{w})} \left\langle \left[i \frac{d}{dt} S_g^+(t), S_w^-(t) \right] \right\rangle \tag{3.16}$$

or

$$\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx} = \frac{1}{2} \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[S_g^+(t), - i \frac{d}{dt} S_w^-(t) \right] \right\rangle \tag{3.17}$$

and

$$\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx} = \frac{1}{2} \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[\left(i \frac{d}{dt} \right)^2 S_g^+(t), - i \frac{d}{dt} S_w^-(t) \right] \right\rangle \tag{3.18}$$

or

$$\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx} = \frac{1}{2} \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left\langle \left[i \frac{d}{dt} S_g^+(t), \left(-i \frac{d}{dt} \right)^2 S_w^-(t) \right] \right\rangle \tag{3.19}$$

After the commutators and traces are performed, the transverse moments are finally found, as follows:

$$
\begin{aligned}
\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx}}{16\beta a^2} = {} & \frac{3}{20} A^2 \left(1 - \frac{1}{4a}\right) + \frac{1}{4} \sum_{\vec{R}} \left[I_+^2 (\vec{R}) + I_0^2 (\vec{R}) \right. \\
& \left. - 2e^{i\vec{K}\cdot\vec{R}} \, I_+(\vec{R}) \, I_0 (\vec{R}) \right]
\end{aligned}
\qquad (3.20)
$$

and

$$
\begin{aligned}
\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx}}{64\beta a^3} = {} & A \left(1 - \frac{1}{4a}\right) \sum_{\vec{R}} \left\{ \frac{3}{5} e^{i\vec{K}\cdot\vec{R}} \left[I_+^3 (\vec{R}) - 2I_0^2 (\vec{R}) \, I_+ (\vec{R}) \right] \right. \\
& \left. + \frac{3}{5} I_+^2 (\vec{R}) \, I_0 (\vec{R}) \right\} \\
& + A^2 \left(1 - \frac{1}{4a}\right) \sum_{\vec{R}} \left\{ - \frac{6}{5} e^{i\vec{K}\cdot\vec{R}} \, I_+ (\vec{R}) \, I_0 (\vec{R}) \right. \\
& \left. + \frac{6}{5} I_+^2 (\vec{R}) + \frac{9}{10} I_0^2 (\vec{R}) \right\} \\
& + A^4 \left(\frac{27}{140} - \frac{39}{280a} + \frac{51}{2240a^2} \right) \\
& + \sum_{\vec{R}} \left\{ - \frac{1}{10} \left(3 + \frac{1}{2a}\right) I_0^4 (\vec{R}) - \frac{1}{2} \left(1 + \frac{1}{4a}\right) I_+^4 (\vec{R}) \right. \\
& + \frac{1}{10} \left(8 + \frac{1}{2a}\right) e^{i\vec{K}\cdot\vec{R}} \, I_+ (\vec{R}) \, I_0^3 (\vec{R}) \\
& - \frac{3}{10} \left(4 + \frac{1}{4a}\right) I_+^2 (\vec{R}) \, I_0^2 (\vec{R})
\end{aligned}
$$

$$
\begin{aligned}
&+\frac{2}{10}\left(6+\frac{1}{a}\right) e^{i\vec{K}\cdot\vec{R}}\, I_+^3(\vec{R})\, I_0(\vec{R}) \Bigg\} \\
&+\sum_{\vec{R}\vec{U}} \Bigg\{ \frac{3}{4}\, I_0^2(\vec{R})\, I_0^2(\vec{U}) + \frac{5}{4}\, I_+^2(\vec{R})\, I_+^2(\vec{U}) \\
&\quad -\frac{1}{2} e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{R})\, I_+(\vec{U})\, I_+(\vec{R}+\vec{U}) \\
&\quad -2\, e^{i\vec{K}\cdot\vec{R}}\, I_+(\vec{R})\, I_0(\vec{R})\, I_0^2(\vec{U}) \\
&\quad - e^{i\vec{K}\cdot\vec{R}}\, I_+(\vec{R})\, I_0(\vec{R})\, I_0(\vec{U})\, I_0(\vec{R}+\vec{U}) \\
&\quad + e^{i\vec{K}\cdot\vec{R}}\, I_+(\vec{R}+\vec{U})\, I_+(\vec{U})\, I_0(\vec{U}) \left[\frac{3}{2}\, I_0(\vec{R}+\vec{U}) + I_0(\vec{U})\right] \\
&\quad +\frac{1}{2} e^{i\vec{K}\cdot\vec{R}}\, I_0^2(\vec{R})\, I_+(\vec{U})\, I_+(\vec{R}+\vec{U}) + \frac{3}{2}\, I_+^2(\vec{R})\, I_0^2(\vec{U}) \\
&\quad - I_+(\vec{R})\, I_+(\vec{U})\, I_+(\vec{R}+\vec{U})\, I_0(\vec{R}+\vec{U}) \\
&\quad + e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{U})\, I_+(\vec{R})\, I_0(\vec{R}+\vec{U}) \\
&\quad - 3\, e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{U})\, I_+(\vec{R})\, I_0(\vec{R}) \Bigg\}
\end{aligned}
\qquad (3.21)
$$

It is noteworthy that two necessary conditions for the correctness of these results for the moments have been satisfied. The first of these is that the moments must be independent of axial anisotropy for a system with S=1/2. In this case, the quantity 4a=1, and the terms in the moments, containing the axial anisotropy constant A, vanish. The second condition is that the moments must contain no terms in N, the number of spin sites. A large number of terms in N and N^2 evolve during the course of calculation but manage to finally cancel.

However, other criteria for attesting to the probable correctness of these final forms of the moments can be applied. For example, in the limit of axial isotropy, i.e., A=0, the transverse and longitudinal moments correspond to the calculations in Reference 7. If, in addition, the exchange becomes isotropic, i.e., $I_+(\vec{R})=I_0(\vec{R})$, the transverse and longitudinal moments become equivalent and correspond to the calculations of Reference 4.

Finally, in the case when axial anisotropy is still finite but only the exchange is isotropic, these results converge to the limiting condition given by Lindgård[27], where all of the moments agree, except the fourth longitudinal moment, where Lindgård gives a value of the $AI^3(\vec{R})$ term of Equation (3.11), which is 1/2 of the present result. However, several checks on this calculation have sustained a conclusion that Equation (3.11) is correct. The exchange-anisotropic nature of the axial anisotropy terms in the moments is entirely new.

It can be noted that in the axially isotropic case the longitudinal moments vanish in the long wavelength limit, i.e., $K << 1$. This does not occur for the transverse moments, except under the additional condition of exchange isotropy.

In the calculation of these moments, a great amount of diagrammatic analysis is still present. However, it is at a lower level of complexity than in direct approaches, since the time-dependence in the Heisenberg representation of the dynamical spin operators has been removed from the structure of the moments. Even so, the moments given here are only about one twentieth as long as their most expanded forms, before combination and cancellation of terms occur.

CORRELATIONS IN ONE DIMENSION

The final forms of the infinite-temperature frequency moments expressed in the "General Form of the Frequency Moments" section are general, in the sense that they apply to any lattice structure, for arbitrary range of the exchange integrals and for any spin magnitude. In order to study the correlations in a certain structure, it is necessary to particularize the various moments for that lattice. To assist in this calculation, Appendix B discusses the geometrical ideas and presents some results. It is convenient to introduce

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4. M.F. Collins & W. Marshall, Proc. Phys. Soc. 92, 367 (1967).
7. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969); B-1, No. 7, 3178 (1970); B-1, No. 9, 3649 (1970); B-1, No. 9, 3671 (1970).
27. P.A. Lindgård, in Neutron Inelastic Scattering, 2, 93 (I.A.E.A., 1968).

a few new definitions. Defining the nearest neighbor distance as (1) and the next nearest neighbor distance as (2), and, in addition, normalizing the energies by the nearest neighbor z-z exchange, $I_0(1)$, that is

$$G = \frac{I_+(1)}{I_0(1)} \; ; \; H = \frac{I_0(2)}{I_0(1)} \; ; \; L = \frac{I_+(2)}{I_0(1)} \; ; \; D = \frac{A}{I_0(1)} \tag{4.1}$$

and introducing, for a linear chain,

$$u_1 = \cos(K) \; ; \; v_1 = \cos(2K) \tag{4.2}$$

we can, with the considerations of Appendix B, recast the longitudinal frequency moments in the following one-dimensional form for a linear chain,

$$\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{zz}}{\beta[4aI_0(1)]^2} = (1-u_1)G^2 + (1-v_1)L^2 \tag{4.3}$$

and

$$\begin{aligned}
\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{zz}}{128\beta a^3 I_0^4(1)} = {} & \frac{6}{5}D\left(1-\frac{1}{4a}\right)\left[(u_1-1)G^2+(v_1-1)L^2H\right] \\
& + \frac{3}{5}D^2\left(1-\frac{1}{4a}\right)\left[(1-u_1)G^2+(1-v_1)L^2\right] \\
& + 2(v_1-u_1)G^2L(1-H)+2(u_1-1)G^2H \\
& + (v_1-1)L^2+\frac{2}{5}\left[(u_1-1)G^2+(v_1-1)L^2H^2\right] \\
& + 2\left[(1-u_1)G^2+(1-v_1)L^2\right](1+H^2)-\frac{8}{5}\left[(1-u_1)G^4+(1-v_1)L^4\right]
\end{aligned}$$

$$+ 5(G^2 + L^2)^2 + 3(u_1 G^2 + v_1 L^2)^2 - 8(u_1 G^2 + v_1 L^2)(G^2 + L^2)$$

$$+ \frac{1}{10a}\left[(u_1 - 1)(G^4 + \frac{3}{2}G^2) + (v_1 - 1)(L^4 + \frac{3}{2}L^2 H^2)\right] \tag{4.4}$$

The transverse moments become

$$\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx}}{16\beta a^2 I_0^2(1)} = \frac{3}{20}D^2\left(1 - \frac{1}{4a}\right) - (u_1 G + v_1 H L)$$

$$+ \frac{1}{2}(1 + H^2 + G^2 + L^2) \tag{4.5}$$

and

$$\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx}}{128\beta a^3 I_0^4(1)} = D\left(1 - \frac{1}{4a}\right)\left[\frac{3}{5}(u_1 G^3 + v_1 L^3)\right.$$

$$\left. + \frac{6}{5}(u_1 G + v_1 H^2 L) + \frac{3}{5}(G^2 + L^2 H)\right]$$

$$+ D^2\left(1 - \frac{1}{4a}\right)\left[-\frac{6}{5}(u_1 G + v_1 HL) + \frac{6}{5}(G^2 + L^2) + \frac{9}{10}(1 + H^2)\right]$$

$$+ 2D^4\left[\frac{27}{560} - \frac{39}{1120a} + \frac{51}{8960a^2}\right]$$

$$+ \frac{1}{20}\left(6 + \frac{1}{a}\right)\left(-1 - H^4 + 4u_1 G^3 + 4v_1 L^3 H\right)$$

$$+ \frac{1}{20}\left(16 + \frac{1}{a}\right)\left(u_1 G + v_1 LH^3 - \frac{3}{2}G^2 - \frac{3}{2}L^2 H^2\right)$$

$$- \frac{1}{8}\left(4 + \frac{1}{a}\right)\left(G^4 + L^4\right) + \frac{3}{2}\left(1 + H^2\right)^2 + \frac{5}{2}\left(G^2 + L^2\right)^2 + 3\left(u_1 G + v_1 LH\right)^2$$

$$- 4(u_1 G + v_1 LH)(1 + H^2) + 3(G^2 + L^2)(1 + H^2)$$

$$- 6(u_1 G + v_1 HL)(G^2 + L^2)$$

$$+GL\left[u_1(2-G^2+L+H)+v_1G(1-\frac{1}{2}L)-GH-2G\right]$$
$$+GH\left[u_1(G^2-2)+v_1(G+\frac{1}{2}H)\right]-v_1HL \tag{4.6}$$

These results exclude exchange interactions beyond the second nearest neighbor.

Therefore the temporal correlations are determined numerically as functions of the energy ratios G, H, L, and D, and of the magnitude of the spin variable, a. The non-dimensionalizing exchange integrals, in this case $I_0(1)$, with its nearest interspin separation, do not give rise to any explicit dependence but rather determine a reduced set of units in which space and time are measured, according to the "law of corresponding states", discussed in Reference 7. This law postulates that there exist appropriate reduced scales for the correlations and for the frequency ω and time t, such that correlations for different spin magnitudes S are approximately the same. This is accomplished for the Fourier transform by plotting $I \langle S_g^{\alpha}(t)\, S_w^{\alpha}(0) \rangle_{\omega}\, (4a)^{-1/2}$ versus $\omega[I(4a)^{1/2}]^{-1}$ and for the time correlation, by plotting $\langle S_g^{\alpha}(t)\, S_w^{\alpha}(0) \rangle\, (a)^{-1}$ versus $It(4a)^{-1/2}$. This law was found to have greater validity as the dimensionality and range of the interaction is increased, since the double sums in the fourth moments given in Equations (3.11) and (3.21) become more important, under these conditions, than the single sums, which are spin-dependent.

For two cases, infinite spin and unity spin, the correlations have been calculated for a linear chain under various conditions. These results are given in Figures 1 through 12. The curves for S=1 are not displayed, except in one case in Figure 2, since they are quite close, usually within a few line thicknesses, to the infinite spin curves, in accordance with the above statements. However, as the axial anisotropic constant, D, increases (or decreases to large negative values), the unity spin curves will show an increased separation from the infinite spin curves, due to the dependence of the D terms in the moments upon the spin parameter, a.

For vanishing axial anisotropy D=0, nearest neighbor exchange only, H=L=0, the line shape is a function of G. Where G, under these conditions, is large, i.e., $I_0 \to 0$, the system approaches the XY model, having only transverse exchange; but when $G \to 0$, i.e., $I_+ \to 0$, the system reduces to a nearest neighbor Ising model with only longitudinal exchange.

Figure 1 displays the infinite spin results for the Fourier transform of the longitudinal self-correlation function under nearest neighbor interactions

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7. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969); B-1, No. 7, 3178 (1970); B-1, No. 9, 3649 (1970); B-1, No. 9, 3671 (1970).

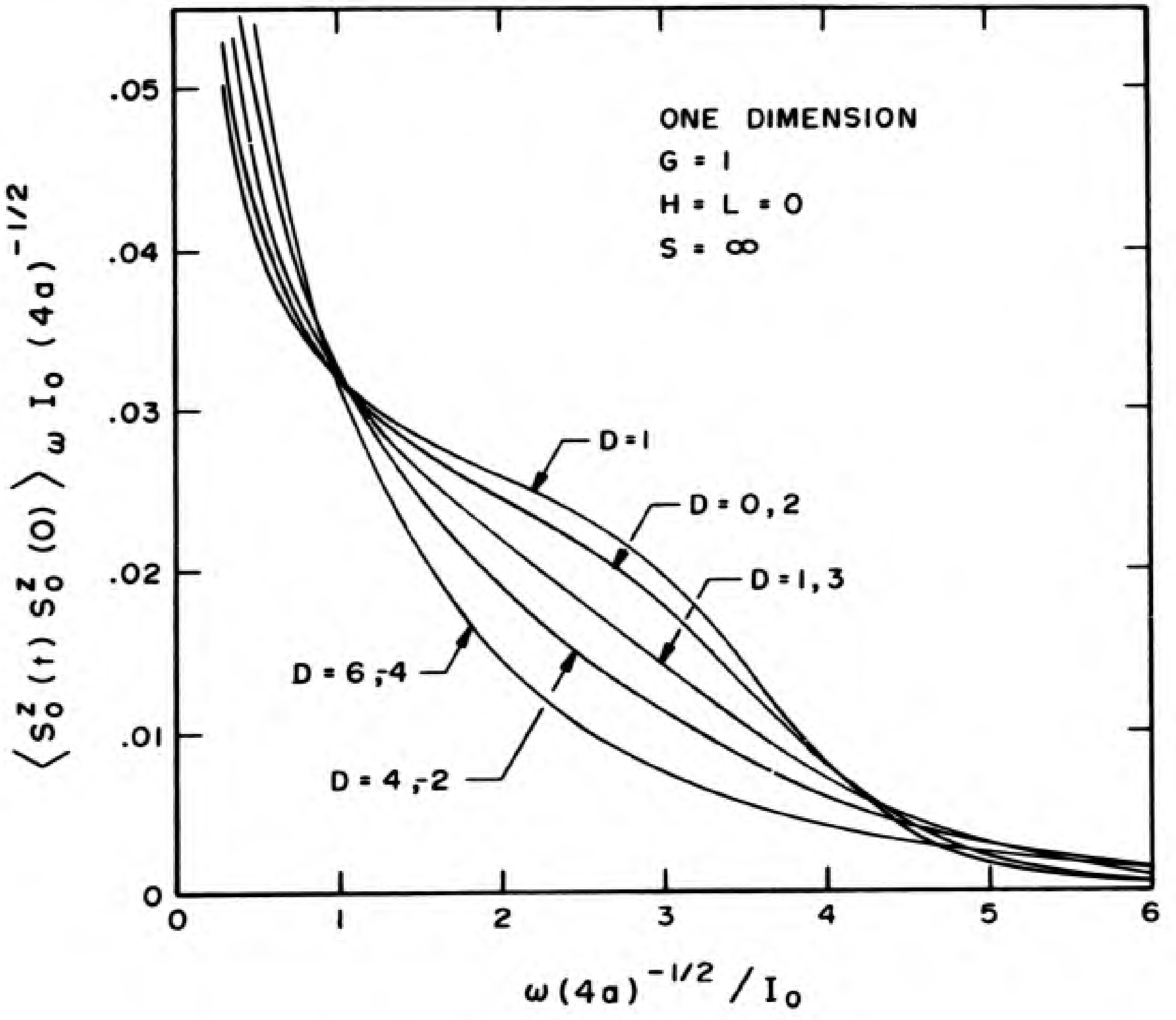

Figure 1. The Frequency Fourier Transforms of the Longitudinal Self-Correlation versus Reduced Frequency and the Axially Anisotropic Parameter D, for a Linear Chain With Nearest Neighbor (H=L=0), Isotropic Exchange (G=1). Only the Infinite Spin Results Are Shown.

