Magnetic susceptibility of disordered one-dimensional systems*†

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The magnetic susceptibility of various one-dimensional (1-D) disordered models is studied. At low temperatures and small transfer integrals the Hubbard model reduces to that of a disordered 1-D Heisenberg antiferromagnet with probability distribution of exchange of the form $P(J) \propto 1/J^{1-c}$. Via a cluster argument we find that the low-temperature magnetic susceptibility behaves as $\chi \propto 1/T^{1-c}$. That is, it has a singularity at $T = 0$°K of the same form as that of the probability distribution. Various exactly soluble model Hamiltonians were also studied using the same probability distribution. From these studies we have inferred that for a sufficiently disordered system the quantum 1-D Heisenberg model can be adequately represented by the classical Heisenberg model.

I. INTRODUCTION

One-dimensional electron systems are the object of increasing interest, especially in connection with the behavior of crystals containing tetracyanoquinodimethanide[1] (TCNQ). Among them a class including the materials N-methyl-phenazinium-tetracyanoquinodimethanide (NMP-TCNQ), quinolinium $(TCNQ)_2$, and acridinium $(TCNQ)_2$ has unusual magnetic properties. Experimental results[2-4] for the magnetic susceptibility of these substances indicate the existence of a singularity at $T = 0$ °K, with the susceptibility behaving at low temperatures as $1/T^\gamma$, $\gamma < 1$. One model extensively used in the study of one-dimensional systems is the Hubbard model. In the case of the infinite periodic Hubbard chain it was proven[5,6] that the magnetic susceptibility at zero temperature is finite. Thus the periodic one-dimensional Hubbard model is inadequate to explain the magnetic-susceptibility data of these materials and a modification of it must be sought.

The noticeable increase in the magnetic susceptibility seems unlikely to be connected with paramagnetic impurities present in the specimen, but it appears to be an intrinsic property of the materials.[2,7] The following experimental facts are in support of this argument: (a) at low temperatures the paramagnetic susceptibility does not obey the Curie law expected from paramagnetic impurities; and (b) the amplitude is too large to be attributed to impurities. We believe that the intrinsic property of the materials to which the behavior of the magnetic susceptibility can be attributed is the structural disorder. Observed variations in the susceptibility in materials of comparable purity could then be attributed to variations in the degree of disorder, but would be difficult to understand in terms of partial charge transfer.[8]

Accordingly, in a previous paper we studied the Hubbard model

$$H = \sum_{i,s} \epsilon_i a^\dagger_{is} a_{is} + U \sum_i a^\dagger_{i\uparrow} a_{i\uparrow} a^\dagger_{i\downarrow} a_{i\downarrow} + t \sum_{i,s} (a^\dagger_{is} a_{i+1,s} + a^\dagger_{i+1,s} a_{is}) , \qquad (1.1)$$

with random single-site energies $\{\epsilon_i\}$. As a result of the randomness, the occupation of single sites at low temperatures is as follows[9]: Sites with energy $\epsilon > \mu$ are empty, those with energy $\mu - U < \epsilon < \mu$ are singly occupied and sites with energy $\epsilon < \mu - U$ are doubly occupied. Here μ is the chemical potential of the system. It was found that in both cases of $t \ll U \ll \sigma$ and $t \ll U \simeq \sigma$ the coupling constant J between the spins of two singly occupied sites separated by n intermediate doubly occupied or empty sites is given by

$$J(n) = D n^\alpha \beta^{2n} , \qquad (1.2)$$

with D, α, and $\beta < 1$ depending on the parameters of the Hamiltonian. Approximate analytic expressions are given for them in Ref. 10 [Eqs. (3.10) and (3.12)]. Furthermore it was found that at low temperatures and small transfer integrals and by ignoring interactions involving more than two spins, the Hamiltonian (1.1) reduces to an effective Hamiltonian

$$H_{\text{eff}} = \sum_i J_i \vec{S}_i \cdot \vec{S}_{i+1} , \qquad (1.3)$$

with J_i a positive random variable whose probability distribution $P(J)$ for $J \ll \sigma$ has the behavior

$$P(J) = \Gamma / (J/D)^{1-c} [\,|\ln(J/D)|\,]^{\alpha c} , \qquad (1.4)$$

where $c = p/2|\ln\beta|$, with p the probability for a site to be singly occupied. We consider p to be sufficiently smaller than unity that $\ln(1-p) \simeq -p$. The analytic formulas for Γ and D as well as for the probability distribution $P(J)$ for arbitrary J are given in Ref. 10. The number of spins involved in Hamiltonian (1.3) is $N_0 = pN$, with N the number of sites of the Hubbard Hamiltonian. From Eq. (1.4) we see that the main feature of $P(J)$ is a singularity at $J=0$, for $c<1$.

