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Comparison of the self-consistent mean-random-field approximation with the $n \longrightarrow 0$ expansion of Sherrington and Kirkpatrick for spin glasses and with experiment

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The magnetic properties of a spin-glass system interacting with a set of Gaussian-distributed exchange potentials are obtained using the self-consistent mean-random-field (MRF) approximation. It is shown that for this distribution the magnetic properties obtained from the MRF approximation are identical with that obtained from the $n \longrightarrow 0$ expansion of the free energy of Edwards and Anderson and of Sherrington and Kirkpatrick. The specific heat from the MRF approximation is also linear in temperature T for low T. For the Ruderman-Kittel-Kasuya-Yosida interaction the n expansion is difficult, however, the therymodynamic properties of the spin glasses are easily obtained in the MRF approximation. Furthermore, the latter gives some of the known properties of the system in excellent agreement with experiment. The MRF approximation predicts the following experimentally measured quantities: the low-temperature specific heat, the low-temperature low-field and high-field magnetization, the cusp in the magnetic susceptibility, the spin dependence of the susceptibility near $T \longrightarrow 0$, and the low-temperature resistivity near $T \longrightarrow 0$. It is furthermore argued that the scaling of the thermodynamic properties with impurity concentration will have a different concentration dependence at high and low temperatures. A possible reason for the agreement of the Ising-like model and the disagreement of the Heisenberg-like model prediction with experiment is given.

I. INTRODUCTION

Several years ago I derived[1,2] a self-consistent method to obtain the probability distribution $P(H)$ of the random molecular field H of a spin-glass system in which the spins interact via a Ruderman-Kittel-Kasuya-Yosida (RKKY)[3] interaction. We shall denote this method as the mean-random-field (MRF) approximation and Ref. 1 will henceforth be denoted as I. The method gives a low-temperature specific-heat linear in temperature T and independent of the fractional impurity concentration c, in agreement with experiments on Cu-Mn.[4] Similarly, the low-temperature magnetic susceptibility predicted from the model is in agreement with the experiments of Franz and Sellmyer[5] and of Dreyfus *et al.*[6] In spite of this agreement with experiment the theory was deficient in the following respects: (i) The results were derived in an Ising model and it was believed that the correct treatment should use a Heisenberg-like distribution of fields.[7] Such a Heisenberg-like distribution gives that the probability for small field goes to zero[8] and results in an internal-field model which does not explain the linear-T dependence of the specific heat. Furthermore, experiments by Murnick *et al.*[9] indicate that the probability for zero fields approaches zero, not unlike the Heisenberg-like distribution of fields.[8] (ii) The molecular-field model did not predict the cusps in the magnetic susceptibility observed by Canella and Mydosh[10] and gave magnetic order for all temperatures, thus resulting in an infinite transition temperature.

A significant step towards proper understanding of the MRF picture has recently been made by Riess and Klein[11] who found that if one restricts the distance of closest approach between the magnetic impurities to be a near-neighbor distance one obtains a finite transition temperature T_c above which the spin-glass system is completely disordered. Riess and Klein[11] also obtain a cusp in the magnetic susceptibility at $T = T_c$.

A completely different and more rigorous approach to the spin-glass problem has been developed by Edwards and Anderson[12] (EA) and by Sherrington and Kirkpatrick[13] (SK) to obtain the free energy for the spin-glass system. In order to evaluate the average of $\langle \ln Z \rangle_c$, where Z is the partition function and $\langle\ \rangle_c$ indicates an average over coordinates, EA[12] use the expression

$$\langle \ln Z \rangle_c = \lim_{n \to 0} \frac{1}{n} \langle (Z^n - 1) \rangle_c ,$$

evaluate $\langle Z^n \rangle_c$ for integer n and continue the function analytically to $n \to 0$. This is denoted as the n expansion. For a set of random exchange potentials J_{ij} having a Gaussian probability distribution $P(J_{ij})$, EA[12] obtain a discontinuous derivative in the magnetic susceptibility. SK[13] evaluated the free energy of the spin-glass system using a Gaussian distribution of exchange potentials and showed that the system may undergo a ferromagnetic (or antiferromagnetic) or spin-glass transition depending upon the parameters entering the Gaussian.

The purpose of this paper is (i) to show that using the SK[13] Gaussian distribution of exchange potentials, the MRF approximation[1] gives identical magnetic properties to that obtained by SK (as well as similar specific heats near $T \rightarrow 0$); (ii) to present a new point of view on the physics of the RKKY system and the MRF approximation; (iii) to argue that the question of the validity of the Ising model or the Heisenberg model is most likely resolved for the thermodynamic properties of the RKKY spin-glass system; and (iv) to demonstrate that the MRF approximation explains many of the experimentally observed data on RKKY spin glasses. Some of these experiments are (a) the low-T specific heat is linear in T and approximately independent of c as was found by Zimmerman and Hoare[4]; (b) the low-temperature magnetic susceptibility χ of the system is $\chi \propto [A_1 + B_1(T/c)]$, where A_1 and B_1 are independent of T and c as was measured by Franz and Sellmyer[5]; (c) the deviation of the low-T resistivity from its $T=0$ value is linear in T and independent of c as was measured by Ramos[14]; (d) the deviation of the very-high-field magnetization $M(T)$ from its saturation value, $M(T)-M(0)$ is proportional to $[1+\alpha(T^2/B^2)]/B$ for low temperatures, where B is the applied magnetic field, in agreement with the experiment of Hou and Coles[15]; (e) there is a cusp in the magnetic susceptibility at temperature T_c, with $T_c \propto c^\alpha$ where $\alpha \approx 0.66$, not unlike the experiments of Canella and Mydosh[10]; (f) furthermore, we find that the second derivative of the magnetic susceptibility with respect to the temperature for very low temperatures may be positive, close to zero, or negative, depending upon the magnitude of the impurity spin; (g) finally, we argue that the μ-meson-depolarization experiments of Fiory *et al.*[9] are likely consistent with the MRF model.

