

GREEN'S-FUNCTION THEORY OF A DISORDERED HEISENBERG FERROMAGNET

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Magnetic properties are calculated for a spin one-half Heisenberg ferromagnet in which the exchange interactions deviate randomly in strength from the mean interaction. Disorder produces a low-energy peak and a high-energy tail in the density of spin-wave states for a simple-cubic geometry. The ferromagnetic Curie temperature is shown to decrease linearly with disorder; the temperature-dependent magnetization is not otherwise markedly altered.

The phenomenon of ferromagnetism in amorphous solids has been observed experimentally in several systems¹ and has been the subject of some theoretical analysis.²⁻⁵ Substitutional impurities in crystalline ferromagnets have received attention,^{3,4} and a molecular-field treatment of amorphous ferromagnetism has been given by Handrich.⁵ This Letter deals with the effects of distributed disorder on the properties of a spin one-half Heisenberg ferromagnet. The disorder is introduced by allowing the exchange interactions between spins to deviate randomly in strength from the mean interaction. The distribution of exchange integrals can, to the order of this calculation, be characterized by two parameters: the mean interaction (for each separation on the lattice) and the mean square deviation from the mean. Temperature-dependent double-time Green's functions have been used to determine the density of spin-wave states, the ferromagnetic Curie temperature, and the temperature-dependent magnetization as functions

of disorder.

A spin one-half Heisenberg ferromagnet in zero magnetic field can be represented by a Hamiltonian of the form⁶

$$\mathcal{H} = 2 \sum_{f,g} J(f,g) [n_f - b_f^\dagger b_g - n_g], \quad (1)$$

where the Pauli spin operators (b_f, b_f^\dagger) obey the commutation relations

$$[b_f, b_g^\dagger] = (1 - 2n_g) \delta_{fg}, \quad (2)$$

and

$$n_g = b_g^\dagger b_g \quad (3)$$

is the spin deviation operator for the g th site. $J(f,g)$ is the (positive, ferromagnetic) exchange interaction between the spins at lattice sites f and g . For a perfect crystal, $J(f,g)$ would depend only on the displacement ($f-g$); in the present treatment, with distributed disorder, the translational symmetry is absent.

Following Zubarev⁶ we define the double-time temperature-dependent Green's function, and its time Fourier transform, by

$$G_{f,g}(t-t') = \langle\langle b_f(t); b_g^\dagger(t') \rangle\rangle = \int_{-\infty}^{\infty} G_{f,g}(\omega) \exp[-i\omega(t-t')] d\omega. \quad (4)$$

The equation of motion for G introduces high-order Green's functions; we decouple the hierarchy of equations by using the approximation^{6,7}

$$\langle\langle n_h(t) b_g(t); b_f^\dagger(t') \rangle\rangle \simeq \frac{1}{2}(1-\sigma) \langle\langle b_g(t); b_f^\dagger(t') \rangle\rangle, \quad (5)$$

where the magnetization

$$\sigma = 1 - 2\langle n \rangle \quad (6)$$

is treated as site-independent. σ is to be determined self-consistently from the so-called "imaginary

part" of G ,

$$\text{Im}G \equiv \lim_{\epsilon \rightarrow 0^+} i[G(\omega + i\epsilon) - G(\omega - i\epsilon)], \quad (7)$$

according to the equation⁶

$$\frac{1}{2}(1 - \sigma) = \int_{-\infty}^{\infty} d\omega N^{-1} \text{Tr} \text{Im}G(\omega) [\exp(\beta\omega) - 1]^{-1}. \quad (8)$$

The equation of motion then takes the form

$$2\pi \sum_h [\delta_{gh} \{(\omega/\sigma) - 2\sum_{h_1} J(g, h_1)\} + 2J(g, h)] G_{hf}(\omega) = \delta_{gf}, \quad (9a)$$

or in simpler notation,

$$AG = 1. \quad (9b)$$

From Eq. (9), the diagonalized form of G for a specific disordered system must be

$$G_{\alpha\beta}(\omega) = \frac{1}{2\pi} \frac{\delta_{\alpha\beta}}{(\omega/\sigma - B_{\alpha})}, \quad (10)$$

where the quantities B_{α} are the eigenvalues of the matrix B whose configuration space matrix elements are

$$B_{gh} = 2\sum_{h_1} J(g, h_1) \delta_{gh} - 2J(g, h). \quad (11)$$

The spin-wave energies are proportional to the magnetization σ . The number of spin-wave states per unit energy may be obtained from the imaginary part of G :