The quantum spin-$\frac{1}{2}$ Heisenberg antiferromagnet has not been solved analytically in one dimension for the periodic case, let alone for the disordered case with a complex distribution of exchange interaction. Accordingly in this paper we study various simple one-dimensional disordered model Hamiltonians for which exact solutions can be obtained. We are particularly interested in the low-temperature region, using Eq. (1.4) as the probability distribution of exchange. We find a simple interpretation of the main features of the susceptibility based on cluster arguments, which leads us to the conclusion that for sufficiently disordered systems, the spin-$\frac{1}{2}$ Heisenberg model is adequately approximated by the classical Heisenberg model in one dimension for all temperatures.

In Sec. II, we derive the behavior of the low-temperature, zero-field magnetic susceptibility of a one-dimensional disordered system from an argument based on the formation of clusters of spins. In Sec. III we derive the zero-field magnetic susceptibility for the following one-dimensional disordered models, using as the probability distribution of exchange the one given by Eq. (1.4): (a) quantum XY model; (b) quantum Ising model with parallel magnetic field; (c) quantum Ising model with perpendicular field; (d) classical Heisenberg model; (e) classical planar model; (f) classical Ising model; and (g) a semiclassical model, where the z component of the spin is treated quantum mechanically and the x and y components are considered components of a classical vector. Finally Sec. IV is devoted to discussion of our results and, in particular, the argument that the spin-$\frac{1}{2}$ Heisenberg model is well represented by the classical Heisenberg model.

II. CLUSTER APPROXIMATION

The coupling constant between spins separated by n intermediate sites is given by Eq. (1.2). At temperature T we consider the coupling to be strong if $J>kT$ and weak if $J<kT$. Therefore the minimum separation n_0 between two spins required in order to consider them weakly coupled is given by

$$D n_0^{\alpha} \beta^{2n_0} \simeq kT \,, \tag{2.1}$$

where n_0 is considered a continuous variable. From Eq. (2.1) we obtain

$$\alpha \ln n_0 + 2n_0 \ln\beta \simeq \ln(kT/D) \,. \tag{2.2}$$

For $KT \ll D$ the separation n_0 becomes large and in that case we can make the approximation

$$\alpha \ln n_0 + 2n_0 \ln\beta \simeq 2n_0 \ln\beta \,. \tag{2.3}$$

Combination of Eqs. (2.2) and (2.3) gives

$$n_0 \simeq \ln(kT/D)/2\ln\beta \,. \tag{2.4}$$

Thus the probability that the spins $\vec{S}_i$ and $\vec{S}_{i+1}$ are weakly coupled is given by

$$Q \simeq e^{-n_0 p} \simeq (kT/D)^{p/2|\ln\beta|} \,, \tag{2.5}$$

and consequently the probability for a strong coupling is

$$W \simeq 1 - (kT/D)^{p/2|\ln\beta|} \,. \tag{2.6}$$

A collection of spins which are coupled strongly among themselves but weakly to the rest is called a cluster. The probability for a given spin to have $n-1$ spins strongly coupled to it from the left and $m-1$ from the right is given by

$$W' = (1-W)^2 W^{m+n-2} \,. \tag{2.7}$$

Therefore the probability for a given spin to belong to a cluster of size k is

$$Q_k = k(1-W)^2 W^{k-1} \,. \tag{2.8}$$

The number of clusters of size k is then equal to

$$N_k = Np\,(1-W)^2 W^{k-1} \,, \tag{2.9}$$

with N the total number of sites of the Hubbard Hamiltonian (1.1).

The susceptibility can be considered as a sum of contributions from each cluster. To estimate the latter, we consider whether internal excitation of the clusters is important. Two effects must be examined. First, as the average size of a cluster increases, its minimum internal excitation energy decreases and may become smaller than kT. We study this effect by replacing the varying exchange coupling within the cluster by an appropriate average value. The average strength $\bar{J}$ of a strong coupling is given by

$$\bar{J} = \sum_{n=0}^{n_0} p(1-p)^n J(n) \,. \tag{2.10}$$

We are interested in estimating the order of magnitude of $\bar{J}$. For this purpose, it is sufficient to approximate $J(n)$ by $D\beta^{2n}$ since these two terms are the ones which dominate the behavior of $J(n)$, the term n^{α} being of minor significance. In that case Eq. (2.10) becomes

$$\bar{J} \simeq p[D - (kT)\beta^2(1-p)^{n_0+1}]/[1-\beta^2(1-p)] \,,$$

and for $kT \ll D$ we obtain

$$\bar{J} \simeq pD/[1-\beta^2(1-p)] \,.$$

Consequently $\bar{J}$ is of the order of D. The average size of a cluster is given by

$$\bar{n} = (1 - W)^{-1} = (D/kT)^{p/2|\ln\beta|}$$

For an infinite periodic chain the excitation spectrum is given by[11]

$$E(q) = \tfrac{1}{2}J\pi \, |\sin q| \,. \tag{2.11}$$

For a large but finite chain we assume that the excitation is approximately given by (2.11) plus the condition $q = (2\pi/N)n$, $n = 0, 1, \ldots, N-1$, with N the size of the chain. In that case the energy gap between the ground and the first excited state is given by

$$\Delta \simeq \tfrac{1}{2}J\pi 2\pi/N \propto J/N \,.$$

Therefore the energy gap existing in a cluster of average size $\bar{n}$, and whose spins are coupled with average strength $\bar{J}$, is

$$\Delta/kT \propto (\bar{J}/kT)/\bar{n} \simeq (D/kT)^{1-p/2|\ln\beta|} ,$$

and for $p/2|\ln\beta| < 1$ and $kT \ll D$ we obtain $\Delta \gg kT$.