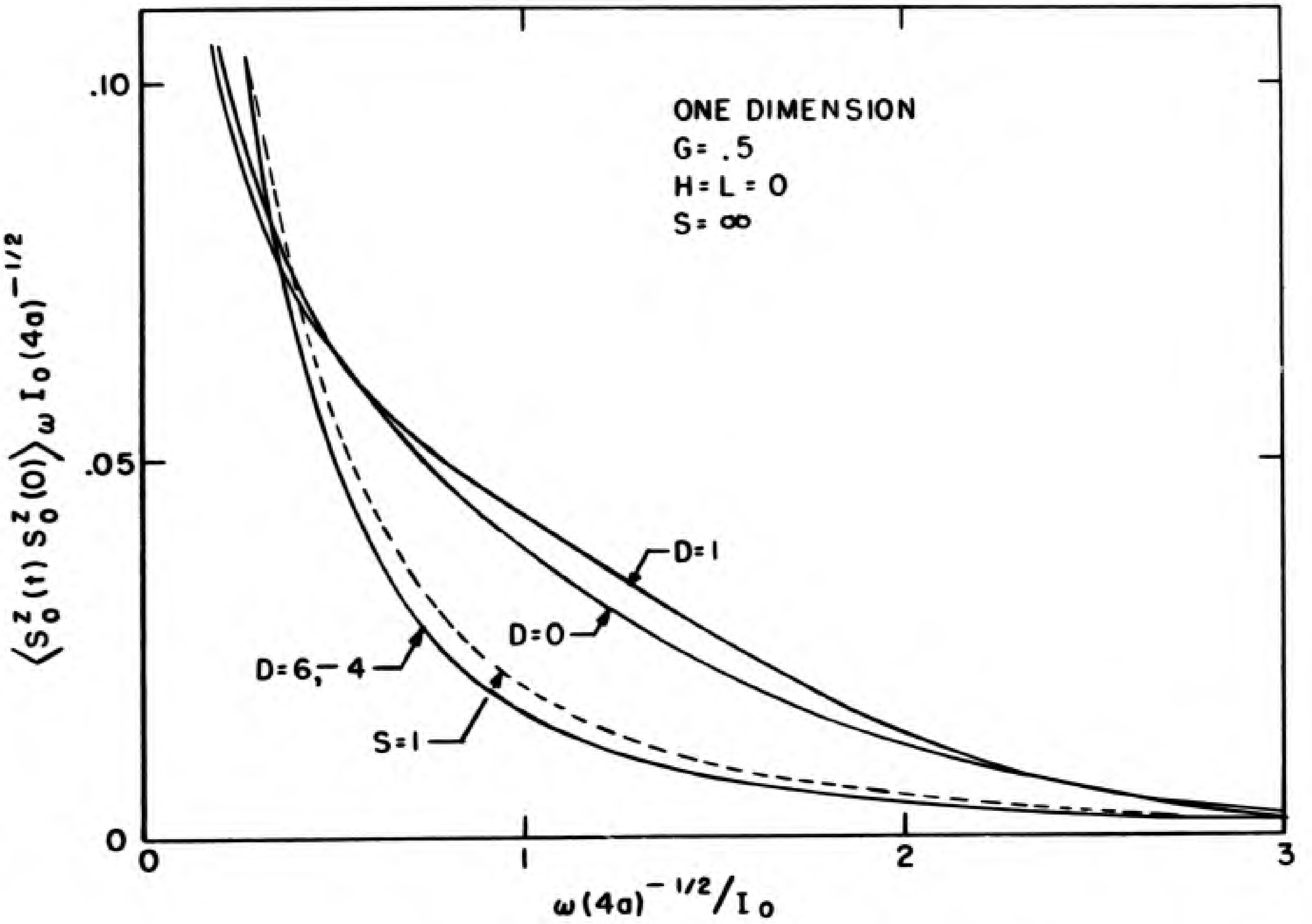

Figure 2. The Same as for Figure 1, Except that the Anisotropic Exchange Parameter G=0.5, i.e, the Ratio of the Transverse Exchange to the Longitudinal Exchange is now 1/2. For D=6, the Broken Line Curve Shows the Unity Spin Transform.

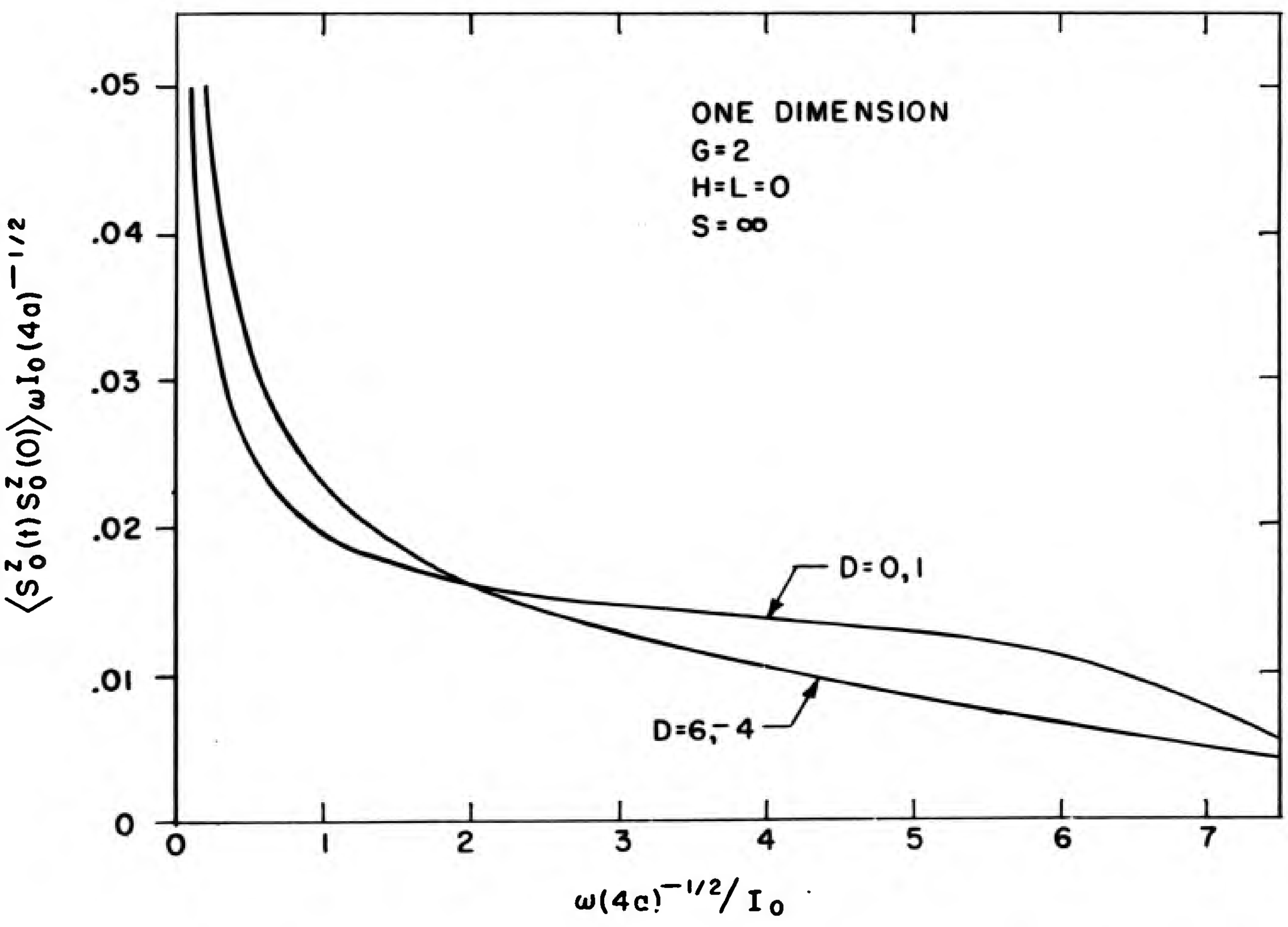

Figure 3. The Frequency Fourier Transforms of the Longitudinal Self-Correlation versus Reduced Frequency and the Axially Anisotropic Parameter D, for a Linear Chain with Nearest Neighbor (H=L=0), Isotropic Exchange (G=2). Only the Infinite Spin Results Are Shown.

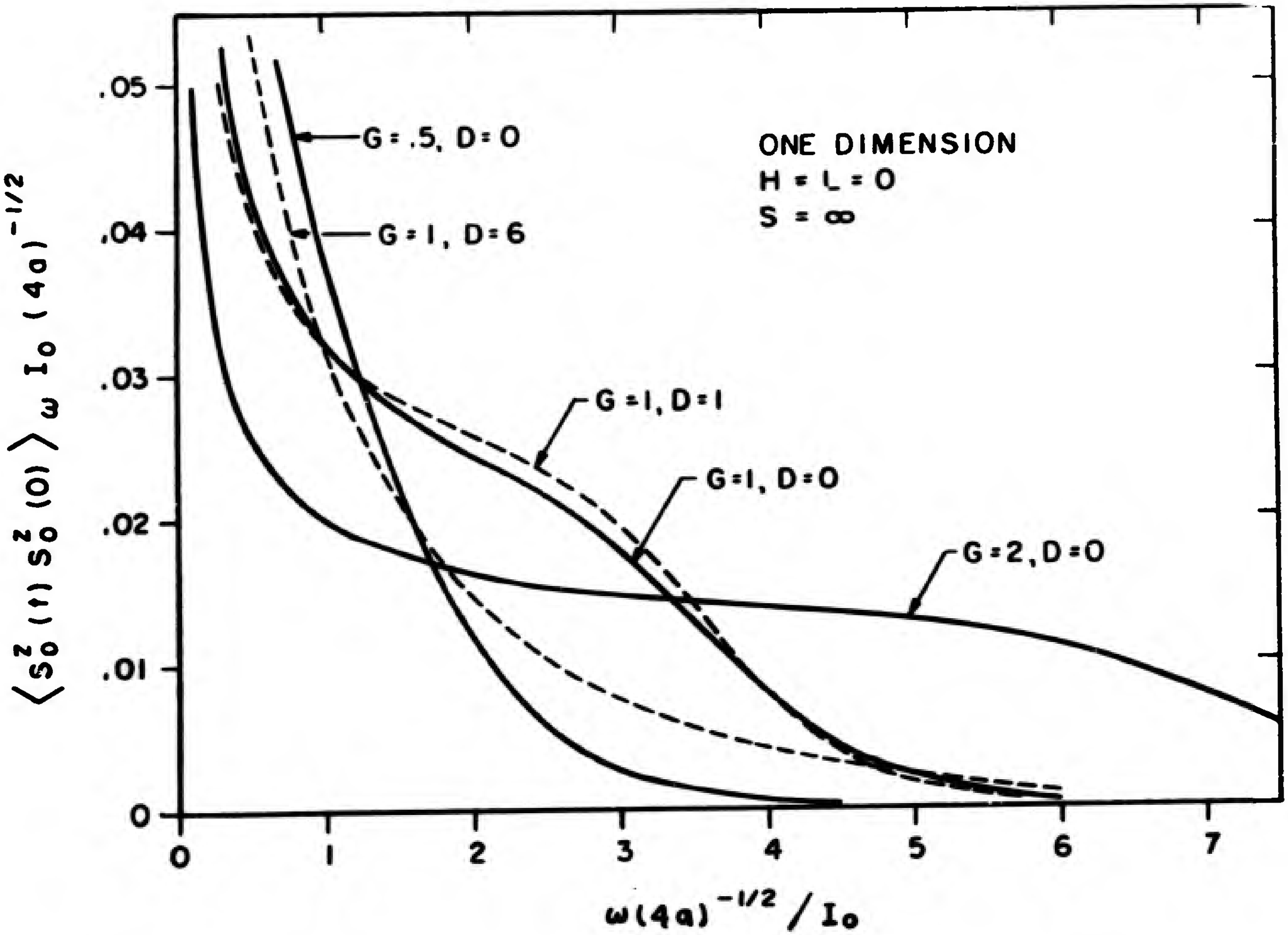

Figure 4. The Frequency Fourier Transforms of the Longitudinal Self-Correlations versus Reduced Frequency, Exchange Anisotropy G With D=0, and Axial Anisotropy D With G=1, for a Linear Chain of Infinite Spins, With Only Nearest Neighbor Interactions (H=L=0).

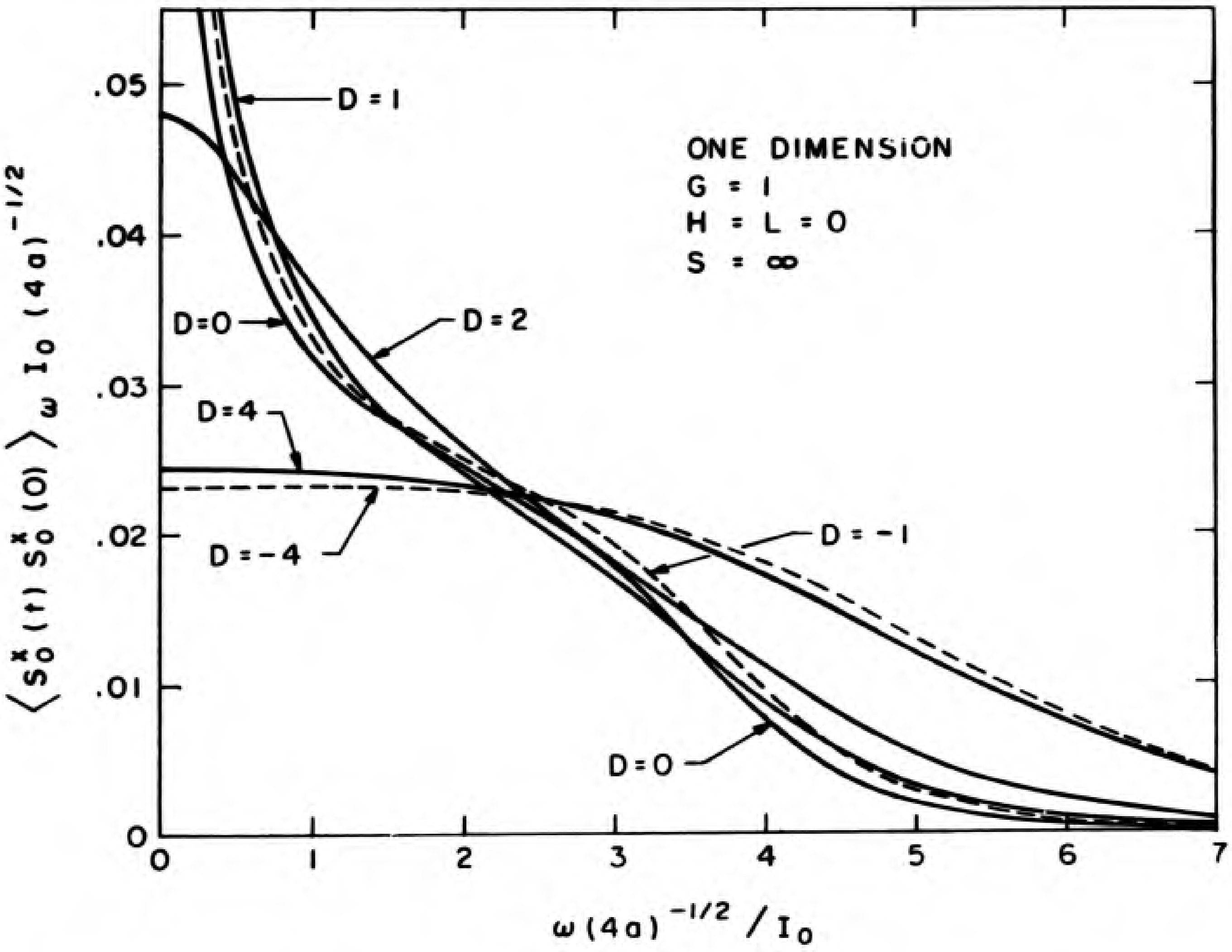

Figure 5. The Effect of the Axial Anisotropy D Upon the Frequency Fourier Transforms of the Transverse Self-Correlation versus Reduced Frequency, for a Linear Chain of Infinite Spins With Nearest Neighbor (H=L=0), Isotropic Exchange (G=1).

