

Static Properties of Spin Glasses

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A recent classical theory for spin glasses of Edwards and Anderson is modified for the quantum mechanical case. One obtains a cusp in the susceptibility $\chi(T)$ and in the specific heat $c_H(T)$ at a characteristic temperature T_f for finite spin S and zero slope for $S \rightarrow \infty$. The specific heat is proportional to T for $T \rightarrow 0$, whereas it tends to the Boltzmann constant k in the classical limit.

Spin glasses are random, fairly dilute magnetic alloys, such as *AuFe* or *CuMn*, with magnetic impurity concentrations c roughly between 10^{-3} and 10^{-1} . They show a variety of surprising properties, the most famous one being a sharp cusp in the static susceptibility at the so-called freezing temperature T_f .¹ These properties are due to exchange couplings of alternating sign between the magnetic atoms, the most important one being the Ruderman-Kittel-Kasuya-Yosida interaction, which lead at low temperatures to a complicated (and not yet completely understood) spin structure. At lower concentrations these interactions are severely influenced by Kondo or spin-fluctuation effects which in the single-impurity problem lead at $T=0$ to vanishing effective impurity moments; at high concentration more conventional (ferromagnetic or antiferromagnetic) order of sufficiently long range sets in and again impairs the characteristic spin-glass properties.

Recently, a new theory of spin glasses was proposed by Edwards and Anderson² (hereafter referred to as EA), in which a classical Heisenberg model with a random distribution of exchange interactions is considered. Assuming that the *local* part of the susceptibility is essential for a spin glass, they derive a self-consistent equation for the quantity $q \equiv \langle \vec{S}_i \rangle_{\text{av}}^2$. Here, $\langle \rangle$ indicates thermal averaging for a fixed configuration of magnetic impurities with spin \vec{S}_i and $[\]_{\text{av}}$ means averaging over a Gaussian distribution of exchange interactions. The quantity q vanishes above the freezing temperature T_f and leads to

discontinuities in the slope of the susceptibility $\chi(T)$ and specific heat $c_H(T)$ for vanishing external field H_z , as one would expect for a third-order phase transition. However, the slope of $\chi(T)$ turns out to be zero just below T_f , in contrast to the cusp found in many systems.¹ Furthermore, the solution of the self-consistent equation for q for $T \rightarrow 0$ leads to $c_H(0) = k$ (k is the Boltzmann constant) which is reminiscent of the behavior of a *classical* spin in an external magnetic field at $T=0$.

This Letter presents a quantum mechanical treatment of the same model. The additional difficulty due to the complicated commutation relations of the spin components is avoided by considering the simple mean-field approach of EA. I find (as expected) $c_H(T)$ to be proportional to T for $T \rightarrow 0$ and $\chi(0)/\chi(T_f) = 0.69$ for $S = \frac{1}{2}$ as compared to 0.92 in the classical limit and 0.6 from experimental data.¹ Most important, a cusp with finite slope just below T_f is obtained for the susceptibility and for the specific heat. For $S \rightarrow \infty$, this slope tends to zero, in agreement with the results of EA.

First I discuss the meaning of the order parameter q . The system is described by a Heisenberg Hamiltonian, including an external field H_z (μ_B is the Bohr magneton, N_i is the number of magnetic atoms, and $g=2$)³:

$$\mathcal{H} = -\frac{1}{2} \sum_{ij} \tilde{J}_{ij} \vec{S}_i \cdot \vec{S}_j + h \sum_i S_{i,z}, \quad h \equiv g\mu_B H_z. \quad (1)$$