The second effect is that some of the exchange couplings may not be significantly larger than kT. Let us evaluate the probability for this to happen. Equation (2.4) gives the minimum separation n_0 required for a coupling to be weak. The coupling in the case where the separation is $n_0 - 1$, this being the minimum coupling which can exist within a cluster, is given by

$$\ln(J_{\min}/D) \simeq 2\ln\beta(n_0 - 1) \,. \tag{2.12}$$

Combining Eqs. (2.12) and (2.4) we obtain

$$J_{\min} \simeq kT/\beta^2 , \tag{2.13}$$

and for $\beta \simeq 0.5$, $J_{\min} = 4kT$. Thus the minimum coupling in a cluster is approximately equal to $4kT$. The probability to have $J_{\min}$ inside a cluster is given by

$$R_1 = p(1-p)^{n_0-1}/[1-(1-p)^{n_0}], \tag{2.14}$$

and the probability for a coupling stronger than $J_{\min}$ to occur is

$$R_2 = [1-(1-p)^{n_0-1}]/[1-(1-p)^{n_0}]. \tag{2.15}$$

For $kT \ll D$ we have $n_0 \gg 1$ and thus $(1-p)^{n_0-1} \simeq 0$. This gives $R_1 \simeq 0$ and $R_2 \simeq 1$. That is, the probability that some of the exchange couplings are not significantly larger than kT goes to zero for $kT/D \to 0$. From the above analysis we conclude that internal excitation of the clusters is not important in determining the magnetic susceptibility.

Thus we can consider that the clusters which contribute significantly to the magnetic susceptibility are in their ground state and have a finite gap of size much greater than kT between ground and first excited state. The total spin of an odd cluster then will be equal to $\frac{1}{2}$, and the total spin of an even cluster will be zero because of the antiferromagnetic coupling. Thus only the odd clusters will contribute to the magnetic susceptibility. Furthermore, since the coupling between clusters is smaller than kT, we can assume to a first approximation that there is no coupling between odd clusters. This argument can be justified better as follows: the number N_{ev} of even clusters is given by

$$N_{\mathrm{ev}} = W N_{\mathrm{odd}} , \tag{2.16}$$

and for $kT \ll D$ we have $N_{\mathrm{ev}} \simeq N_{\mathrm{odd}}$. Since we consider our system to be random, on the average we can expect that between two odd clusters there will be an even one half the time. This further reduces the coupling between odd clusters making it a better approximation to consider it zero. Thus our system consists of free clusters each having a total spin of $\frac{1}{2}$ or zero, and the magnetic susceptibility is given by

$$\chi \simeq N_{\mathrm{odd}} g^2 \mu_B^2/4kT \,. \tag{2.17}$$

The total number of odd clusters is equal to

$$N_{\mathrm{odd}} = Np(1-W)/(1+W) , \tag{2.18}$$

and using (2.6) we obtain

$$N_{\mathrm{odd}} = Np \, \frac{(kT/D)^{p/2|\ln\beta|}}{2-(kT/D)^{p/2|\ln\beta|}} \,. \tag{2.19}$$

Using Eqs. (2.19) and (2.17) we obtain

$$\chi \simeq \frac{Npg^2\mu_B^2}{4D^c} \, \frac{1}{(kT)^{1-c}} \, \frac{1}{2-(kT/D)^c} , \tag{2.20}$$

where $c = p/2|\ln\beta|$. Thus the asymptotic behavior of Eq. (2.20) for $kT \ll D$ is given by

$$\chi \propto 1/T^{1-c} \,. \tag{2.21}$$

From Eq. (2.21) we see that the singularity in the probability distribution of J has as a result the introduction of a singularity of the same type in the magnetic susceptibility.

In what follows, it will be convenient to have, for comparison with (2.21) and with exact results for various models, the contribution to the susceptibility from isolated single spins, coupled weakly to the rest of the chain. The number of such clusters of size one is given by

$$N_1 = NpW(1-W)^2 \,.$$

Therefore their contribution to the magnetic susceptibility is, according to the cluster argument, for $kT \ll D$

$$\chi \propto 1/T^{1-2c} \,. \tag{2.22}$$

III. EXACT RESULTS

Here we examine the consequences of the probability distribution (1.4) for the magnetic susceptibility of some exactly soluble one-dimensional random models.