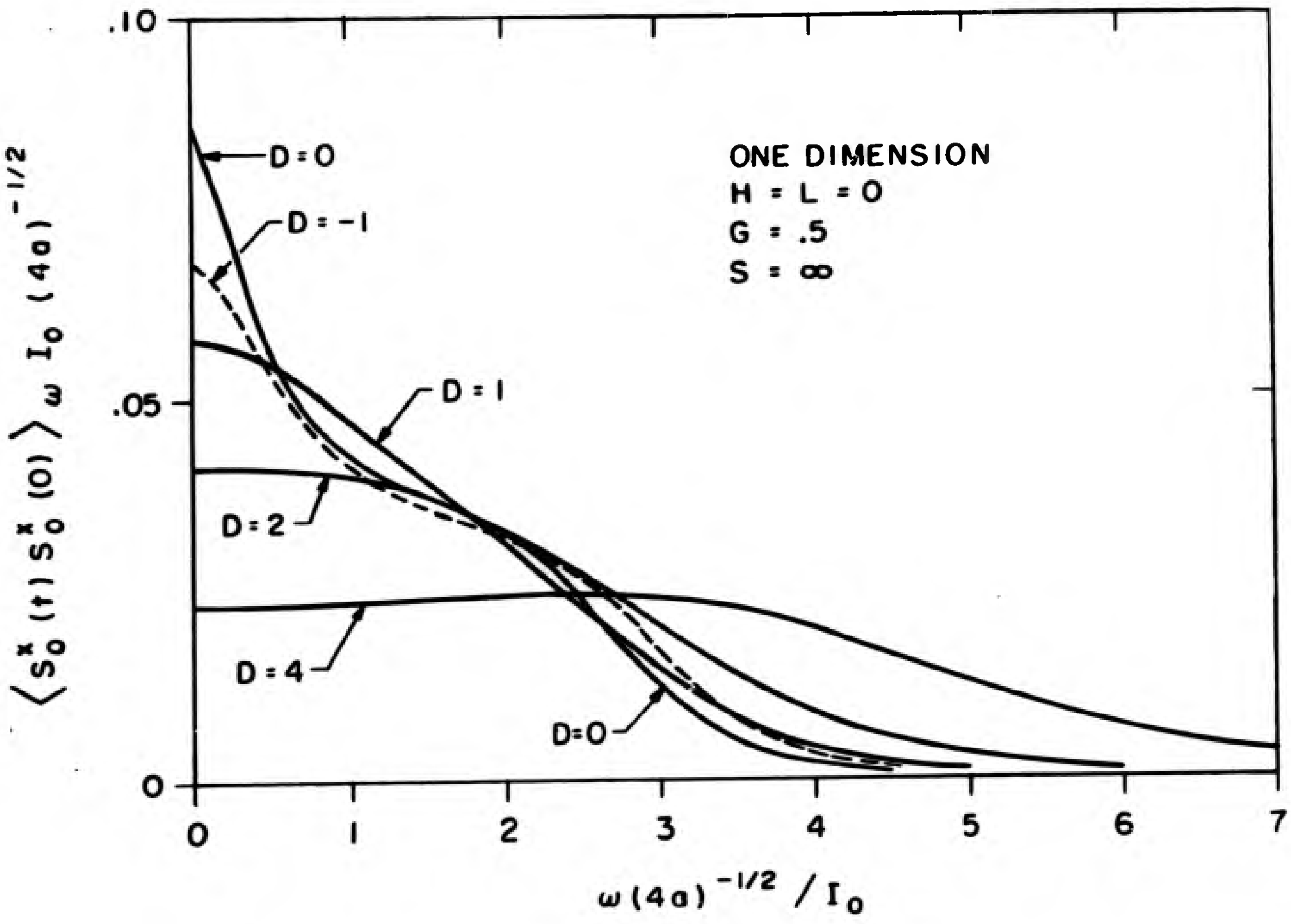

Figure 6. The Effect of the Axial Anisotropy D Upon the Frequency Fourier Transforms of the Transverse Self-Correlation versus Reduced Frequency, for a Linear Chain of Infinite Spins With Nearest Neighbor (H=L=0), Anisotropic Exchange (G=0.5).

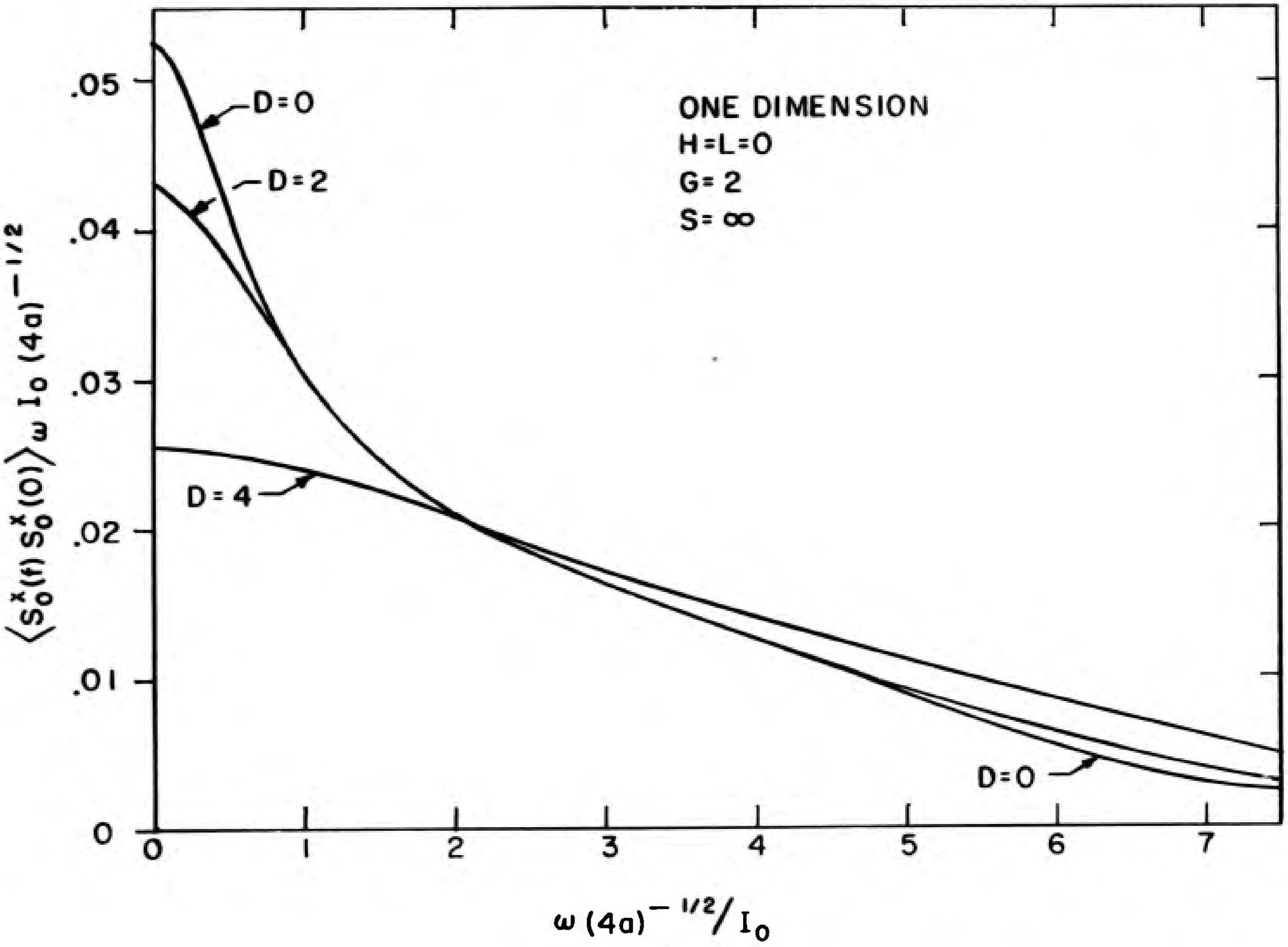

Figure 7. The Effect of the Axial Anisotropy D Upon the Frequency Fourier Transforms of the Transverse Self-Correlation versus Reduced Frequency, for a Linear Chain of Infinite Spins With Nearest Neighbor (H=L=0), Anisotropic Exchange (G=2).

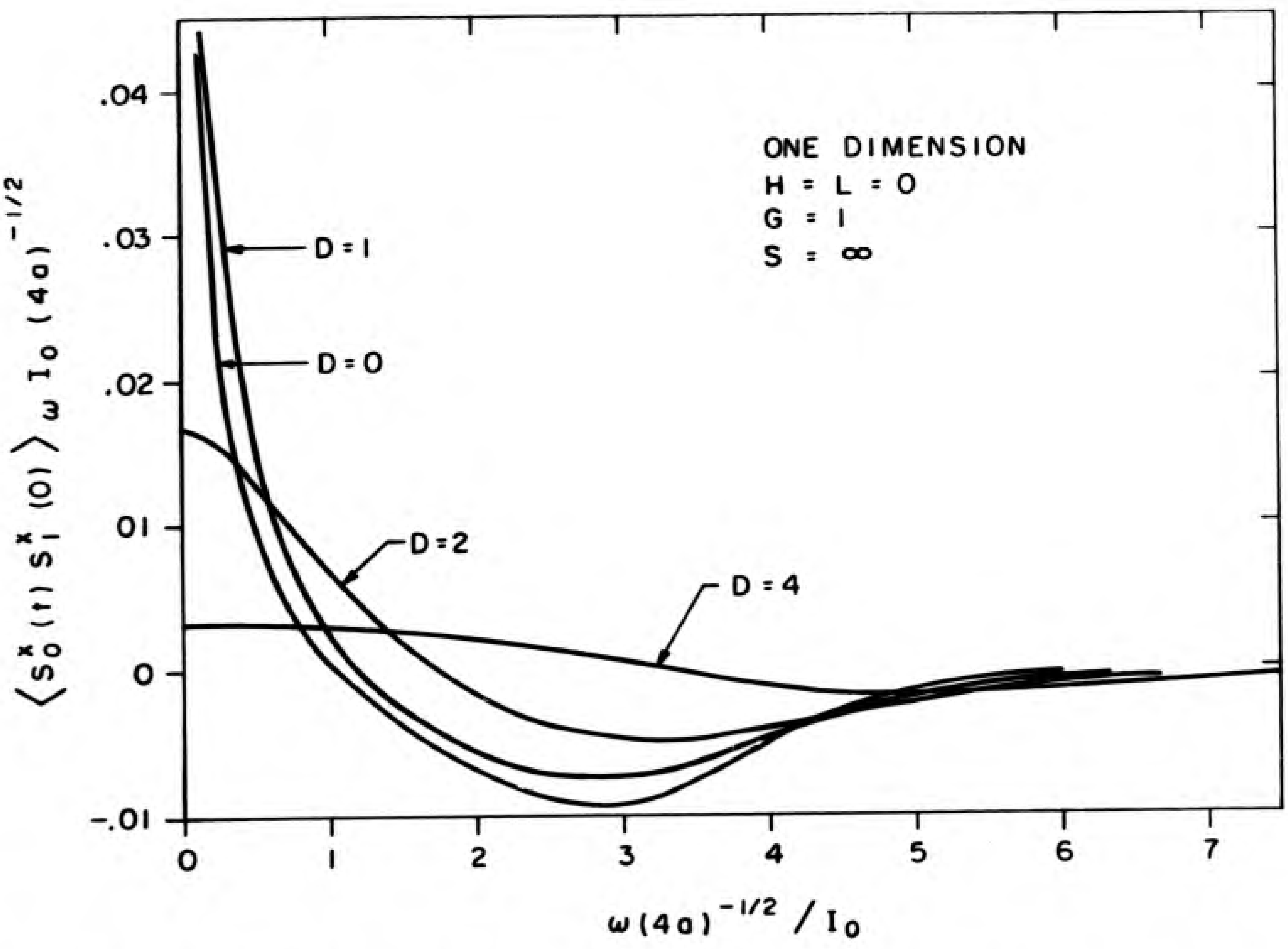

Figure 8. The Frequency Fourier Transforms of the Transverse, First Nearest Neighbor Correlation versus Reduced Frequency, and Their Variation with D, for a Linear Chain of Infinite Spins With Only Nearest Neighbor, Isotropic Exchange.

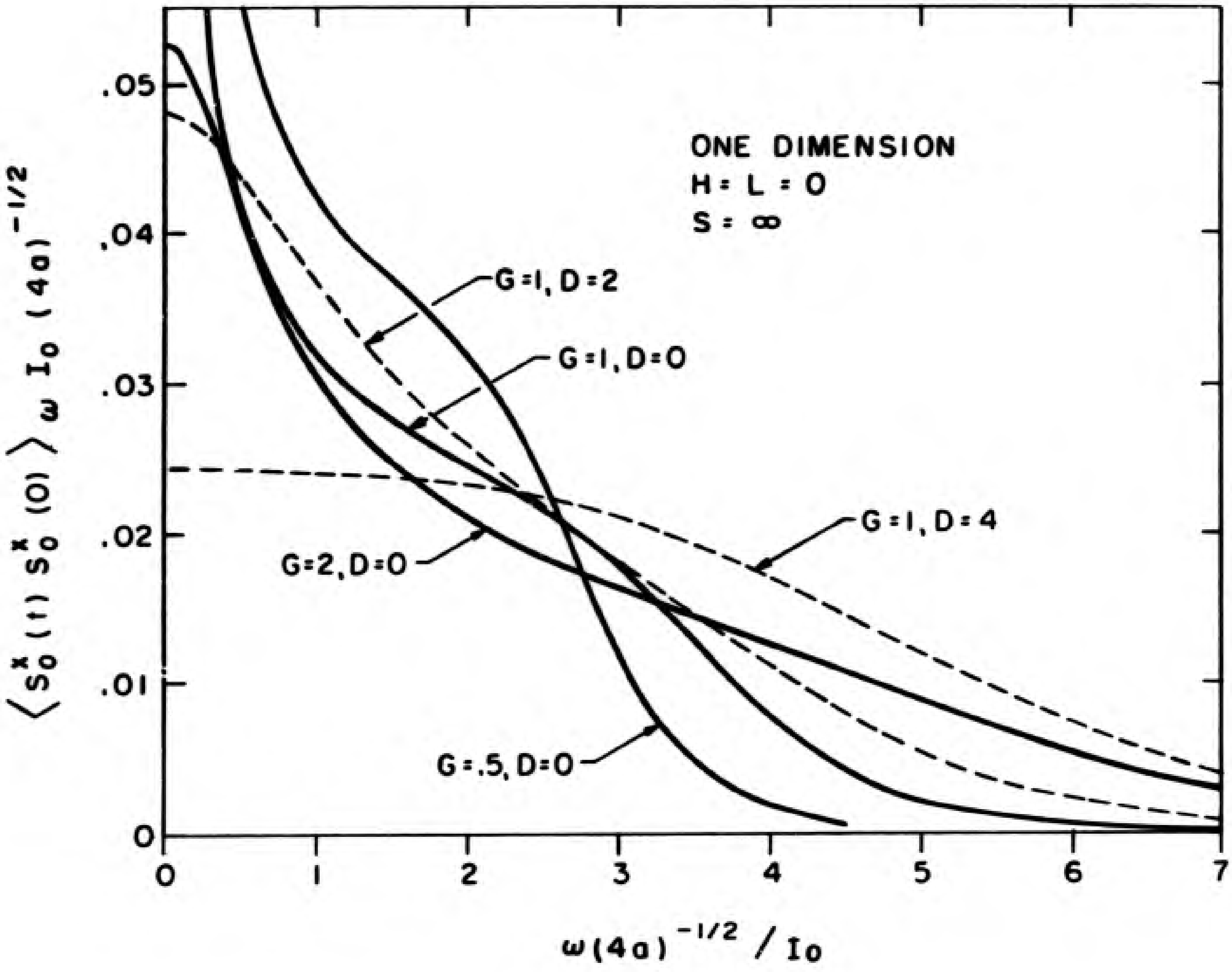

Figure 9. The Variation, With G and D Considered Separately, of the Frequency Fourier Transform of the Transverse, Self-Correlation, for a Linear Chain of Infinite Spins With Nearest Neighbor Interactions.

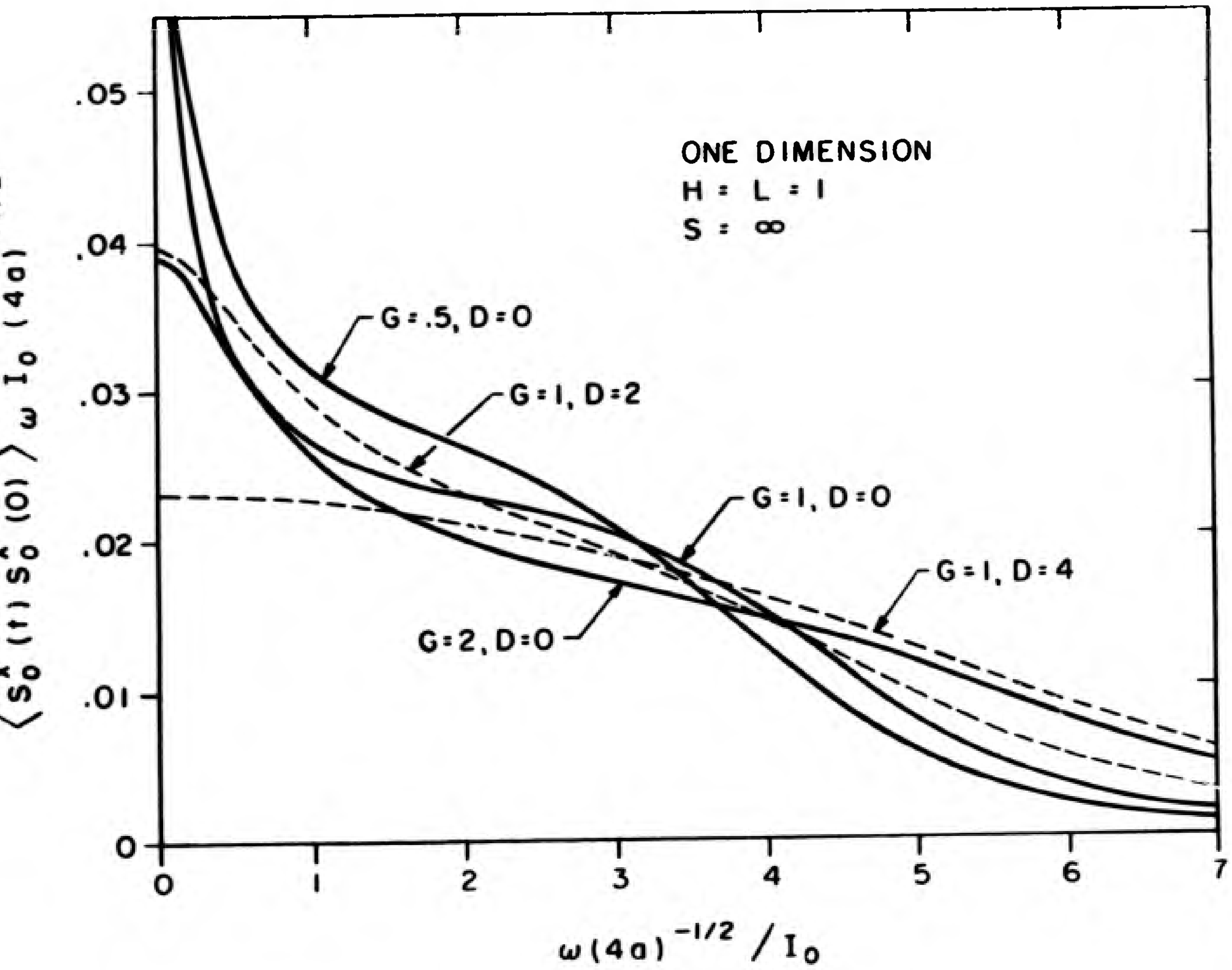

Figure 10. The Variation, with G and D Considered Separately, of the Frequency Fourier Transform of the Transverse, Self-Correlation, for a Linear Chain of Infinite Spins with Next Nearest Neighbor Interactions (H=L=1).

