Time-dependent autocorrelation function for a linear Heisenberg chain at infinite temperature*

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Numerical calculations have been performed to obtain the *exact* infinite-temperature time-dependent spin autocorrelation function $G(t)$ for a linear chain of N spins ($S = 1/2$) interacting by nearest-neighbor Heisenberg exchange for $N = 5,7,9$ and 11 by a method different from that of Carboni and Richards. Exact results for the first 20 moments and estimates for M_{22} to M_{30} of the frequency autocorrelation function for the infinite chain are provided. Excellent agreement is obtained with some recent results obtained by Morita, who, however, gives terms only up to M_{10}. $G(t)$ does not show a simple monotonic diffusive ($\sim t^{-1/2}$) behavior within $4\hbar/J$, the time domain up to which we believe our result for $N = 11$ to be equivalent to that of the infinite chain although it is reduced to about 10% of its value at $t = 0$.

I. INTRODUCTION

The Heisenberg model for a linear chain of N spins, which is described by the Hamiltonian

$$\mathcal{H} = 2J\sum_{i=1}^{N}\vec{S}_i \cdot \vec{S}_{i+1}\,, \tag{1}$$

where $\vec{S}_i$ is the spin vector for the ith particle and J is the exchange constant, has attracted the attention of theoretical physicists for a long time.[1,2] Early authors treated the model primarily as an interesting many-body problem whose solution would enhance our knowledge of the nature of magnetism in more realistic three-dimensional systems. Recently, interest in this problem has been rekindled due to the realization that in several systems[3] the interaction of magnetic ions can be well described by Eq. (1).

The thermodymanic properties of the classical ($S=\infty$) Heisenberg model have been obtained exactly by Fisher.[4] The ground-state ($T=0$) properties of the quantum ($S=\frac{1}{2}$) chain are also amenable to analytic solution and have been studied extensively by various authors.[5-9] des Cloizeaux and Pearson[10] obtained the elementary excitation spectrum for the antiferromagnetic chain. A significant contribution was made by Bonner and Fisher,[11] who diagonalized $\mathcal{H}$ numerically. By considering chains of increasing length, they were able to make accurate extrapolations for the various thermodynamic properties of the infinite chain. These results have been used in determining J from experimental measurements.

Theoretical studies[12-21] have been further stimulated by recent experiments[22-36] probing the spin dynamics of linear magnetic chains. Many of the physical observables in room-temperature experiments involving magnetic inelastic scattering of neutrons,[22-24] EPR,[25-31] and nuclear spin-lattice relaxation[32-36] can be related to the high-temperature time-dependent spin correlation functions[37] $G_r^z(t)$, defined by

$$G_r^z(t) = \mathrm{Tr}(e^{i\mathcal{H}t}S_0^z e^{-i\mathcal{H}t}S_r^z)/\mathrm{Tr}(1)\,, \tag{2}$$

where the trace can be taken over any complete set of states. Although several microscopic theories[38-42] have been proposed to evaluate $G_r^z(t)$, these invariably involve decoupling approximations at some stage of the calculation which lead to errors in the predicted behavior of $G_r^z(t)$ in the long-time region. The first few coefficients in a Taylor-series expansion of $G_r^z(t)$ around $t=0$ are known exactly.[43-46] These are also the moments of $F_r^z(\omega)$, the frequency Fourier transform of $G_r^z(t)$. Various approaches[47-58] to obtain the long-time behavior of $G_r^z(t)$ using these moments, however, have so far not been very successful. The asymptotic behavior of $G_r^z(t)$ as $t\to\infty$ is of considerable interest in itself and is expected to show a $t^{-1/2}$ tail on the basis of spin-diffusion theory.[15,59]

