Molecular-Field Theory of a Random Ising System in the Presence of an External Magnetic Field

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The thermodynamic properties of very dilute concentrations of magnetic impurities in a nonmagnetic system and in the presence of an externally applied magnetic field are examined, using a mean-random-field approximation. The impurities are assumed to interact via a long-range potential which alternates in sign as a function of the position between the magnetic ions. A modified form of Margenau's statistical model is used to obtain the probability distribution $P(\bar{H})$ of the random internal exchange fields $\bar{H}$ in an Ising model. The magnetization and the specific heat are obtained as integrals over the distribution of internal fields. The variation of the magnetization and the low-temperature specific heat as a function of the external magnetic field is obtained for all temperatures. The model, when applied to a dilute alloy system, shows that the excess very-low-temperature specific heat is strongly decreased by the applied magnetic field H_{ext}. The very-low-temperature magnetization per impurity is predicted to be proportional to $\tan^{-1}(H_{\text{ext}}/\Delta)$, where Δ is a temperature- and external-field-dependent width of the probability distribution.

I. INTRODUCTION

THE concept of a random molecular field was introduced by Marshall[1] to obtain the low-temperature specific heat of dilute copper-manganese. The idea behind this random-molecular-field method is as follows: Consider a set of magnetic impurities which are randomly distributed in a nonmagnetic host and which interact via a long-range interaction potential. Each impurity is surrounded by different environment and, therefore, experiences a different effective Weiss molecular field. Since the positions of the impurities are random variables, so is the Weiss molecular field. The objective of the method is to obtain the probability distribution of this random molecular field $P(\bar{H})$, and then to obtain the thermodynamic variables of the system by integrating the expression for the thermodynamic variable of a single spin in a fixed molecular field $\bar{H}$ over the distribution of all fields. Other investigators[2–4] have used the random-molecular-field method to treat various magnetic impurity problems. Most of these approaches have used an Ising model or some other classical model to treat the spin system. They have the deficiency that it is difficult to obtain the probability distribution of the field at all temperatures in a closed form. For this reason, even though the behavior of the random system could be relatively well approximated near $T=0$, it was difficult to obtain even qualitative predictions of the specific heat, the magnetization, and magnetic susceptibility for higher temperatures. To study the temperature dependence of a random Ising system interacting via a Ruderman-Kittel-Yosida[5–7] potential, the author has recently[8] (this paper shall henceforth be referred to as I) used a modified form of Margenau's statistical model to obtain $P(\bar{H})$ for all temperatures in the limit as the impurity concentration approaches zero. In order to obtain analytical expressions for $P(\bar{H})$, the Ruderman-Kittel potential has been replaced by one in which each impurity experiences a random potential of value $\pm v(r_{ij})$, each with probability $\frac{1}{2}$, where $v(r_{ij})=a/r_{ij}^3$, where a is the strength of the interaction and r_{ij} is the distance between the impurities at site i and j. An approximation was also used in which when calculating the field distribution at a particular impurity site, functions of the random fields at other impurity sites are replaced by their mean values. This approximation was called the mean-random-field (MRF) approximation. Using a self-consistency condition with the MRF approximation gave the temperature-dependent probability distribution in a closed form, where the width of the probability distribution was given by an integral equation which was relatively simple to evaluate for some temperatures. The $P(\bar{H})$ derived there gave a low-temperature magnetic susceptibility which is independent of the impurity concentration, and a maximum in the susceptibility as a function of temperature where the temperature of the maximum is proportional to the impurity concentration. Similarly, the very-low-temperature specific heat from the model was independent of the impurity concentration and was approximately linear in temperature for very low temperatures. Even though the results are consistent with the behavior of magnetic

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impurities in nonmagnetic metals[9–12] in the intermediate concentration range, it is not clear whether the theoretical predictions obtained from an Ising model are applicable to a dilute alloy system. Thus, we have reviewed the random Ising model in the absence of externally applied fields.

As far as the author knows, no treatment of the random Ising model in the presence of an externally applied field has been made. The purpose of this paper is to consider the variation of the probability distribution of the internal field, the magnetization, and the specific heat with an externally applied field in the MRF approximation. The results obtained here will hopefully motivate experimentalists to measure the thermodynamic quantities and compare them with the Ising-model predictions.

The method used follows closely that of I and may be considered an extension thereof. In Sec. II, it is shown in the MRF approximation that the application of the externally applied field shifts the probability distribution from being symmetric about a field $\bar{H}=0$ to $\bar{H}=H_{\text{ext}}$, where H_{ext} is the externally applied field. The width of the probability distribution also becomes external-field-dependent. In Secs. II to V the variation of the probability distribution of the field, the specific heat, and the magnetization as a function of the temperature and the external field are examined for all temperatures. It is found that the very-low-temperature specific heat is strongly affected by H_{ext} and is approximately proportional to $\Delta/(\Delta^2+H_{\text{ext}}^2)$, where Δ is the width of the probability distribution. For very low temperatures, Δ is found to be proportional to impurity concentration. The predicted low-temperature magnetization per impurity is approximately proportional to $\tan^{-1}(H_{\text{ext}}/\Delta)$. It is proposed that useful information on the internal-field distribution in dilute alloys could possibly be obtained from a Mössbauer experiment at relatively high temperatures.