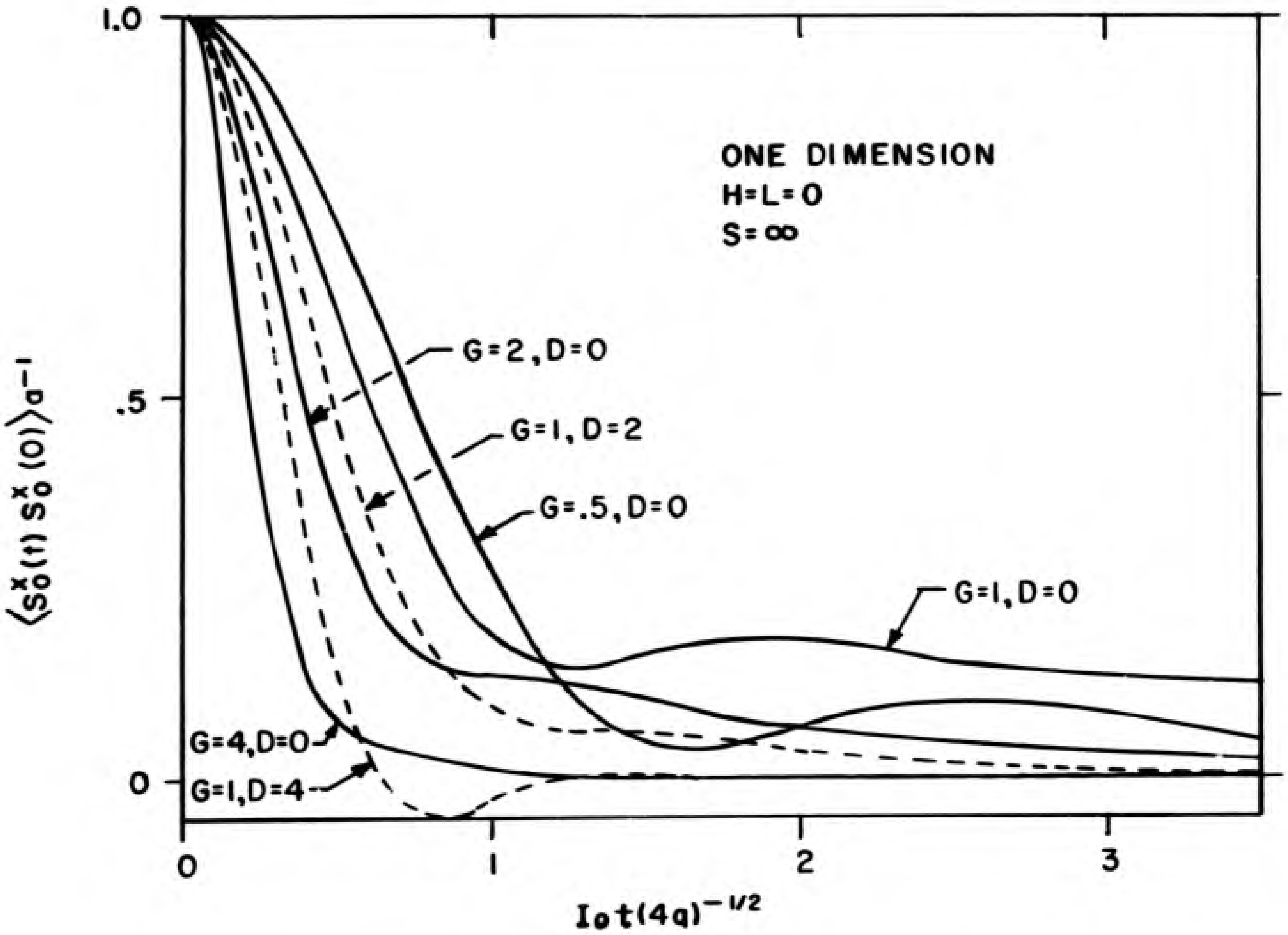

Figure 11. The Variation, with G and D, of the Temporal, Transverse Self-Correlations versus Reduced Time, for the One-Dimensional, Infinite Spin, Linear Chain With Nearest Neighbor Interactions.

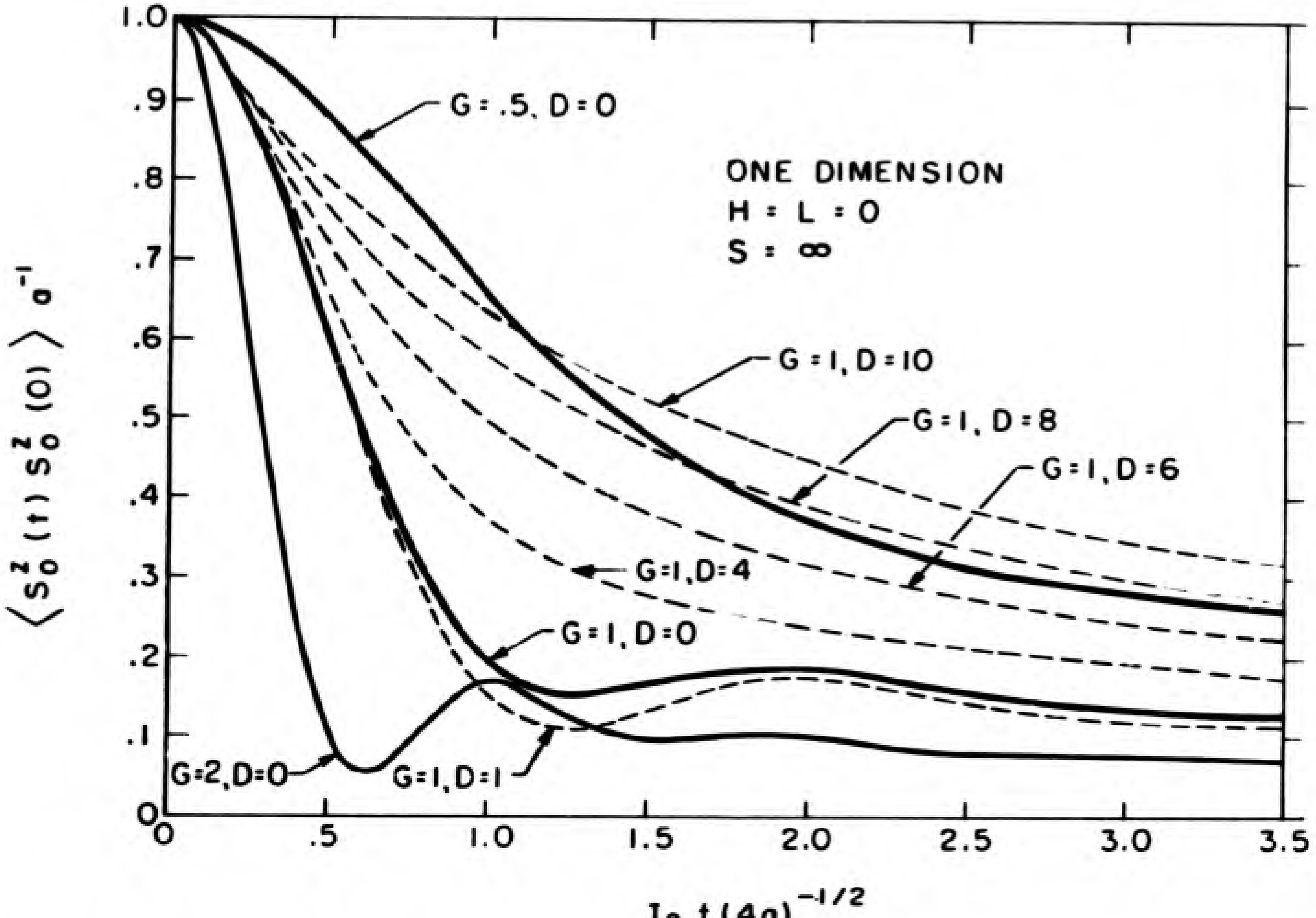

Figure 12. The Variation, with G and D, of the Time Dependent, Longitudinal Self-Correlations Considered for the One-Dimensional, Infinite Spin, Linear Chain With Nearest Neighbor Interactions.

(H=L=0) and isotropic exchange, (G=1) as a function of frequency and the axial anisotropic parameter, D. It is noted that two values of D result in the same Fourier transform. This can be easily explained by observing the quadratic dependence of the D-terms in the fourth longitudinal moment, Equation (5.4). In this case, with G=1, L=H=0, these terms have a dependence upon D, given by the relation, $2D-D^2$=constant, and the D-terms reach a maximum for D=1. As D increases to large values or decreases to large negative values, the frequency spectrum of the Fourier transform becomes compressed toward lower frequencies and appears to be approaching the limiting case for the Ising model, which would show a delta-function at the origin.

In Figure 2, the value of G is changed to 0.5. Here we see that the curves have shifted still more toward lower frequencies. (Note the change in scale.) We also show the separation of the unity spin curve from the infinite spin curve in the case when D=6(or -4). This separation is probably the largest one of all the cases considered herein, and since most unity spin results are considerably closer to those of the corresponding infinite spin cases, the correlations will henceforth not display the S=1 calculations.

In Figure 3, where G=2, it can be seen that for large G, the effects of changes in D are highly mitigated. It has been shown that the curves for large G appear to be approaching the corresponding results for the XY model.

The combined effects of G and D are shown in Figure 4, where, for D=0, curves of G=.5, 1, and 2 are given and for G=1, curves of D=1 and 6(or -4) are included. The results for G=.5, D=0 are quite similar to those for G=1, D=6. It can be concluded for the longitudinal transforms, that large values of axial anisotropy, (positive or negative) yield results similar to those obtained with small exchange anisotropy, whereas the large anisotropic exchange behavior cannot be duplicated by any axial anisotropy.

Note the divergences of these Fourier transforms of the longitudinal correlation for $\omega \to 0$ as discussed in the Section "Formulation" under "The Hydrodynamical Representation of Spin Diffusion." A proof of the logarithmic nature of this divergence has been given by Fernandez and Gersch.[38]

Continuing the study of this nearest neighbor case (H=L=0), we next examine the structure of the transverse self-correlations. First, the transverse correlations will be approximately the same with a large absolute value of D, as can be ascertained by examining the transverse frequency moments in Equations (5.5) and (5.6), where it is seen that the second moment includes a D^2 term and the fourth moment contains terms in D, D^2, and D^4. For large

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[38] J.F. Fernandez and H. Gersch, Phys. Rev. 172, 341 (1968).

$|D|$, the linear term in the fourth moments becomes less important relative to the D^2 and D^4 terms. In Figure 5, the frequency Fourier transforms of the infinite spin, transverse self-correlation are plotted for various values of the axial anisotropy, D=0, 1, 2, 4, -1, -4, under the conditions of exchange isotropy (G=1) and nearest neighbor interaction range. When the system is completely isotropic (D=0, G=1), this resulting curve is identical to the corresponding results for the longitudinal self-correlation. However, when $D \neq 0$, both the small frequency and large frequency behavior of the transverse auto-correlations differ from the corresponding longitudinal results in two ways. First, for large $|D|$, the transverse curves show no divergence for $\omega \rightarrow 0$, whereas the longitudinal cases do diverge (See Figure 1). Secondly, the transverse curves, for large $|D|$, undergo a flattening and the cutoff frequency, i.e., where the correlation approaches zero, is greatly extended, as contrasted to the longitudinal curves.

This behavior can be explained by the fact that the z-component of spin is a constant of the motion. Therefore, this fact will usually give rise to a spin density diffusion, i.e., in the long wave length, long-time limit, the local density of the z component of spin will follow a diffusion equation. In contrast, the x and y components of spin are not constants of the motion unless the system is completely isotropic (G=1, D=0). Therefore the transverse components of spin density do not follow a diffusion equation. In Figure 5, the closeness of the D=4 and -4 curves agree with previous comments, regarding the approximate quadratic dependence upon D, when D is large.

In Figure 6, the only change from Figure 5 is that the exchange anisotropy is introduced, by a decrease with G=.5. Here the transverse curves experience only a slight shift to lower frequencies, (note the change in scale), and for large $|D|$, a broad maximum has appeared. In Figure 7, the exchange anisotropy is increased to G=2, and the effects of changing D, it is seen, are greatly reduced, as expected, by the previous discussion, and by the corresponding results for the longitudinal correlation.

The frequency Fourier transform of the nearest neighbor transverse correlation is displayed in Figure 8. Here many of the above-mentioned trends are evident. In order to preserve the sum rule

$$a\delta_{om} = \lim_{\beta \to 0} \int_{-\infty}^{\infty} \left\langle S_o^a(t) S_m^a(0) \right\rangle_\omega d\omega \tag{4.7}$$

this curve must go below the frequency axis.

It is seen that these nearest neighbor correlations become quite small for large values of axial anisotropy D, as can be expected from the functional dependence of the frequency moments in this limit.

In Figure 9, the effects of varying the nearest neighbor exchange and axial anisotropy are explored in the transverse self-correlation. Here, we allow G=1, 2, .5, for D=0, and, in addition, D=2, 4, for G=1. These effects for the transverse case are quite different from those of the corresponding longitudinal results in Figure 4. First of all, the transverse correlations in the high frequency region for large exchange anisotropy (G=2, D=0) can be simulated with large axial anisotropy (D=2, 4; G=1). Also, at least to a limited extent, the moderately-low frequency results obtained by decreasing G to a value of .5, can also be simulated roughly by a limited increase in the axial parameter D, to a value of approximately 2. In addition, the flattening of the transverse correlation for large |D| is similar to the corresponding results for large G. (It is not shown, but the G=4, D=0 curve lies somewhat below and above the G=1, D=4 curve for low and high frequencies, respectively.)

The effect of next nearest neighbor exchange upon the previous results is examined in Figure 10, where H=L=1. All of the results become highly constricted, as expected, showing that the inclusion of higher range order lessens the effects of both nearest neighbor exchange anisotropy and axial anisotropy.

The time variation of the temporal, transverse self-correlations are shown in Figure 11, for nearest neighbors and various parameters of anisotropy, i.e., G=.5, 1, 2, 4 with D=0, and D=2, 4 with G=1. These curves are the appropriate inverse Fourier transforms of the corresponding curves in Figure 9. It is seen that the decay rate of the time-correlation increases and decreases directly as G increases or decreases from its exchange isotropic value of 1. Variations in the axial parameter D cannot duplicate the decreased decay rate for small G, but the increased decay rate for large G can be simulated with large D.

Finally, in Figure 12, we examine the time-variation of the longitudinal, time-dependent self-correlations for nearest-neighbor exchange and various values of the anisotropic constants, i.e., G=.5, 1, 2 with D=0 and D=1, 4, 6, 8, 10 for G=1. These curves show decay rates which increase or decrease according to the change of G from its isotropic value of 1, similarly to the transverse correlations in Figure 11. The curve for G=.5 is approaching the structure of the Ising limit, where the decay would cease completely, since the z component of each spin would be a constant of the motion. (For the Ising model, i.e., G=0, D=0, the curve would be merely a straight line with the ordinate =1.) The principal result is that increasing the value of / D / to large values also decreases the time-decay of the longitudinal correlations, similar to decreases in G. This expected result is the reverse of the transverse correlations.

An additional limiting case of the present results is to consider the spin correlation for the purely crystal-field Hamiltonian. If the exchange vanishes in Equation (2.1), the Hamiltonian becomes

$$H = -A\sum_f \left(S_f^z\right)^2 \tag{4.8}$$

This simplified spin system admits an exact solution for the transverse spin correlation in the unity spin case. The longitudinal spin correlation vanishes due to the fact that the trace of S_g^z to any odd integer power equals zero.

