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Random Antiferromagnetic Chain

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The quantum spin-$\frac{1}{2}$ Heisenberg antiferromagnet in one dimension with randomly distributed coupling constants is solved approximately. Ground-state energies and low-temperature properties are obtained for several distributions of coupling constants (including both singular and nonsingular distributions). Power-law temperature dependence in specific heat and in susceptibility are found for all distributions studied.

The random one-dimensional Heisenberg antiferromagnet with nearest-neighbor interactions is defined by the Hamiltonian

$$H=\sum_{r=1}^{L} K_r \vec{S}_r\cdot\vec{S}_{r+1} \tag{1}$$

with $L\to\infty$. Here $\vec{S}_r$ are taken to be spin-$\frac{1}{2}$ operators. The coupling constants K_r are different for different r. Their values are distributed randomly following a certain probability $P(K)$, $0<K<J$. The random antiferromagnetic chain does not have the translational invariance of the uniform system (where all K_r's are equal). Consequently, it does not have the spin-wave spectrum of the uniform case, which is difficult enough to have defied analytic solution. Thus, even an approximate solution becomes difficult for the random antiferromagnet, not to mention an exact analytic or numerical solution.

Earlier theoretical investigations were motivated by experimental interest. There are systems which can be modeled by (1). These include poly(metal phosphinates)[1] and $(CD_3)_4NMn_xCu_{1-x}Cl_3$ (TMMC: Cu).[2] More well known are the organic charge-transfer compounds N-methyl-phenazinium tetracyanoquinodimethanide (NMP-TCNQ), quinolinium tetracyanoquinodimethanide $[Qn(TCNQ)_2]$, etc.[3,4] Bulaevskii *et al.*[3] transformed the problem to a Fermi-gas model. In order to explain the singular temperature dependence of the susceptibility of TCNQ, they assumed singular energy dependence of the Fermion density of states even if $P(K)$ is not singular, without theoretical justification. Their results were criticized by Theodorou and Cohen,[4] who argued that only singular $P(K)$ can give rise to a singular susceptibility. A cluster approximation was devised by Theodorou and Cohen to study the cases of singular power-law distributions $P(K)\propto K^{c-1}$, $0<c<1$. Singular temperature dependence in the specific heat C and the magnetic susceptibility χ were found[4,5]:

$$\begin{aligned} &C\propto T^{c},\\ &\chi\propto T^{c-1}. \end{aligned} \tag{2}$$

TABLE I. Ground-state energies for assorted distributions $P(K)$, $0<K<1$.

Label		-Energy/spin
2	1	0.246
1	$2\theta(K-1/2)$	0.341
3	$1.58e^{-K}$	0.210
	$1.34e^{-K^2}$	0.213
	$(2/3)K^{-1/3}$	0.208
4	$(1/2)K^{-1/2}$	0.178
5	$(1/3)K^{-2/3}$	0.142
6	$(1/5)K^{-4/5}$	0.103