II. MATHEMATICAL DEVELOPMENTS

Consider N Ising-model spins randomly and uniformly distributed over N_0 sites in a nonmagnetic system. Let $N\to\infty$, $N_0\to\infty$, such that $N/N_0=c$, the fractional impurity concentration. Let μ_i and μ_j be the Ising-model spin variables at sites r_i and r_j, respectively, and let v_{ij} be the interaction potential between two impurities at sites r_i and r_j, respectively. Let H_{ext} be the externally applied magnetic field. Then the interaction Hamiltonian $\mathcal{H}$ of the system in an Ising model, i.e., a model in which μ_i may take values of ±1 only, is

$$\mathcal{H}=\sum_{i,j} v_{ij}\mu_i\mu_j+\sum_i H_{\text{ext}}\mu_i\,, \tag{2.1}$$

where the potential v_{ij} is assumed to have the form

$$\begin{aligned} v_{ij}&=a/r_{ij}^3\,, &&\text{with probability of } \tfrac{1}{2}\\ v_{ij}&=-a/r_{ij}^3\,, &&\text{with probability of } \tfrac{1}{2} \end{aligned} \tag{2.2}$$

just like in I. This potential is chosen as an approximation to the Ruderman-Kittel potential for very dilute concentration of impurities.

Let the total field at an arbitrary impurity site i be h_i. Then h_i is made up of two parts, the externally applied field H_{ext} and the internal exchange field $\bar{H}$. Thus,

$$h_i=\sum_j v_{ij}\bar{\mu}_j+H_{\text{ext}} \tag{2.3}$$

$$\equiv\bar{H}_i+H_{\text{ext}}\,, \tag{2.4}$$

where $\bar{H}_i$ is the Weiss molecular field experienced by an impurity at site i. The horizontal bars over $\bar{\mu}_i$ and $\bar{H}_i$ indicate that these are thermally averaged quantities consistent with the usual definition of the molecular-field approximation. $\bar{\mu}_j$ is the thermal average value of the Ising spin operator μ_j in an effective field $\bar{H}_j$, where $\bar{H}_j$ is also a random variable. The probability distribution of $\bar{H}_j$ still remains to be determined, and this will be done later on. For the moment, it is assumed that such a field $\bar{H}_j$ exists and the specification of its form is left for later. Then,

$$\bar{\mu}_j=\sum_{\mu_j=\pm1}\mu_j e^{\beta h_j\mu_j}\Big/\sum_{\mu_j=\pm1}e^{\beta h_j\mu_j}=\tanh\beta h_j\,. \tag{2.5}$$

The first objective of this paper is to obtain the probability distribution of the random internal field h_0 at an arbitrary origin 0. In the remaining part of this section we closely follow Sec. II of Paper I.

The formal expression for the probability distribution is obtained using a modified form of Margenau's statistical model,[13] i.e.,

$$P(h_0)=\sum_{v_N}\int_{r_N}\delta(h_0-H_{\text{ext}}-\sum v_{0j}\bar{\mu}_j)\times P(r_{01},r_{02},\cdots,r_{0N})d^3r_{0N}\,, \tag{2.6}$$

where $\sum_{v_N}$ is the sum over each of the potentials $v_{0j}=\pm a/r_{0j}^3$ given in Eq. (2.2) and $\int_{r_N}$ indicates an integral over a $3N$ dimensional volume and $P(r_{01}\cdots r_{0N})\times d^3r_{0N}$ is the joint probability for particle 1 to be in the volume d^3r_{01} at r_1, particle 2 to be in d^3r_{02} at r_2, and particle N to be in d^3r_{0N} at r_N. Rather than summing over discrete sites in the lattice, integration is used. This approximation is reasonable for very dilute concentrations of impurities where the average distance between the impurities is much greater than the lattice

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constant. Upon assuming that the positions of the particles are independent random variables uniformly distributed over the volume V with probability $1/V$ and using Eq. (2.2) in Eq. (2.6) gives

$$P(h_0)=(2\pi)^{-1}\int_{-\infty}^{\infty} d\rho\, e^{i\rho(h_0-H_{\text{ext}})}\int d^3r_1 \times\int d^3r_2\cdots\int d^3r_N \prod_{j=1}^{N-1}\frac{\cos(\rho a\bar{\mu}_j/r_{0j}^3)}{V}. \quad (2.7)$$