In order to study the thermodynamics of this system, we shall use, as in Equation (2.43), the temperature-dependent, double-time retarded Green functions,[39] in the general form

$$\langle\langle C(t); B(t')\rangle\rangle = -i\theta(t-t')\left\langle \left[C(t), B(t')\right]\right\rangle \tag{4.9}$$

The relevant thermodynamic averages or statistical correlations introduced in Equation (2.8), are determined from the Fourier transform of these Green functions in the usual manner, i.e.

$$\left\langle CB\right\rangle = \lim_{\epsilon\to 0}\left[-2\int_{-\infty}^{\infty} d\omega \, \frac{\operatorname{Im}\langle\langle C;B\rangle\rangle_{(\omega+i\epsilon)}}{1-e^{-\beta\omega}}\right] \tag{4.10}$$

Here we have used the notation

$$\langle\langle C;B\rangle\rangle_{(E)} = \frac{1}{2\pi}\int_{-\infty}^{\infty}\langle\langle C(t);B(t')\rangle\rangle \, e^{iE(t-t')} \, d(t-t'), \operatorname{Im} E > 0 \tag{4.10a}$$

The quantity $\langle\langle C;B\rangle\rangle_{(E)}$ is the analytic extension of the Fourier transform of the Green function into the upper half of the complex energy plane.

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[39] D.N. Zubarev, Soviet Physics Uspekhi, 3, 320 (1960).

It is convenient to define two different Green functions,[40] namely $G^{(n)}(t-t')$ with n=0 and 1, where

$$G^{(n)}(t-t') = \langle\langle C^{(n)}(t); S^-(t')\rangle\rangle, \quad C^{(n)}(t) = \left[S^z(t)\right]^n S^+(t) \tag{4.11}$$

The equations of motion of these Green functions are now determined under the Hamiltonian (4.8) and their energy transforms are found according to Equation (4.10a). With the property common to all unity spin Ising systems,[41] that is

$$G^{(2)}(t-t') = G^{(1)}(t-t') \tag{4.12}$$

one obtains the following relationships,

$$G^{(0)}(E)\left[E + A\right] = \frac{\sigma}{\pi} + 2AG^{(1)}(E) \tag{4.13}$$

$$G^{(1)}(E)\left[E - A\right] = \frac{1}{2\pi}(3m + \sigma - 2) \tag{4.14}$$

where $G^{(n)}(E)$ is the Fourier transform of $G^{(n)}(t-t')$, that is

$$G^{(n)}(t-t') = \int_{-\infty}^{\infty} G^{(n)}(E)\, e^{-iE(t-t')}\, dE \tag{4.15}$$

and

$$\sigma = \langle S^z \rangle \tag{4.16}$$

$$m = \langle \left[S^z\right]^2 \rangle \tag{4.17}$$

Now Equations (4.13) and (4.14) are readily solved, and the use of Equation (4.9) along with the unity spin kinematic relations leads to the following[40]

$$\sigma = 0 \tag{4.18}$$

$$m = \frac{2}{2 + e^{-\beta A}} \tag{4.19}$$

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[40] G.B. Taggart and R.A. Tahir-Kheli, Physica 44, 321 (1969).

[41] R.A. Tahir-Kheli, Phys. Rev. 169, 517 (1968).

Using these two simple results for the purely uniaxial case in Equations (4.13) and (4.14), we obtain for $G^{(0)}(E)$, the Fourier transform of the Green function equivalent to the transverse correlation of interest

$$G^{(o)}(E) = \frac{A}{\pi}\left(\frac{6}{2+e^{-\beta A}} - 2\right)\frac{1}{(E+A)(E-A)} \tag{4.20}$$

In the form of partial fractions, this becomes,

$$G^{(o)}(E) = \frac{1}{\pi}\left(\frac{3}{2+e^{-\beta A}} - 1\right)\left(-\frac{1}{E+A} + \frac{1}{E-A}\right) \tag{4.21}$$

For the transverse spin operators, Equation (4.10a) can be written as

$$\begin{aligned} G^{(o)}(E) &= \langle\langle S^{+};S^{-}\rangle\rangle_{(E)} \\ &= \frac{1}{2\pi}\int_{-\infty}^{\infty}\langle\langle S^{+}(t);S^{-}(t')\rangle\rangle\, e^{iE(t-t')}\, d(t-t') \end{aligned} \tag{4.22}$$

The specific form of the statistical spin correlation is then given by Equation (4.10), in the following form

$$\langle S^{+}(t')S^{-}(t)\rangle = \lim_{\epsilon\to 0}\, i\int_{-\infty}^{\infty}\left\{\frac{\langle\langle S^{+};S^{-}\rangle\rangle_{(E+i\epsilon)} - \langle\langle S^{+};S^{-}\rangle\rangle_{(E-i\epsilon)}}{e^{\beta E}-1}\right\} e^{-iE(t-t')}\, dE \tag{4.23}$$

where, by Equation (4.21)

$$\begin{aligned} \langle\langle S^{+};S^{-}\rangle\rangle_{(E+i\epsilon)} &= \frac{1}{\pi}\left(\frac{3}{2+e^{-\beta A}} - 1\right)\left(-\frac{1}{E+i\epsilon+A} + \frac{1}{E+i\epsilon-A}\right) \\ \langle\langle S^{+};S^{-}\rangle\rangle_{(E-i\epsilon)} &= \frac{1}{\pi}\left(\frac{3}{2+e^{-\beta A}} - 1\right)\left(-\frac{1}{E-i\epsilon+A} + \frac{1}{E-i\epsilon-A}\right) \end{aligned} \tag{4.24}$$

Applying the delta function

$$\delta(x) = \frac{1}{2\pi i}\left(\frac{1}{x-i\epsilon} - \frac{1}{x+i\epsilon}\right) \tag{4.25}$$

in the limit of $\epsilon \to 0$ to the integral of Equation (4.23), we find that the time-dependent transverse correlation is finally

$$\left\langle S^{+}(t)S^{-}(0)\right\rangle = 2\left(\frac{3}{2+e^{-\beta A}} - 1\right)\left(-\frac{e^{iAt}}{e^{-\beta A}-1} + \frac{e^{-iAt}}{e^{\beta A}-1}\right) \tag{4.26}$$

where the times t and t' have been changed to 0 and t, respectively. In the limit of infinite temperature, this equation becomes simply

$$\left\langle S^{+}(t)S^{-}(0)\right\rangle = \frac{4}{3}\cos(At) \tag{4.27}$$

and the transverse correlation, according to Equation (3.5) is

$$\left\langle S^{x}(t)\,S^{x}(0)\right\rangle = \frac{2}{3}\cos(At) \tag{4.28}$$

Note that the amplitude of 2/3 is the spin parameter, a, for unity spin.

This exact harmonic result can then be compared to the transverse correlation based on the approximate two-parameter Gaussian diffusivity with the derived frequency moments. To do this, the frequency moments of Equations (4.5) and (4.6) must be renormalized with powers of A instead of I_0. Then the transverse frequency moments, with the exchange terms vanishing, become

$$\frac{\left\langle \omega^{2}\right\rangle^{xx}}{16\beta a^{2}A^{2}} = \frac{3}{20}\left(1 - \frac{1}{4a}\right) \tag{4.29}$$

and

$$\frac{\left\langle \omega^{4}\right\rangle^{xx}}{128\beta a^{3}A^{4}} = \frac{27}{280} - \frac{39}{560a} + \frac{51}{4480a^{2}} \tag{4.30}$$

Note that these moments are independent of the wave-vector so that only the self-correlation is non-zero.

With the same procedure, the longitudinal moments go to zero.

In Figure 13, the time-dependent transverse correlations for the purely uniaxial Hamiltonian are presented. The exact result of Equation (4.28) is followed reasonably well by the approximate diffusivity-frequency moment result for short times. However, the long-time behavior of the approximate correlation only qualitatively reproduces the exact result.

CORRELATIONS IN TWO DIMENSIONS

In a manner similar to the one dimensional case in the previous chapter, the frequency moments may be particularized for a two-dimensional, simple quadratic net lattice structure. We still maintain the general conditions of axial and exchange anisotropy, and next nearest neighbor interactions. With redefinitions in two-dimensional wave vectors,

$$u_2 = \cos k_X + \cos k_Y \; ; \; v_2 = \cos k_X \cos k_Y \tag{5.1}$$

the longitudinal frequency moments, by Equations (3.10) and (3.11) and the two-dimensional results of Appendix B, become

$$\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{zz}}{16\beta a^2 I_0^2(1)} = (2 - u_2)\, G^2 + 2(1 - v_2)\, L^2 \tag{5.2}$$

and

$$\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{zz}}{128\beta a^3 I_0^4(1)} = \frac{6}{5} D \left(1 - \frac{1}{4a} \right) \left[(u_2 - 2)\, G^2 + 2(v_2 - 1)\, L^2 H \right] + \frac{3}{5} D^2 \left(1 - \frac{1}{4a} \right) \left[(2 - u_2)\, G^2 + 2\,(1 - v_2)\, L^2 \right]$$

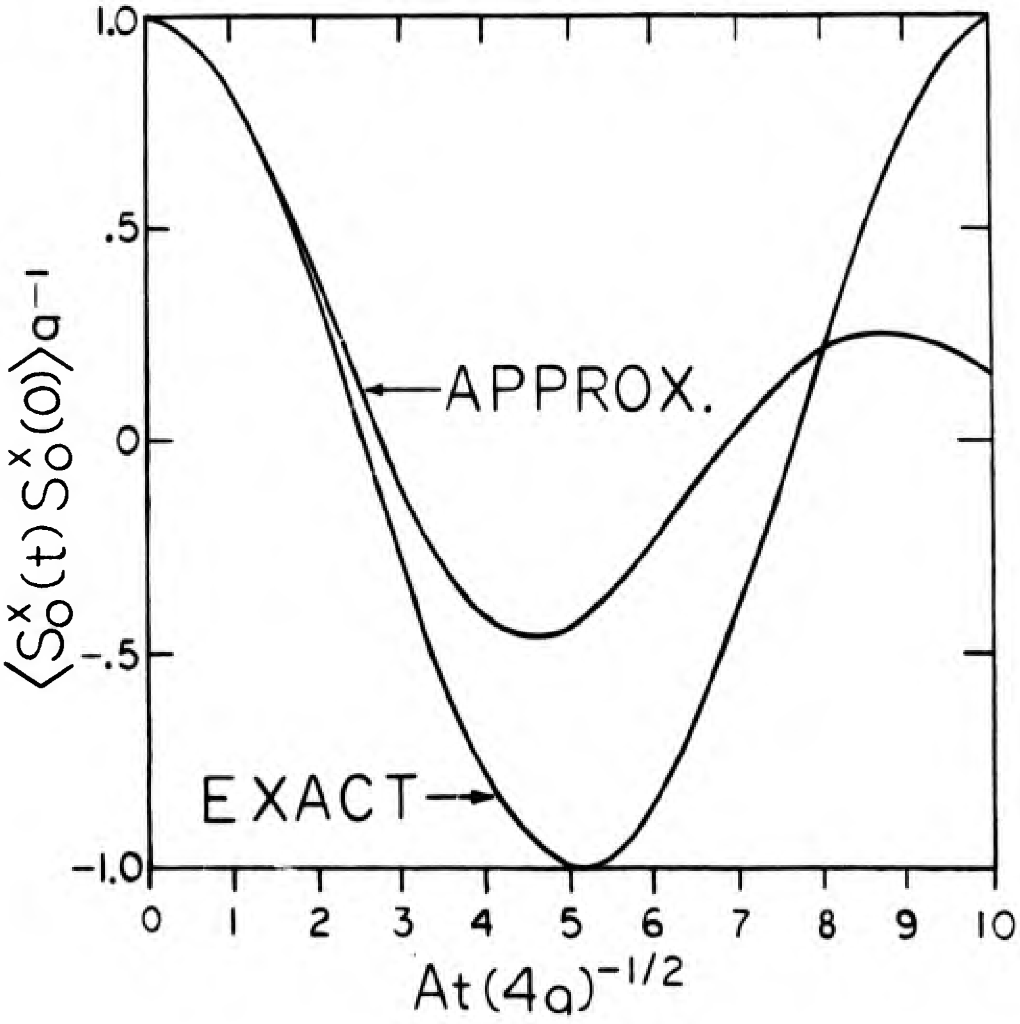

Figure 13. A Comparison of the Unity Spin, Time-Dependent Transverse Correlations of the Exact Harmonic Solution of the Pure Uniaxial Hamiltonian and of the Approximate Two-Parameter Gaussian Diffusivity Based on the Zero-Exchange Frequency Moments Retaining Only the Uniaxial Terms.

$$
\begin{aligned}
&+ 4G^2 L(1-H)(2v_2-u_2) + 4\left[(u_2-2)G^2 H + (v_2-1)L^2\right] \\
&+ \frac{2}{5}\left[(u_2-2)G^2 + 2(v_2-1)L^2H^2\right] - \frac{8}{5}\left[(2-u_2)G^4 + 2(1-v_2)L^4\right] \\
&+ 4\left[(2-u_2)G^2 + 2(1-v_2)L^2\right](1+H^2) + 20(G^2+L^2)^2 \\
&+ 3(u_2G^2 + 2v_2L^2)^2 - 16(u_2G^2 + 2v_2L^2)(G^2+L^2) \\
&+ \frac{1}{10a}\left[(u_2-2)\left(G^4 + \frac{3}{2}G^2\right) + 2(v_2-1)\left(L^4 + \frac{3}{2}L^2H^2\right)\right] \qquad (5.3)
\end{aligned}
$$

The transverse moments are

$$
\begin{aligned}
\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx}}{16\beta a^2 I_0^2(1)} &= \frac{3}{20} D^2\left(1 - \frac{1}{4a}\right) - (u_2G + 2v_2LH) \\
&+ 1 + G^2 + H^2 + L^2 \qquad (5.4)
\end{aligned}
$$

and

$$
\begin{aligned}
\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx}}{128\beta a^3 I_0^4(1)} &= \frac{3}{5}D\left(1 - \frac{1}{4a}\right)\Big\{u_2G^3 + 2v_2L^3 - 2(u_2G + 2v_2H^2L) \\
&+ 2(G^2 + L^2H)\Big\} \\
&+ \frac{6}{5}D^2\left(1 - \frac{1}{4a}\right)\left\{-(u_2G + 2v_2LH) + 2(G^2+L^2) + \frac{3}{2}(1+H^2)\right\} \\
&+ 2D^4\left\{\frac{27}{560} - \frac{39}{1120a} + \frac{51}{8960a^2}\right\} \\
&+ \frac{1}{10}\left(6 + \frac{1}{a}\right)\left(-1 - H^4 + 2u_2G^3 + 4v_2L^3H\right) \\
&+ \frac{1}{20}\left(16 + \frac{1}{a}\right)\left(u_2G + 2v_2LH^3 - 3G^2 + 3L^2H^2\right)
\end{aligned}
$$

$$\begin{aligned}
&+ 12(G^2 + L^2)(1 + H^2) - 12(u_2 G + 2v_2 LH)(G^2 + L^2) \\
&- \frac{1}{4}\left(4 + \frac{1}{a}\right)\left(G^4 + L^4\right) + 10\left(G^2 + L^2\right)^2 + 6(1 + H^2)^2 \\
&- 8(u_2 G + 2v_2 LH)(1 + H^2) + 3(u_2 G + 2v_2 LH)^2 \\
&+ GL[u_2(-2G^2 + 4 + 2H + 2L) + v_2 G(4 - 2L) - 4GH - 8G] \\
&+ GH[u_2(2G^2 - 4) + v_2 G(4 + 2H)] - 4v_2 LH \qquad (5.5)
\end{aligned}$$

The correlations based on these moments are now calculated under many conditions of the dimensionless parameters. For nearest neighbor exchange, some of these results are shown in Figures 14 through 17. Many of the features observed for the one-dimensional case carry over into the quadratic lattice. The behavior of the Fourier transforms and time-dependent forms of the longitudinal self-correlations in Figures 14 and 15 (compare to Figures 4 and 12) possess many of the features of the one-dimensional case. The Fourier transform, for example: (1) still diverges for the zero frequency limit, (2) shows a similarity of behavior for small G and large D in its narrowing toward the origin, and (3) exhibits a decreasing divergence at zero frequency and a broadening of its spectrum to higher frequencies, when the exchange anisotropy is increased, with no possibility of approach to this result via the axial anisotropy. However, two slight differences in the transforms in two-dimensions, as compared to the linear chain, are that the zero-frequency divergence is less severe and that the results for unity spin and infinite spin are even closer in two dimensions than in one dimension. Hence these results agree with the previous statements concerning the divergence and with the aforementioned "law of corresponding states". The unity spin results are again not shown because of this reason.

The time-dependent, longitudinal self-correlation in Figure 15 again exhibits: (1) a similar behavior for large D and small G in decreasing the decay rate from the isotropic case, (2) a large increase in the decay rate for large G, a result which cannot be simulated by variations in D. However, the effect of the higher dimensionality is to make the longitudinal time correlations less sensitive to increases in D, relative to corresponding changes in G. More specifically, the G=1, D=6, curve in two dimensions is much closer to the G=1, D=0 (isotropic) curve and farther from the G=.5, D=0 curve than the corresponding one-dimensional results.