The static susceptibility χ is obtained⁴ from the free energy F :

$$\chi \equiv \sum_{ij} \chi_{ij} = - (g\mu_B)^2 (\partial^2 F / \partial h^2) |_{h=0} = \sum_{ij} \{ [\langle \vec{S}_i \cdot \vec{S}_j \rangle]_{\text{av}} - [\langle \vec{S}_i \rangle \cdot \langle \vec{S}_j \rangle]_{\text{av}} \} \chi^0 / N_i S(S+1), \quad (2)$$

with the susceptibility of independent spins $\chi^0 = N_i S(S+1) (g\mu_B)^2 / 3kT$. The first term in the curly brackets of (2), being essential for a phase transition in a ferromagnet (and also in a recent theory of spin glasses by Adkins and Rivier⁵), is thought to be rather irrelevant for spin glasses. It is believed that

the basic properties of spin glasses are already seen in the *local* susceptibility χ_{ii} for which this term reduces to $S(S+1)$, whereas the second term reduces to the quantity q defined above. Thus it is characteristic for spin glasses that the thermal average $\langle \vec{S}_i \rangle \neq 0$ for $T < T_f$ and for a given impurity configuration, corresponding to a time-independent random internal field which acts on the spin at the site i . As is well known, in many cases a system of interacting particles can formally be described by a system of independent particles in a fictitious randomly fluctuating field.⁶ In spin glasses this field is real, and the corresponding interactions lead to a well-defined transition temperature T_f .

Following EA, I calculate the free energy F of the system from

$$-\beta F = [\ln Z]_{\text{av}} = \lim_{m \rightarrow 0} \{ ([Z^m]_{\text{av}} - 1)/m \}, \quad (3)$$

with $(\alpha = 1, \dots, m)$

$$\begin{aligned} [Z^m]_{\text{av}} &= [\{\text{Tr} \exp(-\beta H)\}^m]_{\text{av}} = [\text{Tr}_\alpha \exp(-\beta \sum_\alpha H^\alpha)]_{\text{av}} \\ &\equiv \prod_{ij} [d\vec{J}_{ij} P(\vec{J}_{ij})] \sum_{m_1^\alpha} \dots \sum_{m_i^\alpha} \langle m_i^\alpha | \dots \exp(-\beta \sum_\alpha H^\alpha) \dots | m_i^\alpha \rangle. \end{aligned} \quad (4)$$

The trace is performed with the $mN_i(2S+1)$ eigenstates $|m_i^\alpha\rangle$ of $S_{i,z}^\alpha$, where

$$\sum_\alpha H^\alpha = -\frac{1}{2} \sum_{ij} \vec{J}_{ij} \sum_\alpha \vec{S}_i^\alpha \cdot \vec{S}_j^\alpha + h \sum_{i,\alpha} S_{i,z}^\alpha.$$

Thus, formally, the three degrees of freedom per spin are replaced by $3m$ degrees of freedom with $[\vec{S}_i^\alpha, \vec{S}_i^\beta] = 0$ for $\alpha \neq \beta$, where $m \rightarrow 0$. Since the analytic continuation of Z^m from $m=1, 2, \dots$ to $m \rightarrow 0$ might not be unique, I checked Z^m in Eq. (4) for $m \rightarrow 0$ classically by comparing it with the linked-cluster (high-temperature) expansion of $\ln Z$.⁷ Both expansions agree at least to order \vec{J}^4 . However, a general proof of (4) for $m \rightarrow 0$ seems to be difficult.

To proceed, I assume for the exchange interactions \vec{J}_{ij} a Gaussian distribution with different widths $\Delta_{ij} = \Delta_{i-j}$ for different neighbors of the impurity i :

$$P(\vec{J}_{ij}) = (2\pi)^{-1/2} \Delta_{ij}^{-1} \exp(-\vec{J}_{ij}^2 / 2\Delta_{ij}^2). \quad (5)$$

In simplest approximation one introduces a cutoff for Δ_{ij} . The assumption² $\Delta_{ij} = \Delta$ for all i, j would lead to Δ_{ij}^2 proportional to N_i^{-1} , which is very unsatisfactory.