II. MOLECULAR-FIELD THEORY FOR THE GAUSSIAN-DISTRIBUTED EXCHANGE POTENTIAL

In this section we show that for the Gaussian-distributed exchange potentials used by SK,[13] the random-molecular-field model gives identical magnetic properties to that determined by SK.

Consider the Hamiltonian $\mathcal{H}$ of the form

$$\mathcal{H} = -\frac{1}{2} \sum_{i \neq j} J_{ij} S_i S_j , \tag{2.1}$$

where J_{ij} is a random variable. SK[13] use a probability distribution of J_{ij} of the form

$$P(J_{ij}) = (2\pi J^2)^{-1/2} \exp[-(J_{ij} - J_0)^2/2J^2] \tag{2.2}$$

for each J_{ij}.

Using the molecular-field approximation[1] we have for the internal field at site i, H_i,

$$H_i = \sum_j J_{ij} \langle S_j \rangle , \tag{2.3}$$

where $\langle S_j \rangle$ is the thermal average of the spin at site j in the field H_j, as is discussed in I. Using the statistical model of Margenau[16] in the form developed in Eq. (2.7) of I we obtain

$$P(H_i) = \int P(R)\, d^3R\, \delta\left(H_i - \sum J_{ij} \langle S_j \rangle\right) , \tag{2.4}$$

where for the case considered by SK[13]

$$P(R)\, d^3R = \prod_{i \neq j} P(J_{ij}) dJ_{ij} , \tag{2.5}$$

where $P(J_{ij})$ is given by Eq. (2.2)

Substituting Eq. (2.5) in Eq. (2.4) and letting $\delta(x) = (2\pi)^{-1} \int e^{i\rho x} d\rho$ gives

$$\begin{aligned} P(H_i) &= \frac{1}{2\pi} \int e^{i\rho H_i} d\rho \prod_{j=1}^{N} \int P(J_{ij}) dJ_{ij}\, e^{-i\rho J_{ij} \langle S_j \rangle} \\ &= \frac{1}{2\pi} \int \exp\left[i\rho \left(H_i - J_0 \sum_j \langle S_j \rangle \right) \right] \\ &\quad \times e^{-J^2 \rho^2/2} \sum_j \langle S_j \rangle^2 , \end{aligned} \tag{2.6}$$

where the dependence of $\langle S_j \rangle$ on J_{ij} is neglected.

We define the average magnetization M by the relation,

$$M = \langle \langle S_i \rangle \rangle_c \equiv \int P(H_i) \tanh \beta H_i \, dH_i \tag{2.7}$$

where $\langle \langle S_i \rangle \rangle_c$ indicates an average of $\langle S \rangle$ over the coordinates of the system. We also define the spin-glass order parameter m

$$m = \langle \langle S_i \rangle^2 \rangle_c \equiv \int P(H_i) \tanh^2 \beta H_i \, dH_i , \tag{2.8}$$

where as a self-consistency condition we require that m_i, M_i, and $P(H_i)$ are independent of i.

We remark that our M corresponds to SK's magnetization and our m corresponds to SK's order parameter q. Using the definitions Eqs. (2.7) and (2.8) in Eq. (2.6) and letting $\sum_i \langle S_i \rangle = zM$, $\sum_i \langle S_i \rangle^2 = zm$, and integrating gives

$$P(H) = \frac{1}{(2\pi m z)^{1/2} J} e^{-(H - J_0 z M)^2 / 2J^2 m z} , \tag{2.9}$$

where z is the effective number of neighbors.

Using Eq. (2.9) in Eq. (2.7), and changing variables of integration gives

$$M = \frac{1}{(2\pi)^{1/2}} \int e^{-x^2/2} dx \tanh \beta [J(zm)^{1/2} x + J_0 z M] . \tag{2.10}$$

Using Eq. (2.9) in Eq. (2.8) and letting $\tanh^2 x = 1 - \mathrm{sech}^2 x$, we obtain

$$m = 1 - \frac{1}{(2\pi)^{1/2}} \int e^{-x^2/2} dx\, \mathrm{sech}^2 \beta [J(zm)^{1/2} x + J_0 z M] . \tag{2.11}$$

The results for the magnetization M and the spin-glass order parameter m arising from the MRF approximation are thus identical with that obtained by SK[13] from the evaluation of the free energy using the n expansion.

We next obtain the low-temperature specific heat from the molecular-field approximation. We recall that for any distribution of exchange potentials the free energy is

$$-\beta F = \int P(R)\, d^3R \ln Z(R) , \tag{2.12}$$

where for the case considered by SK, $P(R)\, d^3R$ is given by Eq. (2.5). The internal energy U is

$$U = + \frac{\partial(\beta F)}{\partial\beta} = \int P(R)\, d^3R \left(\frac{d \ln Z(R)}{\partial\beta} \right) , \tag{2.13}$$

since $P(R)$ is independent of T. The specific heat C_v is given by

$$\begin{aligned} C_v &= \int P(R)\, d^3R\, \frac{d^2 \ln Z}{\partial\beta^2} \\ &= \int P(R)\, d^3R C_v(R) . \end{aligned} \tag{2.14}$$

In the molecular-field approximation $C_v(R) \equiv C_v(\sum_j J_{ij} \langle S_j \rangle)$. Thus we write Eq. (2.14) in the form

$$\begin{aligned} C_v(T) = N \int \left[\int \delta\left(H - \sum J_{ij} \langle S_j \rangle \right) P(R)\, d^3R \right] \\ \times C_v(H)\, dH , \end{aligned} \tag{2.15}$$

where N is the number of spins in the system.