Equation (2.7) gives the probability distribution of the field h_0 at site 0 in terms of functions of the total field at site j. The right-hand side of Eq. (2.7) involves $\bar{\mu}_j=\tanh\beta h_j$, where in principle h_j is a random variable. h_j itself may be written as

$$h_j=\sum_k v_{jk}\bar{\mu}_k+H_{\text{ext}}, \quad (2.8)$$

where $\bar{\mu}_k$ is a function of the total field at site k. Thus, the arguments of the spin variables $\bar{\mu}_j$ are position-dependent, which makes the evaluation of the integral in Eq. (2.7) difficult, and using Eq. (2.8) in Eq. (2.7) brings one no closer towards the solution of Eq. (2.7). Equation (2.8) is only exhibited to show the structure of the equations obtained. In order to solve Eq. (2.7), a so-called mean random field (MRF) approximation is used, in which when solving for the field distribution at site 0, functions of the fields at all other sites than 0 are replaced by their average values, i.e.,

$$\cos\frac{\rho a\bar{\mu}(h_j)}{r_{0j}^3}\xrightarrow{\text{MRF}}\int_{-\infty}^{\infty}P(h_j)\cos\frac{\rho a\bar{\mu}(h_j)}{r_{0j}^3}dh_j, \quad (2.9)$$

where the arrow with MRF indicates that in the MRF approximation $\cos[\rho a\bar{\mu}(h_j)/r_{0j}^3]$ is replaced by its average value over a probability distribution of the fields h_j. Thus, Eq. (2.7) becomes

$$P(h_0)=(2\pi)^{-1}\int_{-\infty}^{\infty} d\rho\, e^{i\rho(h_0-H_{\text{ext}})}\prod_{j=1}^{N-1}\int_{-\infty}^{\infty}P(h_j)dh_j \times\int_V d^3r_{0j}\frac{\cos[\rho a\bar{\mu}(h_j)/r_{0j}^3]}{V}. \quad (2.10)$$

Next, as a self-consistency condition, the requirement is imposed that $P(h_j)$ be independent of the site j under consideration. Then, Eq. (2.10) becomes

$$P(h)=(2\pi)^{-1}\int_{-\infty}^{\infty} d\rho\, e^{i\rho(h-H_{\text{ext}})}\left(\int_{-\infty}^{\infty}\frac{1}{V}P(h)dh\int_V d^3r \times\cos\frac{\rho a\bar{\mu}(h)}{r^3}\right)^{N-1}. \quad (2.11)$$

Let

$$V'=\int_V d^3r\int_{-\infty}^{\infty}P(h)dh\left[1-\cos\frac{\rho a\bar{\mu}(h)}{r^3}\right], \quad (2.12)$$

then Eq. (2.11) becomes[8]

$$P(h)=(2\pi)^{-1}\int_{-\infty}^{\infty} d\rho\, e^{i\rho(h-H_{\text{ext}})-n_0V'(\rho)c}, \quad (2.13)$$

where n_0 is the number of sites per unit cell, $n_0=4$ for an fcc lattice, V' is defined by Eq. (2.12), and c is the fractional impurity concentration. For a $1/r^3$ potential, V' becomes

$$V'=\tfrac{2}{3}\pi^2|\rho a|\times\|\bar{\mu}\|, \quad (2.14)$$

where

$$\|\bar{\mu}\|=\int_{-\infty}^{\infty}P(h)|\tanh\beta h|dh, \quad (2.15)$$

where the single vertical bar in Eqs. (2.14) and (2.15) indicate absolute values. Substituting Eq. (2.15) into Eq. (2.13) gives

$$P(h)=\frac{1}{\pi}\frac{\Delta(\beta,H_{\text{ext}},c)}{[\Delta(\beta,H_{\text{ext}},c)]^2+[h-H_{\text{ext}}]^2}, \quad (2.16)$$

where

$$\Delta(\beta,H_{\text{ext}},c)=\tfrac{2}{3}\pi^2|a|n_0c\|\bar{\mu}(\beta,H_{\text{ext}},c)\|\equiv\gamma c\|\bar{\mu}\|. \quad (2.17)$$

Equation (2.16) shows that in the MRF approximation the effect of the external field is (a) to shift the center of the probability distribution from $h=0$ to $h=H_{\text{ext}}$ and (b) to modify the width Δ of the distribution function. From Eq. (2.4), one finds that

$$\bar{H}=h-H_{\text{ext}}, \quad (2.18)$$

where in Eq. (2.18) the index i is again suppressed because of the self-consistency requirement. Using Eq. (2.18) in Eq. (2.16) gives

$$P(h)=\frac{1}{\pi}\frac{\Delta(\beta,H_{\text{ext}},c)}{[\Delta(\beta,H_{\text{ext}},c)]^2+\bar{H}^2}\equiv P(\bar{H}). \quad (2.19)$$