The \vec{J} integration in (4) with (5) can easily be carried out for a single pair by expansion. However, it is not straightforward for the general case, since the commutator $[(\vec{S}_i \cdot \vec{S}_j), (\vec{S}_i \cdot \vec{S}_n)] \neq 0$. Therefore I restrict myself to a mean-field approximation. Resorting to the Feynman variation principle,⁸ one writes (compare EA)⁹

$$\begin{aligned} [Z^m]_{\text{av}} &\geq \text{Tr}_\alpha \left(\exp[-\beta h \sum_{i,\alpha} m_i^\alpha + \frac{1}{2} \zeta \sum_{i,\alpha \neq \beta} (\vec{S}_i^\alpha \cdot \vec{S}_i^\beta) + C] \int \prod_{ij} (\pi^{-1/2} \Delta_{ij} \vec{J}_{ij}) \exp(-\sum_{ij} \vec{J}_{ij}^2) \right. \\ &\quad \left. \times \{ 1 + \frac{1}{2} \beta^2 \sum_{ij} \Delta_{ij}^2 \vec{J}_{ij}^2 (\sum_\alpha \vec{S}_i^\alpha \cdot \vec{S}_j^\alpha)^2 - \frac{1}{2} \zeta \sum_{i,\alpha \neq \beta} \vec{S}_i^\alpha \cdot \vec{S}_i^\beta - C \} \right). \end{aligned} \quad (6)$$

Here, ζ and C are variation parameters, and $\vec{J}_{ij} \equiv \vec{J}_{ij} / 2^{1/2} \Delta_{ij}$. The Ansatz (6) for the trial free energy F_0 [the first term in the curly brackets is $m(1 - \beta F_0)$ with $m \rightarrow 0$] is motivated, of course, by the assumption that correlations $\langle \vec{S}_i \cdot \vec{S}_j \rangle$ with $i \neq j$ are not essential. I checked this point, replacing $\zeta \delta_{ij}$ by ζ_{ij} with $\partial [Z^m]_{\text{av}} / \partial \zeta_{ij} = 0$, and found $\chi(T)$ and $c_H(T)$ essentially to be unchanged near T_f if $[\langle \vec{S}_i \cdot \vec{S}_j \rangle]_{\text{av}}$ varies smoothly in this temperature range.

The remaining calculation is completely analogous to EA. One obtains with

$$\langle (\sum_\alpha \vec{S}_i^\alpha \cdot \vec{S}_j^\alpha)^2 \rangle_0 = \sum_{\alpha\beta\lambda} \langle S_{i\lambda}^\alpha S_{i\lambda}^\beta \rangle_0 \langle S_{j\lambda}^\alpha S_{j\lambda}^\beta \rangle_0 = \frac{1}{3} \sum_{\alpha\beta} \langle \vec{S}_i^\alpha \cdot \vec{S}_i^\beta \rangle_0 \langle \vec{S}_j^\alpha \cdot \vec{S}_j^\beta \rangle_0$$

the variation parameter

$$C = \frac{1}{2} N_i m \left\{ \frac{1}{3} \beta^2 \Delta^2 [S^2(S+1)^2 + (m-1)q^2] - \zeta q(m-1) \right\}, \quad (7)$$

where $\Delta^2 \equiv \sum_j \Delta_{ij}^2$ and where $q = z_0 N_i \langle \vec{S}_i^\alpha \cdot \vec{S}_i^\beta \rangle_0 = 2m^{-1}(m-1)^{-1} \partial \ln z_0 / \partial \zeta$ with $\alpha \neq \beta$. The thermal average $\langle \dots \rangle_0$ is defined as

$$\langle \dots \rangle_0 = \text{Tr}_\alpha \left[\dots \exp \frac{1}{2} \zeta \sum_{i,\alpha \neq \beta} (\vec{S}_i^\alpha \cdot \vec{S}_i^\beta) \right] / z_0^{N_i}, \quad (8)$$

where

$$z_0 \equiv \text{Tr}_\alpha [\exp \frac{1}{2} \xi \sum_{\alpha \neq \beta} (\vec{S}_i^\alpha \cdot \vec{S}_i^\beta)]$$

$$= \exp[-\frac{1}{2} m \xi S(S+1)] (2\pi)^{-3/2} \int d^3 r \exp(-\frac{1}{2} r^2) \{ \sinh[(S + \frac{1}{2}) \xi^{1/2} r] / \sinh(\frac{1}{2} \xi^{1/2} r) \}^m. \tag{9}$$