The expression in the large square brackets of Eq. (2.15) is by definition $P(H)$, as is seen from Eq. (2.4). We then have

$$C_v(T) = N \int P(H) C_v(H)\, dH . \tag{2.16}$$

The internal energy U is $U = -\frac{1}{2} H \tanh\beta H$ and $C_v(H) = (H^2/k_B T^2) \operatorname{sech}^2\beta H$. Using this result in Eq. (2.16) and changing variables of integration gives

$$\begin{aligned} C_v(T) = \frac{N}{2k_B T^2} \int_{-\infty}^{\infty} \frac{e^{-x^2/2}}{(2\pi)^{1/2}} [J(mz)^{1/2}x + J_0 zM]^2 \\ \times \operatorname{sech}^2\beta[J(mz)^{1/2}x + J_0 zM] dx . \end{aligned} \tag{2.17}$$

For the spin-glass transition with $M = 0$ we obtain,[17] for very low temperatures, for the leading term in the specific heat

$$C_v = Nk_B \left(\frac{k_B T}{z^{1/2} J} \right) \left(\frac{2}{\pi} \right)^{1/2} \left(\frac{\pi^2}{12} \right) . \tag{2.18a}$$

The expression obtained by SK[13] for T near zero is

$$C_v = Nk_B \left(\frac{k_B T}{z^{1/2} J} \right) \left(\frac{2}{\pi} \right)^{1/2} \left(\frac{\pi^2}{12} - \frac{1}{2\pi} \right) . \tag{2.18b}$$

In spite of the fact that we obtain from the MRF approximation a leading term similar to SK, our equations point out the difficulty which arises in deriving the thermodynamic properties of the system in the MRF approximation. Equations (2.12)–(2.14) follow from each other. Had, we, however, integrated Eqs. (2.12) or (2.13) and had obtained the specific heat from differentiating the resulting integral for U with respect to T, we would have obtained an extra contribution for $dP(H)/dT$ which is not in the original expression, Eq. (2.14), for the specific heat. Such difficulties always arise when one evaluates thermodynamic quantities using an approximation method, in our case the molecular-field approximation. It is still gratifying that the simple molecular-field approximation gives reasonable agreement with the n expansion. The agreement for the specific heat is limited to low temperatures, for at high temperatures SK[13] found that $C_v \propto T^{-2}$ and the molecular-field approximation gives $C_v = 0$ for $T > T_c$, where T_c is the transition temperature to be discussed later on. However, the magnetic properties from the two methods are identical.

Even though the results obtained from the distribution $P(J_{ij})$ of Eq. (2.2) are interesting by themselves, in order to compare the RKKY system with experiment one would have to derive $P(J_{ij})$ for the RKKY system self-consistently, and then use the n expansion to obtain the free energy. The latter is difficult for the RKKY interaction; however, thermodynamic properties of the spin-glass system are easily obtained in the MRF approximation.

III. MRF APPROXIMATION FOR THE RKKY INTERACTION

In this section a comparison between the predictions of the MRF approximation for the RKKY interaction and experiment is made. It is found that there is very good agreement between theory and experiment for many of the thermodynamic properties. One wonders why the molecular-field approximation should give results which are in such good agreement with experiment. The most likely reason for this is that the RKKY potential is a very-long-range interaction, and at least for the ferromagnet the molecular-field approximation becomes exact when the range of interaction approaches infinity.[18] Before we make a detailed comparison mentioned above, we comment on the validity of the Ising model calculations of I.

It was found recently by Riess and Klein[11] that the RKKY spin-glass system undergoes a phase transition and exhibits a cusp in the magnetic susceptibility at $T=T_c$, where T_c is the spin-glass transition temperature. Below T_c the system is in an ordered state in which the magnetization M (in the absence of an externally applied magnetic field) is zero. However m, the average of the square of the local magnetization, is nonzero. We shall argue later that whether the order parameter m is Ising-like (up or down only) or has some complicated structure (some random spirallike structure which a Heinsenberg-like model may predict) may be of no consequence as far as the thermodynamic properties of the system are concerned. The suggested qualitative reason for this is that once the system undergoes a phase transition, the orientations of the local spins are fixed by the local internal fields experienced by the individual spins. Therefore, when one calculates the probability distribution of the internal field at a particular site one should not allow all other spins to have arbitrary orientations, but only orientations which are in accordance with the direction which the spin-glass order parameter makes locally. We will come back to a discussion of this point later. Thus we use an Ising distribution, and will show later that a *simple* application of the Heisenberg distribution gives thermodynamic properties in complete disagreement with experiment.

The probability distribution $P(H)$ of the internal field H in an Ising model is given by Eq. (2.14) of I, i.e.,

$$P(H)=(2\pi)^{-1}\int e^{i\rho H-(N/V)V'}\,d\rho\,, \tag{3.1}$$

where N/V is the number of particles per unit volume, and

$$V'=4\pi\int_{-\infty}^{\infty}P(H)\,dH\int_0^{\infty}[1-\cos(\rho a\mu/r^3)]\,r^2\,dr\,, \tag{3.2}$$

where in Eq. (3.2) the distance of closet approach between the impurities is allowed to go to zero. Since in a realistic physical situation the distance of closest approach between the impurities is limited by y_0, where y_0 is the near-neighbor distance in the solid, Eq. (3.2) exaggerates the importance of large fields and results in a Lorentzian distribution of fields. All moments, except the zeroth, of the Lorentzian diverge, thus a high-temperature expansion of the magnetization would give an infinite transition temperature (in other words such an expansion is not valid). To bring Eq. (3.2) in accordance with the appropriate physical situation, we let the lower limit of the second integral in Eq. (3.2) be y_0 instead of zero. Changing variables of integration and letting n_0 be an effective number of sites within a unit cell, Eq. (3.2) becomes

$$V'=\tfrac{1}{3}(4\pi\,|\rho a|)\int P(H)\,|\mu(H)|\,dH \times\int_0^{n_0 a|\mu(H)|\rho}(1-\cos z)/z^2, \tag{3.3}$$

where $n_0=(d/y_0)^3$, where d is the lattice constant and a is the strength of the RKKY interaction at a distance of one lattice constant. The vertical brackets $|x|$ indicate absolute values of x, and

$$|\mu(H)|=S\,|B_S(x)|\,, \tag{3.4}$$

where

$$B_S(x)=\left(\frac{2S+1}{2S}\right)\coth\left(\frac{2S+1}{2S}\right)x-\frac{1}{2S}\coth\frac{x}{2S}\,, \tag{3.5}$$

with $x=S(g\mu_B)(H+B)/k_BT$, where g is the gyromagnetic ratio, μ_B is the Bohr magneton and H is the internal and B is the external field.