Equation (2.19) coupled with Eqs. (2.15) and (2.17) gives an integral equation for the width of the probability distribution Δ. Using Eq. (2.19), one finds that

$$\int_{-\infty}^{\infty}P(h)F(h)dh=\int_{-\infty}^{\infty}P(\bar{H})F(\bar{H}+H_{\text{ext}})d\bar{H}, \quad (2.20)$$

where $P(\bar{H})$ is given in Eq. (2.19). Equation (2.20) allows one to evaluate all thermodynamic functions for the system by integrating over the random variable $\bar{H}$ and the probability distribution $P(\bar{H})$ defined in Eq. (2.19). In particular, the width of the distribution function Δ may be obtained by solving the integral equation

$$\|\bar{\mu}\|=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\Delta}{\Delta^2+\bar{H}^2}|\tanh\beta(\bar{H}+H_{\text{ext}})|d\bar{H}, \quad (2.21)$$

where the relationship between Δ and $\|\bar{\mu}\|$ is given in Eq. (2.17).

III. WIDTH OF THE DISTRIBUTION FUNCTION

A. Evaluation of Δ for Low Temperatures

The width of the probability distribution in Eq. (2.21) depends upon the temperature, the impurity concentration, and a parameter a, given in Eq. (2.2), which characterises the strength of the impurity-impurity interaction. Let γc in Eq. (2.17) be $\Delta(\infty)$. It will be shown later on that $\Delta(\infty)$ is the width of the probability distribution as $\beta\rightarrow\infty$. Let

$$\bar{H}/\Delta(\infty)=y,\quad H_{\rm ext}/\Delta(\infty)=y_0,\quad \beta\Delta(\infty)=\alpha,\tag{3.1}$$

and let

$$P^{\pm}(y)=\frac{1}{\pi}\frac{\|\bar{\mu}\|}{\|\bar{\mu}\|^2+(y\pm y_0)^2},\tag{3.2}$$

where the variables $H_{\rm ext}$ and c have been suppressed in the expression for Δ, i.e., $\Delta\equiv\Delta\ (c,\beta,H_{\rm ext})$ in Eq. (3.1). Using Eqs. (3.1) and (3.2) in Eq. (2.21) gives

$$\|\bar{\mu}\|=\int_0^\infty |\tanh\alpha y|[P^+(y)+P^-(y)]dy.\tag{3.3}$$

Writing

$$\tanh\alpha y=1-\frac{2e^{-2\alpha y}}{1+e^{-2\alpha y}}=1+2\sum_{k=1}^{\infty}(-1)^k e^{-2k\alpha y}$$

in Eq. (3.3) gives

$$\|\bar{\mu}\|=1+2\sum_{k=1}^{\infty}(-1)^k\int_0^\infty e^{-2k\alpha y}\times[P^+(y)+P^-(y)]dy.\tag{3.4}$$

For very low temperatures $P^+(y)$ and $P^-(y)$ in Eq. (3.4) may be expanded in the form

$$P^{\pm}(y)\approx\frac{\|\bar{\mu}\|}{\pi[\|\bar{\mu}\|^2+y_0^2]}\times\left(1-\frac{y(y\pm 2y_0)}{\|\bar{\mu}\|^2+y_0^2}+\frac{[y(y\pm 2y_0)]^2}{[\|\bar{\mu}\|^2+y^2]^2}\right).\tag{3.5}$$

Using Eq. (3.5) in Eq. (3.4) gives

$$\|\bar{\mu}\|\approx 1-\frac{2\ln 2\|\bar{\mu}\|}{\pi\alpha[\|\bar{\mu}\|^2+y_0^2]}+\frac{4\|\mu\|(3y_0^2-\|\mu\|^2)}{\pi[\|\bar{\mu}\|^2+y_0^2]^3}\times\sum_{k=1}^{\infty}(-1)^k\int_0^\infty y^2e^{-2\alpha k y}dy\tag{3.6}$$

$$=1-\frac{2\ln 2\|\bar{\mu}\|}{\pi\alpha[\|\bar{\mu}\|^2+y_0^2]}-\frac{\|\mu\|(3y_0^2-\|\mu\|^2)}{2\pi[\|\bar{\mu}\|^2+y_0^2]^3\alpha^3}\sum_{k=0}^{\infty}(-1)^k\frac{1}{(k+1)^2}+O\left(\frac{1}{\alpha^5}\right).\tag{3.7}$$