A. Quantum models

1. Random one-dimensional XY model

The Hamiltonian for a disordered one-dimensional XY model with the magnetic field placed along the z direction is given by

$$H=\sum J_i(S_i^+S_{i+1}^-+S_i^-S_{i+1}^+)+g\mu_B H\sum S_i^z, \qquad (3.1)$$

with $\vec{S}_i$ the spin operator. Smith[12] pointed out that the random XY model with $S=\frac{1}{2}$ is isomorphic to Dyson's[13] model of a linear chain of identical atoms coupled by random springs. In the case for which the probability distribution of the coupling constant is given by

$$P_n(J)=[2n^n/(n-1)!](J/J_0)^{2n-1}e^{-nJ^2/J_0^2},$$

$$n=1,2,\ldots .$$

Dyson's model can be solved exactly. The solution reveals that there is a singularity in the density of states at zero energy of the following form[2]:

$$n(E)\propto 1/E\,|\ln(E/J_0)|^3 . \qquad (3.2)$$

Therefore the susceptibility of this model will have for $T\to 0$ the behavior[2]

$$\chi(T)\propto 1/T\,|\ln(T/T_0)|^2 . \qquad (3.3)$$

That is, an infinitesimal amount of disorder produces a $1/T$ singularity in the magnetic susceptibility. In our case, where the probability distribution of J has a singularity at the origin, we have a greater number of spins weakly coupled. Therefore a strong $1/T$ singularity in the magnetic susceptibility will occur. This seems to contradict the result of the cluster argument according to which χ should have a singularity of the form $1/T^{1-c}$. We can, however, explain why the cluster argument cannot be invoked for this specific case as follows:

Application of the well-known canonical transformation[14]

$$S_l^+=C_l^+\exp\left(i\pi\sum_{j=1}^{l-1}C_j^+C_j\right),$$
$$S_l^-=(S_l^+)^+,\quad S_l^z=C_l^+C_l-\tfrac{1}{2}, \qquad (3.4)$$

which relates the spin operators associated with site l to Fermi operators C_r^+ and C_r associated with sites $r=1,2,\ldots,\ l-1,\ l$ reduces Hamiltonian (3.1) to that of spinless fermions with nearest-neighbor interaction and off-diagonal disorder only

$$H=\sum_i J_i(C_i^+C_{i+1}+C_{i+1}^+C_i). \qquad (3.5)$$

Theodorou and Cohen have shown in that case, that there exist strong amplitude fluctuations in the wave function of the state of the middle of the band.[15] These fluctuations are too large to allow decomposition of the spins clearly into separate clusters and this prevents us from applying the cluster argument. Therefore the $1/T$ behavior of the magnetic susceptibility of the random XY model is a special characteristic of this model which, however, does not hold in the case of the random Heisenberg model.

In what follows we will briefly outline why the cluster argument can be made for the case of the random antiferromagnetic Heisenberg model. Making the canonical transformation (3.4) and subtracting a constant term Hamiltonian (3.1) becomes

$$H=\frac{1}{2}\sum J_l(C_l^\dagger C_{l+1}+C_{l+1}^\dagger C_l)-\frac{1}{2}\sum(J_l+J_{l-1})C_l^\dagger C_l$$
$$+\sum J_l C_l^\dagger C_l C_{l+1}^\dagger C_{l+1} . \qquad (3.6)$$

We separate it into the following two parts:

$$H_0=\frac{1}{2}\sum J_l(C_l^\dagger C_{l+1}+C_{l+1}^\dagger C_l)-\frac{1}{2}\sum(J_l+J_{l-1})C_l^\dagger C_l$$

and

$$V=\sum J_l C_l^\dagger C_l C_{l+1}^\dagger C_{l+1} . \qquad (3.7)$$

H_0 is equivalent to a Hamiltonian of spinless fermions with nearest-neighbor interaction only, and with diagonal and off-diagonal randomness. According to a general theorem,[16-18] all states are localized in that case. Let us now examine the effect of V upon the localization. This term introduces an interaction between particles sitting on neighboring sites. Since $J>0$, they tend to avoid each other. Thus the presence of V will further increase the localization and reduces the effect of the fluctuations. The cluster argument can then once again be made, and any singularity in the density of states will be a consequence of a singularity in $P(J)$.

2. Random one-dimensional Ising model with parallel field

The Hamiltonian of this Model is given by

$$H=\tfrac{1}{4}\sum J_i\,\sigma_i^z\sigma_{i+1}^z+\tfrac{1}{2}g\mu_B H\sum\sigma_i^z , \qquad (3.8)$$

where the σ_i are the Pauli matrices.

The zero-field magnetic susceptibility for this model was calculated by Cabib and Mahanti[19] and was found equal to

$$\chi_\parallel^I/N_0=(g^2\mu_B^2/4kT)(1+\langle u\rangle)/(1-\langle u\rangle) , \qquad (3.9)$$

with

$$u=\tanh(-J/4kT)\ . \tag{3.10}$$

From this we get that if J is a positive random variable with a probability distribution having a singularity of the form $J^{-(1-c)}$ at the origin, the magnetic susceptibility will have the behavior

$$\chi_{\parallel}^{I}\propto 1/T^{1-c}\ . \tag{3.11}$$

This result is in agreement with the cluster argument.

