q-dependent magnetic susceptibility of a disordered linear chain*

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The q-dependent magnetic susceptibility for disordered Ising and classical Heisenberg chains is calculated. The coupling between spins is of the form $J(n) = J_0 e^{-\beta(n-1)}$, $\beta > 0$, with n the random number of interatomic units between neighboring spins. The cases where J is ferromagnetic or antiferromagnetic or of spin-glass type are examined.

I. INTRODUCTION

In studying magnetic properties of physical systems the Heisenberg and Ising spin Hamiltonians have been widely used. Exact results are scarce in two or three dimensions.[1] In one dimension, however, the possibility of obtaining analytic solutions is greater, since the mathematics involved are less complicated than in the two- or three-dimensional cases. Interest in the one-dimensional systems is further enhanced by the existence of some real crystals[2] where the dominant spin interactions occur between atoms arranged in linear chains.

Much of the theoretical study based on the Heisenberg model has been devoted to the periodic case.[3] Recently the disordered case received considerable attention, and the static magnetic properties of the disordered classical Heisenberg linear chain have been examined.[4-7] The classical Hamiltonian is expected to yield results close to the quantum ones for large values of the spin $\vec{S}$. In addition, for the case of a sufficiently disordered system the present authors,[5,6] using a specific probability distribution, have provided strong evidence that the classical Heisenberg Hamiltonian can describe adequately well the quantum case even for small $\vec{S}$ ($S=\frac{1}{2}$). Consequently the study of the disordered classical Heisenberg Hamiltonian is of physical interest, and its results can be quite instructive.

In previous work[5,8] we have demonstrated that the zero-field magnetic susceptibility of the organic conductors N-methylphenazinium-tetracyanoquinodimethanide (NMP-TCNQ), quinolinium$(TCNQ)_2$ and acridinium$(TCNQ)_2$ can be adequately well described by the classical antiferromagnetic $S=\frac{1}{2}$ disordered Heisenberg model provided that the probability distribution of the coupling constant J is of the form $P(J) \propto 1/J^{1-c}$, that is, it is singular at the origin for $c<1$. The singularity in the probability distribution of the coupling constant has profound effects on the thermodynamic properties of the above materials. Examination of the response, χ_q, of the classical disordered Heisenberg model, to a field of the form $H(\vec{r}) = H\cos(\vec{q}\cdot\vec{r})$, for the case where $P(J)\propto 1/J^{1-c}$, will thus be of physical importance, since it will provide information on the outcome of neutron-scattering experiments performed on NMP-TCNQ, quinolinium$(TCNQ)_2$, or acridinium$(TCNQ)_2$. Accordingly, the study of the q-dependent susceptibility of the disordered classical Heisenberg linear chain will be the subject of this paper. We also provide results for the Ising model using the same probability distribution.

Thorpe[7] has studied $\chi(q)$ for a random classical Heisenberg model considering the following two cases: (a) Spins along a topologically-linear polymer chain, that is, all sites are magnetic with the same coupling for all nearest neighbors but the adjacent monomers form random angles; (b) Spins located on every site of a periodic chain containing two kinds of atom A and B, the exchange interactions being $J_{AA}, J_{AB}=J_{BA}, J_{BB}$. He also examined the special case where one of the two kinds of atoms is nonmagnetic, but assumed that there is a nonzero coupling between spins, only when they occupy nearest-neighbor sites. The physical systems considered by Thorpe are not the proper ones to describe the q-dependent susceptibilities of the materials we are interested in since they lead to a $q=0$ susceptibility which at low temperatures is either constant or behaves like $1/T$, whereas the experimentally observed $q=0$ susceptibilities of NMP-TCNQ, quinolinium$(TCNQ)_2$, or acridinium$(TCNQ)_2$ behave like[9] $1/T^{\alpha}$ with $\alpha<1$. In what follows we introduce disorder in a way that produces the singular probability distribution of the coupling constant required to obtain the experimental low-temperature behavior $\chi(q=0)\propto 1/T^{\alpha}$.

We consider a system of localized spins randomly placed on a one-dimensional array of atoms or molecules. We assume that we have interactions only between spins separated by an arbitrary number of intermediate sites of zero magnetic moment. These spins are called nearest-neighbor spins and the distance between them is a random variable. Denoting p to be the probability that a site will have one spin localized on it, we find that the probability of having a distance of n interatomic units between two nearest-neighbor spins is given by

$$P(n)=p(1-p)^{n-1}. \tag{1.1}$$

The interaction between the localized spins was analyzed in detail in Ref. 10 and has the form

$$J(n)=J_0 e^{-\beta(n-1)}, \quad \beta>0 \tag{1.2}$$

The above outlined process produces a Hamiltonian[10]

$$\mathcal{H}=\sum_i J_i \vec{S}_i \cdot \vec{S}_{i+1} \tag{1.3}$$

with J_i a random variable. The probability distribution[10] of J for $J \ll J_0$ has the behavior $P(J) \propto 1/J^{1-c}$, where $c=|\ln(1-p)|/\beta$. We note that the subscript i in Eq. (1.3) represents the ith localized spin and not the ith site of the one-dimensional lattice.

The structure of the paper is as follows: In Sec. II we develop the formalism for the solution of the problem for both the Ising and the classical Heisenberg chains. Section III contains the calculation of $\chi(q)$ at $T=0°$ for the cases where the interaction is ferromagnetic or antiferromagnetic or of spin-glass type. The calculation of $\chi(q)$ at finite temperatures is presented in Sec. IV. Finally, Sec. V is devoted to discussion of the results.