In the limit as $\alpha=\beta\Delta(\infty)\rightarrow\infty$, Eq. (3.7) gives

$$\lim_{\alpha\to\infty}\|\bar{\mu}\|=1.\tag{3.8}$$

Combining Eq. (2.17) with Eq. (3.8) gives

$$\lim_{\beta\to\infty}\Delta\equiv\Delta(\infty)=\gamma c.\tag{3.9}$$

Using the relation $\Delta=\gamma c\|\bar{\mu}\|=\Delta(\infty)\|\bar{\mu}\|$ given in Eq. (2.17) and solving Eq. (3.7) for $\|\bar{\mu}\|$ at very low temperatures gives

$$\|\bar{\mu}\|=1-\frac{2\ln 2}{\pi\beta\Delta(\infty)(1+y_0^2)}-\left(\frac{2\ln 2}{\pi\beta\Delta(\infty)(1+y_0^2)}\right)^2\tag{3.10}$$

$$=1-q/\beta-q^2/\beta^2,\tag{3.11}$$

where

$$q=2\ln 2/\pi\Delta(\infty)(1+y_0^2).\tag{3.12}$$

It is interesting to compare Eq. (3.12) with the corresponding Eq. (3.9) of I. It is found that up to order T^3 the very low-temperature expression for $\|\bar{\mu}\|$ in the presence of an external field may be obtained by replacing $[\pi\Delta(\infty)]^{-1}$ in Eq. (3.9) of I by $[\pi\Delta(\infty)]^{-1}(1+y_0^2)^{-1}$.

B. Evaluation of Δ for High Temperatures

Equation (2.21) may be rewritten

$$\|\bar{\mu}\|=\int_{-\infty}^{\infty}P(y)|\tanh\alpha(y+y_0)|dy$$

$$=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{\beta\Delta}{(\beta\Delta)^2+x^2}|\tanh(x+x_0)|dx,\tag{3.13}$$

where $x=\beta\bar{H}$, $x_0=\beta H_{\rm ext}$. For very high temperatures $\alpha y_0=\beta H_{\rm ext}\ll 1$, and

$$\tanh(x+x_0)=\tanh x_0\,{\rm sech}^2x+\tanh x\,{\rm sech}^2x_0+O(\tanh^3x_0)\tag{3.14}$$

and

$$\lim_{\beta\Delta\to 0}\frac{1}{\pi}\frac{\beta\Delta}{(\beta\Delta)^2+x^2}\rightarrow\delta(x).\tag{3.15}$$

Using Eqs. (3.14) and (3.15) in Eq. (3.13) gives

$$\lim_{\beta\Delta\to 0}\|\bar{\mu}\|\approx\tanh x_0\int_{-\infty}^{\infty}\delta(x)\,{\rm sech}^2x\,dx+{\rm sech}^2x_0\int_{-\infty}^{\infty}\delta(x)|\tanh x|dx$$

$$\approx\tanh\beta H_{\rm ext}.\tag{3.16}$$

Thus, for very high temperatures when $\beta H_{\rm ext}\ll 1$ and $\beta\Delta\rightarrow 0$, $\|\bar{\mu}\|\approx\tanh\beta H_{\rm ext}$.

The physical meaning of Eq. (3.16) is as follows. At very high temperatures the width of the distribution function goes to zero when no external field is applied to the system as is found in Eq. (3.12) of Ref. 8. The reason for this is that the width of the distribution function is proportional to the thermal average value of the (average) magnetic moment $||\bar{\mu}||$ which goes to zero because of thermal fluctuation at high temperatures.

As an external field is applied the width of the distribution function increases approximately proportionally to $\tanh\beta H_{ext}$. Thus, the broadening increases with increasing fields. This broadened distribution of fields should reflect itself in the conduction electron polarization of a metal as well as in the broadening of the paramagnetic resonance line and the Mössbauer experiment on the hyperfine field at the nucleus of the magnetic ion. See further discussion of the Mössbauer effect in Sec. V.

It should be remarked that the approximation that $\beta\Delta \rightarrow 0$ has made it relatively easy to evaluate Eq. (3.13) and resulted in Eq. (3.16). However, at intermediate temperatures where more detailed calculations for $||\bar{\mu}||$ are needed, these can in principle be obtained from Eqs. (2.16) and (3.3). The integral equation for $||\bar{\mu}||$, Eq. (3.3), was solved using a computer for several values of the external magnetic field. The results are shown in Fig. 1, which gives $||\bar{\mu}||$ as a function of $\Delta(\infty)/k_BT=\alpha$ for several values of y_0.

IV. APPLICATIONS

A. Expression for Specific Heat

Before the expression for the low-temperature specific heat C_v is obtained it will be argued that the specific heat should be obtained as a derivative of the entropy, rather than the energy, for the system.