The classical Heisenberg model has been applied by several authors[15,16] to explain observations in $(CH_3)_4NMnCl_3$(TMMC) in which the Mn^{2+} ions ($S=\frac{5}{2}$) form a linear chain. For the more difficult $S=\frac{1}{2}$ case, Carboni and Richards[60] (CR) performed *ab initio* calculations for $F_r^z(\omega)$ for finite chains of N spins with N as large as 10. Particular attention was focused on $F_0^z(\omega)$, the autocorrelation function, because it could be calculated more accurately compared to other correlation functions. They obtained histograms for $F_0^z(\omega)$ for chains of increasing length, performed extrapolations for $N\to\infty$, fitted their result to a smooth linear function by neglecting a low-frequency divergence, and finally obtained an analytic expression for $G_0^z(t)$ valid for t up to $2\hbar/J$. Their results stimulated further investigations[61-63] into the problem. We have extended their work on $G_0^z(t)$ to $N=11$. Further, we have obtained *exact* results for $G_0^z(t)$ for $N=5$, 7,

9, and 11 by adopting a method[64] which is quite different from that of CR, as is noted in the following points. (i) We do not coarse grain the discrete frequency spectrum obtained to form histograms. (ii) We evaluate $G_0^z(t)$ directly and demonstrate that our result for $N=11$ is expected to reproduce that of the infinite chain up to $t \sim 4\hbar/J$. (iii) As a by-product, we also obtain the first 20 moments and estimate the next 10 moments of $F_0^z(\omega)$ for the infinite chain, whereas only the first 10 moments have been previously determined.

Our method of calculation is explained in Sec. II, the results are discussed in Sec. III, and Sec. IV provides some concluding remarks.

II. CALCULATION

Let us consider a linear chain of N exchange-coupled spins ($S=\frac{1}{2}$) where the Hamiltonian $\mathcal{H}$ is given in Eq. (1).

Baxter[65] has recently diagonalized a more general Hamiltonian analytically but the matrix elements necessary to obtain $G_r^z(t)$ have not yet been evaluated. We therefore adopt the standard method[11,60,66-69] of numerical diagonalization of $\mathcal{H}$.

We first confine the spins to a ring and impose a periodic boundary condition

$$\vec{S}_{i+N}=\vec{S}_i \tag{3}$$

for each i.

One can now introduce a translation operator T which moves each spin one lattice spacing towards its left and another operator S^z, the z component of total spin. T, S^z, $\mathcal{H}$ form a set of commuting operators which can be simultaneously diagonalized. All the eigenvalues and eigenvectors of $\mathcal{H}$, in a representation in which T and S^z are diagonal, can be obtained by numerical diagonalization in a straightforward manner. The eigenvalue equations are listed below:

$$\begin{aligned} T|n, m\rangle &= e^{2\pi i n/N}|n, m\rangle , \\ S^z|n, m\rangle &= m|n, m\rangle , \\ \mathcal{H}|n, m\rangle &= \epsilon_{n,m}|n, m\rangle , \end{aligned} \tag{4}$$

where n takes the values 1, 2, ..., N and m takes the values

$$+\tfrac{1}{2}N,\ \tfrac{1}{2}N-1,\ \ldots,\ -\tfrac{1}{2}N.$$

For room-temperature experiments in most linear chain systems the condition

$$J \ll kT \tag{5}$$

holds.[3] Under this condition, one can make the usual high-temperature approximation[70] for the equilibrium density matrix. The relevant spin correlation functions can be shown to be given by $G_r^z(t)$ for different values of r. We now focus our attention on $G_0^z(t)$ defined by

$$G_0^z(t)=\mathrm{tr}(e^{i\mathcal{H}t}S_0^z e^{-i\mathcal{H}t}S_0^z)/\mathrm{tr}(1) . \tag{6}$$

Evaluating the trace in a representation in which $\mathcal{H}$ is diagonal yields

$$G_0^z(t)=\left(\sum_{\substack{n,m\\p,q}} e^{i(\epsilon_{n,m}-\epsilon_{p,q})t}\,|\langle p, q|S_0^z|n, m\rangle|^2\right)\Big/\mathrm{tr}(1). \tag{7}$$

One now makes the following observations:

(i) $$\langle p, q|S_0^z|n, m\rangle=\delta_{m,q}\langle p, m|S_0^z|n, m\rangle . \tag{8}$$

(ii) $$[\mathcal{H}, R]=0 , \tag{9}$$

where R is the spin-inversion operator. This gives

$$\langle n, m|\mathcal{H}|n, m\rangle=\langle n, -m|\mathcal{H}|n, -m\rangle . \tag{10}$$