Except for the near-neighbor cutoff Eq. (3.3) is the same as that used in I and Ref. 11. However, we now use the Brillouin function, instead of $\mu=\tanh\frac{1}{2}x$. In this way we can obtain the spin dependence of the thermodynamic quantities. We now let $B\rightarrow 0$ and delete the quantities g and μ_B whenever convenient and put them back in the last step of our derivation.

In order to evaluate Eq. (3.3) we replace the value of $|\mu(H)|$ in the upper limit of the integral by its average value m, where

$$m=\int P(H)\,|\mu(H)|\,dH =\int P(H)S\,|B_S(x)|\,dH\,. \tag{3.6}$$

The approximation by which we replace the average value of $|\mu(H)|$ by m we denote as the modified-MRF approximation. For the distribution function with no cutoff the two are identical, however, with a cutoff the two are somewhat different. The physical meaning of the modified-approximation is that the spin which occurs as an argument of the cosine in Eq. (3.2) is replaced by its average magnitude.

Substituting Eq. (3.6) into Eq. (3.3) and the result used in Eq. (3.1) gives

$$P(H)=\frac{1}{\pi}\int_0^{\infty}\cos\rho H\,\exp[-(\tfrac{4}{3}\pi)cn_0am\rho f(\rho n_0am)]\,d\rho\,, \tag{3.7a}$$

$$f(\rho n_0am)=\int_0^{\rho n_0am}(1-\cos z)/z^2\,dz\,. \tag{3.7b}$$

Let $y = \rho n_0 a m$, changing variables we have

$$P(H) = (\pi a n_0 m)^{-1} \int_0^{\infty} \cos y(H/n_0 a m) \times \exp[-(\tfrac{4}{3}\pi) c y f(y)]\, dy\,. \tag{3.7c}$$

Using Eq. (3.7c) to evaluate the nth moment of the distribution function η_n gives

$$\eta_n = \int P(H)\, |H^n|\, dH \tag{3.8a}$$

$$= (\pi a n_0 m)^{-1} \int dy \int dz\, P(z) z^n \cos yz \times \exp[-(\tfrac{4}{3}\pi) c y f(y)]$$

$$= (a n_0 m)^n g_n(c)\,, \tag{3.8b}$$

where η_n is the magnitude of the nth moment of $P(H)$ and $g_n(c)$ is a function of the impurity concentration c only. This result will become important when we consider the behavior of the system near the spin-glass transition.

For small values of H the behavior of Eq. (3.7) is approximately Lorentzian in the sense that the behavior of $P(H)$ for small H (and low c) is approximately given by

$$P_1(H) \approx \frac{1}{\pi} \frac{\Delta_1}{\Delta_1^2 + H^2}\,, \quad H \ll n_0 a m \tag{3.9}$$

where $\Delta_1 = 2\pi^2 a n_0 c m = \gamma_1 c m$, with m given by Eq. (3.6).

Equation (3.9) is identical with Eq. (3.3) or I where no cutoff was introduced in the exchange potential. Equation (3.7) was solved using a computer, and for small H and for impurity concentrations of the order of 1%, Eq. (3.3) of I differs from Eq. (3.9) by only a few percent. The difference could, however, be possibly detected when measuring low-temperature quantities which strongly depend upon the very-low-field behavior of $P(H)$. We note that m in Eq. (3.6) is analogous to the order parameter of SK.[13]

For very high fields ($H \gg n_0 a m$), Eq. (3.7) drops off approximately like a Gaussian and is of the form

$$P_2(H) \propto \Delta_2^{-1} \exp[-\tfrac{1}{2}(H/\Delta_2)^2]\,, \quad H \gg n_0 a m \tag{3.10}$$

where $\Delta_2 = (\frac{2}{3}\pi)^{1/2} a n_0 c^{1/2} m$, where m is again given by Eq. (3.6).

One important difference between the low-field and high-field probability distribution is the concentration dependence of their width. This will have an important effect on the scaling of the thermodynamic properties of the system with impurity concentration. The very-low temperature properties of the spin glass are primarily determined by very small fields, where Eq. (3.9) applies. Since Δ_1 is proportional to c, these low-T properties will scale differently with the impurity concentration than the high-T properties. The latter depend more on the high-field distribution, whose width is proportional to $c^{1/2}$. Thus our results show that the generally accepted scaling argument, according to which all thermodynamic properties of the Ruderman-Kittel spin glass depend upon T/c only, are incorrect. Rather, the scaling with T/c should be reasonably good for very low T, but fails completely near the spin-glass transition temperature T_c where we find that $T_c \propto c^{\alpha}$, where $\alpha \approx 0.66$.