3. Random Ising model with perpendicular field

The Hamiltonian of the model is given by

$$H=\frac{1}{2}\sum J_i\sigma_i^z\sigma_{i+1}^z+\tfrac{1}{2}g\mu_B H\sum\sigma_i^x\ . \tag{3.12}$$

The zero-field magnetic susceptibility of this model is given by[19]

$$\frac{\chi_{\perp}^{I}}{N_0}=\frac{g^2\mu_B^2}{4N_0kT}\sum_{i=1}^{N}\frac{(J_i/2kT)\tanh(J_i/2kT)-(J_{i-1}/2kT)\tanh(J_{i-1}/2kT)}{(J_i/2kT)^2-(J_{i-1}/2kT)^2}\ . \tag{3.13}$$

For large N_0 we can apply the central-limit theorem and obtain

$$\frac{\chi_{\perp}^{I}}{N_0}=\frac{g^2\mu_B^2}{4kT}\left\langle\frac{(J/2kT)\tanh(J/2kT)-(J'/2kT)\tanh(J'/2kT)}{(J/2kT)^2-(J'/2kT)^2}\right\rangle\ . \tag{3.14}$$

For J and J' independent positive random variables with probability distributions having a singularity of the form $J^{-(1-c)}$ at the origin, we learn by using Eq. (3.14) that the magnetic susceptibility has the behavior

$$\chi_{\perp}^{I}\propto 1/T^{1-2c}\ . \tag{3.15}$$

This result again seems to contradict the cluster argument, but this is not the case. We can indeed explain the behavior of $\chi_{\perp}^{I}$, given by Eq. (3.15), in terms of the cluster argument as follows: A cluster of size $n>1$, with the spins pointing in the z direction has energy of the order $-(n-1)\bar{J}$, where $\bar{J}$ is the average coupling between the spins. Rotation of the spins to the x direction will increase the energy of the cluster from $-(n-1)\bar{J}$ to zero. Therefore for $\bar{J}\gg kT$ the spins belonging to clusters of size $n>1$ will not contribute to $\chi_{\perp}^{I}$ since it is energetically unfavorable for them to rotate from the z to the x direction. The only clusters which will contribute to $\chi_{\perp}^{I}$ are clusters of size one. Their contributions will be proportional to N_1/T and using Eq. (2.22) we get $\chi_{\perp}^{I}\propto 1/T^{1-2c}$, a result identical to (3.15).

B. Classical models

In addition to the quantum models we consider the case of a chain consisting of randomly interacting ν-component classical spins. For $\nu=1,2,3$ the model reduces to Ising, planar, and Heisenberg, respectively.

1. Classical Heisenberg model

The Hamiltonian of a classical isotropic Heisenberg chain is given by

$$H=\sum J_i\,\vec{S}_i\cdot\vec{S}_{i+1}+g\mu_B H\sum S_i^z\ , \tag{3.16}$$

with $\vec{S}$ a classical vector of magnitude $[S(S+1)]^{1/2}$. For the case of a periodic chain the zero-field magnetic susceptibility was calculated by Fisher.[20] Fisher's method, with only minor modifications, can be applied for the case of the classical random chain. The zero-field magnetic susceptibility of the random chain is given by[21]

$$\chi=\frac{N_0g^2\mu_B^2S(S+1)}{3kT}\,\frac{1+\langle u\rangle}{1-\langle u\rangle}\ , \tag{3.17}$$

with

$$u=kT/JS(S+1)-\coth[JS(S+1)/kT]\ .$$

For $S=\frac{1}{2}$ the average value of u is given by

$$\langle u\rangle=-1+\langle 1+4kT/3J-\coth(3J/4kT)\rangle\ . \tag{3.18}$$

when $J\gg kT$,

$$1+4kT/3J-\coth(3J/4kT)\simeq 4kT/J\ ,$$

and therefore the region $J\gg kT$ will not contribute significantly to the average. In the case where $kT\ll\sigma$ the major contribution to the average will come from $J\ll\sigma$. Consequently we can use as $P(J)$ the asymptotic form given by (1.4). Furthermore we can approximate $\coth x$ by

$$\coth x\simeq\begin{cases}1/x+\frac{1}{3}x & \text{for } x\lesssim 1.5\ ,\\ 1 & \text{for } x>1.5\ .\end{cases} \tag{3.19}$$

Using these approximations we obtain

$$\langle u\rangle\simeq -1+\frac{\Gamma(kT)^cD^{1-c}}{[\,|\ln(kT/D)|\,]^{\alpha c}}\left(\frac{2^c}{c}+\frac{4}{3}\frac{1}{2^{1-c}(1-c)}-\frac{2^{1+c}}{4(1+c)}\right)\cdot \tag{3.20}$$

Thus the magnetic susceptibility of the chain for $kT \ll \sigma$ is given by

$$\chi \simeq \frac{\mu_B^2}{2} \frac{\Gamma N_0}{(kT/D)^{1-c}[\,|\ln(kT/D)|\,]^{\alpha c}} \times \left(\frac{2^c}{c} + \frac{4}{3}\frac{1}{2^{1-c}(1-c)} - \frac{2^{1+c}}{4(1+c)}\right). \quad (3.21)$$

Equation (3.21) shows that the magnetic susceptibility of a random classical Heisenberg chain has a singularity at $T=0$ identical to the singularity in the probability distribution of the coupling constant. Result (3.21) is in agreement with the cluster argument. This is because the cluster argument does not make a distinction between classical and quantum cases.