(iii) $$\langle n, m|\mathcal{H}|n, m\rangle=\langle N-n, m|\mathcal{H}|N-n, m\rangle . \tag{11}$$

Using these properties Eq. (7) can be simplified to

$$G_0^z(t)=2^{-N}\sum_{\substack{n,p,m\\(p\le n)}} f_{n,p}\cos[(\epsilon_{n,m}-\epsilon_{p,m})t]\,|\langle p, m|S_0^z|n, m\rangle|^2, \tag{12}$$

where f_{np} is the degeneracy parameter which weighs each transition probability properly. Using Eq. (12) one now evaluates $G_0^z(t)$ for any given time.

A Taylor-series expansion of $G_0^z(t)$ around $t=0$ gives

$$G_0^z(t)=\mu_0+\sum_{k=1}^{\infty}\frac{(-1)^k}{(2k)!}\mu_{2k}t^{2k} , \tag{13}$$

where $\mu_0=\frac{1}{4}$ and μ_{2k} is the $(2k)$th moment of the frequency Fourier transform of $G_0^z(t)$.

From Eq. (12) one gets

$$\mu_{2k}=2^{-N}\sum_{\substack{n,p,m\\(p\le n)}} f_{np}(\epsilon_{n,m}-\epsilon_{p,m})^{2k}\,|\langle p, m|S_0^z|n, m\rangle|^2 . \tag{14}$$

We have evaluated $G_0^z(t)$ for t lying between 0 and $4\hbar/J$ and μ_{2k} for $k=1, 2, \ldots, 15$ when $N=5$, 7, 9, 11.

The computer was programmed to evaluate the matrix elements of $\mathcal{H}$ and also the transition probabilities, $|\langle p, m|S_0^z|n, m\rangle|^2$. The largest matrix that had to be diagonalized was 84×84 for $N=11$ and the total number of different frequencies in that case was about 40 000. All computations were performed on the University of Pittsburgh's PDP-10 computer, which precluded use of double precision for the complex eigenvectors, although eigenvalues obtained are in double precision. The relative difficulty involved in going from $N=9$ to $N=11$ is illustrated by the total running time in each case, 6 min for $N=9$ to almost 5 h for $N=11$. We have checked our eigenvalues against those of Bonner and Fisher[11] and they agree to within seven significant digits. A second check is a comparison of our μ_{2k} for $k=1-5$ with those obtained by Morita[46]

TABLE I. Moments for the autocorrelation function.

	μ_{2k}/J^{2k}				
$2k$	$N=5$	$N=7$	$N=9$	$N=11$	$N=\infty$
2	1.000 00	1.000 00	1.000 00	1.000 00	1.000 00
4	11.000 0	11.000 0	11.000 0	11.000 0	11.000 0
6	163.000	163.000	163.000	163.000	163.000
8	2905.00	2909.00	2909.00	2909.00	2909.00
10	57 728.0	60 704.0	60 704.0	60 704.0	60 704.0
12	$1.223\,00\times10^{6}$	$1.473\,93\times10^{6}$	$1.474\,05\times10^{6}$	$1.474\,05\times10^{6}$	$1.474\,05\times10^{6}$
14		$4.162\,59\times10^{7}$	$4.174\,72\times10^{7}$	$4.174\,72\times10^{7}$	$4.174\,72\times10^{7}$
16		$1.360\,26\times10^{9}$	$1.380\,66\times10^{9}$	$1.380\,67\times10^{9}$	$1.380\,67\times10^{9}$
18		$5.080\,67\times10^{10}$	$5.316\,95\times10^{10}$	$5.317\,29\times10^{10}$	$5.317\,29\times10^{10}$
20		$2.132\,04\times10^{12}$	$2.369\,24\times10^{12}$	$2.370\,02\times10^{12}$	$2.370\,02\times10^{12}$
22			$1.211\,23\times10^{14}$	$1.212\,58\times10^{14}$	1.2126×10^{14}
24			$7.032\,53\times10^{15}$	$7.053\ 15\times10^{15}$	7.053×10^{15}
26			$4.584\,19\times10^{17}$	$4.614\ 00\times10^{17}$	4.61×10^{17}
28				$3.356\ 34\times10^{19}$	3.36×10^{19}
30				$2.685\ 46\times10^{21}$	2.7×10^{21}

for the infinite linear chain in an entirely different manner.