A. Cusp in the magnetic susceptibility

For high temperatures Δ_1 and Δ_2 of Eqs. (3.9) and (3.10) approach zero. Riess and Klein[11] examined the behavior of the order parameter m in the presence of small applied fields B and find that $\lim_{B\to 0}(m/B) \to \infty$ as $T \to T_c$, indicating the existence of a nonzero m below T_c, where Riess and Klein used the MRF approximation to obtain T_c. They found that

$$T_c = \tfrac{1}{3} S(S+1)[\eta_k(0)]^{1/k}\,,$$

where k in the MRF approximation was either 1 or 2. Thus in the MRF approximation there is an ambiguity is the determination of the transition temperature. In the modified-MRF approximation no such ambiguity arises. When going through the same procedure as was done in Ref. 11, we obtain

$$T_c = \tfrac{1}{3}(S+1)\eta_1(0)\,, \tag{3.11}$$

where $\eta_k(0)$ is the magnitude of the kth moment of the distribution function evaluated at $T = 0$. Or

$$\eta_k(0) = 2 \int_0^{\infty} P(H,0) H^k\, dH\,, \tag{3.12}$$

where $P(H,0)$ is the expression for the probability distribution at $T = 0$. The value of $\eta_k(0)$ is proportional to S^k as can be seen by substituting Eq. (3.6) into Eq. (3.8b).

The magnetization per impurity $M^1(B)$ is

$$M^1(B) = \int_{-\infty}^{\infty} P(H) S\, B_S[g\mu_B\beta\, |H + B|\,]\, dH\,. \tag{3.13}$$

and the single-spin susceptibility $\chi_1(\beta)$ is

$$\chi^1(\beta) = \lim_{B\to 0} \frac{d}{dB} \int_{-\infty}^{\infty} P(H) S B_S(x)\, dH$$

$$= \lim_{B\to 0} \int_{-\infty}^{\infty} P(H) \frac{d}{dB}\, [S B_S(x)]\, dH\,. \tag{3.14}$$

Expanding Eq. (3.14) in a power series of β for high temperatures we obtain.

$$\chi^1(\beta)=\frac{P^2_{\text{eff}}}{3k_BT}\left(1-\frac{2S^2+2S+1}{10(k_BT)^2}\,\eta_2(0)m^2\right), \quad (3.15)$$

where $P^2_{\text{eff}}=[S(S+1)\,(g\mu_B)^2]$. For $T>T_c, m=0$ and we obtain

$$\chi^1=P^2_{\text{eff}}/3k_BT\,, \quad T>T_c\,. \quad (3.16)$$

We thus obtain *a* T^{-1} dependence of the paramagnetic susceptibility rather than the usual $(T-T_c)^{-1}$ dependence for ferromagnets. The slope of the susceptibility for $T>T_c$ is

$$\frac{d\chi^1}{dT}=\frac{-P^2_{\text{eff}}}{(3k_BT)^2}\,. \quad (3.17)$$

Solving Eq. (3.6) and (3.10) for m^2 near the transition temperature and substituting the result into Eq. (3.15) gives for the derivative of the susceptibility just below T_c

$$\frac{d\chi^1}{dT}=\frac{P^2_{\text{eff}}}{3k_BT_c^2}\left(\frac{3\eta_2(0)\eta_1(0)}{\eta_3(0)}-1\right)\,. \quad (3.18)$$

We note that since $\eta_k(0)$ is proportional to S^k, the spin-dependent factors in the large parentheses of Eq. (3.18) cancel, and the only remaining spin dependence is in P^2_{eff}.

A comparison of the slope of the susceptibility above T_c from Eq. (3.17) to that just below T_c in Eq. (3.18) shows that even though the susceptibility is continuous at $T=T_c$ it has a discontinuous derivative at T_c. For our distribution the slope of the susceptibility still remains negative below T_c. Thus the MFR theory gives a cusp in the susceptibility with a concentration-dependent slope on the two sides of T_c. The maximum in χ occurs, however, at a temperature below T_c.

After this paper was prepared for publication it came to the authors attention that Rivier and Adkins [*Amorphous Magnetism*, edited by H. O. Hooper and A. M. de Graaf (Plenum, New York, 1973), p. 215] obtained an analogous result to that of Held and Klein.[8] In their result m is a constant and thus agrees with our $T=0$ derivation only, whereas our solution is for all T. Similarly, Adkins and Rivier[26] obtain a cusp in the susceptibility. Their[26] cusp arises from introducing an average magnetization which exists only within a correlation length, thus being of definite short-range nature. In our derivation the cusp occurs because of long-range order in the spin-glass order parameter m.

B. Comparison with experiment

We have calculated the various moments of the distribution function given by Eq. (3.3) directly using a computer. A log-log plot of the moments as a function of the impurity concentration is shown in Fig. 1. Since the transition temperature is proportional to the first moment $\eta_1(0)$ it is interesting to examine its behavior. We find

$$\begin{aligned}&\eta_1(0)\approx 0.12(n_0aS)c^{2/3}, \quad 1\%<c<5\%\\ &\eta_1(0)\approx 0.38(n_0aS)c^{1/2}, \quad c>6\%.\end{aligned} \quad (3.19\text{a})$$

Similarly, we have

$$\eta_2(0)\approx 0.042(n_0aS)^2c, \quad 1\%<c<19\% \quad (3.19\text{b})$$

and

$$\begin{aligned}&\eta_3(0)\approx 0.024(n_0aS)^3c^{1.2}, \quad 1\%<c<4\%\\ &\eta_3(0)\approx 0.22(n_0aS)^3c^{1.5}, \quad c>5\%.\end{aligned} \quad (3.19\text{c})$$

For $1\%<c<5\%$ we have

$$T_c=\tfrac{1}{3}\,S(S+1)\,(0.120)\,(n_0a)c^{2/3}\,, \quad 1\%<c<5\%. \quad (3.20)$$