Variation of F_0 with respect to ξ yields $\xi = \frac{1}{3} (\beta \Delta)^2 q$. One finds with (9) for $m \rightarrow 0$ and $h = 0$

$$S(S+1) - q = (2\pi)^{-3/2} \int d^3 r \exp(-\frac{1}{2} r^2) r \xi^{-1/2} \{ (S + \frac{1}{2}) \coth[(S + \frac{1}{2}) \xi^{1/2} r] - \frac{1}{2} \coth(\frac{1}{2} \xi^{1/2} r) \}. \tag{10}$$

The expression in the curly brackets is identical with the Brillouin function $SB_S(S\xi^{1/2}r)$. Equation (10) reduces in the classical limit with $S^2 = 1$ to the result of EA. Since the argument of the Brillouin function contains $\xi^{1/2} \sim q^{1/2}$, rather than q , the susceptibility near T_f behaves differently from that in an ideal antiferromagnet.

Expanding (10) for $q \rightarrow 0$ yields

$$q \cong \frac{1}{4} (1-t) [(2S+1)^4 - 1]^2 / [(2S+1)^6 - 1],$$

which reduces to the classical result $q \cong 1-t$ for $S \rightarrow \infty$. Here, $t \equiv T/T_f$ with

$$kT_f = \frac{1}{12} \Delta [(2S+1)^4 - 1]^{1/2} \tag{11}$$

as compared to the classical value $kT_f = \Delta/3$. The susceptibility, Eq. (2), $\chi(T) = \chi^0 [1 - q/S(S+1)]$ shows a cusp with positive slope for $t \leq 1$ for all finite spin values S . Since

$$\Delta^2 = \sum_j [\vec{J}_{ij}^2]_{av} = \sum_j J_{ij}^2 [c_j^2]_{av} \propto c$$

one has $T_f \propto c^{1/2}$. However, in view of the crudeness of the model, the agreement with the experimental values¹ $T_f \propto c^n$, with $0.55 \leq n \leq 0.75$, seems to be rather fortuitous. For $t \rightarrow 0$ and $S = \frac{1}{2}$ one has $q = \frac{3}{4} - (5/6\pi)^{1/2} t - (5/9\pi) t^2$, as compared to the classical spin with $S^2 = 1$, where $q_c = 1 - (8/3\pi)^{1/2} t - \frac{1}{3} (4/\pi - 1) t^2$. For $t > 1$ one has the susceptibility of free spins. Since all lattice effects have been neglected, the present model should be more suitable for amorphous spin glasses where one observes¹⁰ above T_f a Curie-Weiss law with

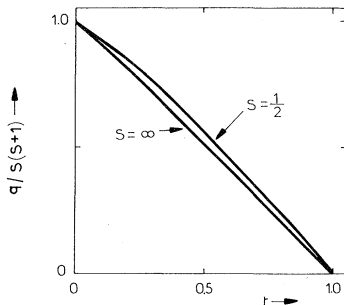


FIG. 1. The parameter q versus reduced temperature $t \equiv T/T_f$.

$\Theta \approx 0$, in agreement with theoretical results.¹¹ The functions $q(t)$ and $\chi(T)/\chi(T_f)$ are indicated in Figs. 1 and 2(a) for $S = \frac{1}{2}$ and for the classical limit.

The zero-field specific heat $c_H = du/dT$ per spin is obtained from the internal energy

$$u = kT^2 N_i^{-1} \partial [\ln Z]_{av} / \partial T$$

$$= -\Delta^2 (6kT)^{-1} [S^2(S+1)^2 - q^2]. \tag{12}$$

For $S = \frac{1}{2}$ and $t \rightarrow 0$ we have, besides exponential terms, $u/kT_f = -(8/5\pi)^{1/2} (\frac{3}{4} q^{-1/2} + q^{1/2})$ which leads to $c_H/k = (8/9\pi) t [1 + (160/27\pi)^{1/2} t + \dots]$ and in the classical limit to $c_H/k = 1 + (8/3\pi)^{1/2} (1 - 2/\pi) t + O(t^2)$. Near $t = 1$ one has $c_H/k = 0.9 - 0.75(1-t)$ and $c_H/k = \frac{3}{2} [1 - 1.2(1-t)^2]$, respectively. The temperature dependence of the specific heat for both cases is indicated in Fig. 2(b). The value $c_H = k$ at $t = 0$ agrees with that for a classical spin