III. RESULTS

The μ_{2k}'s for $k=1, 10$ obtained here are expected to be the same as those for the infinite chain. A simple proof of this is as follows.

The conventional expression for μ_{2k} can be obtained directly from Eq. (6), and contains $2k$ $\mathcal{H}$:

$$\mu_{2k}=\mathrm{tr}([\mathcal{H},[\mathcal{H},\ldots,[\mathcal{H},S_0^z]]\cdots]S_0^z)/\mathrm{tr}(1). \quad (15)$$

The traces are usually evaluated in a representation in which S_i^z is diagonal for each i.[46] However, except for μ_2 and μ_4,[43,44] a straightforward analytic evaluation of μ_{2k} becomes rapidly unmanageable. Only recently Morita[46] has obtained terms up through μ_{10} for the linear-chain case and terms up through μ_8 for other lattices.

The so-called moment expansion [Eq. (13)] is a particular form of linked-cluster expansion and a study of Eq. (15) reveals the well-known fact that clusters containing only up to $k+1$ particles can contribute to μ_{2k}.[46] If the spins are arranged on a ring, all possible diagrams connecting, 2, 3, ..., $k+1$ particles for the infinite chain are also allowed for the ring if we are dealing with a nearest-neighbor Hamiltonian—provided, of course, that the ring contains $k+1$ particles. Therefore the moments obtained from an exact solution of the N-spin problem are expected to be the same as those for the infinite chain up to and including μ_{2N-2}. These moments are shown in Table I.

Comparison with Morita's calculation[46] shows excellent agreement for all moments up to μ_{10}, including μ_6, which does not agree with that of McFadden and Tahir-Kheli,[45] and we believe one should be cautious in using their result. The higher moments, to our knowledge, have not been published before.[71–73]

An interesting feature emerges from the fact that the N-spin solution not only gives moments up to μ_{2N-2} exactly (except a small discrepancy in μ_{2N-2} for $N=5$ and $N=7$, arising because of the small number of spins) but also gives fairly good estimates of μ_{2N}, μ_{2N+2}, etc., as well. This result implies that in one-dimensional systems $(k+1)$-particle diagrams do *not* dominate μ_{2k}. Therefore a few of the higher moments for $N=11$ are expected to be good estimates for those of the infinite chain.

Various methods[47–58] have been used to obtain $G_r^z(t)$ from a knowledge of the lower moments. Some of these[47–50,53,54] assume a smooth function $f(t)$, usually a Gaussian or its product with a power series with respect to time, for either the correlation function itself[53] or the so-called "memory

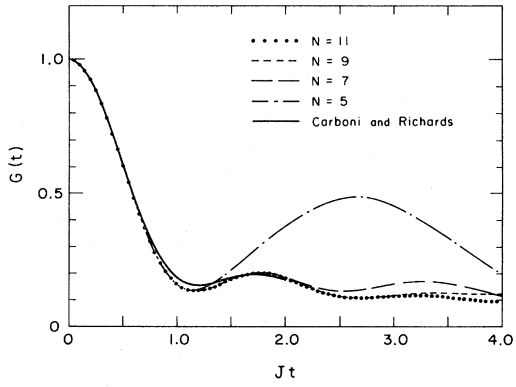

FIG. 1. Normalized infinite-temperature autocorrelation function for a linear Heisenberg chain for different values of N. Carboni-Richards (Ref. 60) results are also shown.

function."[54] One then determines the parameters in the theory by assuming $f(t)$ to have the correct short-time behavior. Another approach is to form rigorous bounds[56,57] for $G_\tau^z(t)$ or to first form a set of "modified moments,"[58] make use of any known asymptotic behavior of $G_\tau^z(t)$, and then estimate $G_\tau^z(t)$. Instead, we have evaluated $G_0^z(t)$ directly using Eq. (12), which amounts to summing to infinite order all diagrams in the moment expansion which can be embedded in a finite ring of spins.