This concentration dependence of T_c is in reasonable agreement with the experiments of Mydosh[10] (who quotes $T_c\propto c^{\alpha_M}$, where $0.55<\alpha_M<0.75$ for $0.1\%<c<10\%$). For $c>6\%$ the character of the first moment becomes more Gaussian with $T_c \propto c^{1/2}$. The value of χ at $T=T_c$ is predicted from our model (taking $k=1$ for low c)

$$\chi(T_c)=N_0cP^2_{\text{eff}}/3k_BT_c\propto c^{1/3}\,, \quad 1\%<c<5\% \quad (3.21)$$

and the slope of the susceptibility just above T_c $(T=T_c^+)$ is

$$\frac{d\chi(T_c^+)}{dT}=-\frac{N_0cP_{\text{eff}}}{3k_BT_c^2}\propto c^{-1/3}\,, \quad 1\%<c<5\%. \quad (3.22\text{a})$$

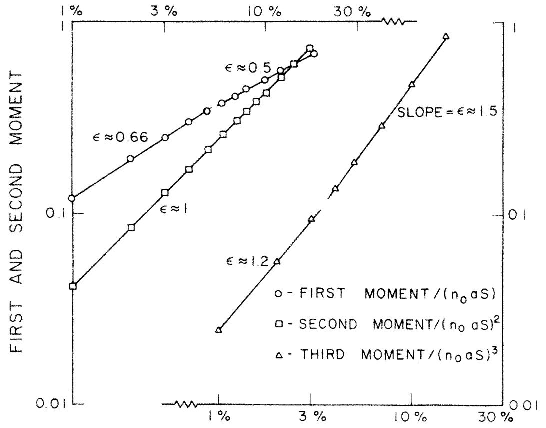

FIG. 1. Computer-calculated values of the first three moments of probability distribution as a function of the fractional impurity concentration c. Note the break in the slope ϵ of the first and third moments near $c=5\%$.

Using Eqs. (3.19) in Eq. (3.18) gives

$$\frac{d\chi(T_c^-)}{dT}=\frac{N_0 c P_{\text{eff}}}{3k_B T_c^2}(0.63c^{0.46}-1),\ 1\%<c<5\%. \tag{3.22b}$$

Thus the MRF model perdicts for the RKKY interaction that there is a discontinuous derivative of the susceptibility at $T=T_c$; however, the predicted slope of χ from our calculation continues to be negative at least for some c below T_c. Thus the MRF model predicts a maximum in χ for $1\%<c<2.5\%$ below T_c. The temperature of the maximum, $T_{\max}\propto c$ as was found in I. To our knowledge, such a maximum was not yet observed experimentally except possibly in the measurements of Lutes and Schmit,[19] but in this case[19] the magnetization was measured in appreciably large external fields, which suppresses the cusps as is discussed in Sec. III D.

For a Gaussian $P(H)$ we obtain that $d\chi(T_c^-)/dT=0$, in agreement with the results of SK[13] for the Gaussian. Whereas one expects reasonable agreement with experiment for the first moment and the transition temperature, it would be surprising if Eq. (3.22b) were to agree well with experiment since it involves three different quantities, $\eta_1(0)$, $\eta_2(0)$, and $\eta_3(0)$, the value of each of which is obtained only from the molecular-field approximation. Also, our powers of c were obtained from a graphical analysis, and the reader is cautioned not to jump to the conclusion that our exponents of c are rational numbers.

C. Low-temperature low-field magnetization

The expression for the low-temperature low-field magnetization for the spin-$\frac{1}{2}$ Ising system is given by Eq. (4.8) of Ref. 2. In this section we give the result for general spin S obtained by Fischer and Klein.[20] The expression for the total magnetization of $M(B)$ for $N=N_0c$ impurities, where N_0 is the total number of sites in the solid, B is the external field, is given by Eq. (15) of Ref. 20. For $\mu_B B\ll k_B T$ and $k_B T\ll\gamma c$, where γ is given by Eq. (3.9) we have

$$M(B,T)=\frac{N_0B(g\mu_B)^2}{\gamma}\left\{\frac{2}{\pi}+\frac{4}{\pi^2}\ln(2S+1)\left(\frac{k_BT}{\gamma cS}\right)+\left[\frac{4}{\pi}\left(\frac{2\ln(2S+1)}{\pi}\right)^2-\frac{4\pi}{3(2S+1)}\right]\left(\frac{k_BT}{\gamma cS}\right)^2\right\}+O(T^3). \tag{3.23}$$

For T near zero we have

$$M(B,0)\equiv\lim_{T\to 0}M(B)=NB(g\mu_B)^2/\gamma. \tag{3.24}$$

Thus the $T=0$ magnetization is predicted to be independent of impurity concentration and of the impurity spin and is inversely proportional to the RKKY interaction strength. For $T\neq 0$ the slope of the magnetization is proportional to T/c. This result as well as Eq. (3.24) has been found to agree with the experiments of Franz and Sellmyer[5] on dilute magnetic alloys.

The coefficient of the T^2 term gives the most information on the spin dependence of the impurity, since everywhere *else* in Eq. (3.24) only the product $S\gamma$ appears, thus S can only be determined when γ is known. We note that for small values of S the coefficient of the T^2 term is negative, but becomes positive for large values of S. Let $y=k_BT/\gamma cS$, we then define the quantity

$$t=\frac{\gamma}{2N\mu_BB}\frac{d^2M}{dy^2}=\frac{16}{\pi^3}[\ln(2S+1)]^2-\frac{4\pi}{3(2S+1)}, \tag{3.25}$$

we find that $t=-1.85$ for $S=\frac{1}{2}$, $t=-0.773$ for $S=1$, $t=-0.0563$ for $S=\frac{3}{2}$, $t=+0.50$ for $S=2$, and $t=+0.96$ for $S=\frac{5}{2}$. Thus for spin $\frac{3}{2}$ the curvature of the very-low-temperature susceptibility should be close to zero, whereas for spin $\frac{5}{2}$ the curvature is positive. We remark that Mydosh[9] found a positive curvature for Ag-Mn and a curvature close to zero for Au-Cr. Recent measurements of the susceptibility of Au-Fe by Guy[21] show clearly that for this system the curvature of χ below T_c is close to zero. This again is in agreement with our prediction if we assume that the spin of the Fe impurity dissolved in Au is $\frac{3}{2}$.