2. *Classical planar model*

The Hamiltonian of the system consisting of randomly coupled classical planar spins with the magnetic field placed in the spin plane is given by

$$H = \sum J_i(S_i^x S_{i+1}^x + S^y S_{i+1}^y) + g\mu_B H \sum S_i^x . \quad (3.22)$$

The zero-field magnetic susceptibility can be calculated in a way similar to that of the three-component classical model previously outlined. The result is

$$\chi_{\text{planar}} = (N_0 g^2 \mu_B^2 S^2/2kT)(1+\langle u\rangle)/(1-\langle u\rangle) , \quad (3.23)$$

with

$$u = -\frac{I_1(JS^2/kT)}{I_0(JS^2/kT)} .$$

S is the magnitude of the spins and I_0, I_1 are the modified Bessel functions of order zero and one. Using the asymptotic behavior of the modified Bessel functions we can prove that the contribution to $\langle u\rangle$ from $J \gg kT$ is unimportant. Thus for $kT \ll \sigma$ we can use the asymptotic probability distribution given by (1.4) and obtain

$$\langle u\rangle + 1 \propto (kT)^c . \quad (3.24)$$

The zero-field magnetic susceptibility will then have the behavior

$$\chi_{\text{planar}} \propto 1/T^{1-c} . \quad (3.25)$$

3. *Classical Ising model*

For classical spins lying along the z axis and with the magnetic field placed in the same direction the magnetic susceptibility of the system is identical to that of the quantum Ising model. Therefore for a probability distribution of the coupling constant given by Eq. (1.4) the magnetic susceptibility at low temperatures will have the behavior

$$\chi \propto 1/T^{1-c} . \quad (3.26)$$

C. Semiclassical model

In this model we consider that the z component of the spin can take the values $\pm\frac{1}{2}$, while the x and y components are components of a classical vector of magnitude $S^2 = S(S+1) - S_z^2 = \frac{1}{2}$ which rotates in the XY plane. We also assume that the components of the spin commute among themselves. The Hamiltonian of the system in the presence of a magnetic field which lies in the XZ plane and makes an angle θ with the z axis is given by

$$H = \sum J_i \vec{S}_i \cdot \vec{S}_{i+1} + g\mu_B H\left(\cos\theta \sum_i S_i^z + \sin\theta \sum_i S_i^x\right). \quad (3.27)$$

The magnetization then can be written as

$$\begin{aligned}\langle M\rangle = & -g\mu_B \cos\theta\, \mathrm{Tr}\left[\sum S_i^z \exp\left(-\beta\sum_i J_i S_i^z S_{i+1}^z - \beta g\mu_B H\cos\theta \sum_i S_{i+1}^z\right)\right] \\ & \times \left[\mathrm{Tr}\exp\left(-\beta\sum_i J_i S_i^z S_{i+1}^z - \beta g\mu_B H \cos\theta \sum_i S_i^z\right)\right]^{-1} \\ & - g\mu_B \sin\theta\, \mathrm{Tr}\left[\sum S_i^x \exp\left(-\beta\sum_i J_i(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) - \beta g\mu_B H \sin\theta \sum_i S_i^x\right)\right] \\ & \times \left[\mathrm{Tr}\exp\left(-\beta\sum_i J_i(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) - \beta g\mu_B H\sin\theta \sum_i S_i^x\right)\right]^{-1} . \end{aligned} \quad (3.28)$$

From Eq. (3.28) we obtain that the zero-field magnetic susceptibility is given by

$$\chi_{\text{semiclassical}} = \cos^2\theta\, \chi_{\parallel}^I + \sin^2\theta\, \chi_{\text{planar}} . \quad (3.29)$$

Averaging over all possible orientations of the magnetic field we obtain

$$\chi_{\text{semiclassical}} = \tfrac{1}{3}\chi_{\parallel}^I + \tfrac{2}{3}\chi_{\text{planar}} , \quad (3.30)$$

where $\chi_{\parallel}^I$ is given by Eq. (3.9) and χ_{planar} by Eq. (3.23). Making use of Eqs. (3.11) and (3.25) we get that the low-temperature behavior of $\chi_{\text{semiclassical}}$ is