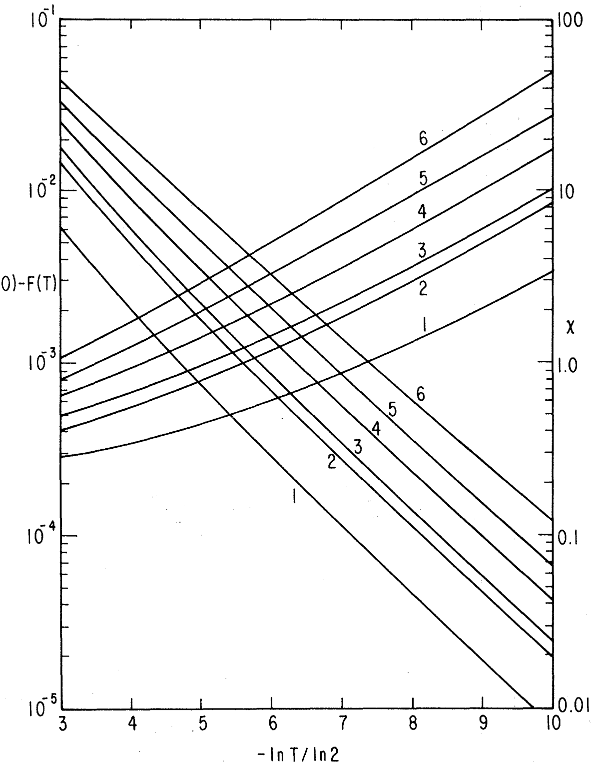

FIG. 1. Free energies (with ground-state energy subtracted) and zero-field susceptibilities for assorted distributions. The labels 1 through 6 denote the distributions given in Table I.

In this Letter we summarize an approximate solution to the problem for arbitrary $P(K)$. This approximation allows at least semiquantitative evaluations of physical properties as well as a simple qualitative picture. The full details are available elsewhere.[6]

We have calculated the ground-state energy for a set of $P(K)$ (see Table I), and the low-temperature free energy, specific heat, and susceptibility (see Fig. 1). The most important feature we find is the approximate power-law dependence on T:

$$C \propto T^{\gamma_c}, \qquad \chi \propto T^{\gamma_s - 1}. \tag{3}$$

The exponents γ_c and γ_s are slowly decreasing functions of $\ln(1/T)$ (see Fig. 2). Their values depend on $P(K)$. Such power-law T dependence is a *universal feature, i.e., it holds for nonsingular as well as for singular P(K).* Further discussion of these results will follow the description of the method of calculation.

Our approximation is based on successive elimination transformations, each of which eliminates a pair of neighboring spins with the maximum coupling J. Such a transformation is defined as follows. Consider the pair of spins interacting with coupling J. Call them $\vec{S}_1$ and $\vec{S}_2$. Let $\vec{S}_1'$ be the other spin interacting with $\vec{S}_1$ with coupling K_1 and $\vec{S}_2'$ be the other spin interacting with $\vec{S}_2$ with coupling K_2. The part of the Hamiltonian involving $\vec{S}_1$ and $\vec{S}_2$ is

$$H_{12} = J\vec{S}_1\cdot\vec{S}_2 + K_1\vec{S}_1'\cdot\vec{S}_1 + K_2\vec{S}_2\cdot\vec{S}_2'. \tag{4}$$

Now we calculate the free energy for H_{12} keeping $\vec{S}_1'$ and $\vec{S}_2'$ as fixed vectors:

$$H_{12}' \equiv -T\ln\operatorname{Tr}\exp(-H_{12}/T) = F_{12}' + K'\vec{S}_1'\cdot\vec{S}_2', \tag{5}$$

where the trace is taken over $\vec{S}_1$ and $\vec{S}_2$. We find

$$F_{12}' = -\tfrac{3}{4}J - T\ln(1+3e^{-J/T}) - \frac{3}{16J}(K_1^2+K_2^2)V\left(\frac{J}{T}\right), \tag{6}$$
$$K' = K_1K_2W(J/T)/2J.$$

The functions V and W are defined by

$$V(y) = [1-e^{-y}(1-y)]/(1+3e^{-y}), \qquad W(y) = [1-e^{-y}(1+y)]/(1+3e^{-y}). \tag{7}$$

These results are correct only up to the second order in K_1 and K_2. Since J is the maximum, we expect K_1 and K_2 to be quite a bit smaller than J and the second-order calculation should be a good approximation if $P(K)$ is not concentrated near $K=J$. Now we remove H_{12} from H and add back $K'\vec{S}_1'\cdot\vec{S}_2'$ to obtain the new Hamiltonian H'. This completes an elimination transformation. $\vec{S}_1$ and $\vec{S}_2$ have been eliminated. The free energy $F(H')$ for the system described by H' is related to the old free energy $F(H)$ by