D. High-field low-temperature magnetization

As the external field B becomes very high it is expected that the magnetization of the system will reach its saturation value, provided $\mu_BB\gg k_BT$. However, because of the competing interaction in the RKKY potential there will be important deviation from saturation. The magnetization in the presence of large internal fields for a spin-$\frac{1}{2}$ Ising system has already been derived previously and is given by Eqs. (4.8) and (4.9) of Ref. 2. For a general spin S the low-temperature low-concentration magnetization is given in Eq. (16) of Ref. 20, and is

$$M=N_0 c g\mu_B S\left\{1-\frac{2\gamma S c}{\pi g\mu_B B}\left[1+\frac{2\pi^2}{3(2S+1)}\left(\frac{k_B T}{g\mu_B B}\right)^2\right]\right.$$
$$\left.+\ln(2S+1)\left(\frac{2}{\pi}\right)^2\left(\frac{\gamma c S}{g\mu_B B}\right)^2\left[\frac{k_B T}{g\mu_B B}+\frac{2\pi^2}{3(2S+1)}\left(\frac{k_B T}{g\mu_B B}\right)^3\right]+O(c^4T)+O(cT^4)\right\}. \qquad (3.26)$$

In order to compare with experiment, we substitute the value of γ in Eq. (3.26) and keeping terms to order c^2 *only* we have

$$M=N_0 c\mu_B S\left\{1-\frac{4}{3}\,\frac{\pi(an_0)cS}{g\mu_B B}\left[1+\frac{2\pi^2}{3(2S+1)}\left(\frac{k_B T}{g\mu_B B}\right)^2\right]\right\}. \qquad (3.27)$$

Recalling that a is the average RKKY interaction at a distance of one lattice constant, then

$$an_0=V_0\left(\frac{2}{\pi}\right)\int_0^{\pi/2}\cos x\,dx=\frac{2V_0}{\pi},$$

where V_0 is the strength of the interaction used by Larkin and Khmel'nitsky,[22] we obtain for spin $\frac{1}{2}$ the identical numerical value (but not the same S dependence) as Larkin.[22]

Equation (3.27) is to be compared with the experimental results of Hou and Coles.[15] We find that for spin $\frac{1}{2}$ our expression agrees exactly with Hou and Coles's second equation. However for spin $S\neq\frac{1}{2}$ our results are different from theirs. Since the spin was not varied in the Hou-Coles[15] experiment our Eq. (3.27) agrees, at least qualitatively, with their experiments on Ag-Mn.

Considering the term proportional to c^3 in Eq. (3.26) we find that this term gives an increase in the magnetization with temperature. At first this may seem contradictory to physical expectations, since one normally expects that as T increases the magnetization decreases. But a more careful analysis will show that our results make good physical sense. The spin-glass properties of the RKKY system arise because of the antiparallel nature of spin alignment resulting from the oscillating RKKY interaction. Thus two spins will, on the average, give zero contribution to the magnetization. However, a third spin, whose contribution will be proportional to c^3 in our self-consistent treatment, will tend to interfere with the antiparallel alignment and thus give an increase in the magnetization. It may be useful to perform experiments at somewhat higher concentrations than was done by Hou and Coles[15] to see whether our prediction is observed. Before leaving this subject we wish to comment on Smith's[23] comparison with our Eq. (3.27) as shown in Eq. (4.6) of Ref. 2. We obtain a term linear in T but proportional to c^3, whereas Smith's[23] term is proportional to c^2.

E. Magnetization and its derivative for finite fields

The cusp in the susceptibility is obtained only in the limit as the externally applied field is zero. However, when the magnetization and its derivative are measured in reasonably large fields, the order parameter m defined in Eq. (3.7) is nonzero for all T, and the cusp in χ is absent. Such is the case in the experiments of Lutes and Schmit.[19] In this case one observes a well-rounded maximum in the susceptibility. The behavior of the distribution function for large fields is unimportant for this case since m is always nonzero and $P(H)$ can be approximated by Eq. (3.9). This case was examined in detail in Refs. 1 and 2, where it was found that the temperature of the maximum in χ, $T_{\max}\propto c$. Similarly, $\chi(T_{\max})\propto c^0$ for very small c. The deviation of the susceptibility for $T>T_{\max}$ from its Curie-law behavior was found by Klein and Shen[24] to be approximately proportional to c^2/T^2 in agreement with the experiments of Lutes and Schmit[23] on Au-Fe and Au-Cr.

F. Heisenberg versus Ising model and the muon depolarization experiments on dilute alloys

The Ising model[1,25] gives an internal field where $P(H)$ is finite for $H=0$. The Heisenberg distribution of fields[8] gives, on the other hand, that $P(H)\to 0$ for $H\to 0$. The latter is consistent with polarized μ-meson experiments on dilute alloys by Fiory *et al.*[9] Fischer and Klein[20] have examined the detailed behavior of the high-B and the low-T magnetization of spin glasses in the simple Heisenberg and Ising models and found that in each case the Ising distribution gives results which are in excellent agreement with experiment, whereas the Heisenberg distribution always disagrees with experiment. Similarly the low-temperature specific-heat experiments agree with the Ising prediction and disagree with the Heisenberg-model results.[8] It was also shown recently by Riess and Klein[11] that the MRF approximation gives a well-defined transition temperature and cusps in the susceptibility of the spin-glass system.

These facts force us to give a new physical interpretation to the spin-glass state which indicates why the Ising-model calculation gives the correct properties of the system even though the fields are Heisenberg-like distributed.