II. FORMALISM

The system of interest described in the introduction is shown in Fig. 1. The chain consists of $N+1$ localized spins. We start measuring the spins from zero, the zeroth spin being placed at the origin of the chain. The distance between the $i-1$ and ith spins is denoted by n_i. The average distance between neighboring spins is a/p, with a the interatomic distance. Thus the average length of the chain we are considering is Na/p. The Hamiltonian of the system in the presence of a magnetic field is given for the Heisenberg case by

$$\mathcal{H}=\sum_{i=1}^{N} J_i \vec{S}_{i-1}\cdot \vec{S}_i - g\mu_B \sum_{i=1}^{N} H_i S_i^z \tag{2.1}$$

and for the Ising case by

$$\mathcal{H}=\frac{1}{4}\sum_{i=1}^{N} J_i \sigma_{i-1}^z \sigma_i^z - \tfrac{1}{2} g\mu_B \sum_{i=1}^{N} H_i \sigma_i^z , \tag{2.2}$$

with H_i the magnetic field at the position of the ith spin and σ^z the Pauli matrix. We consider the case where H_i has the form

$$H_i = H_q e^{iqaL_i} , \tag{2.3}$$

where L_i is the distance of the ith spin from the origin of the chain, $L_i=\sum_{i=1}^{i} n_i$. Here q is in the first Brillouin zone of the chain of molecules, $-\pi/a \le q \le \pi/a$.

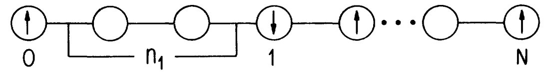

FIG. 1. Chain consisting of $N+1$ localized spins. The empty circles denote sites of zero magnetic moment.

The magnetic moment of site i is

$$M_i = g\mu_B \frac{\mathrm{Tr}(S_i^z e^{-\beta\mathcal{H}})}{\mathrm{Tr}e^{-\beta\mathcal{H}}} . \tag{2.4}$$

We define the magnetic susceptibility χ_{ij} by the following formula

$$\chi_{ij}=\left(\frac{\partial M_i}{\partial H_j}\right)_0 = \frac{g^2\mu_B^2}{kT}\langle S_i^z S_j^z\rangle_0 , \tag{2.5}$$

where the subscript zero denotes that the quantity is calculated for zero magnetic field. Eq. (2.5) also indicates that $\chi_{ij}=\chi_{ji}$. For small magnetic field we can write

$$M_i=\sum_j \chi_{ij} H_j . \tag{2.6}$$

Using the magnetic field given by Eq. (2.3), Eq. (2.6) becomes

$$M_i = H_q \sum_j \chi_{ij} e^{iqaL_j} \equiv \sum_{q'} M_{q'} e^{iq'aL_i} . \tag{2.7}$$

Fourier inversion of Eq. (2.7) leads to

$$M_{q'} = H_q \frac{p}{N+1}\sum_{ij} e^{-iq'aL_i}\chi_{ij} e^{iqaL_j} .$$

Consequently the magnetic susceptibility per site is given by

$$\chi_{q',q}=\frac{\partial M_{q'}}{\partial H_q}=\frac{p}{N+1}\sum_{ij} e^{-iq'aL_i}\chi_{ij}e^{iqaL_j} \tag{2.8}$$

in q space.

From Eq. (2.8) it is evident that the susceptibility has off-diagonal as well as diagonal matrix elements. We further note that Eq. (2.8) gives the zero-field susceptibility for a specific configuration of the $N+1$ spins. In nature, however, an isolated chain does not exist. What we have instead is collection of parallel chains weakly interacting among themselves. Thus to give our formalism physical meaning we have to study the configuration average $\langle\langle \chi_{q',q}\rangle\rangle$. In what follows, $\langle\langle\ \rangle\rangle$ will denote configuration average while $\langle\ \rangle$ will represent thermal average.

From Eq. (2.8) we see that knowledge of the quantity χ_{ij} is essential in finding the susceptibility $\langle\langle \chi_{q',q}\rangle\rangle$. Our inability to calculate χ_{ij} for the case of the quantum Heisenberg model forces us to treat the Ising and the classical Heisenberg chains. In both the latter cases χ_{ij} can be calculated exactly and is given by

$$\chi_{ij}=\frac{g^2\mu_B^2S(S+1)}{3kT}\begin{cases}\prod_{l=\min(i,j)+1}^{\max(i,j)} U\left(\frac{J_l}{kT}\right) & \text{for } i\neq j,\\ 1 \text{ for } i=j.\end{cases} \tag{2.9}$$

For the classical Heisenberg chain[3]

$$U\left(\frac{J}{kT}\right)=\frac{kT}{JS(S+1)}-\coth\left(\frac{JS(S+1)}{kT}\right) \tag{2.10}$$

and $\vec{S}^2=S(S+1)$, while for the Ising chain we have[11]

$$U(J/kT)=\tanh(-J/kT) \tag{2.11}$$

and $S=\frac{1}{2}$.