$$\chi_{\text{semiclassical}} \propto 1/T^{1-c} . \tag{3.31}$$

IV. DISCUSSION

In this paper we were confronted with the difficult problem of finding the magnetic susceptibility of the disordered one-dimensional Heisenberg antiferromagnet for a probability distribution of the coupling constant which has a singularity at the origin. Since we were unable to find an analytical solution, we developed instead a cluster argument which gives the low-temperature behavior of the magnetic susceptibility of the above mentioned system. The main feature of the susceptibility is that it diverges at $T=0$ and its divergence is identical to that of the probability distribution of the coupling constant at $J=0$. Since the cluster argument makes no distinction between classical and quantum cases, this means that the magnetic susceptibility for both the classical and quantum disordered models, has the same low-temperature behavior. In order to test the correctness of the cluster argument we examined the susceptibility of various exactly soluble disordered models always using as the probability distribution of exchange the one given by Eq. (1.4). Our examination proved that indeed the behavior of the low-temperature magnetic susceptibility for most of these exactly soluble models was identical to that provided by the cluster argument. Two quantum models, namely the Ising model with perpendicular magnetic field and the XY model raised doubts as to the validity of the argument. We were able to resolve them by proving that the first case had the special characteristic that only clusters of size one were contributing to the magnetic susceptibility and thus we should apply our cluster argument only for them, while for the XY case the strong fluctuations of the state at the middle of the band prohibited the application of a cluster argument. Furthermore we gave strong evidence that for the case of the one-dimensional quantum disordered Heisenberg model, which is of our main interest, the cluster argument can be applied.

From the arguments thus far, however, we have been able to obtain only the low-temperature behavior of the magnetic susceptibility of the quantum Heisenberg model. Our main purpose in analyzing the susceptibility of the Heisenberg model is the application of our findings to describe the magnetic properties of a real material. For this purpose the knowledge of only the behavior of the susceptibility at low temperatures is not enough; we need an analytic formula holding throughout the entire experimental temperature range in order to undertake a systematic fitting of the data. Since, as we mentioned above, we cannot obtain an analytic expression for the susceptibility of the quantum Heisenberg model, our next task is to find another model to take its place which must fulfill the following requirements: (a) to be analytically soluble; (b) the behavior of its low-temperature susceptibility to be identical to that derived for the quantum Heisenberg model from the cluster argument; and (c) to be a good approximation to the quantum-mechanical model in the whole temperature region. Our choice for such a model is the one-dimensional classical Heisenberg model. The low-temperature behavior of its susceptibility agrees with the results of the cluster argument, and it is analytically soluble. In order to test how close it is to the quantum Heisenberg model we must have an approximate formula for the latter. Since numerical results are available for the periodic quantum Heisenberg model, we thought that if we can make a connection between the susceptibility formulas for the periodic and disordered quantum models, we can then succeed in obtaining an approximate formula for the disordered Heisenberg model. The best way to make such a connection is to look at the exactly soluble models. For the periodic Ising as well as for all the periodic classical models, the susceptibility has the general form

$$\chi_{\text{per}} = \chi_0 (1+u_{\text{per}})/(1-u_{\text{per}}) , \tag{4.1}$$

with

$$\chi_0 = N_0 \mu_B^2 g^2/4kT, \quad u_{\text{per}} = -\tanh(J/4kT) ,$$
$$I_1(J/2kT)/I_0(J/2kT), \quad 4kT/3J - \coth(3J/4kT)$$

for the Ising, classical planar, and classical Heisenberg, respectively. For the disordered models, formula (4.1) becomes

$$\chi_{\text{dis}} = \chi_0 (1+\langle u_{\text{per}}\rangle)/(1-\langle u_{\text{per}}\rangle) , \tag{4.2}$$

with u_{per} the same as before. Therefore, the only difference is to replace u by its mean value. Equation (4.2) can also be written

$$\chi = \chi_0 \frac{1+\langle(\chi_{\text{per}}-\chi_0)/(\chi_{\text{per}}+\chi_0)\rangle}{1-\langle(\chi_{\text{per}}-\chi_0)/(\chi_{\text{per}}+\chi_0)\rangle} . \tag{4.3}$$

Thus Eq. (4.3), using as χ_{per} the susceptibility of the quantum Heisenberg model as calculated by Bonner and Fisher,[22] provides us with an approximate formula for the disordered quantum Heisenberg model.

In a previous paper[10] we calculated the probability distribution of the coupling constant for the values $t=0.055$ eV, $U=0.130$ eV, and $\sigma=0.136$ eV, where t, σ, and U are the parameters of the Hub-

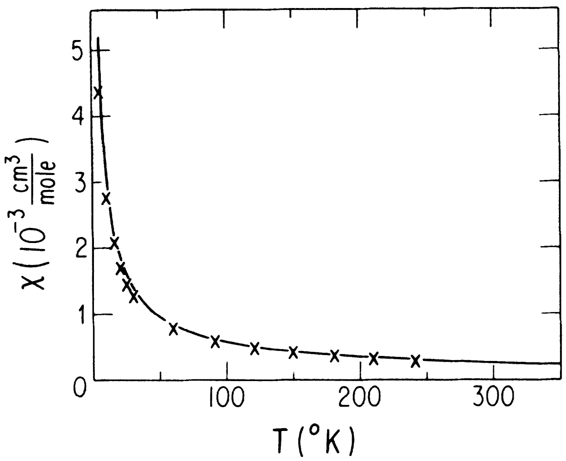

FIG. 1. Magnetic susceptibility of (a) the classical Heisenberg model (solid line); and (b) the approximate quantum model (×), given by Eq. (4.3), where the probability distribution of the coupling constant for both cases is given by Eq. (1.4) with $t = 0.055$ eV, $U = 0.130$ eV, and $\sigma = 0.136$ eV.