$$F(H) = F(H') + F_{12}'. \tag{8}$$

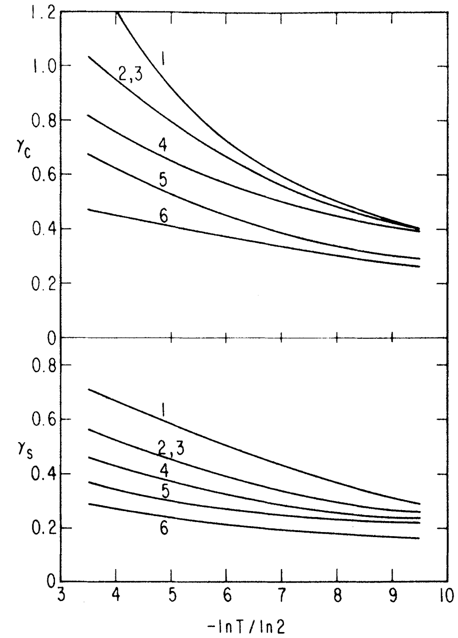

FIG. 2. The specific heat exponent γ_c and the susceptibility exponent γ_s [see (3)] for assorted distributions. γ_c and γ_s are obtained by respectively differentiating the free energy and the susceptibility. The labels are the same as in Fig. 1.

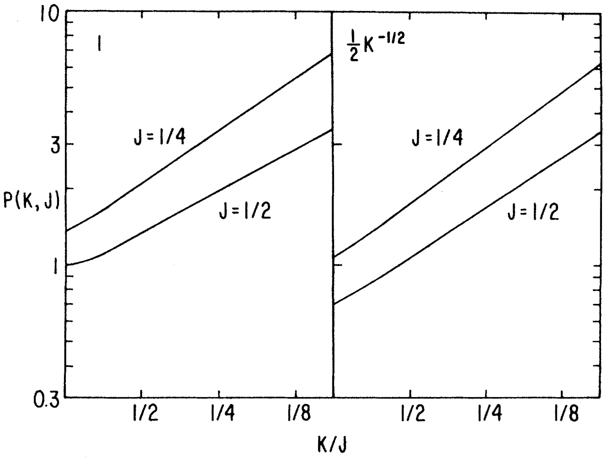

FIG. 3. Transformed distributions $P(K,J)$ for $P(K,1) = 1$, and $\frac{1}{2}K^{-1/2}$.

Of crucial importance is that the elimination transformation generates a new coupling K' between the two neighbors of the eliminated pair. Successive transformations not only lower J but also modify the form of $P(K)$. The modification of P is determined by solving the equation

$$-\frac{\partial}{\partial J}P(K,J)=P(J,J)\int_0^J dK_1\,dK_2\,P(K_1,J)P(K_2,J)\times\delta(K-K_1K_2W(J/T)/2J), \qquad (9)$$

which follows from (6). $P(K,J)$ is the probability distribution when the maximum coupling is lowered to J. The free energy can readily be obtained using (8) and P.

Since $W\leq 1$ [see (7)], we see that, as J is lowered, $P(K,J)$ increases sharply for small K, i.e., very weak couplings are generated. One can see this qualitatively by setting $P=1$ in the integral in (9), and finding a term $\ln(1/K)$, which indicates the building up of a negative power in K. In fact, calculations show that an approximate power-law form is quickly approached:

$$P(K,J)\sim K^{\alpha-1} \qquad (10)$$

as J is lowered. The exponent $\alpha=\alpha(\ln(1/J))$ is a slowly decreasing function of $\ln(1/J)$. One obtains from (9) an approximate equation

$$d\alpha/d\ln(1/J)=-\alpha^2(2/W)^\alpha[1+O(\alpha^2)] \qquad (11)$$

valid for small α. This approach to a power law is a universal feature shared by all the $P(K)$ we have studied, and accounts for the universal low-temperature behavior (3).

For the power law (3) to be valid, T has to be low enough so that $P(K,J)$ has reached a power law (10) for $J\sim T$. For the initially non-power-law $P(K)$ which we have studied, $P(K,J)$ reaches a power law readily before $J\sim\frac{1}{4}$ from the initial $J=1$. The step distribution $\theta(K-\frac{1}{2})$, which is zero for $K<\frac{1}{2}$, takes a smaller J.