III. $T\to 0\,°\mathrm{K}$ CASE

Our aim is to calculate the quantity

$$\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2S(S+1)}\langle\langle\chi_{q'q}(T)\rangle\rangle =\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2S(S+1)}\;\frac{p}{N+1}\sum_{ij}\langle\langle e^{-i(q'aL_i-qaL_j)}\chi_{ij}\rangle\rangle. \tag{3.1}$$

In both the Ising and the classical Heisenberg models we have that $\lim_{T\to 0°K}U(J/kT)=\mathrm{sgn}(-J)$. Using this fact and also Eq. (2.9), Eq. (3.1) becomes

$$\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2S(S+1)}\langle\langle\chi_{q'q}(T)\rangle\rangle =\frac{p}{N+1}\sum_{ij}[\mathrm{sgn}(-J)]^{|i-j|}\langle\langle e^{-i(q'aL_i-qaL_j)}\rangle\rangle \tag{3.2}$$

for the ferromagnetic or antiferromagnetic cases. The spin glass will be analyzed later.

A. Diagonal matrix elements

In order to calculate the average $\langle\langle\exp[-iqa(L_i-L_j)]\rangle\rangle$ for $q'=q$, we distinguish the following cases: (a) $i<j$. In that case

$$L_j-L_i=\sum_{l=i+1}^{j}n_l$$

and

$$\langle\langle\exp[-iqa(L_i-L_j)]\rangle\rangle=\prod_{l=i+1}^{j}\langle\langle\exp(iqan_l)\rangle\rangle =\langle\langle\exp(iqan)\rangle\rangle^{j-i}. \tag{3.3}$$

The average in Eq. (3.3) can be evaluated with the use of Eq. (1.1) and is equal to

$$\langle\langle e^{iqan}\rangle\rangle=p\sum_{n=1}^{\infty}(1-p)^{n-1}e^{iqan}=\frac{pe^{iqa}}{1-(1-p)e^{iqa}}\equiv A_q, \tag{3.4}$$

so that

$$\langle\langle\exp[-iqa(L_i-L_j)]\rangle\rangle=A_q^{j-i}; \tag{3.5}$$

(b) $i=j$. In that case

$$\langle\langle\exp[iqa(L_i-L_j)]\rangle\rangle=1; \tag{3.6}$$

(c) $i>j$.

$$\langle\langle\exp[-iqa(L_i-L_j)]\rangle\rangle=\langle\langle\exp[iqa(L_i-L_j)]^*\rangle\rangle =(A_q^*)^{i-j}. \tag{3.7}$$

Denoting $A_q\,\mathrm{sgn}(-J)$ as B_q and making use of Eqs. (3.4), (3.6), and (3.7), in the thermodynamic limit, $N\to\infty$, Eq. (3.2) becomes

$$\lim_{T\to 0}\frac{3kT}{pg^2\mu_B^2S(S+1)}\langle\langle\chi_q(T)\rangle\rangle=\frac{1-|B_q|^2}{1+|B_q|^2-(B_q+B_q^*)}. \tag{3.8}$$

1. Antiferromagnetic chain

Antiferromagnetic coupling leads to $\mathrm{sgn}(-J)=-1$ and Eq. (3.8) becomes

$$\lim_{T\to 0}\frac{3kT}{pg^2\mu_B^2S(S+1)}\langle\langle\chi_q(T)\rangle\rangle =\frac{(1-p)[1-\cos(qa)]}{(1-p)[1-\cos(qa)]+p[2p-1+\cos(qa)]} \tag{3.9}$$

The maximum occurs at $qa=\pi$ and the minimum at $q=0$, the values being $1/(1-p)$ and zero, respectively. For $p\ll 1$ and $q\neq 0$ we have

$$\lim_{T\to 0}\frac{3kT}{pg^2\mu_B^2S(S+1)}\langle\langle\chi_q(T)\rangle\rangle\simeq\frac{1-p}{1-2p}\simeq 1+p. \tag{3.10}$$

In Fig. 2 the quantity $\lim_{T\to 0}[3kT/pg^2\mu_B^2S(S+1)]\times\langle\langle\chi_q(T)\rangle\rangle$ is plotted versus qa, for different values of p. The striking feature is that the maximum occurs always at $q=\pi/a$. Consider a periodic antiferromagnetic spin chain where the distance between the spins is equal to the average spin distance a/p of the random chain. In this chain the maximum will occur at $q=p(\pi/a)$, which is different from our result for the ensemble averaged random chain. For $p=1$ we recapture the periodic result, that is

$$\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2S(S+1)}\langle\langle\chi_q(T)\rangle\rangle=\begin{cases}0 & \text{for } q\neq\pi/a,\\ +\infty & \text{for } q=\pi/a.\end{cases} \tag{3.11}$$

2. Ferromagnetic chain

In this case $\mathrm{sgn}(-J)=1$ and Eq. (3.8) becomes

$$\lim_{T\to 0}\frac{3kT}{pg^2\mu_B^2S(S+1)}\langle\langle\chi_q(T)\rangle\rangle=\begin{cases}1-p & \text{for } q\neq 0,\\ +\infty & \text{for } q=0.\end{cases} \tag{3.12}$$

As in the antiferromagnetic case, for $p=1$ we obtain the periodic result:

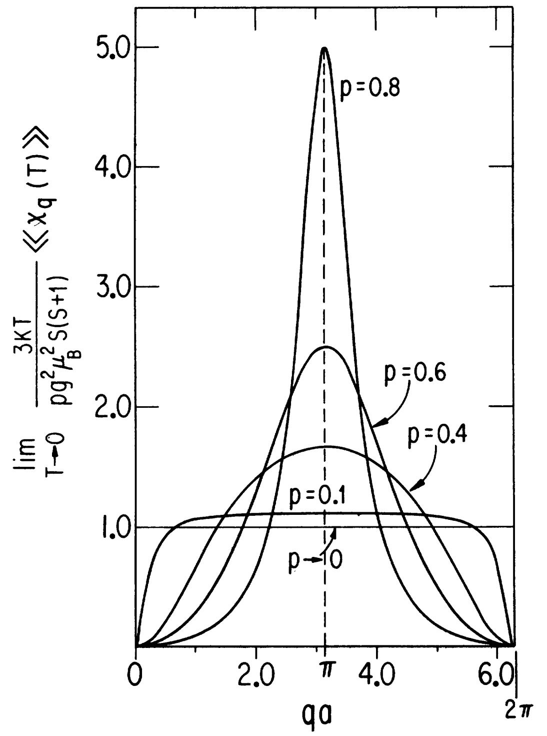

FIG. 2. Quantity $\lim_{T\to 0}\,[3kT/g^2\mu_B^2 pS(S+1)]\,\langle\langle\chi_q(T)\rangle\rangle$ is plotted vs qa for an antiferromagnetic chain and for $p=0$, 0.1, 0.4, 0.6, and 0.8.

$$\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2 S(S+1)}\langle\langle\chi_q(T)\rangle\rangle=\begin{cases}0 & \text{for } q\neq 0,\\ +\infty & \text{for } q=0.\end{cases}$$

3. Spin-glass-like case

Besides an antiferromagnetic or ferromagnetic interaction among the spins, a combination of these two former cases can exist. We assume the interaction to be ferromagnetic if the number n_i of interatomic units between the spins is even and antiferromagnetic for n_i odd. In this system $\lim_{T\to 0}U(J_i/kT)=(-1)^{n_i}$. The configuration average we need to calculate in order to find the diagonal matrix elements of $\langle\langle\chi_{q,q}\rangle\rangle$ is given by

$$\langle\langle(-1)^n e^{iqan}\rangle\rangle=p\sum_{n=1}^{\infty}(1-p)^{n-1}(-e^{iqa})^n$$
$$=\frac{-pe^{iqa}}{1+(1-p)e^{iqa}}\equiv C_q. \qquad (3.13)$$

From this point on the calculations are similar to that of the antiferromagnetic or ferromagnetic cases and the result is the following:

$$\lim_{T\to 0}[3kT/pg^2\mu_B^2 S(S+1)]\langle\langle\chi_q(T)\rangle\rangle$$
$$=(1-|C_q|^2)/[1+|C_q|^2-(C_q+C_q^*)], \qquad (3.14)$$

where C_q^* is the complex conjugate of C_q. Inserting into Eq. (3.14) the value of C_q as given by Eq. (3.13), we obtain

$$\lim_{T\to 0}[3kT/pg^2\mu_B^2 S(S+1)]\langle\langle\chi_q(T)\rangle\rangle=1-p. \qquad (3.15)$$

For $q\neq 0$ we note that the result in the spin-glass case is the same as that in ferromagnetic case. At $q=0$, however, the spin-glass model produces a nonsingular $\chi_0(T)$ in contrast with the ferromagnetic case.

From our examination of the ferromagnetic, the antiferromagnetic and the spin-glass-like chains we see that in all cases $\lim_{T\to 0}T\langle\langle\chi_q(T)\rangle\rangle=$ constant for $q\neq 0$. Thus the behavior of $\langle\langle\chi_q(T)\rangle\rangle$ at low temperatures and for $q\neq 0$ is given by

$$\langle\langle\chi_q(T)\rangle\rangle\propto 1/T\ . \qquad (3.16)$$

B. Off-diagonal matrix elements

1. Ferro and antiferromagnetic cases

In order to evaluate the off-diagonal matrix elements of $\langle\langle\chi_{q',q}(T)\rangle\rangle$ we need to know the configuration average $\langle\langle\exp[-i(q'aL_i-qaL_j)]\rangle\rangle$. We distinguish the following cases:

Case 1: $i=j$. In this particular case

$$\langle\langle\exp[-i(q'-q)aL_i]\rangle\rangle=\langle\langle\exp[-i(q'-q)an]\rangle\rangle^i.$$

Since

$$\langle\langle\exp[-i(q'-q)an]\rangle\rangle=p\sum_{n=1}^{\infty}(1-p)^{n-1}\exp[-i(q'-q)an]$$
$$=\frac{p\exp[-i(q'-q)a]}{1-(1-p)\exp[-i(q'-q)a]},$$

we finally obtain

$$\langle\langle\exp[-i(q'-q)aL_i]\rangle\rangle=\left(\frac{p\exp[-i(q'-q)a]}{1-(1-p)\exp[-i(q'-q)a]}\right)^i. \qquad (3.17)$$

Case 2: $i>j$.

$$\langle\langle\exp[-i(q'L_i-qL_j)a]\rangle\rangle$$
$$=\langle\langle\exp[-i(q'-q)aL_j]\exp[-iq'a(L_i-L_j)]\rangle\rangle$$
$$=\left(\frac{p\exp[-i(q'-q)a]}{1-(1-p)\exp[-i(q'-q)a]}\right)^i$$
$$\times\left(\frac{p\exp(-iq'a)}{1-(1-p)\exp(-iq'a)}\right)^{i-j}. \qquad (3.18)$$