$$\text{Im}G_{\alpha\beta}(\omega) = \delta_{\alpha\beta} \delta(\omega/\sigma - B_{\alpha}), \quad (12)$$

so that

$$N^{-1} \text{Tr} \text{Im}G = N^{-1} \sum_{\alpha} \delta(\omega/\sigma - B_{\alpha}) \equiv g(\omega/\sigma), \quad (13)$$

the "density of spin-wave states." It is easily seen that $g(x)$ is normalized,

$$\int g(x) dx = 1, \quad (14a)$$

and that its centroid,

$$\int xg(x) dx = N^{-1} \text{Tr}B, \quad (14b)$$

depends only on the average coupling. Moreover, the Curie temperature, at which the magnetization vanishes [see Eq. (8)], is inversely proportional to

$$\frac{\int g(x) dx}{x} = \frac{1}{N} \sum_{\alpha} \frac{1}{B_{\alpha}} = -\frac{2\pi}{N} \text{Tr}G(0). \quad (14c)$$

For a perfect crystal the solution to Eq. (9) can be obtained by a Fourier transform to the reciprocal lattice space. In a disordered system we must perform a suitable ensemble average over systems with similar disorder to obtain an expression for the ensemble-averaged Green's function $\langle G_{gf} \rangle$. From $\langle G \rangle$ we can derive a density

of spin-wave states, Curie temperature, and magnetization for the disordered system.

To obtain $\langle G \rangle$ we decompose the A matrix into two parts

$$A = \Gamma^{-1} - \Delta, \quad (15)$$

where Γ depends only on the mean exchange interactions and is the Green's function for a "corresponding perfect crystal," to be defined more exactly below, while Δ contains all deviations of the exchange interactions from their mean values. It is then easy to show that

$$G = \Gamma + \Gamma \Delta G, \quad (16)$$

so that the ensemble-averaged G obeys a "Dyson equation"

$$\langle G \rangle = \Gamma + \Gamma \Sigma \langle G \rangle, \quad (17)$$

and can be expressed in terms of the "self-energy" Σ as

$$\langle G \rangle = (1 - \Gamma \Sigma)^{-1} \Gamma. \quad (18)$$

The decomposition of A is chosen to make the ensemble average of the fluctuation matrix $\langle \Delta \rangle$ vanish. Thus, we write the exchange integral $J(f, g)$ as

$$J(f, g) = J^0(f-g) + j(f, g), \quad (19)$$

where $J^0(f-g)$ is the ensemble average of the exchange coupling between spins at sites whose vector separation is $(f-g)$ and $j(f, g)$ is the deviation from this mean value. Γ is then the Green's function for a perfect crystal with couplings $J^0(f-g)$.

The distributed disorder we consider is assumed to be such that deviations from the mean of couplings between different pairs of sites are uncorrelated—an assumption which would be inappropriate for describing, e.g., substituted magnetic impurities. For simplicity we also assume that fluctuations are symmetric about the mean, so that ensemble averages of odd powers of Δ all vanish. We can then approximate the

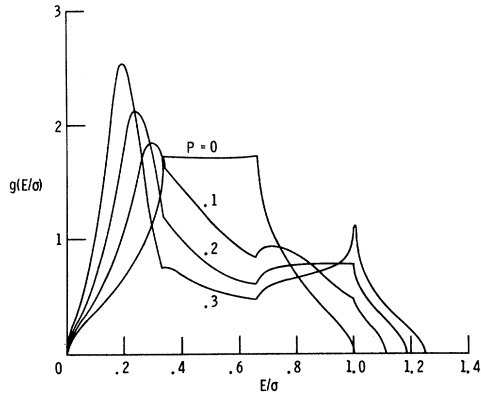


FIG. 1. Density of spin-wave states. Number of spin-wave energies at E vs E for a simple-cubic lattice with varying disorder. The energies are proportional to the magnetization σ in this approximation so that E/σ is temperature independent. The disorder parameter P is proportional to the mean-square fluctuation of the couplings about the average coupling strength.

self-energy as

$$\Sigma \approx \Sigma^0 \equiv \langle \Delta \Gamma \Delta \rangle. \quad (20)$$