The normalized autocorrelation functions $G(t)$, defined by

$$G(t)=\frac{G_0^z(t)}{G_0^z(0)}=4G_0^z(t) \tag{16}$$

for $N=5$, 7, 9, 11, are shown in Fig. 1. As expected, addition of spins on the chain does not alter the short-time region but modifies the function at longer times. Physically, this corresponds to a finite speed of propagation of information which prohibits a spin from sensing spins far away from itself for a long time. We know, from the nine-spin solution, that the time domain τ up to which our finite-chain results are the same as that for the infinite chain is $3\hbar/J$, and the 11-spin solution is believed to extend τ to a time $\sim 4\hbar/J$. $G(t)$ obtained by CR is also shown in the figure and is in good agreement with our result.

Recently Lurie, Huber, and Blume[20] (LHB) obtained numerical results for $G_\tau^z(t)$ for the classical Heisenberg chain by performing computer experiments. In order to compare our results for $G_0^z(t)$ with theirs, we scale the exchange integrals by the relation

$$J=J_{\mathrm{LHB}}/\sqrt{3}\,, \tag{17}$$

so that both the results will have the same second moment. The comparison is shown in Fig. 2. Because of the quantum nature, our result shows more structure. The similiarity between these two extreme cases in the long-time region implies that for the Heisenberg model $G_0^z(t)$ is not very sensitive to the actual spin value S and is in accord with some previous observations.[49,50]

Considerable attention[61-63] has been focused on the low-frequency divergence in $F_0^z(\omega)$, first suggested by CR. Fernandez and Gersch[61] subsequently proved that the divergence is faster than logarithmic, while spin-diffusion theory predicts[15,59] $F_0^z(\omega)\sim\omega^{-1/2}$ as $\omega\to 0$. Although some recent experiments on TMMC[26,32,34] support this diffusive behavior, the situation is not so clear for the $S=\frac{1}{2}$ chains, such as $Cu(NH_3)_4SO_4\cdot H_2O$ (CTS),[28] $CuCl_2\cdot 2NC_5H_5$ (CPC),[28,29] or the organic free radical 2, 2, 6, -tetramethyl-4-piperidinol-1-oxyl (Tanol),[35,36] where the failure to observe such a divergence is usually interpreted to be due to finite interchain interactions.

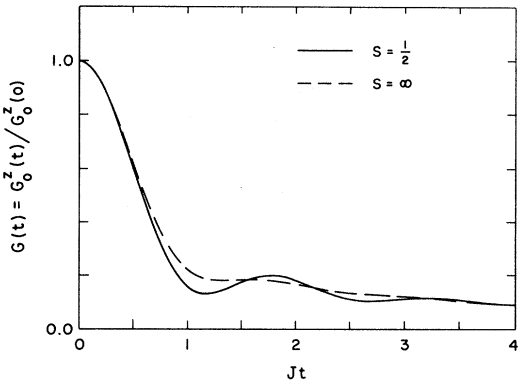

FIG. 2. Comparison of normalized infinite-temperature autocorrelation function for a linear Heisenberg chain for different values of S. The continuous curve is the present calculation for $S=\frac{1}{2}$ and $N=11$. The dashed curve is taken from Ref. 20, after proper normalization and change of scale.

Although we cannot draw any quantitative conclusions regarding the nature of this zero-frequency divergence, the slow decay of $G(t)$ (Fig. 1) does indicate its presence. A simple monotonic ($\sim t^{-1/2}$) damping of $G(t)$ is not seen within $4\hbar/J$, perhaps because contributions from nondiffusive modes are not negligible within this time domain, although $G(t)$ has decayed to about 10% of its value at $t=0$.

IV. CONCLUDING REMARKS

The present analysis shows that much more information can be obtained from an exact solution of a finite spin Hamiltonian by calculating the time correlation functions directly rather than coarse graining the discrete frequency spectrum obtained, the method used by CR. Although we have explicitly considered the autocorrelation function for a Heisenberg chain, our method can be applied to time correlation functions involving any set of operators for more general Hamiltonians. Spin dynamics of a general anisotropic Heisenberg chain is currently under investigation.