Case 3: $i<j$. In this case we have

$$\begin{aligned}\langle\langle\exp[-i(q'L_i-qL_j)a]\rangle\rangle &= \langle\langle\exp[-i(q'-q)aL_i]\rangle\rangle\,\langle\langle\exp[-iqa(L_i-L_j)]\rangle\rangle\\ &=\left(\frac{p\exp[-i(q'-q)a]}{1-(1-p)\exp[-i(q'-q)a]}\right)^i\\ &\quad\times\left(\frac{p\exp(-iqa)}{1-(1-p)\exp(-iqa)}\right)^{j-i}. \end{aligned}\tag{3.19}$$

Using the notation

$$\begin{aligned}A_1(q'-q)&=pe^{-i(q'-q)a}/[1-(1-p)e^{-i(q'-q)a}],\\ B_1(q')&=p\{e^{-iq'a}/[1-(1-p)e^{-iq'a}]\}\operatorname{sgn}(-J),\\ C_1(q)&=p\{e^{-iqa}/[1-(1-p)e^{-iqa}]\}\operatorname{sgn}(-J),\end{aligned}\tag{3.20}$$

and also making use of Eqs. (3.17), (3.18), and (3.19) we obtain that for $q\neq q'$ the susceptibility at $T\to 0\,°\mathrm{K}$ is given by

$$\begin{aligned}\lim_{T\to 0}\frac{3kT}{g^2\mu_B^2S(S+1)}\langle\langle\chi_{q'q}(T)\rangle\rangle &=\frac{p}{N+1}\left[\frac{A_1}{A_1-B_1}\left(\frac{A_1^{N+1}-A_1}{A_1-1}-\frac{B_1^{N+1}-B_1}{B_1-1}\right)\right.\\ &\quad\left.+\frac{C_1A_1}{C_1-1}\left(\frac{A_1^N-C_1^N}{A_1-C_1}-\frac{A_1^N-1}{A_1-1}\right)\right].\end{aligned}\tag{3.21}$$

Provided that all denominators in Eq. (3.21) are different from zero and going to the thermodynamic limit, $N\to\infty$, we obtain

$$\lim_{T\to 0}[3kT/g^2\mu_B^2S(S+1)]\,\langle\langle\chi_{q'q}(T)\rangle\rangle=0. \tag{3.22}$$

Equation (3.21) also implies that

$$\lim_{T\to 0}(\lim_{N\to\infty}\langle\langle\chi_{q'q}(T)\rangle\rangle)=0 \tag{3.23}$$

if we allow $N\to\infty$ first and then take the limit $T\to 0\,°\mathrm{K}$. Therefore, the off-diagonal matrix elements of the susceptibility are zero.

We examine now the special cases in which at least one of the denominators of Eq. (3.21) is zero. From Eq. (3.20) we see that $A_1=1$ only if $q=q'$. This corresponds to diagonal matrix elements and was examined before. For the off-diagonal matrix elements ($q\neq q'$) we always have $|A|<1$ and thus the denominator A_1-1 never becomes zero. In the examination of the possibility that one of the remaining denominators is zero we distinguish the following cases:

(a) Antiferromagnetic Case ($J>0$). For $p\neq 0,1$, B_1 and C_1 are different from zero and thus B_1-1 and C_1-1 never become zero. In the case where $A_1=B_1$ we note the following: Since $|A_1|<1$ this implies $|B_1|<1$. For $|B_1|<1$ it is easy to prove that Eqs. (3.22) and (3.23) hold in the thermodynamic limit. The same is true for the case $C_1=A_1$. In conclusion, for antiferromagnetic coupling, relation (3.23) always holds.

(b) Ferromagnetic Case ($J<0$). The analysis for the cases $B_1=A_1$ and $C_1=A_1$ is similar to that of the antiferromagnetic case. We consider now the cases $B_1=1$ and $C_1=1$ which occur for $q'=0$ and $q=0$, respectively. For the case $q'=0$ and for small but finite temperatures, we have

$$B_1(T)=\langle\langle U(J/kT)\rangle\rangle \tag{3.24}$$

with U given by Eq. (2.10) for the case of the classical Heisenberg model. Using Eq. (1.2) and calculating the average in Eq. (3.24) we obtain that the behavior of $B_1(T)$ at low T is given by

$$B_1(T)=\begin{cases}1-\delta_1kT/J_0 & \text{if } |\ln(1-p)|>\beta\\ 1-\delta_2(kT/J_0)^{|\ln(1-p)|/\beta} & \text{if } |\ln(1-p)|<\beta\end{cases}$$

with δ_1,δ_2 positive. Thus the limit of $[1/(N+1)]\times 1/[B_1(T)-1]$, allowing first N to go to infinity and then $T\to 0$, is zero. Consequently relation (3.23) is valid.

A similar analysis applies to the $q=0$ case. Examining the Ising model shows that the behavior is the same as that of the classical Heisenberg chain.

2. *Spin-glass case*

As regards the off-diagonal matrix elements, following the same procedure as in the ferromagnetic and antiferromagnetic cases we find that the relations of (3.20) change to

$$\begin{aligned}A_1(q'-q)&=p\,\frac{e^{-i(q'-q)a}}{1-(1-p)e^{-i(q'-q)a}},\\ B_1(q')&=-p\,\frac{e^{-iq'a}}{1+(1-p)e^{-iq'a}},\\ C_1(q)&=-p\,\frac{e^{-iqa}}{1+(1-p)e^{-iqa}}.\end{aligned}\tag{3.25}$$

Otherwise everything remains the same and thus, in the thermodynamic limit, Eq. (3.23) holds.