At $T > T_c$ the order parameter m given by either Eq. (3.7) or Eq. (3.10) is zero. For $T < T_c$ the order parameter m is nonzero, which shows that the magnitudes of the spins become correlated to each other and become aligned in some complicated fashion (may be some complicated set of random spiral structures) and the direction of the local field at a particular site is random and varies from site to site. Thus if $\vec{u}(r)$ is a unit vector in the direction of the order parameter at site r $\vec{u}(r)$ is expected to vary from site to site. The internal field at site r will be $\vec{H}(r_i) = \sum v_{0j} m_j \vec{u}_j$ and will have a well-defined value and direction at site r. Next we want to consider the internal field distribution from two reference frames: (a) a direction fixed in the laboratory, as in the case of the polarized-muon experiment; (b) reference frame of any one impurity in the system. With respect to a fixed external direction, the fields are all oriented randomly (in direction as well as in magnitude) and the probability is similar to that found for the Heisenberg model.[8] This explains why the probability for zero fields is close to zero in the polarized μ-menson experiment. The sudden increase of the field as the temperature is lowered is, on the other hand, explained by the existence of the spin-glass transition.[11]

Next we consider the field distribution with respect to the local impurity. A spin-glass transition occurs at some temperature T_c, and the transition occurs in the Heisenberg- as well as in the Ising-model MRF approximation. Now in the Heisenberg model, the direction of the vector spin changes from site to site, however, each spin, regardless what its direction is locally, contributes to the buildup of the nonvanishing order parameter below T_c. At T near zero the spins are frozen-in in their random directions. The magnetization is a function of $\vec{r}$ and is a vector quantity $\vec{M}(\vec{r})$ depending upon the local position $\vec{r}$ such that $\int \vec{M}(\vec{r})\, d\vec{r} = 0$. Now consider the spin at $\vec{r} = \vec{r}_i$. $\vec{M}(\vec{r}_i)$ has a certain well-defined direction, because the field at $\vec{r}_i$ has a well-defined direction, the direction being dictated by the $N_0 c - 1$ other spins. The Heisenberg distribution was obtained[8] by assuming that each spin may be oriented in an arbitrary direction, thus resulting in a degeneracy of the field as a function of the angle, and thus the probability of the field H is proportional to $H^2\, dH$. When the system orders, some particular component $\vec{H}(\vec{k})$ of the vector field $\vec{H}$ predominates the system, presumably resulting in some complicated structure of the spins. Since the system is now ordered, the local spin at a particular site $\vec{r}_i, \vec{M}(\vec{r}_i)$, will not have an arbitrary direction of orientation (i.e., will not have all possible orientations with equal probability) but will be frozen-in in the direction determined locally by the order parameter. Thus when one calculates the probability distribution, the volume element will no longer be $H^2\, dH$, but rather closer to an Ising distribution. Clearly these arguments are hand-waving and *no proof* for a realistic system.

G. Low-temperature specific heat

That the MRF approximation gives a low-temperature specific heat C_v which is linear in T and independent of c has been discussed in detail in I. This argument still holds since the major contribution to the low-T specific heat comes from small internal fields, where Eq. (3.9) is the right approximation to $P(H)$ provided $k_B T/\Delta_1 \ll 1$.

Had we used the Gaussian, Eq. (3.10), to calculate C_v we would have obtained $C_v \propto Tc^{1/2}$ in disagreement with experiment. Similarly, the distribution used by EA[11] and SK[12] as well as the calculation of Fischer[27] should give a low-T specific heat proportional to $Tc^{1/2}$. The fact that the very-high-field contribution depends upon $\Delta_2 \propto c^{1/2}$ and results in a $T_c \propto c^{0.66}$, and that the low-temperature specific heat and magnetization depend upon small fields for which $\Delta_1 \propto c$, indicates that the scaling of the thermodynamic properties of the spin glass has a different concentration dependence for high T and low T.

H. Low-temperature resistivity

The low-temperature resistivity $\rho(T)$ was calculated in the molecular-field approximation by Harrison and Klein[28,29] using an Ising-like distribution of fields. They found that

$$\lim_{T\to 0}\left(\frac{d\rho}{dT}\right) \approx \frac{AScJ^2}{\Delta_1}\left[1+\frac{3J}{E_F}\left(2+\ln\frac{k_B}{4E_F}\right)\right] , \tag{3.28}$$

and since Δ_1 is proportional to c, Harrison and Klein obtain that the deviation of the resistivity from its $T = 0$ value is independent of the impurity concentration and is proportional to the temperature. Recent measurements of the resistivity by Ramos[14] agree with this prediction of the molecular-field model.

IV. CONCLUSION

We find that the mean-random-molecular-field approximation used with the Gaussian-distribution exchange potentials of SK[13] gives identical magnetic properties to the n expansion of EA[12] and SK.[13] The very-low-T specific heat is linear in T as in the n expansion, however, at high temperatures the n expansion gives a specific heat C_v proportional to T^{-2}, whereas $C_v = 0$ for $T > T_c$ from the MRF approximation.

The MRF approximation applied to the RKKY system gives the following properties in excellent agreement with experiment: (i) the concentration and temperature dependence of the low-temperature specific heat; (ii) the concentration and temperature dependence of the low-T low-field and high-field magnetization; (iii) the low-temperature susceptibility; (iv) the T^{-1} dependence of the susceptibility for high T. Further experiments which are in good qualitative agreement with the MRF approximation are (v) the concentration dependence of the temperature of the cusp in the susceptibility; (vi) the spin dependence of the curvature of the low-T magnetization; (vii) the concentration and temperature dependence of the low-T resistivity; (viii) the concentration and temperature dependence of dM/dH in reasonably large fields; (ix) finally, the data on the μ-meson polarization experiment in dilute alloys seem to be consistent with the MRF approximation.

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