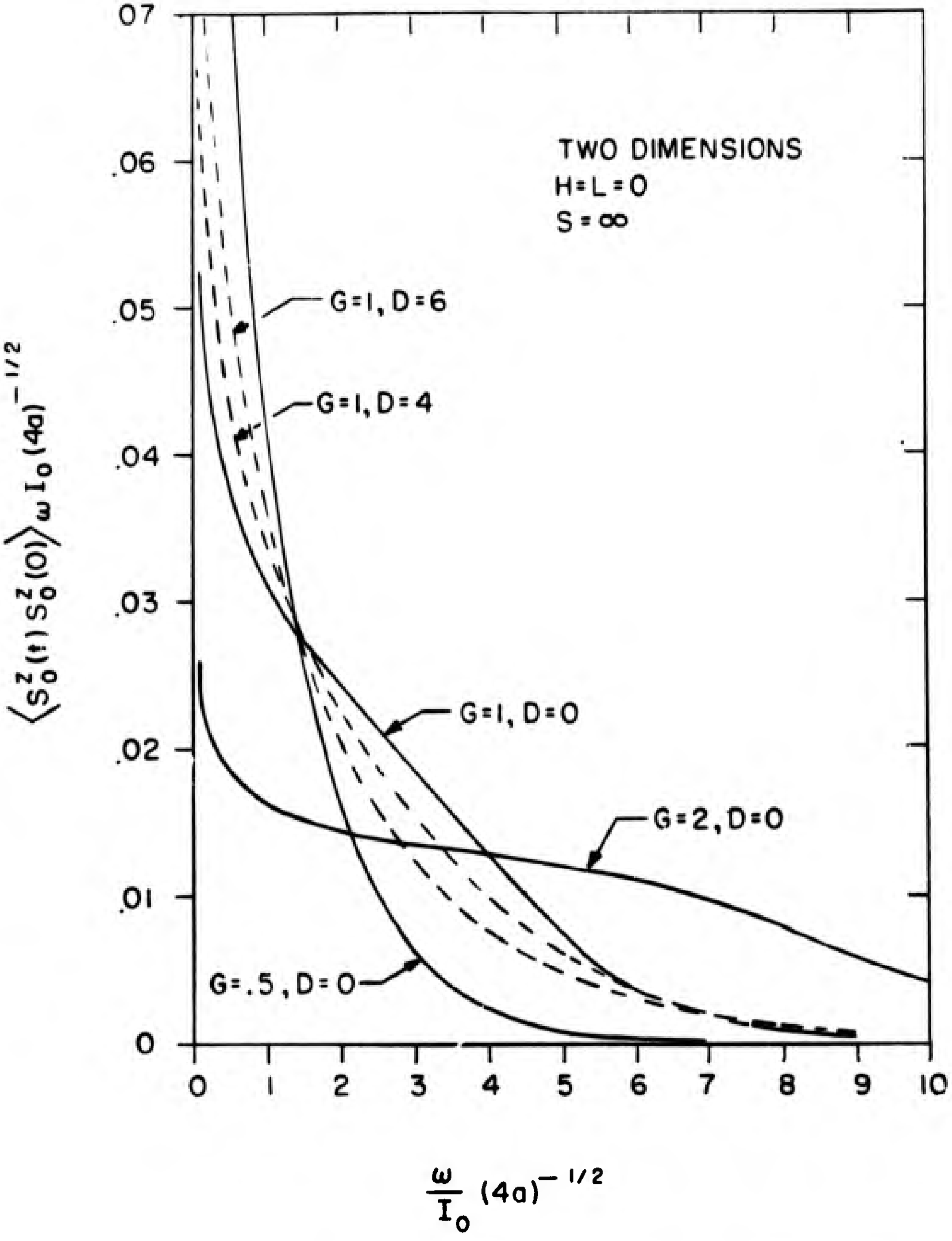

Figure 14. The Frequency Fourier Transforms of the Longitudinal Self-Correlations versus Reduced Frequency, Exchange Anisotropy G With D=0, and Axial Anisotropy D With G=1, for a Simple Two-Dimensional Net of Infinite Spins, With Only Nearest Neighbor Interactions (H=L=0).

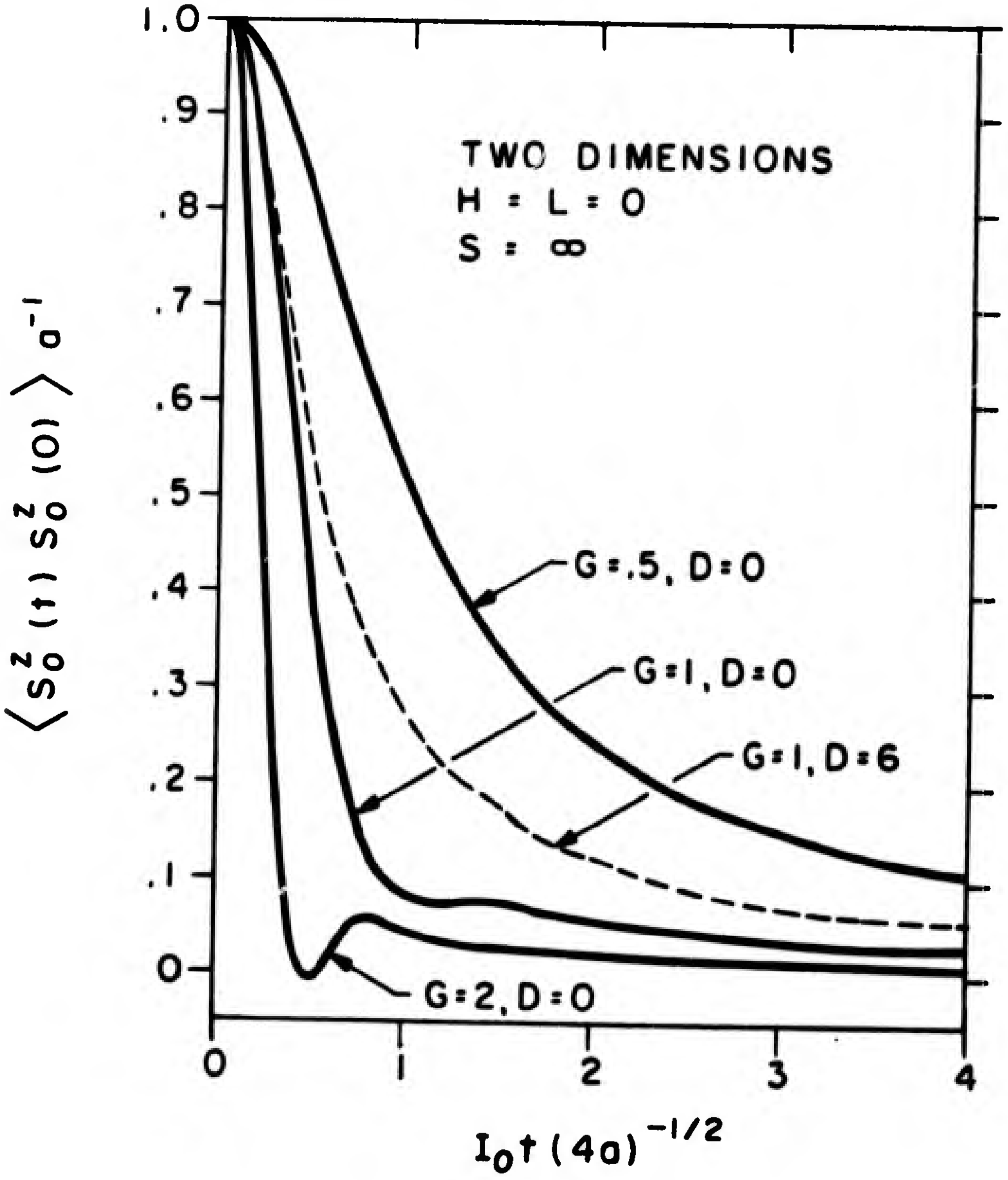

Figure 15. The Variation, With G and D, of the Temporal, Longitudinal Self-Correlations versus Reduced Time, for the Simple Two-Dimensional Net With Nearest Neighbor Exchange and Infinite Spin.

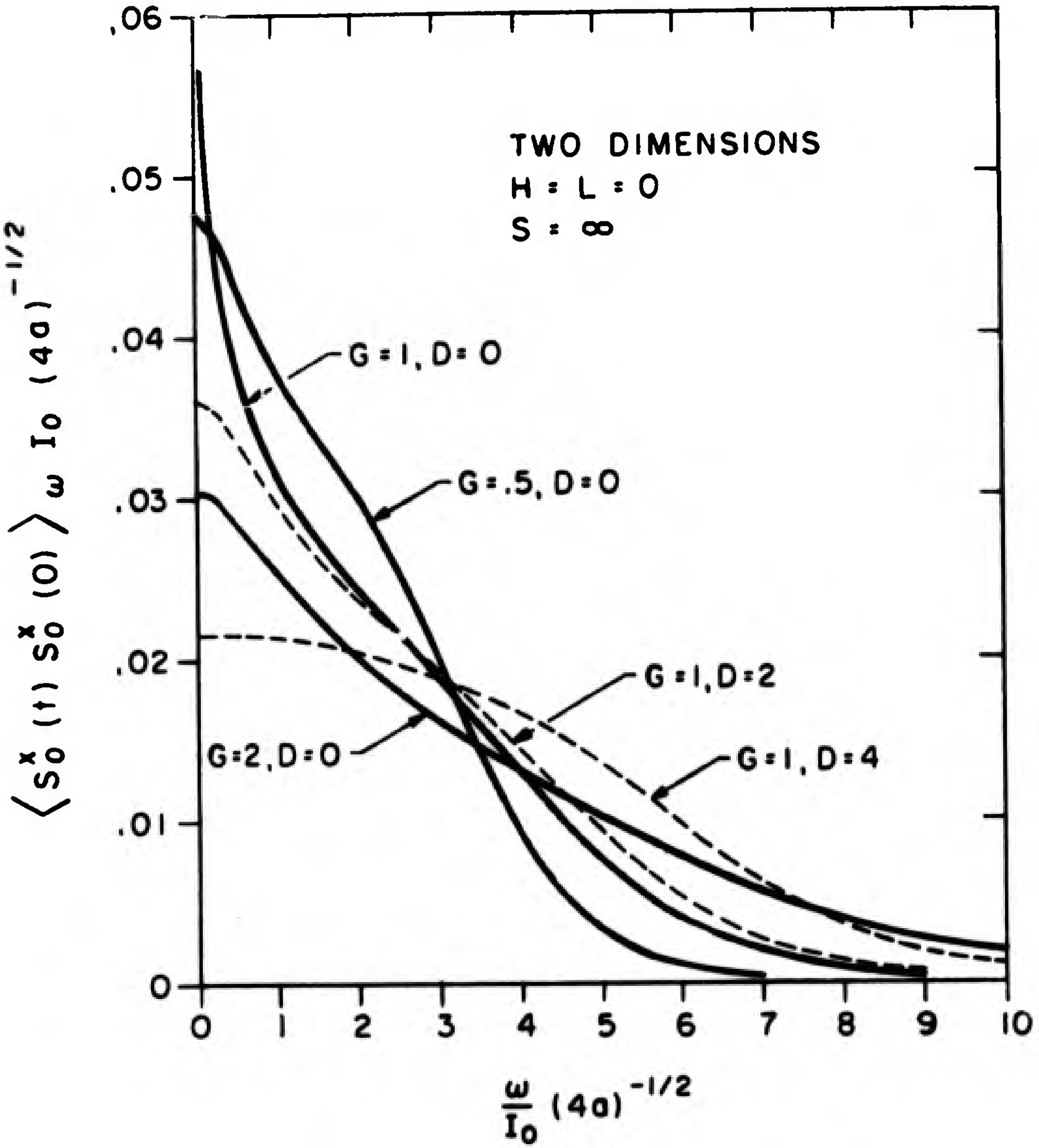

Figure 16. The Variation, With G and D Considered Separately, of the Frequency Fourier Transform of the Transverse, Self-Correlation, for a Simple Two-Dimensional Net of Infinite Spins With Nearest Neighbor Interactions.

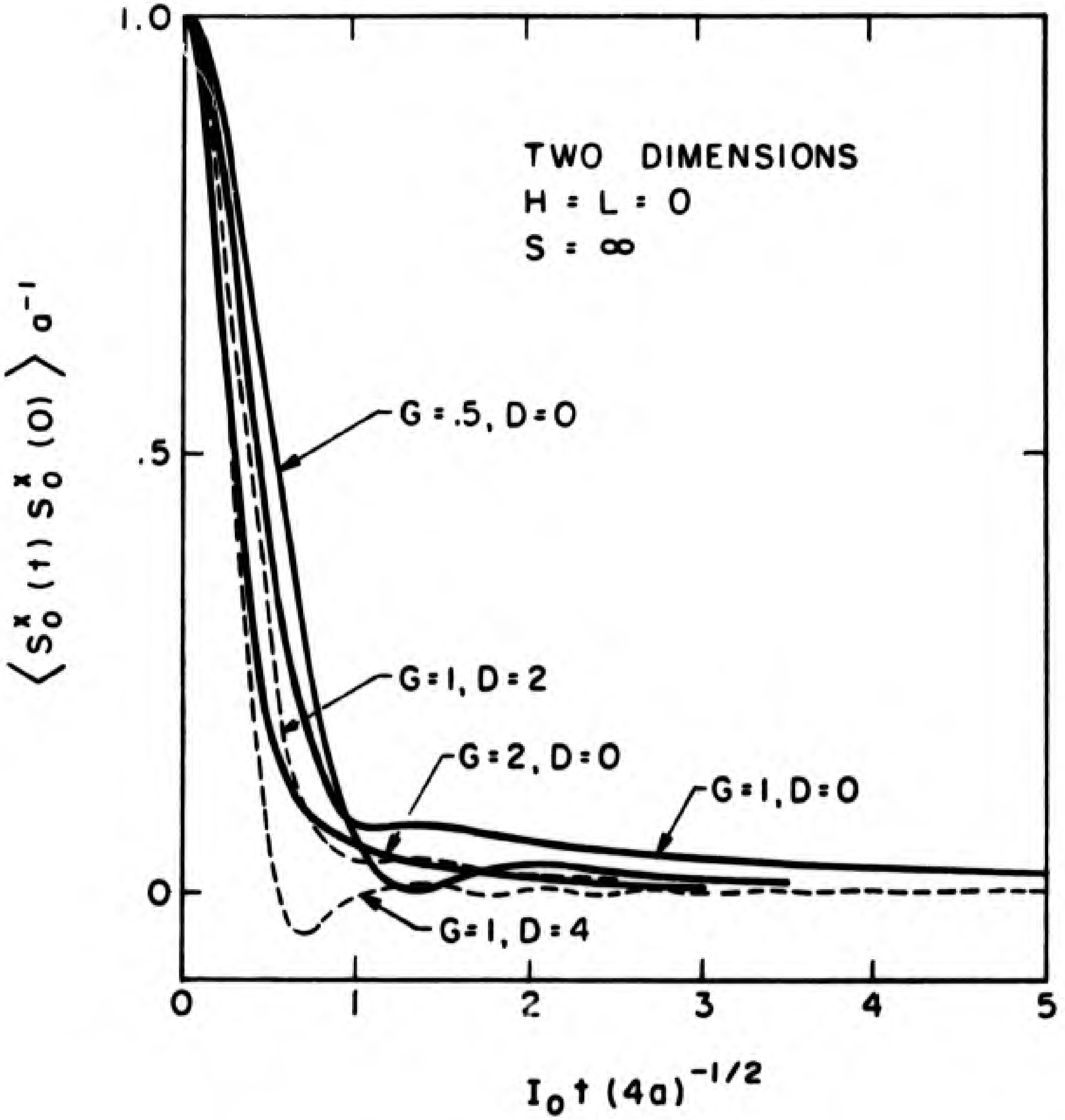

Figure 17. The Variation, With G and D, of the Temporal, Transverse Self-Correlations versus Reduced Time, for the Two-Dimensional, Infinite Spin, Simple Net With Nearest Neighbor Interactions.

The behavior of the frequency Fourier transform of the transverse correlation in two dimensions is shown in Figure 16, and also exhibits features analogous to its one-dimensional counterpart. (See Figure 9.) The transforms at zero frequency are smaller than in the linear chain, and the cutoff frequencies are slightly larger. The transforms for large exchange anisotropy can be simulated with large axial anisotropy.

The temporal transverse correlations are exhibited in Figure 17, where it is seen, in a comparison with the linear chain (see Figure 11), that the time decays are generally more rapid for the same conditions on the parameters, and that the large axially anisotropic cases tend toward similarity with the results for large exchange anisotropy. One interesting feature, however, is that the time decay of the G=1, D=4 curve is more rapid than the G=2, D=0 curve, contrasting with the corresponding results of the linear chain.