The idea of the probability distribution of the field conceptually considers the whole system made up of a large number of independent subsystems, with the spin of an impurity at an arbitrary center of each of the subsystems, say i, having a well-defined internal field $\bar{H}_i$. Then the energy associated with a spin at the center of subsystem i, U_i is

$$U_i = -\bar{H}_i \tanh\beta\bar{H}_i, \tag{4.1}$$

where the field is given in units of energy. The total energy for the whole system is then

$$U = -\sum_i \bar{H}_i \tanh\beta\bar{H}_i, \tag{4.2}$$

where in Eq. (4.1) the following implicit assumptions are present: (a) It is assumed that surface interactions between subsystems are negligible. (b) It is assumed that the internal fields of the different impurities are independent of each other. (c) It is assumed that the energy of the impurities is the additive sum of N terms. This results in an overcounting of the number of terms in Eq. (4.2) by a factor of 2. The latter is usually corrected for by introducing a factor of $\frac{1}{2}$ multiplying[14] Eq. (4.2). The normalization of the probability distribution introduces a constraint between the internal fields of the various subsystems, thus the energy of the individual impurities (subsystems) is not in general an additive quantity. In order to evaluate the specific heat C_v the physical requirement is imposed that the entropy of the individual subsystems (impurities) shall be additive (i.e., an extensive quantity) and C_v will be obtained as a derivative of the entropy. However, to make the specific heat of the system be the same as that obtained from the molecular-field approximation when the impurities are independent[14] a factor of $\frac{1}{2}$ is introduced in the expression for the specific heat arising from the entropy.

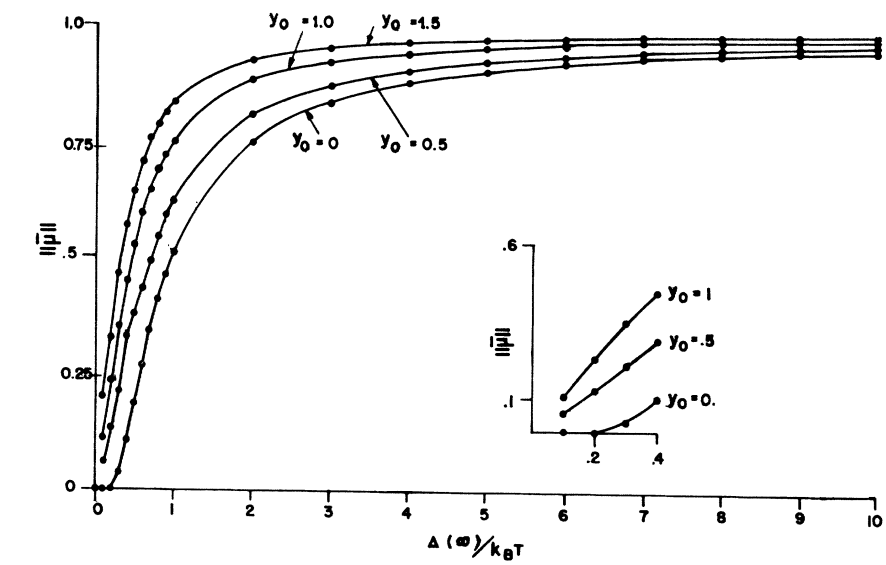

FIG. 1. Graph for $||\mu(x)||$ as a function of $x=\beta\Delta(\infty)$ for several values of $y_0=H_{ext}/\Delta(\infty)$, where H_{ext} is the externally applied field $\beta=(k_BT)^{-1}$, and $\Delta(\infty)$ is the width of the probability distribution as $\beta\rightarrow\infty$ and $H_{ext}\rightarrow 0$.

[14] See, for example, Ref. 1.

It is shown in Appendix B, Eq. (B10) that the expression for the specific heat C_v is

$$C_v=\frac{N_0ck_B}{2\alpha}\int_0^\infty(\alpha y)^2[P^+(y)+P^-(y)]\times\operatorname{sech}^2\alpha y\left(1+\frac{\beta\Delta'}{\Delta}\right)d(\alpha y)+\frac{\beta\Delta'}{\Delta}(\alpha y_0)\times\int_0^\infty(\alpha y)[P^+(y)-P^-(y)]\operatorname{sech}^2\alpha y\,d(\alpha y)\,, \qquad (4.3)$$

where α, y, y_0, and $P^\pm(y)$ are defined in Eqs. (3.1) and (3.2) and $\beta\Delta'/\Delta$ is given by Eq. (A5) of Appendix A, where $\Delta'=(d\Delta/d\beta)$.