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[1]For an account of the historical development in linear magnetic chains see E. H. Lieb and D. C. Mattis, *Mathematical Physics in One Dimension* (Academic, New York, 1966).

[2]For a review on thermodynamic properties of linear magnetic models see C. J. Thompson, in *Phase-Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 1, pp. 177–226.

[3]For a systematic survey, see the recent review article by L. J. de Jongh and A. R. Miedma, Adv. Phys. 23, 1 (1974).

[4]M. E. Fisher, Am. J. Phys. 32, 343 (1964).

[5]H. A. Bethe, Z. Phys. 71, 205 (1931).

[6]L. Hulthen. Arkiv Mat. Astron. Fysik 26A, No. 11 (1938).

[7]R. B. Griffiths, Phys. Rev. 133, A768 (1964).

[8]C. N. Yang and C. P. Yang, Phys. Rev. 147, 303 (1966); 150, 321 (1966); 150, 327 (1966); 151, 258 (1966).

[9]R. J. Baxter, Ann. Phys. (N.Y.) 70, 323 (1972).

[10]J. des Cloizeaux and J. Pearson, Phys. Rev. 128, 2131 (1962).

[11]J. C. Bonner and M. E. Fisher, Phys. Rev. 135, A640 (1964).

[12]P. M. Richards, Phys. Rev. Lett. 27, 1800 (1971).

[13]S. A. Scales and H. A. Gersch, Phys. Rev. Lett. 28, 917 (1972).

[14]S. W. Lovesey and R. A. Meserve, Phys. Rev. Lett. 28, 614 (1972).

[15]F. B. McLean and M. Blume, Phys. Rev. B 7, 1149 (1973); 7, 5017(E) (1973).

[16]H. Tomita and H. Mashiyama, Prog. Theor. Phys. 48, 1133 (1972).

[17]K. Tomita and K. Kawasaki, Prog. Theor. Phys. 49, 1858 (1973).

[18]T. Todani and K. Kawasaki, Prog. Theor. Phys. 50, 1216 (1973).

[19]C. G. Windsor, *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1968), Vol. II, p. 83.

[20]N. A. Lurie, D. L. Huber, and M. Blume, Phys. Rev. B 9, 2171 (1974).

[21]P. C. Hohenberg and W. F. Brinkman, Phys. Rev. B 10, 128 (1974).

[22]M. T. Hutchings, G. Shirane, R. J. Birgeneau, and S. L. Holt, Phys. Rev. B 5, 1999 (1972).

[23]H. Ikeda and K. Harakawa, J. Phys. Soc. Jpn. 35, 722 (1973).

[24]Y. Endoh, G. Shirane, R. J. Birgeneau, P. M. Richards, and S. L. Holt, Phys. Rev. Lett. 32, 170 (1974).

[25]For a review on magnetic resonance studies on one-dimensional systems see D. Hone, AIP Conf. Proc. 5, 413 (1971).

[26]R. E. Dietz, F. R. Merrit, R. Dingle, D. Hone, B. G. Silbernagel, and P. M. Richards, Phys. Rev. Lett. 26, 1186 (1971).

[27]R. R. Bartkowski and B. Morosin, Phys. Rev. B 6, 4209 (1972).

[28]M. J. Hennessy, C. D. McElwee, and P. M. Richards, Phys. Rev. B 7, 930 (1973).

[29]W. Duffy, J. E. Venneman, D. L. Strandburg, and P. M. Richards, Phys. Rev. B 9, 2220 (1974).

[30]Z. G. Soos, T. Z. Huang, J. S. Valentine, and R. C. Hughes, Phys. Rev. B 8, 993 (1973).

[31]T. Z. Huang and Z. G. Soos, Phys. Rev. B 9, 4981 (1974).

[32]F. Borsa and M. Mali, Phys. Rev. B 9, 2215 (1974).

[33]D. Hone, C. Scherer, and F. Borsa, Phys. Rev. B 9, 965 (1974).