CORRELATIONS IN THREE DIMENSIONS

We now particularize the general form of the frequency moments to a simple cubic lattice structure. With the aid of Appendix B and the following redefinitions in terms of three-dimensional wave-vectors

$$u_3 = \cos k_x + \cos k_y + \cos k_z$$

$$v_3 = \cos k_x \cos k_y + \cos k_x \cos k_z + \cos k_y \cos k_z \tag{6.1}$$

the longitudinal frequency moments become

$$\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{zz}}{16\beta a^2 I_0^2(1)} = (3 - u_3)G^2 + 2(3 - v_3)L^2 \tag{6.2}$$

and

$$\begin{aligned}\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{zz}}{128\beta a^3 I_0^4(1)} = {} & \frac{6}{5} D \left(1 - \frac{1}{4a}\right)\left[(u_3 - 3)G^2 + 2(v_3 - 3)L^2 H\right] \\ & + \frac{3}{5} D^2 \left(1 - \frac{1}{4a}\right)\left[(3 - u_3) G^2 + 2(3 - v_3) L^2\right] \\ & + 8(u_3 - v_3)G^2 L(1 - H) + 8(u_3 - 3)G^2 H + 4(v_3 - 3)L^2 (1 + 2H^2) \\ & + \frac{2}{5}\left[(u_3 - 3)G^2 + 2(v_3 - 3)L^2 H\right] + 6\left[(3 - u_3) G^2 + 2(3 - v_3)L^2\right] \\ & \times (1 + 2H^2) - \frac{8}{5}\left[(3 - u_3)G^2 + 2(v_3 - 3)L^4\right] + 45(G^2 + 2L^2) \\ & + 3(u_3 G^2 + 2v_3 L^2)^2 - 24(u_3 G^2 + 2v_3 L^2)(G^2 + 2L^2) \\ & + \frac{1}{10a}\left[(u_3 - 3)\left(G^4 + \frac{3}{2} G^2\right) + 2(v_3 - 3)\left(L^4 + \frac{3}{2} L^2 H^2\right)\right] \end{aligned} \tag{6.3}$$

The transverse moments are

$$\begin{aligned}\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{xx}}{16\beta a^2 I_0^2(1)} = {} & \frac{3}{20} D^2 \left(1 - \frac{1}{4a}\right) - (u_3 G + 2v_3 LH) \\ & + \frac{3}{2}(1 + 2H^2 + G^2 + 2L^2) \end{aligned} \tag{6.4}$$

and

$$\begin{aligned}\frac{\left\langle \omega^4 \right\rangle_{\vec{K}}^{xx}}{128\beta a^3 I_0^4(1)} = {} & \frac{3}{5} D \left(1 - \frac{1}{4a}\right)\left[u_3 G^3 + 2v_3 L^2 - 2(u_3 G + 2v_3 H^2 L) + 3(G^2 + 2L^2 H)\right] \\ & + \frac{6}{5} D^2 \left(1 - \frac{1}{4a}\right)\left[- u_3 G - 2v_3 LH + 3(G^2 + 2L^2) + \frac{9}{4}(1 + 2H^2)\right] \end{aligned}$$

$$
\begin{aligned}
&+ 2D^4\left[\frac{27}{560} - \frac{39}{1120a} + \frac{51}{8960a^2}\right] \\
&+ \frac{3}{20}\left(6+\frac{1}{a}\right)\left(\frac{4}{3}u_3G^3 + \frac{8}{3}v_3L^3H - 2H^2 - 1\right) \\
&+ \frac{1}{20}\left(16+\frac{1}{a}\right)\left(u_3G + 2v_3LH^3 - \frac{9}{2}G^2 - 9L^2H^2\right) \\
&- \frac{3}{8}\left(4+\frac{1}{a}\right)\left(G^4 + 2L^4\right) + \frac{27}{2}\left(1+2H^2\right)^2 + \frac{45}{2}\left(G^2+2L^2\right)^2 \\
&- 12\left(u_3G + 2v_3LH\right)\left(1+2H^2\right) + 3\left(u_3G+2v_3LH\right)^2 \\
&+ 27\left(G^2+2L^2\right)\left(1+2H^2\right) - 18\left(u_3G+2v_3LH\right)\left(G^2+2L^2\right) \\
&+ GL\left[4u_3\left(2-G^2+L+H\right) + 2v_3G\left(2-L\right) - 12G\left(H+2\right)\right] \qquad (6.5)
\end{aligned}
$$

For the case of nearest neighbor exchange only (H=L=0), the correlations are presented in Figures 18 through 21, where the results follow the general pattern established in one and two dimensions.

The Fourier transform of the longitudinal self-correlation is shown in Figure 18, where it can be seen that the zero-frequency values are now finite for G≠0 or D≠0. (The G=.5, D=0, curve does actually intersect the ordinate axis at value of approximately .122, which is off the scale.) Therefore the higher dimensionality has flattened the curves and extended them toward higher frequencies. The similarity of the behavior of the transforms for decreased G and for increased D is again present. The curve for G=2, D=0 is now quite flat, varying slowly over the indicated frequency range, and showing the approach to the XY model for large G. In this limit, the transform would approach very small values, maintained constant but extending over increased frequencies.

In Figure 19, the temporal, longitudinal correlations are exhibited. The time decays are somewhat more rapid in three dimensions than in two, but the similarity of results obtained by decreasing the exchange anisotropy and by increasing the axial anisotropy can again be observed.

In Figure 20, we note that the Fourier transforms of the transverse, self-correlations are also compressed toward the frequency axis and extend to higher frequencies. The simulation of large G behavior with large D behavior is again noted; e.g., the G=2, D=0, curve is fairly close to the G=1, D=4 curve. This same similarity can be observed in Figure 21, where the time-transverse

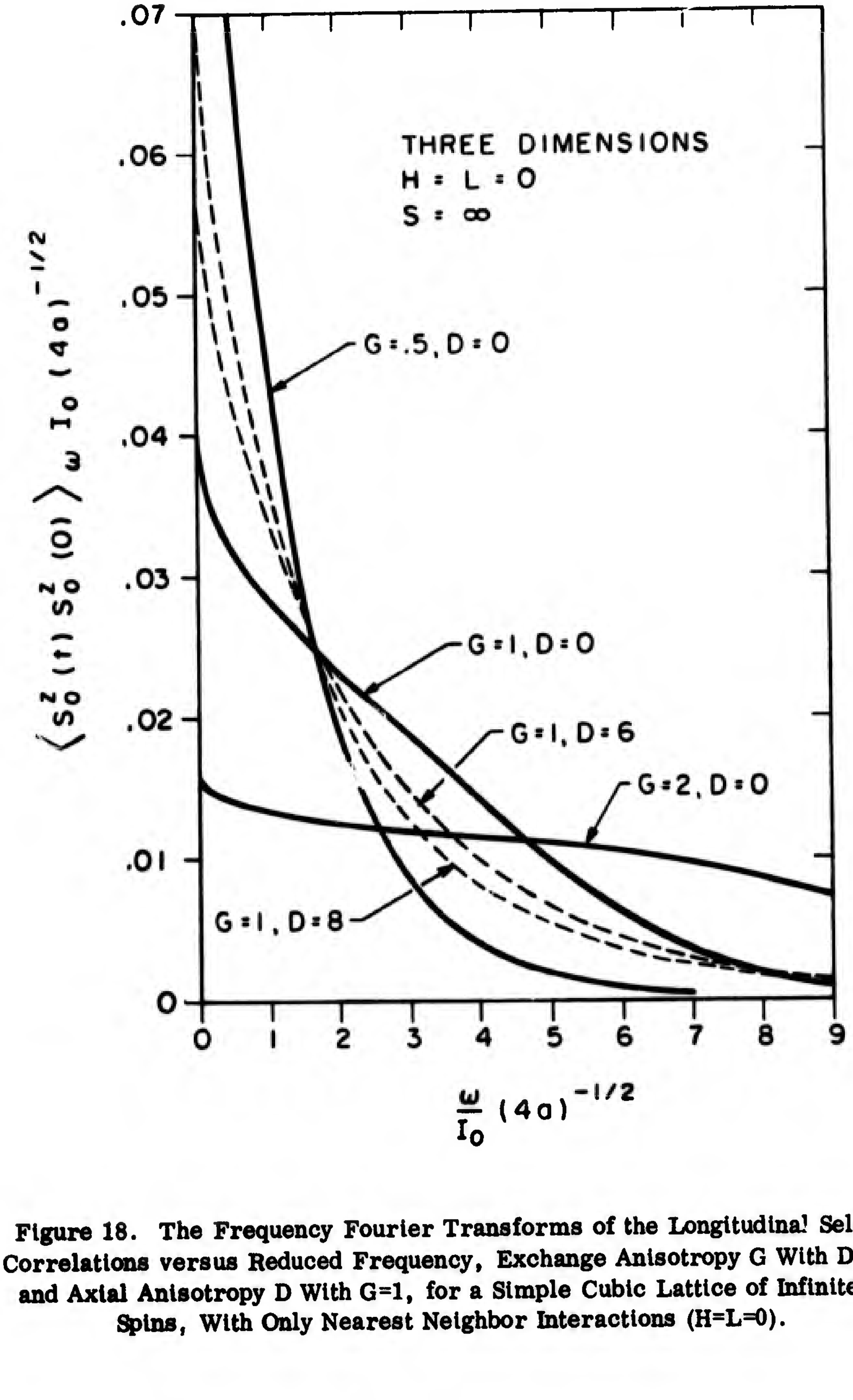

Figure 18. The Frequency Fourier Transforms of the Longitudinal Self-Correlations versus Reduced Frequency, Exchange Anisotropy G With D=0, and Axial Anisotropy D With G=1, for a Simple Cubic Lattice of Infinite Spins, With Only Nearest Neighbor Interactions (H=L=0).

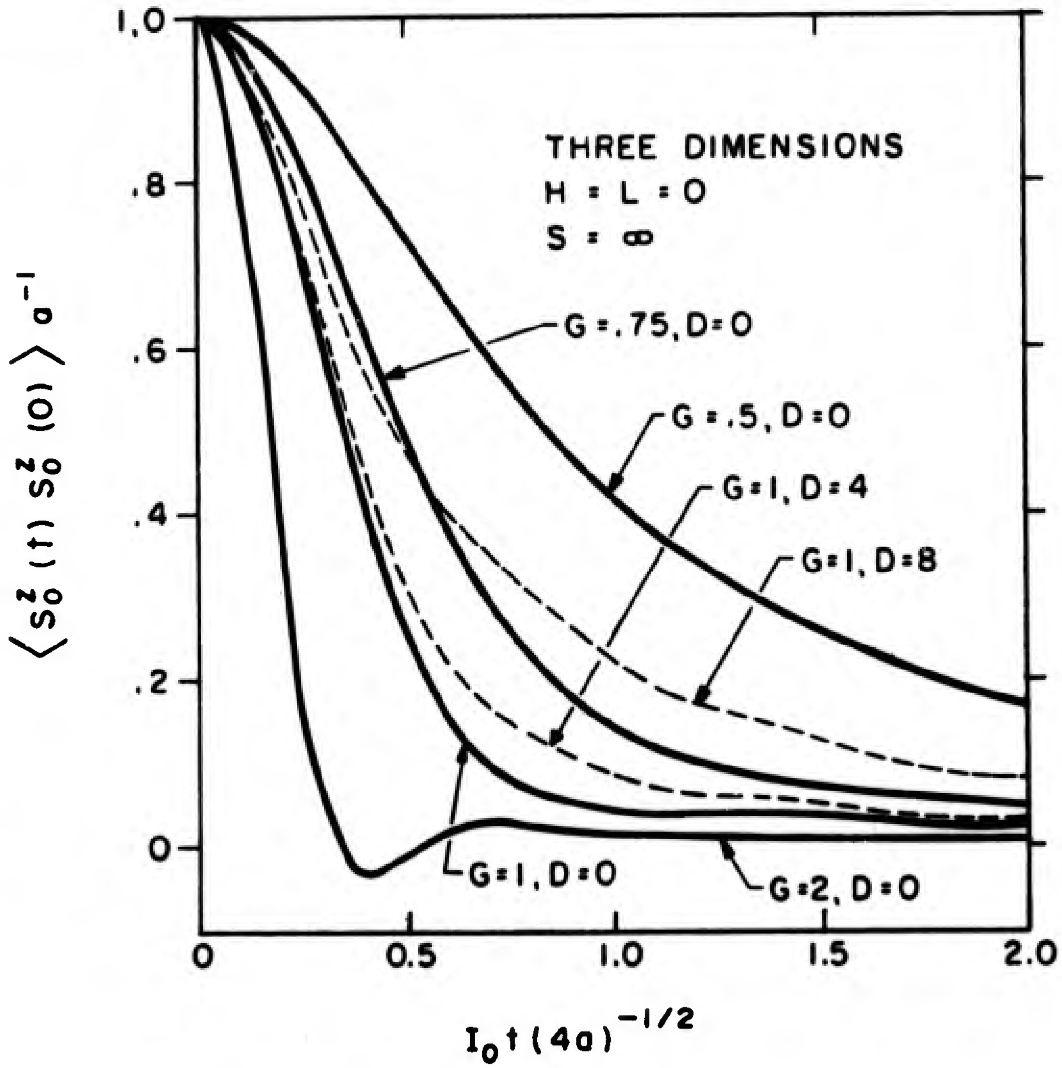

Figure 19. The Variation, With G and D, of the Time Dependent, Longitudinal Self-Correlations are Considered for the Three-Dimensional Infinite Spin, Simple Cubic Lattice With Nearest Neighbor Interactions.

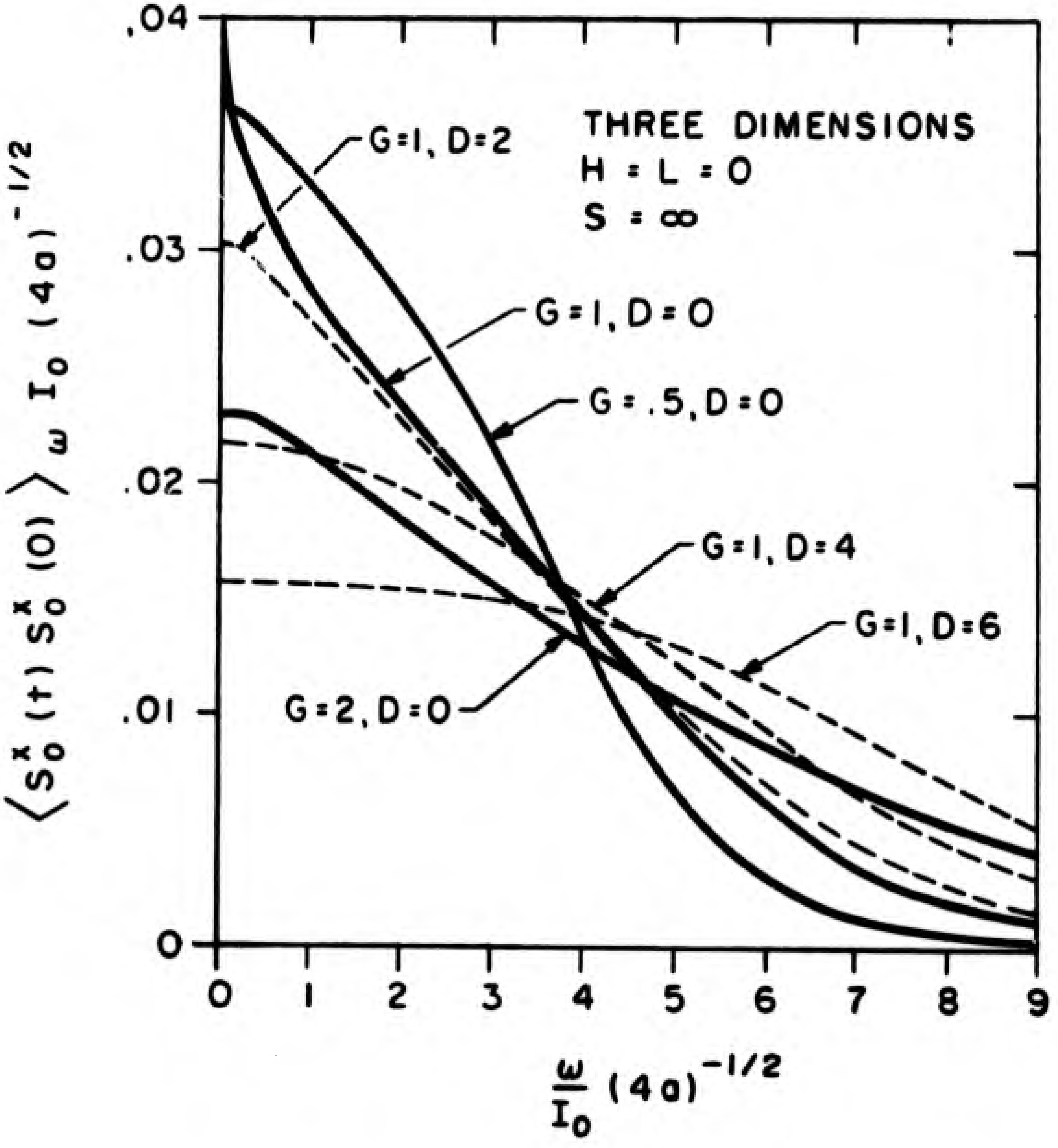

Figure 20. The Variation, With G and D Considered Separately, of the Frequency Fourier Transform of the Transverse, Self-Correlation, for a Simple Cubic Lattice of Infinite Spins With Nearest Neighbor Interactions.