In summary, from our analysis of Sec. III B we conclude that in the thermodynamic limit, the off-diagonal matrix elements of $\langle\langle\chi_{q'q}(T)\rangle\rangle$, with $T\to 0\,°\mathrm{K}$, are zero for the antiferromagnetic, ferromagnetic, and spin-glass case.

IV. FINITE TEMPERATURES

A. Diagonal matrix elements

For the calculation of χ_q at finite temperatures we need to calculate the quantity

$$D_q(T)=\langle\langle U[J(n_i)/kT]e^{iqan_i}\rangle\rangle, \tag{4.1}$$

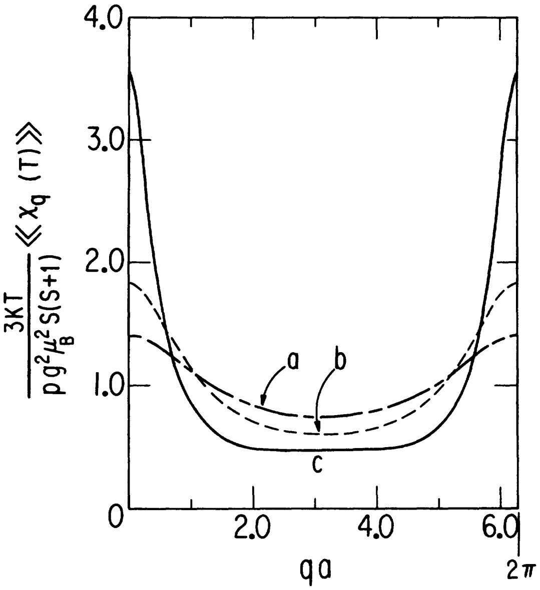

FIG. 3. Ferromagnetic classical Heisenberg chain $[3kT/g^2\mu_B^2 pS(S+1)]\langle\langle\chi_q(T)\rangle\rangle$ is plotted vs qa, for $kT/S(S+1)J_0 =$ (a) 1.0, (b) 0.5, (c) 0.1, $p=0.5$ and $\beta=2.0$.

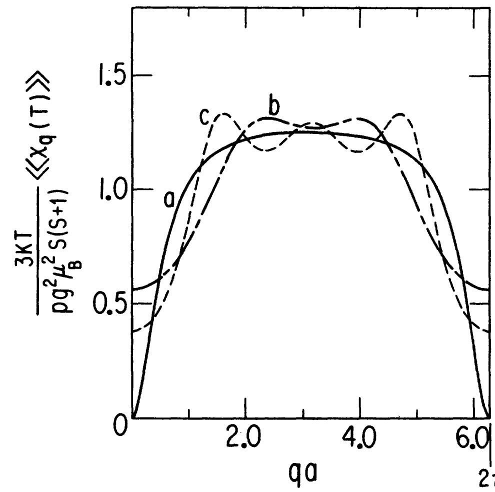

FIG. 4. Antiferromagnetic classical Heisenberg chain $[3kT/g^2\mu_B^2 pS(S+1)]\ \langle\langle\chi_q(T)\rangle\rangle$ is plotted vs qa, for $kT/S(S+1)J_0 =$ (a) 0, (b) 10^{-2}, (c) 10^{-4}, $p=0.2$ and $\beta=4.0$.

in terms of which we have

$$\frac{3kT}{pg^2\mu_B^2 S(S+1)}\langle\langle\exp[iqa(L_i-L_j)]\chi_{ij}\rangle\rangle=\begin{cases}[D_q(T)]^{i-j} & \text{for } i>j\,,\\ 1 & \text{for } i=j\,,\\ [D_{-q}(T)]^{j-i} & \text{for } i<j\,.\end{cases} \tag{4.2}$$

Proceeding in a way similar to the zero-temperature case, we obtain in the thermodynamic limit, $N\to\infty$,

$$[3kT/pg^2\mu_B^2 S(S+1)]\langle\langle\chi_q(T)\rangle\rangle=[1-|D_q(T)|^2]/\{1+|D_q(T)|^2-[D_q(T)+D_{-q}(T)]\}\,. \tag{4.3}$$

$D_q(T)$ is calculated numerically. $\langle\langle\chi_q\rangle\rangle$ is shown as a function of qa for different temperatures in Figs. 3 and 4 for the ferromagnetic and antiferromagnetic classical Heisenberg chains, respectively. Figures 5 and 6 are the corresponding results for the Ising chain. From Fig. 4 it is apparent that in the antiferromagnetic case at low temperatures there exist q fluctuations in $\langle\langle\chi_q\rangle\rangle$, while from Fig. 7 we see that such fluctuations are absent at high temperatures. In order to understand the origin of these fluctuations we examine the behavior of $\langle\langle\chi_q(T)\rangle\rangle$ for the limiting case of $T\ll J_0$. We focus on the classical antiferromagnetic Heisenberg chain only, the behavior of the Ising model being similar. $U(J)$ for the classical Heisenberg model is given by Eq. (2.10). Using the approximation

$$\coth x=\begin{cases}1/x+\tfrac{1}{3}x & \text{for } 0<x<1.5\,,\\ 1 & \text{for } x\geq 1.5\,.\end{cases} \tag{4.4}$$