This is equivalent to replacing $\langle (\Gamma \Delta)^{2n} \rangle$ by $\langle (\Gamma \Delta)^2 \rangle^n$ in the ensemble average of the iterated solution to Eq. (8). Since ensemble-averaged quantities are translationally invariant, Γ , $\langle G \rangle$, and Σ^0 are all diagonalized by a space Fourier transform, and Eq. (18) becomes

$$\langle G \rangle_q = [(\Gamma_q)^{-1} - \Sigma_q^0]^{-1}. \quad (21)$$

These results have been applied to isotropic systems with nearest-neighbor interactions. When all spin sites are equivalent (as in cubic lattices, for example), the two parameters are the average nearest-neighbor exchange, J_0 , and the mean-square deviation j^2 . The latter is introduced through the ensemble averaging:

$$\langle j(f, g)j(h, k) \rangle = j^2 [\delta_{fh} \delta_{gk} + \delta_{fk} \delta_{gh}]. \quad (22)$$

For these systems the dependence of the Curie temperature on disorder is found to be linear in the mean-square deviation:

$$T_C = T_0 [1 - (2/z)j^2/J_0^2], \quad (23)$$

where z is the number of nearest neighbors for each site.

For the case of a simple cubic lattice, the averaged Green's function, spin-wave state density $g(x)$, and magnetization $\sigma(T)$ have been calculated, using Eqs. (8), (13), (20), and (21). The results are illustrated in Figs. 1 and 2. The disorder parameter P with which the curves are

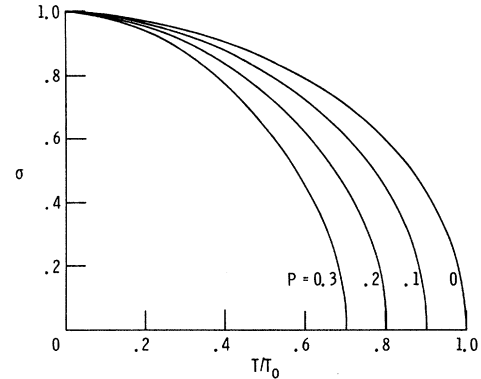


FIG. 2. Magnetization of a disordered ferromagnet. The magnetization σ is plotted versus temperature T for various disorder parameters P . T_0 is the ferromagnetic Curie temperature for a system with no disorder. The Curie-point depression is proportional to P .

labeled is simply

$$P = j^2/3J_0^2, \quad (24)$$

a measure of the width of the distribution of interactions.

The density of states is seen to change markedly even for rather small disorder. The striking features are (1) the appearance of states at energies above the band-edge for the crystal, (2) the emergence of a low-energy peak in the state density, and (3) the decrease in state density for energies in the upper half of the original bands. It should be observed that in spite of these shape changes the mean energy remains unaltered.

The predominant effect of disorder on the magnetization is the Curie-point depression, which varies linearly with the disorder parameter. Since even in the disordered systems the magnetization curves have zero slope at $T=0$ and (negative) infinite slope at $T=T_C$, there are no marked shape changes, even for high degrees of disorder. For a given ratio of T/T_C the magnetization decreases slowly with increasing disorder—the curves are merely flattened slightly from the perfect crystal shape.

The approach outlined in this Letter is being extended to include the effects of external magnetic fields and of higher spins (using treatments similar to those of Callen⁸ and Praveczi⁹). Also under study are the results of using alternate schemes⁷ for decoupling the Green's function hierarchy. These extensions, with details of the calculations, will be reported elsewhere.

¹S. Mader and A. S. Norwick, *Appl. Phys. Lett.* **7**, 57

(1965); B. Elschner and H. Gartner, *Z. Angew. Phys.* **20**, 342 (1966); P. Duwez and S. C. H. Lin, *J. Appl. Phys.* **38**, 4096 (1967); K. Tamura and H. Endo, *Phys. Lett.* **29A**, 52 (1969).

²A. I. Gubanov, *Fiz. Tverd. Tela* **2**, 502 (1960) [*Sov. Phys. Solid State* **2**, 468 (1961)].

³D. Hone and K. Vogelsang, *J. Appl. Phys.* **39**, 1356 (1968); K. Vogelsang and D. Hone, *J. Appl. Phys.* **40**, 1118 (1969).

⁴W. J. De Bonte and H. Callen, *Phys. Rev.* **188**, 831 (1969).

⁵K. Handrich, *Phys. Status Solidi* **32**, K55 (1969).