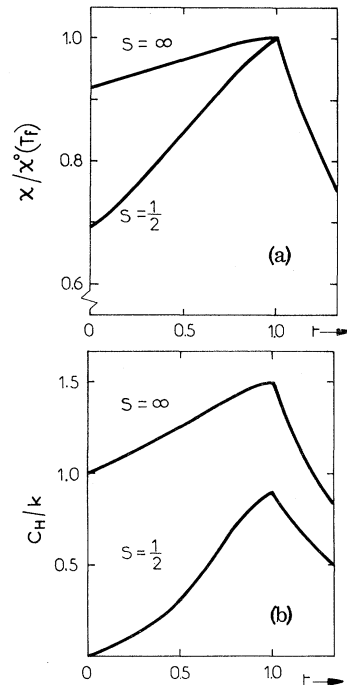


FIG. 2. (a) Susceptibility and (b) specific heat versus temperature t .

in an external magnetic field. For $t > 1$ one has in all cases $c_H \propto T^{-2}$, as is also obtained for independent spins in a magnetic field $h \ll kT$.

Results for the magnetic field dependence of $\chi(T)$ and $c_H(T)$ and for the entropy will be published elsewhere.

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³I use the notation of EA with $\tilde{J}_{ij} = J_{ij}c_i c_j$, where $c_i = 0, 1$, depending on whether a lattice site i is occupied by a magnetic atom or not. In the following i, j label impurity sites only.

⁴I assume that there is no preferred direction in spin glass for $h \rightarrow 0$, that is $\chi_{zz} = \chi_{xx} = \chi_{yy}$, and use $[H, \sum_i S_{i,\alpha}] = 0$.

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⁹Actually the application of the Feynman variation principle is complicated because of the fact that we have to go to second order in $H' = H - H_0$, where H_0 is the trial Hamiltonian, since first-order terms in \tilde{J} vanish after the \tilde{J} integrations. However, following Ref. 8 one can show that to order m

$$\text{Tr}_\alpha \exp(-\beta H_0 - \beta H')$$

$$\geq \text{Tr}_\alpha \exp(-\beta H_0)(1 - \beta H' + \frac{1}{2}\beta^2 H'^2),$$

which still remains correct if one drops in H' the term proportional to ζ^2 . After performance of the \tilde{J} integrations this leads to an expression which is analogous to the classical expression (3.13) of EA. Following EA we use the variation principle in the more restrictive form $F \approx F_0(\xi)$ and $-\beta \langle H' \rangle_0 + \frac{1}{2}\beta^2 \langle H'^2 \rangle_0 = 0$ with $\partial F_0 / \partial \xi = 0$. The constant C in Eq. (6) is then defined by the second equation which leads after performance of the \tilde{J} integrations to Eq. (7) of the text.

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Polyatomic-Ion Implantation Damage in Silicon

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We have investigated the number, N_d , of displaced lattice atoms in room-temperature implantation in Si of polyatomic-carbon-ion beams (C_n^+ and $C_{n-x}O_x^+$) for several values of n , using the backscattering-channeling-effect technique. For each ion species the same energy (8.8 keV) per carbon and the same atomic fluence and flux were used. N_d increases rapidly with increasing n , indicating that the deposited-energy density within the collision cascade is a key factor in determining how much damage is created and retained.

The use of polyatomic- (or molecular-) ion beams in ion implantation provides a simple, direct method of varying the deposited-energy density within each individual collision cascade, while maintaining constant most other cascade parameters such as ion range or cascade volume. It thus provides a powerful technique for investigating some of the mechanisms involved in radiation-damage studies: for example, the role of "energy-spike" effects as recently discussed by

Sigmund,¹ or the possible existence and magnitude of significant annealing stages during implantation.

Moore, Carter, and Tinsley² found that diatomic heavy ions (As_2^+) implanted into GaAs at room temperature produced about 30% more damage than the same number of As atoms introduced as monatomic ion species (As^+). In silicon, an even larger "molecular effect" has been observed,³ with the diatomic heavy ions (As_2^+ , Sb_2^+ , Te_2^+ ,