$U[J(n)/kT]$, for $S=\frac{1}{2}$, becomes

$$U\left(\frac{J(n)}{kT}\right)=\begin{cases}-\tfrac{1}{3}[J_0S(S+1)/kT]e^{-\beta(n-1)} & \text{for } n>n_0\,,\\ [kT/J_0S(S+1)]e^{\beta(n-1)}-1 & \text{for } n\leq n_0\,,\end{cases} \tag{4.5}$$

where n_0 is defined by $[J_0S(S+1)/kT]e^{-\beta(n_0-1)}=1.5$. For $kT\ll J_0$, so that $n_0\gg 1$, we have

$$\langle\langle e^{iqan}U[J(n)]\rangle\rangle\simeq p\sum_{n=1}^{n_0}(1-p)^{n-1}e^{iqan}\left(\frac{kT}{J_0S(S+1)}e^{\beta(n-1)}-1\right)-\frac{p}{3}\sum_{n=n_0+1}^{\infty}(1-p)^{n-1}e^{iqan}\frac{J_0S(S+1)}{kT}e^{-\beta(n-1)}\,. \tag{4.6}$$

Since $n_0=(1/\beta)\ln\{[J_0S(S+1)/1.5kT]e^{\beta}\}$, Eq. (4.6) becomes

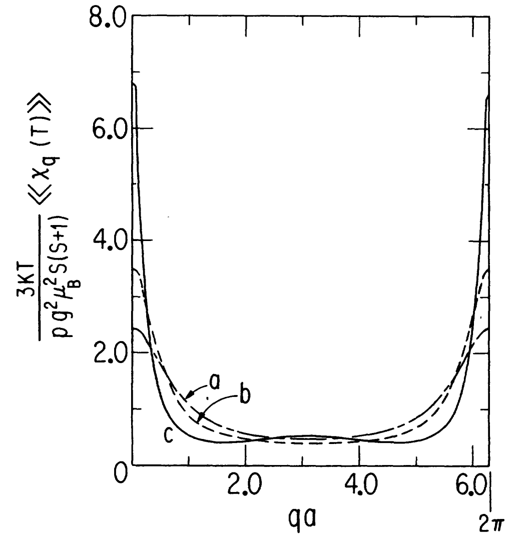

FIG. 5. Ferromagnetic Ising chain $[3kT/g^2\mu_B^2 pS(S+1)] \times \langle\langle \chi_q(T)\rangle\rangle$ is plotted vs qa, for $kT/S(S+1)J_0=$ (a) 1.0, (b) 0.5, (c) 0.1, $p=0.5$ and $\beta=2.0$.

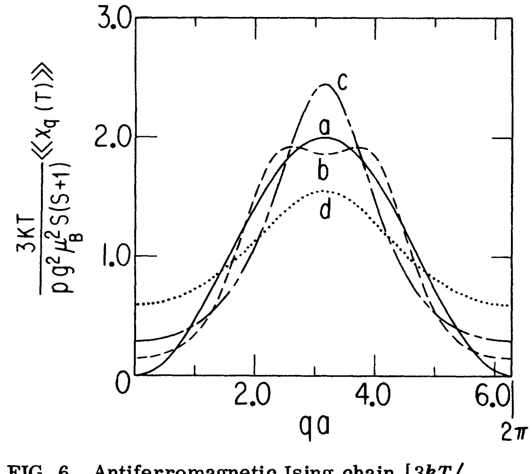

FIG. 6. Antiferromagnetic Ising chain $[3kT/g^2\mu_B^2 pS(S+1)]\ \langle\langle \chi_q(T)\rangle\rangle$ is plotted vs qa, for $kT/S(S+1)J_0$ = (a) 0, (b) 0.1, (c) 0.5, (d) 2.0, $p=0.5$ and $\beta=2$.

$$\langle\langle e^{iqan}U[J(n)]\rangle\rangle \simeq -\frac{pe^{iqa}}{1-(1-p)e^{iqa}}+\frac{pkT}{S(S+1)J_0}\,\frac{e^{iqa}}{1-(1-p)e^{iqa}e^{\beta}}+pe^{iqa(n_0+1)}\left(\frac{1.5k}{S(S+1)}\,\frac{T}{J_0}\,e^{-\beta}\right)^{|\ln(1-p)|/\beta}$$
$$\times\left(\frac{1}{1-(1-p)e^{iqa}}-\frac{1}{2}\,\frac{1}{e^{\beta}-(1-p)e^{iqa}}-\frac{2}{3}\,\frac{1}{e^{-\beta}-(1-p)e^{iqa}}\right). \quad (4.7)$$

For $|\ln(1-p)|/\beta<1$ the behavior of $\langle\langle e^{iqan}UJ(n)/kT\rangle\rangle$ at low temperatures will be the following:

$$\left\langle\left\langle e^{iqan}U\left(\frac{J(n)}{kT}\right)\right\rangle\right\rangle \simeq -\frac{pe^{iqa}}{1-(1-p)e^{iqa}}+pe^{iqa(n_0+1)}\left(\frac{1.5k}{S(S+1)}\,\frac{T}{J_0}\,e^{-\beta}\right)^{|\ln(1-p)|/\beta}$$
$$\times\left(\frac{1}{1-(1-p)e^{iqa}}-\frac{1}{2}\,\frac{1}{e^{\beta}-(1-p)e^{iqa}}-\frac{2}{3}\,\frac{1}{e^{-\beta}-(1-p)e^{iqa}}\right). \quad (4.8)$$

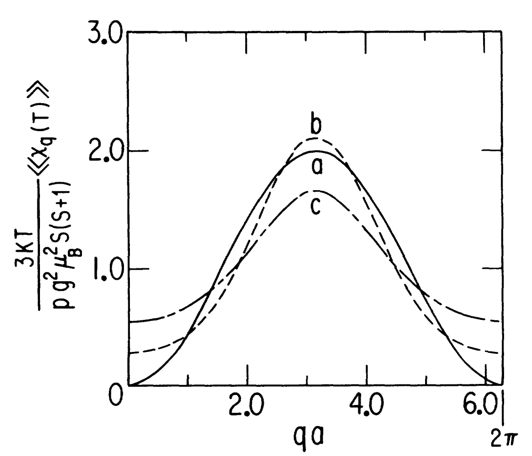

FIG. 7. Antiferromagnetic classical Heisenberg chain $[3kT/g^2\mu_B^2 p\, S(S+1)]\ \langle\langle \chi_q(T)\rangle\rangle$ is plotted vs qa, for $kT/S(S+1)J_0=$ (a) 0, (b) 0.1, (c) 0.5, $p=0.5$ and $\beta=2.0$.