⁶S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Plenum, New York, 1967); D. N. Zubarev, *Usp. Fiz. Nauk* **71**, 71 (1960) [*Sov. Phys. Usp.* **3**, 320 (1960)]. We follow the notation of this latter paper; see especially Eq. (7), p. 366.

⁷Alternative decouplings have been proposed: K. Moorjani and T. Tanaka, *Phys. Lett.* **28A**, 645 (1969); H. B. Callen, *Phys. Rev.* **130**, 890 (1963).

⁸Callen, Ref. 7.

⁹E. Praveczi, *Phys. Status Solidi* **35**, 63 (1969).

PERTURBATION TECHNIQUE FOR THE ANDERSON HAMILTONIAN*

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A new strong-coupling diagrammatic perturbation treatment of the Anderson Hamiltonian confirms in finite order Kondo divergencies for the partition function, associated with an effective s - d coupling J as predicted by the Schrieffer-Wolff transformation. This holds true also for the symmetric case. By summing the diagrams with two conduction-electron intermediate states in the limit $U \rightarrow \infty$, a Kondo temperature with J near diverging is obtained.

Similarities between the physical properties of the Anderson¹ and the s - d exchange Hamiltonians for dilute magnetic alloys were suggested when Schrieffer and Wolff² established the approximate equivalence of the two models most elegantly by means of a canonical transformation. Other approaches, confirming this equivalence, have been based on finite-order perturbation theory.^{3,4} However, recent treatments of the Anderson model by means of the Green's functions equations-of-motion decoupling scheme^{5,6} seem to show some important differences between the two models. In particular, Ref. 5 obtains a value for the effective s - d coupling J which is half the value of the J in the Schrieffer-Wolff transformation.⁷ In Ref. 6 it is proven that the decoupled equations of motion lead to vanishing of all Kondo singularities in the symmetric case $\epsilon_d + \frac{1}{2}U = 0$.⁸ Since for that particular case the Green's function perturbation theory carried out in Ref. 4 breaks down, while the functional-integral approach leads to the usual Kondo effect,⁹ the situation is sufficiently unclear to be re-examined from the viewpoint of direct perturbation theory.

For that purpose we follow Scalapino³ up to his Eq. (4), which we rewrite as

$$Z/Z_0 = 1 + \sum_{n=1}^{\infty} \int_0^{\beta} d\lambda_1 \int_0^{\lambda_1} d\lambda_2 \cdots \int_0^{\lambda_{n-1}} d\lambda_n \sum \cdots V_{k'd}^* \cdots V_{k'd} \times \langle k\text{-electron operators} \rangle \langle d\text{-electron operators} \rangle, \quad (1)$$

where the second sum extends over momenta, spins, and all sequences of n d -electron and n k -electron operators. The first bracket, representing the normalized thermodynamic expectation value of the k -electron operators written in the interaction representation, can be evaluated in the usual way, using Wick's theorem for thermodynamic averages. Wick's theorem cannot be used for the second bracket since the zero-order d -electron Hamiltonian H_d^0 contains the part $Un_{d\uparrow}n_{d\downarrow}$. Evaluating the trace in the second bracket in the representation where H_d^0 is diagonal, we obtain

$$\begin{aligned} \langle d\text{-electron operators} \rangle = & n_0 \langle 00 | \cdots c_{ds}(\lambda_j) \cdots c_{ds'}^\dagger(\lambda_k) \cdots | 00 \rangle + n_{d\uparrow} \langle 01 | \cdots c_{ds}(\lambda_j) \cdots c_{ds'}^\dagger(\lambda_k) \cdots | 10 \rangle \\ & + n_{d\downarrow} \langle 10 | \cdots c_{ds}(\lambda_j) \cdots c_{ds'}^\dagger(\lambda_k) \cdots | 01 \rangle \\ & + n_{dd} \langle 11 | \cdots c_{ds}(\lambda_j) \cdots c_{ds'}^\dagger(\lambda_k) \cdots | 11 \rangle. \end{aligned} \quad (2)$$

Here n_0 , $n_{d\uparrow}$, $n_{d\downarrow}$, n_{dd} are the occupation probabilities for the corresponding d states (see Scalapino³).

Taking the set of d -electron operators $c_{d\uparrow}(\lambda_1)c_{d\uparrow}^\dagger(\lambda_2)c_{d\downarrow}^\dagger(\lambda_3)c_{d\downarrow}(\lambda_4)c_{d\uparrow}(\lambda_5)c_{d\downarrow}^\dagger(\lambda_6)$ as an example in