Exact analytic solution to (9) is difficult but numerical solution of (9) is straightforward. Figure 3 exemplifies the approach of $P(K,J)$ to a power law. The elimination transformations can also be carried out without going through (9). One can generate a few thousand values of K_r in a computer calculation following a given $P(K)$ and numerically carry out the transformations one by one. When an external magnetic field is present, the elimination transformation becomes much more complicated than (6). A transformation will also generate a random magnetic field and Ising-type interactions.[6] The direct numerical transformation without use of an integral equation is then more effective for obtaining thermal and magnetic properties. The numerical accuracy of the results in Table I and Figs. 1, 2, and 3 is ~5%. Since $P(K,J)$ is singular, better accuracy in solving (9) requires better programing. Larger samples are needed for better statistics in the direct elimination method. No special difficulty is expected to improve the accuracy when needed.

There are some difficult unanswered questions about the accuracy of the transformation formulas (5) and (6) themselves. Note that improving them means keeping not only higher-order terms but also more complicated forms of interactions such as next-nearest neighbor and longer-range ones. As a rudimentary check, we solved the four-spin problem defined by (4) with $K_1 = K_2$ exactly and by the elimination process. The agreement is very good if $\frac{1}{2}J \gtrsim K$. One thus expects good answers if $P(K)$ is well spread out for $K \sim J$, i.e., not concentrated near the maximum value. The transformation was applied to a uniform model (i.e., all K_r's are equal) to eliminate two of every three spins. The ground-state energy so obtained was surprisingly good.[6]

In closing, we note that if $P(K)$ is nonsingular, the power-law T dependence (3) is absent for the *classical* model,[1,7] which is defined by replacing the operators $\vec{S}_r$ in (1) by classical unit vectors. The classical susceptibility $\chi_{cl} \sim P(0) \ln(1/T)$ is only weakly divergent for $T \to 0$. [For a singular $P(K) \sim K^{c-1}$, however, $\chi_{cl} \sim T^{c-1}$ (see Ref. 5).] We are aware of the belief in the literature that χ_{cl} should always be an upper bound for the quantum mechanical χ. This belief is, however, incorrect, as is easily checked by explicitly calculating χ and χ_{cl} for the four-spin system with the Hamiltonian H_{12} in Eq. (4). One finds that when $K_1K_2/2TJ$ and T/J are small, $\chi > \chi_{cl}$. The physical reason is that the quantum mechanical states are discrete. For small T, $\vec{S}_1$ and $\vec{S}_2$ are "frozen" in a singlet state and $\vec{S}_1'$ and $\vec{S}_2'$ become essentially free. This does not happen for the classical case where the excitation energy has a continuous distribution.

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Sputter-Induced Subsurface Segregation in a Cu-Ni Alloy

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Measurements of the surface composition of a Cu-Ni alloy using Auger electrons with different average escape depths reveal sputter-induced subsurface compositional changes at elevated temperatures which are larger than those produced directly at the surface by preferential sputtering. The compositional gradients which are produced extend to significantly greater depths than have been considered previously.

Sputtering with low-energy ions is routinely used to produce clean substrates for surface-analysis studies and, in conjunction with many different techniques, to profile the chemical composition of multicomponent alloys beneath an exposed surface.[1] Ion sputtering at elevated temperatures will also play an important role in determining the plasma contaminate level for advanced fusion devices.[2] An understanding of compositional changes induced by ion sputtering is thus of quite general interest and importance. In this Letter, we report surface and subsurface compositional changes induced in a Cu-Ni (40-at.% Ni) alloy during ion sputtering at temperatures between 50 and 600 °C.

Surface compositional changes during ion sputtering near room temperature have been observed in many alloy systems, an effect which is called preferential sputtering. Sputtering of Cu-Ni alloys at room temperature is known to preferentially remove Cu to a degree which is consistent with the sputtering yield ratios of pure Cu and Ni.[3,4] However, only very limited information exists on elevated-temperature sputtering effects, and no direct experimental information is available on subsurface changes[5] at elevated tempera-