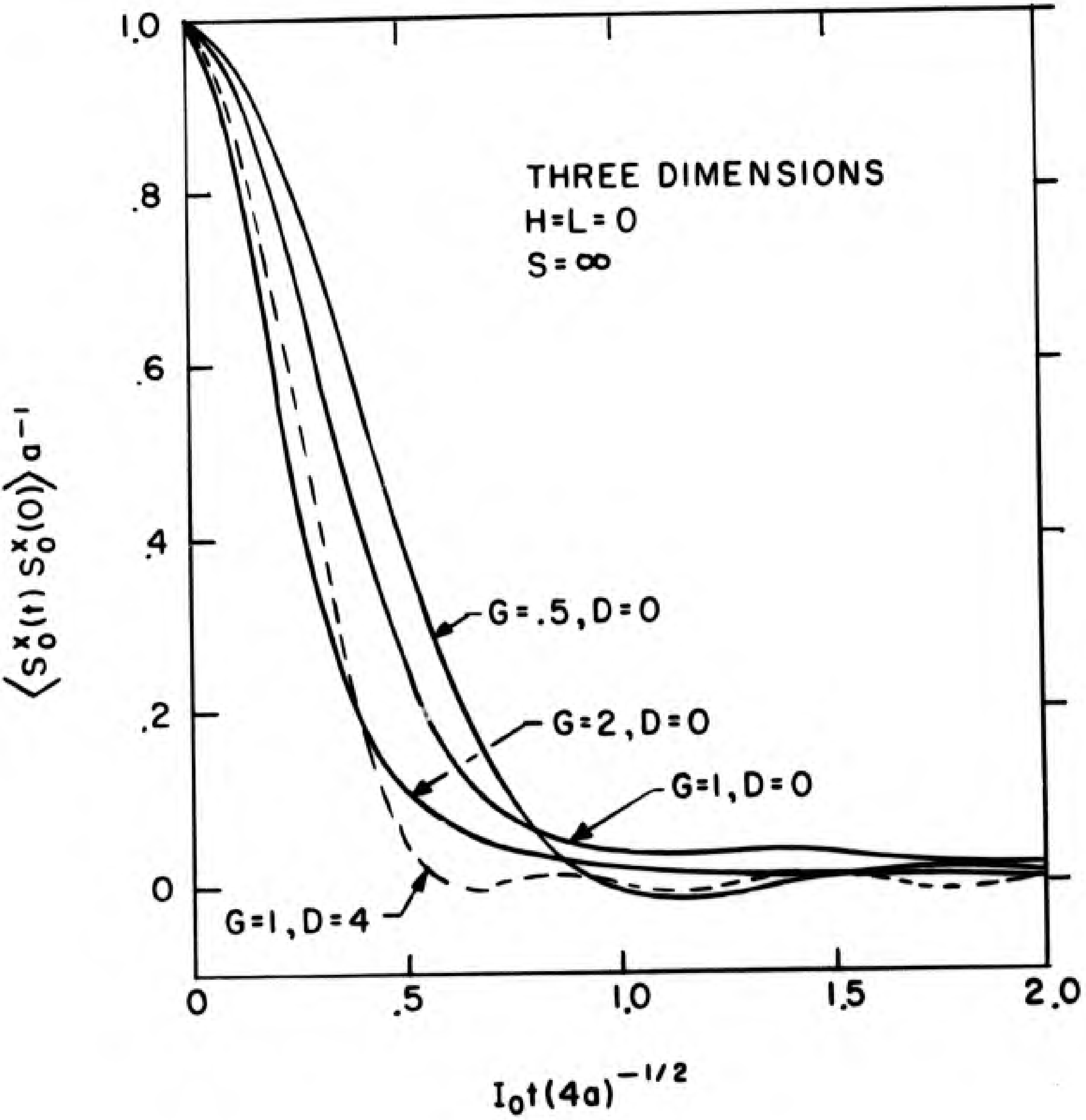

Figure 21. The Variation, With G and D, of the Temporal, Transverse Self-Correlations versus Reduced Time, for the Infinite Spin, Simple Cubic Lattice With Nearest Neighbor Interactions.

correlations are presented and show approximately the same increase in the decay rate for the two curves alluded to in the previous statement. These results show that the decay rates for corresponding cases, in three dimensions are somewhat larger than in two dimensions. (Compare with Figure 17 and note the scale change.)

Finally it should be mentioned that the infinite spin correlations for vanishing axial and exchange anisotropy in the frequency moment equations have been studied in Reference 7 and yield excellent agreement with the classical limit of Windsor,[11,14] as applied to a simple cubic structure.

CONCLUDING REMARKS

In these calculations in one, two, and three dimensions, two approximations frequently encountered in similar studies in the literature have not been used. The assumption of a spherical continuum model was not applied in any calculations. Instead, the precise contributions from the exact lattice sites were computed. The random phase approximation, which simplifies the long calculations of the thermal averages by factorization or decoupling procedures was also avoided. The thermal averages of the spin operator products were, instead, calculated precisely for their given length. Therefore, a fair assessment of this study is that it involves a relatively exact calculation of an approximate phenomenological representation of space-time-dependent correlations for a very general Heisenberg spin system at high temperatures.

In this study we have evaluated the longitudinal and transverse spin correlations of Heisenberg spin systems with both uniaxial and exchange anisotropy in the limit of elevated temperature by the method of frequency moments, utilizing the zeroth, second, and fourth moments in the frequency wave-vector spectral function, based on the hydrodynamical two-parameter Gaussian diffusivity. The moment equations are valid for all spins for arbitrary range of the exchange interactions and for arbitrary dimensionality and lattice structure. These moments are judged to be likely correct, even though a discrepancy was noted in the literature, because of the agreements with certain necessary conditions and approaches to various limiting cases. These moments were particularized to one, two, and three dimensions, including the next nearest neighbor

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11. C.G. Windsor, Proc. Phys. Soc. 87, 501 (1966); 89, 825 (1966); 91, 353 (1967).
14. C.G. Windsor, in Inelastic Scattering of Neutrons, 2, 83 (I.A.E.A. 1968).

interactions; and the resulting time-dependent correlations and their Fourier transforms were calculated, exhibiting features predicted by theoretical considerations, and pointing to several conclusions:

(1) The effect of increasing the axial anisotropy upon the longitudinal correlations is similar to that of decreasing the exchange anisotropy.

(2) In contrast, the behavior of the transverse correlations is similar for increasing axial anisotropy or for increasing exchange anisotropy.

(3) The anisotropic effects on the correlations are decreased as the strength of the exchange interaction is increased or as its range is extended to the next nearest neighbors.

The fact that the present approximate phenomenological theory appears to give a satisfactory representation of some limiting cases of isotropic Heisenberg systems gives hope that the present results are equally meaningful for more general cases for which direct solutions are not available, since they cannot be carried out, in any practical sense.

APPENDIX A

TRACES OF SPIN OPERATOR PRODUCTS

The traces of the spin operator products tabulated by Ambler et al.[37] are reformulated, as explained in the section "General Form of the Frequency Moments."

All factors of (2S+1) are omitted, since they cancel with the Tr(1) term in the denominators of the thermal expansion. With the previously used definition of

$$a = \frac{1}{3} S(S+1)$$

some of the traces repeatedly encountered in these calculations are listed:

$$\mathrm{Tr}\,(S^z)^2 = a$$

$$\mathrm{Tr}\,S^+ S^- = 2a$$

$$\mathrm{Tr}\,S^+ S^- S^z = a$$

$$\mathrm{Tr}\,S^- S^+ S^z = -a$$

$$\mathrm{Tr}\,S^+ S^- S^+ S^- = \frac{4}{5} a(6a+1)$$

$$\mathrm{Tr}\,S^+ S^- S^- S^+ = \frac{6}{5} a(4a-1)$$

$$\mathrm{Tr}\,S^+ S^- S^z S^z = \frac{a}{5}(6a+1) = \mathrm{Tr}\,S^- S^+ S^z S^z$$

$$\mathrm{Tr}\,(S^z)^4 = \frac{a}{5}(9a-1)$$

$$\mathrm{Tr}\,S^z S^+ S^- S^- S^+ = 0 = \mathrm{Tr}\,S^z S^- S^+ S^+ S^-$$

$$\mathrm{Tr}\,S^z S^+ S^- S^+ S^- = \frac{2}{5} a(6a+1)$$

$$\mathrm{Tr}\,(S^z)^3 S^- S^+ = -\frac{a}{5}(9a-1)$$

$$\mathrm{Tr}\,(S^z)^3 S^+ S^- = \frac{a}{5}(9a-1)$$

$$\mathrm{Tr}\,(S^z)^6 = \frac{a}{7}(27a^2 - 9a + 1)$$

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[37] E. Ambler, J.C. Eisenstein, and J.F. Schooley, J. Math. Phys. 3, 118 (1962).

$$\mathrm{Tr}\,(S^z)^4\, S^+ S^- = \frac{a}{35}\,(54a^2 + 24a - 5)$$

$$= \mathrm{Tr}\,(S^z)^4\, S^- S^+$$

$$\mathrm{Tr}\, S^+ S^- S^+ S^- (S^z)^2 = \frac{2}{35}\, a\, (36a^2 + 30a - 1)$$

$$\mathrm{Tr}\, S^- S^+ S^+ S^- (S^z)^2 = \frac{2}{35}\, a\, (36a^2 - 33a + 6)$$

APPENDIX B

FREQUENCY MOMENTS APPLIED TO LATTICE STRUCTURES

In applying the frequency moments in their general forms derived in "General Form of the Frequency Moments" to specific lattice structures, it is necessary to consider the range of exchange interaction to the desired n^{th} nearest neighbor. In this study, n=2, so that interactions extending to first and second nearest neighbors are considered to be dominant, in agreement with considerable experimental data.

In one dimension, there are two first nearest neighbors and two second nearest neighbors. In the two dimensional net, these numbers are four and four, respectively; and for the simple cubic case, they are six and twelve, respectively. The symmetries which exist reduce the distinct $\vec{K}$-vector terms to the u_i , v_i types given previously.

The following three summations will now be taken as examples, applying to the one, two, and three dimensional cases: $\sum_{\vec{R}} I_+^2(\vec{R})\, I_0^2(\vec{R})$; $\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0^2(\vec{U})$; $\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0(\vec{U})\, I_0(\vec{R}+\vec{U})$. We shall also illustrate these three expressions with the $e^{i\vec{K}\cdot\vec{R}}$ factor.

With the same definitions introduced in "Correlations in One Dimension" for (1) and (2) indicating the separations from the reference site to the first and second nearest neighbors, the above summations become, for a linear chain

$$\sum_{\vec{R}} I_+^2(\vec{R})\, I_0^2(\vec{R}) = 2\, I_+^2(1)\, I_0^2(1) + 2\, I_+^2(2)\, I_0^2(2)$$

$$\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0^2(\vec{U}) = \left[2I_+^2(1) + 2I_+^2(2)\right]\left[2I_0^2(1) + 2I_0^2(2)\right]$$

$$\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0(\vec{U})\, I_0(\vec{R}+\vec{U}) = 2 \sum_{\vec{R}} \left\{ I_+^2(\vec{R}) \left[I_0(1)\, I_0(\vec{R}+1) + I_0(2)\, I_0(\vec{R}+2) \right] \right\}$$

$$= 2I_+^2(1)\,I_0(1)\,I_0(2) + 2I_+^2(1)\,I_0(2)\,I_0(1) + 2I_+^2(2)\,I_0(1)\,I_0(1)$$

$$= 4I_+^2(1)\,I_0(1)\,I_0(2) + 2I_+^2(2)\,I_0^2(1)$$

The other sums with the wave-vector are, by the definitions of Equation (4.2)

$$\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{R})\, I_0^2(\vec{R}) = 2u_1\, I_+^2(1)\, I_0^2(1) + 2v_1\, I_+^2(2)\, I_0^2(2)$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{R})\, I_0^2(\vec{U}) = \left[2u_1\, I_+^2(1) + 2v_1\, I_+^2(2)\right]\left[2I_0^2(1) + 2I_0^2(2)\right]$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{R})\, I_0(\vec{U})\, I_0(\vec{R}+\vec{U}) = 2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I_+^2(\vec{R})\left[I_0(1)\, I_0(\vec{R}+1) + I_0(2)\, I_0(\vec{R}+2)\right]$$

$$= 2u_1\, I_+^2(1)\, I_0(1)\, I_0(2) + 2u_1\, I_+^2(1)\, I_0(2)\, I_0(1) + 2v_1\, I_+^2(2)\, I_0(1)\, I_0(1)$$

$$= 4u_1\, I_+^2(1)\, I_0(1)\, I_0(2) + 2v_1\, I_+^2(2)\, I_0^2(1)$$

For the two dimensional net, we have

$$\sum_{\vec{R}} I_+^2(\vec{R})\, I_0^2(\vec{R}) = 4I_+^2(1)\, I_0^2(1) + 4I_+^2(2)\, I_0^2(2)$$

$$\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0^2(\vec{U}) = \left[4I_+^2(1) + 4I_+^3(2)\right]\left[4I_0^2(1) + 4I_0^2(2)\right]$$

$$\sum_{\vec{R}\vec{U}} I_+^2(\vec{R})\, I_0(\vec{R})\, I_0(\vec{R}+\vec{U}) = 4\sum_{\vec{R}} \left\{ I_+^2(\vec{R})\left[I_0(1)\, I_0(\vec{R}+1) + I_0(2)\, I_0(\vec{R}+2)\right]\right\}$$

$$= 8I_+^2(1)\, I_0(1)\, I_0(2) + 8I_+^2(1)\, I_0(2)\, I_0(1) + 8I_+^2(2)\, I_0(1)\, I_0(1)$$

$$= 16I_+^2(1)\, I_0(1)\, I_0(2) + 8I_+^2(2)\, I_0^2(1)$$

By the definitions of Equation (5.1), the other sums, involving the wave-vectors, become

$$\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I_{+}^{2}(\vec{R})\; I_{0}^{2}(\vec{R}) = 2u_{2}\, I_{+}^{2}(1)\, I_{0}^{2}(1) + 4v_{2}\, I_{+}^{2}(2)\, I_{0}^{2}(2)$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}}\, I_{+}^{2}(\vec{R})\, I_{0}^{2}(\vec{U}) = \left[2u_{2}\, I_{+}^{2}(1) + 4v_{2}\, I_{+}^{2}(2)\right] \left[4I_{0}^{2}(1) + 4I_{0}^{2}(2)\right]$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}}\, I_{+}^{2}(\vec{R})\, I_{0}(\vec{U})\, I_{0}(\vec{R}+\vec{U}) = 4 \sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I_{+}^{2}(\vec{R}) \left[I_{0}(1)\, I_{0}(\vec{R}+1) + I_{0}(2)\, I_{0}(\vec{R}+2)\right]$$

$$= 4u_{2}\, I_{+}^{2}(1)\, I_{0}(1)\, I_{0}(2) + 4u_{2}\, I_{+}^{2}(1)\, I_{0}(2)\, I_{0}(1)$$

$$+ 8v_{2}\, I_{+}^{2}(2)\, I_{0}(1)\, I_{0}(1)$$

$$= 8u_{2}\, I_{+}^{2}(1)\, I_{0}(1)\, I_{0}(2) + 8v_{2}\, I_{+}^{2}(2)\, I_{0}^{2}(1)$$

For the simple cubic case, we have

$$\sum_{\vec{R}} I_{+}^{2}(\vec{R})\, I_{0}^{2}(\vec{R}) = 6I_{+}^{2}(1)\, I_{0}^{2}(1) + 12I_{+}^{2}(2)\, I_{0}^{2}(2)$$

$$\sum_{\vec{R}\vec{U}} I_{+}^{2}(\vec{R})\, I_{0}^{2}(\vec{U}) = \left[6I_{+}^{2}(1) + 12I_{+}^{2}(2)\right]\left[6I_{0}^{2}(1) + 12I_{0}^{2}(2)\right]$$

$$\sum_{\vec{R}\vec{U}} I_{+}^{2}(\vec{R})\, I_{0}(\vec{U})\, I_{0}(\vec{R}+\vec{U}) = \sum_{\vec{R}} I_{+}^{2}(\vec{R})\left[6I_{0}(1)\, I_{0}(\vec{R}+1) + 12I_{0}(2)\, I_{0}(\vec{R}+2)\right]$$

$$= 24I_{+}^{2}(1)\, I_{0}(1)\, I_{0}(2) + 24I_{+}^{2}(1)\, I_{0}(2)\, I_{0}(1)$$

$$+ 24I_{+}^{2}(2)\, I_{0}(1)\, I_{0}(2) + 48I_{+}^{2}(2)\, I_{0}(2)\, I_{0}(2)$$

and with the definitions of Equation (6.1)

$$\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}} I_+^2(\vec{R})\, I_0^2(\vec{R}) = 2u_3 I_+^2(1)\, I_0^2(1) + 4v_3 I_+^2(2)\, I_0^2(2)$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}} I_+^2(\vec{R})\, I_0^2(\vec{U}) = \left[2u_3 I_+^2(1) + 4v_3 I_+^2(2)\right]\left[6I_0^2(1) + 12I_0^2(2)\right]$$

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}} I_+^2(\vec{R})\, I_0(\vec{U})\, I_0(\vec{R}+\vec{U}) = \sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}} I_+^2(\vec{R}) \left[4I_0(1)\, I_0(\vec{R}+1) + 4I_0(2)\, I_0(\vec{R}+2)\right]$$

$$= 8u_3\, I_+^2(1)\, I_0(1)\, I_0(2) + 8u_3 I_+^2(1)\, I_0(2)\, I_0(1)$$

$$+ 8v_3 I_+^2(2)\, I_0(1)\, I_0(1) + 16v_3 I_+^2(2)\, I_0(2)\, I_0(2)$$

To assist in these summations, Table I presents a listing of the multiplicity of terms of the above kind as a function of dimensionality and of the allowable permutation of vectors. When the summation includes $e^{i\vec{K}\cdot\vec{R}}$ as a factor, then the number of terms are determined by the number of cos terms. For instance, u_2 includes two terms but v_2, only one term. In three dimensions, both u_3 and v_3 contain three cos terms, and the last term given above, the $+16v_3 I_+^2(2) I_0(2) I_0(2)$, is in agreement with the value of 48, given for the correspondingly structured summation for the simple cubic case in Table I.

TABLE I.
Multiplicity of Exchange Terms

Exchange Sums	$\lvert\vec{R}\rvert$	$\lvert\vec{U}\rvert$	$\lvert\vec{R}+\vec{U}\rvert$	Number of Terms Linear Chain	 2-Dim. Net	 Simple Cubic
$\sum_{\vec{R}} I(\vec{R})$	1	-	-	2	4	6
	2	-	-	2	4	12
$\sum_{\vec{R}\vec{U}} I(\vec{R})I(\vec{U})$	1	1	-	4	16	36
	1	2	-	4	16	72
	2	1	-	4	16	72
	2	2	-	4	16	144
$\sum_{\vec{R}\vec{U}} I(\vec{R})I(\vec{U})I(\vec{R}+\vec{U})$	1	1	2	2	8	24
	1	2	1	2	8	24
	2	1	1	2	8	24
	2	2	2	0	0	48

Table I. The multiplicity of terms in three kinds of exchange interaction summations, given for various dimensionalities and for specific, non-vanishing combinations of first and second nearest neighbor distances. The notation here is that $\lvert\vec{R}\rvert = 1$ and 2 denotes the first and second nearest neighbor distances, respectively.

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