Since n_0 depends on the temperature, the presence of the factor $e^{iqa(n_0+1)}$ will produce strong q fluctuations on the second term of Eq. (4.8). Thus $\langle\langle\chi_q(T)\rangle\rangle$ will also exhibit fluctuations, and we are able to understand Fig. 4 which is the result of *exact* calculations for $p=0.2$, $\beta=4$, and $|\ln(1-p)|/\beta=0.0558$. In the case $|\ln(1-p)|/\beta>1$, the linear term in Eq. (4.7) dominates the temperature dependence for $kT\ll J_0$ and $\langle\langle\chi_q(T)\rangle\rangle$ does not exhibit oscillations due to the absence of the $e^{iqa(n_0+1)}$ term. These arguments suggest that oscillations of $\langle\langle\chi_q(T)\rangle\rangle$ with q must occur also for the ferromagnetic case at low temperature, but will be confined to the high peak in $\langle\langle\chi_q(T)\rangle\rangle$ near $q=0$, where the susceptibility is large. Because the slope of $\langle\langle\chi_q(T)\rangle\rangle$ with q is very large there, it will be difficult to notice the fluctuations.

For completeness we also examine the case

$T \gg J_0$. In that limit we have

$$U[J(n)/kT] \simeq -\tfrac{1}{3}[J_0 S(S+1)/kT]e^{-\beta(n-1)}$$

and thus

$$\left\langle\left\langle e^{iqan} U\left(\frac{J(n)}{kT}\right)\right\rangle\right\rangle \simeq \frac{-pJ_0S(S+1)}{3kT}\,\frac{e^{iqa}}{1-(1-p)e^{-\beta}e^{iqa}}\ .$$

The susceptibility $\langle\langle \chi_q(T)\rangle\rangle$ is then given by

$$\frac{3kT}{g^2\mu_B^2 S(S+1)}\langle\langle \chi_q(T)\rangle\rangle \simeq \frac{p}{N+1}\sum_{i=j} 1 \simeq p\,. \qquad (4.9)$$

Equation (4.9) indicates that in the case $T \gg J_0$ we get the same result as for free spins.

B. Off-diagonal matrix elements

The analysis for the off-diagonal matrix elements at finite temperatures is essentially the same as that of the $T \to 0$ °K case. The only changes are the replacement of $B_1(q')$ by $D_{-q'}(T)$ and $C_1(q)$ by $D_{-q}(T)$. In fact, the finite T case is much simpler since $|D_{-q}(T)|<1$.

V. DISCUSSION

In this paper we have calculated the q-dependent susceptibility for disordered Ising and classical Heisenberg chains. Our investigation included the cases where the interaction between the spins is ferromagnetic or antiferromagnetic or of a spin-glass type. The most interesting result of our calculations occurred for the antiferromagnetic case where at $T \to 0$ °K and also at finite T, $\langle\langle\chi_q\rangle\rangle$ exhibits a maximum at $qa=\pi$, instead of $qa=p\pi$ which one would expect by analogy with a periodic chain with interatomic distance equal to the average distance of the random chain. The reason that the maximum occurs at $qa=\pi$ is that a is the most probable distance between the spins in this one-dimensional case. What is different in our result from that of the periodic chain with interatomic distance a is that our peak at $qa=\pi$ exhibits a width. The width is due to the disorder, and its value will depend on the amount of the latter. The implications of this result are the following: The scattering cross section in the quasielastic approximation is given by[12]

$$\frac{d\sigma}{d\Omega} \propto \chi(q). \qquad (5.1)$$

Since the position of the maxima does not depend on the amount, $1-p$, of the disorder present in the system, the scattering angles will not change with disorder. The effect of disorder will be the introduction of a width in the scattering angles.

Another interesting phenomenon is the oscillation of the susceptibility with q for the antiferromagnetic case at low temperatures. These oscillations arise from the existence of a separation $n_0(T)$ beyond which the exchange becomes significantly less than T in magnitude. The oscillations occur only when $|\ln(1-p)|/\beta$ is less than unity, which corresponds to the case where the probability distribution of exchange is singular at[5,6] $J=0$. In that case there is a significant number of exchanges smaller than T, and $n_0(T)$ introduces a meaningful temperature-dependent cut off into the exchange distribution. The $q=0$ susceptibility changes from nonsingular to singular behavior as $T\to 0$ °K for precisely the same reason.[5,6]

We expect our theory to be applicable to the case of quasi-one-dimensional disordered materials such as NMP-TCNQ, quinolinium$(TCNQ)_2$, and acridinium$(TCNQ)_2$. However, until now, neutron-scattering experiments have not been performed in these materials. We believe that such experiments will be the source of valuable information.

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