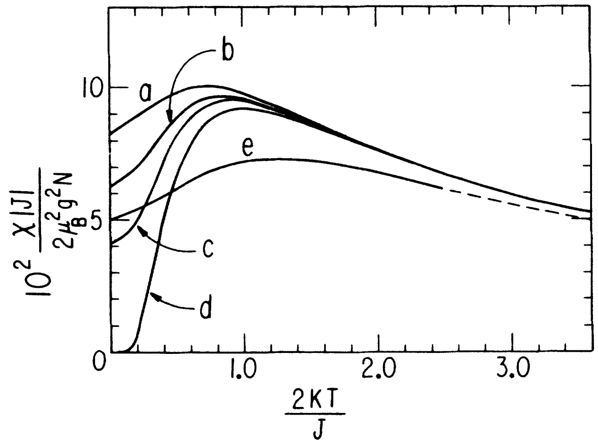

FIG. 2. Susceptibility vs temperature for periodic one-dimensional antiferromagnetic models: (a) classical Heisenberg; (b) classical planar; (c) semiclassical; (d) Ising; and (e) quantum Heisenberg.

bard Hamiltonian (Eq. 1.1). Using this probability distribution we evaluated the magnetic susceptibility for the classical Heisenberg model and the approximate quantum one. The results of our calculations are shown in Fig. 1. The results of our calculations are shown in Fig. 1. We note that there is a difference of $\simeq 11\%$ between the two models at $T = 5$ °K and this difference remains practically the same as the temperature increases. This indicates that it is a rather good approximation to represent the unknown quantum Heisenberg model with the classical one for this particular case where we are dealing with a sufficiently disordered system. Indeed, one can argue that the disordered classical Heisenberg susceptibility bounds the disordered spin-$\frac{1}{2}$ Heisenberg susceptibility from above because the zero-point motion of the spins reduces the susceptibility in the latter case. Similarly, one can argue that the use u_{per}, calculated by Bonner and Fisher,[22] bounds the disordered spin-$\frac{1}{2}$ Heisenberg susceptibility from below because the use of u_{per} overemphasizes this zero-point effect, underemphasizing the disruptive effect of disorder on the propagation of spin deviations. The percentage of error in using the classical Heisenberg model is therefore less than 11% throughout the entire temperature range, as the temperature dependence must be the same A/T^γ for all three cases below 5 °K. For comparison we have calculated the magnetic susceptibility of various exactly soluble spin-$\frac{1}{2}$ periodic models. The results of our calculations are shown in Fig. 2. In the same figure the results of calculations of Bonner and Fisher for the quantum spin-$\frac{1}{2}$ case are also shown. These calculations show that the susceptibility for the quantum Heisenberg model starts to deviate significantly from the classical models at temperatures $kT < J$. Also the quantum susceptibility is always smaller than the classical, due to zero-point motion. We note that Fisher has already given a comparison of the classical and quantum Heisenberg models.[20] However the classical model he used was one in which $\vec{S}/S$ was treated as a classical unit vector, whereas we have used $\vec{S}/[S(S+1)]^{1/2}$. The latter normalization is more accurate for small spin. In the particular case of interest to us, $S = \frac{1}{2}$, the temperature scale used by Fisher for the classical case is contradicted by a factor of 3 relative to the quantum case and to our classical case. Our choice of $\vec{S}/[S(S+1)]^{1/2}$ leads to an asymptotic approach of the susceptibilities of the two models at high temperatures.

Turning now to the question of the appropriate range of values of the parameters U, σ, and t, all of the previous analysis of the susceptibility of the disordered Hubbard model was for the case $U \lesssim \sigma$, for which we found that the susceptibility has a singularity at $T = 0$. Let us briefly examine the case where U is considerably larger than σ. According to the analysis of Ref. 10, in that case all sites are singly occupied and the exchange coupling between two nearest-neighboring spins has a minimum value J_{min} for $J < J_{min}$. For the case of the disordered classical Heisenberg model it can be easily derived that a singularity in the susceptibility at $T = 0$ is the consequence of a singularity in the probability distribution of the coupling constant at $J = 0$. This must also be true for the susceptibility of the spin-$\frac{1}{2}$ Heisenberg model, since as we argued before the latter is bounded from above by the classical Heisenberg susceptibility. For the case where U is considerably larger than σ, the probability distribution of the coupling constant is not singular at $J=0$, therefore the magnetic susceptibility of the system will have a finite value at $T=0$. The above analysis also casts strong doubts on the argument of Bulaevskii *et al.*[2] that a nonsingular probability distribution of the coupling constant can produce a singularity in the magnetic susceptibility of the quantum Heisenberg model with $S=\frac{1}{2}$. They justified their argument by reference to the XY model, which as we argued in Sec. III is different in that regard from the Heisenberg model.

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