[34]M. Ahmed-Bakheit, Y. Barjhoux, F. Ferrieu, M. Nechtschein, and J. P. Boucher, Solid State Commun. 15, 25 (1974).

[35]S. Clement and Y. H. Tchao, J. Phys. (Paris) 34, 583 (1973).

[36]J. P. Boucher, F. Ferrieu, and M. Nechtschein, Phys. Rev. B 9, 3871 (1974).

[37]For a review on spin dynamics of exchange-coupled system see W. Marshall and R. D. Lowde, Rept. Prog. Phys. 31, 705 (1968).

[38]H. Mori and K. Kawasaki, Prog. Theor. Phys. 27, 529 (1962).

[39]P. Resibois and M. DeLeener, Phys. Rev. 152, 305 (1966).

[40]M. DeLeener and P. Resibois, Phys. Rev. 152, 318 (1966).

[41]M. Blume and J. Hubbard, Phys. Rev. B 1, 3815 (1970).

[42]G. F. Reiter, Phys. Rev. B 5, 222 (1972); 7, 3325 (1973).

[43]P. G. deGennes, J. Phys. Chem. Solids 4, 223 (1967).

[44]M. F. Collins and W. Marshall, Proc. Phys. Soc. Lond. 92, 390 (1967).

[45]D. G. McFadden and R. A. Tahir-Kheli, Phys. Rev. B 1, 3671 (1970).

[46]T. Morita, J. Math. Phys. 12, 2062 (1971).

[47]H. S. Bennet and P. C. Martin, Phys. Rev. 138, A608 (1965).

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

[49]R. A. Tahir-Kheli and D. G. McFadden, Phys. Rev. 182, 604 (1969).

[50]D. G. McFadden and R. A. Tahir-Kheli, Phys. Rev. B 1, 3649 (1970).

[51]S. W. Lovesey and R. A. Meserve, J. Phys. C 6, 79 (1973).

[52]T. Gaskell and J. W. Tucker, J. Phys. C 6, 3315 (1973).

[53]T. Morita, J. Math. Phys. 13, 714 (1972).

[54]T. Horiguchi and T. Morita, Phys. Rev. B 7, 1949 (1973).

[55]R. G. Gordon, J. Math. Phys. 9, 1087 (1968).

[56]O. Platz and R. G. Gordon, Phys. Rev. B 7, 4764 (1973).

[57]O. Platz and R. G. Gordon, Phys. Rev. Lett. 30, 264 (1973).

[58]J. C. Wheeler, Phys. Rev. A 9, 825 (1974).

[59]T. Morita, Phys. Rev. B 6, 3385 (1972).

[60]F. Carboni and P. M. Richards, Phys. Rev. 177, 889 (1969). Some additional work is reported in P. M. Richards and F. Carboni, Phys. Rev. B 5, 2014 (1972).

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

[62]H. A. Gersch, Phys. Rev. B 1, 2270 (1970).

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

[64]The same method has been used in a recent study of dipolar-coupled spins and briefly reported in A. Sur and I. J. Lowe, Bull. Am. Phys. Soc. 19, 251 (1974). Also, we have considered odd values of N only, because

at infinite temperature the even-N and odd-N results converge to the same function (see Ref. 11).
[65]R. J. Baxter, Ann. Phys. (N.Y.) 76, 1 (1973); 76, 48 (1973).
[66]J. C. Bonner, Ph. D. thesis (University of London, 1968) (unpublished).
[67]F. Carboni, Ph. D. thesis (University of Kansas, 1967) (unpublished).
[68]C. K. Majumdar, K. Krishan, and V. Mubyai, J. Phys. C 5, 2896 (1972).
[69]C. K. Majumdar, V. Mubyai, and C. S. Jain, Chem. Phys. Lett. 21, 175 (1973).
[70]G. E. Pake, *Paramagnetic Resonance* (Benjamin, New York, 1962).
[71]After the completion of our present investigation we were informed (Ref. 72) of some independent work by Cook (Ref. 73), who gives terms up to μ_{18} which agree with ours in the first three significant digits.
[72]P. M. Richards (private communication).
[73]A. C. Cook, Ph. D. thesis (University of Kansas, 1972) (unpublished).