B. Evaluation of Very Low-Temperature Specific Heat

For very low temperatures, the expression for $P^\pm(y)$ in Eq. (3.5) coupled with the expression for $\|\bar{\mu}\|$ given in Eq. (3.10) is used in Eq. (4.3) to give

$$C_v=\frac{N_0ck_B\pi T}{12\Delta(\infty)(1+y_0^2)}\left[1+\frac{k_BT}{\Delta(\infty)}\left(\frac{2\ln4(1-y_0^2)}{\pi(1+y_0^2)^2}\right)-\left(\frac{k_BT}{\Delta(\infty)}\right)^2\left(\frac{7\pi^2}{20}\frac{(1-3y_0^2)}{(1+y_0^2)^2}-\frac{(5-7y_0^2)(\ln4)^2}{\pi^2(1+y_0^2)^4}\right)+O(T^3)\right]. \qquad (4.4)$$

The fact that the coefficient of T^2 term in Eq. (4.4) is positive shows that for zero applied field C_v/T has a slight maximum as a function of T. This has been discussed previously.[8] Equation (4.4) also shows that

$$\lim_{T\to0}(C_v/T)\propto1/(1+y_0^2)\,. \qquad (4.5)$$

Thus, for very low temperatures, the specific heat decreases with increasing applied field according to Eq. (4.5). The Lorentzian shape is not expected to be valid for very large applied fields, i.e., $H_{\text{ext}}\gg\Delta$. In fact, the probability distribution should be a truncated Lorentzian when one considers the physical restriction imposed on the integral Eq. (2.10), that the distance of closest approach between the impurities shall be a near-neighbor distance. One can now solve Eq. (4.4) to find the temperature at which the specific heat is a maximum. Let $T_{\max}(0)$ be the temperature of the maximum when $H_{\text{ext}}=0$, and let $T_{\max}(y_0)$ be the temperature of the maximum as a function of $y_0=H_{\text{ext}}/\Delta(\infty)$. $T_{\max}(0)$ was found previously[8] to be approximately given by $k_BT_{\max}(0)\approx\Delta(\infty)(21\pi^2/20)^{-1/2}\approx\Delta(\infty)/\pi$. In the presence of small external fields such that $y_0=H_{\text{ext}}/\Delta(\infty)\ll1$, the temperature of the maximum is found from Eq. (4.4) to be approximately given by

$$k_BT_{\max}(y_0)\approx[\Delta(\infty)/\pi](1+2.5y_0^2)\approx k_BT_{\max}(0)(1+2.5y_0^2)\,. \qquad (4.6)$$

Equation (4.6) gives that the temperature of the maximum increases with increasing external fields. Equation (4.6) is only valid when $H_{\text{ext}}\ll\Delta(\infty)$ and should therefore be used with caution in interpreting experiments at low concentrations. Using a computer, the variation of the specific heat as a function of tem-

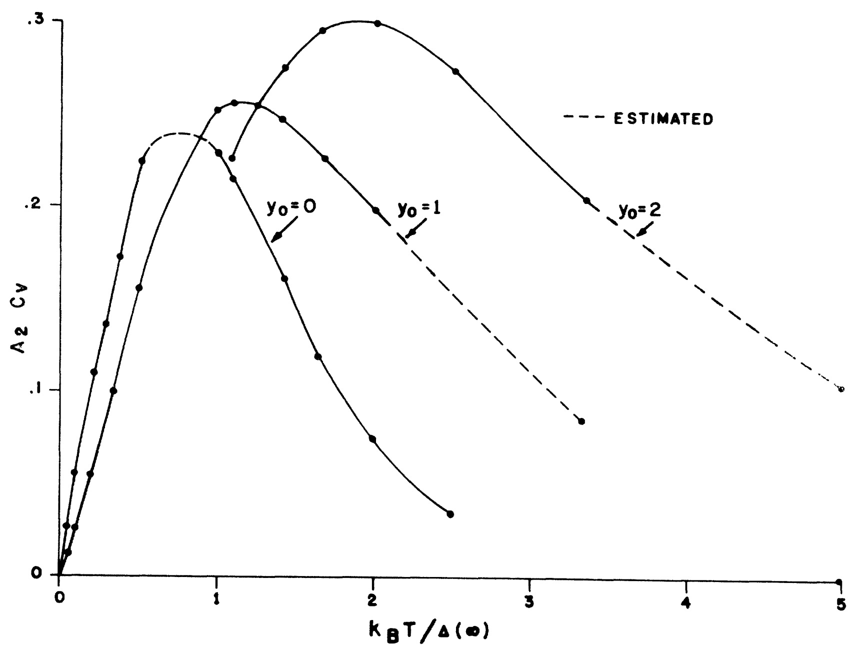

FIG. 2. Calculated results for the low-temperature specific heat as a function of $[\beta\Delta(\infty)]^{-1}=k_BT/\Delta(\infty)$ for several values of $y_0=H_{\text{ext}}/\Delta(\infty)$, where H_{ext} is the externally applied field and $\Delta(\infty)$ is the width of distribution function as $\beta\to\infty$ and $y_0\to0$. $A_2=(N_0ck_B\pi/12)^{-1}$, where N_0 is the number of sites per unit volume and c is the fractional impurity concentration.

perature for several applied fields has been calculated by solving Eq. (4.3) as a function of $\Delta(\infty)$, c, T. The results are shown in Fig. 2. Figure 3 shows the calculated values of the specific heat divided by T as a function of temperature for several values of the magnetic fields. Figure 4 shows the values of C_v as a function of applied fields for several temperatures. It is interesting to note that the maximum in C_v/T increases with magnetic field. The reason for this is that the maximum height of $P(\bar{H})$ is shifted from zero field to the field $H_{\rm ext}$, causing the probability of finding an average spin in very small fields (small energies) to decrease.

C. Magnetization

Next, we obtain the expression for the magnetization $M_1(h,\beta)$ for a single impurity in an effective field

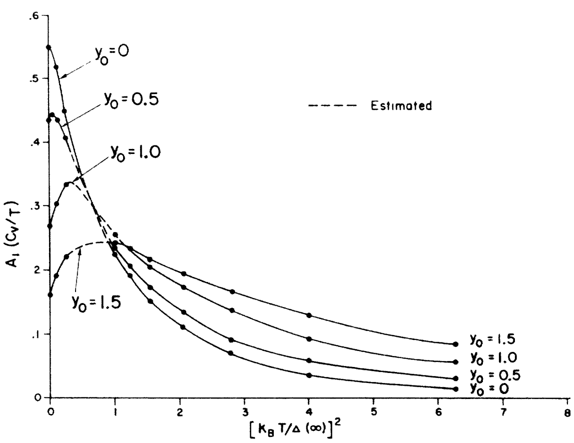

FIG. 3. Calculated results for C_v/T, where C_v is the specific heat at a constant field as a function of $k_BT/\Delta(\infty)$ for several values of $y_0=H_{\rm ext}/\Delta(\infty)$, where $H_{\rm ext}$ is the external field and $\Delta(\infty)$ is the width of the probabilities distribution as $\beta\to\infty$, $y_0\to 0$. $A_1=[N_0ck_B{}^2\pi/12\Delta(\infty)]^{-1}$.

$h=\bar{H}+H_{\rm ext}$:

$$M_1(h,\beta)=\mu_B\tanh\beta(H_{\rm ext}+\bar{H}) =\mu_B\tanh\alpha(y+y_0)\,. \tag{4.6'}$$

The total magnetization is obtained by averaging over the internal fields of the impurities using the probability distribution of the fields given in Eq. (2.19). Thus,

$$M=N_0c\mu_B\int_{-\infty}^{\infty}P(y)\tanh\alpha(y+y_0)dy =N_0c\mu_B\left(\frac{2}{\pi}\tan^{-1}\left(\frac{H_{\rm ext}}{\Delta}\right) -2\int_0^\infty\frac{e^{-2\alpha y}}{1+e^{-2\alpha y}}[P^-(y)-P^+(y)]dy\right), \tag{4.7}$$

where $P^{\pm}(y)$ is given by Eq. (3.5) and $P(y)$ is $P^{\pm}(y)$

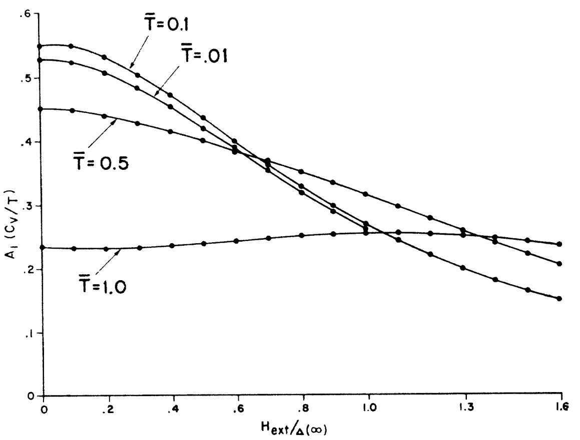

FIG. 4. Calculated values for C_v/T, where C_v is the specific heat as a function of the external field $H_{\rm ext}$ for several values of $\bar{T}$, where $\bar{T}=k_BT/\Delta(\infty)$ and $\Delta(\infty)$ is the width of the distribution function at $\beta\to\infty$ and $y_0\to 0$. A_1 is defined in the caption for Fig. 2.

with $y_0=0$. Equation (4.7) is easily evaluated for low temperatures; one obtains

$$M=N_0c\mu_B\left[\frac{2}{\pi}\tan^{-1}\left(\frac{H_{\rm ext}}{\Delta}\right) -\frac{H_{\rm ext}/\Delta}{(\beta\Delta)^2[1+(H_{\rm ext}/\Delta)^2]^2}\frac{\pi}{6}+O\left(\frac{1}{\beta\Delta}\right)^4\right]. \tag{4.8}$$

Using the relation $\Delta=\Delta(\infty)\|\bar{\mu}\|$, where $\|\bar{\mu}\|$ for low temperatures is given by Eq. (3.10), one finds that

$$M=\frac{2}{\pi}N_0c\mu_B\left\{\tan^{-1}y_0+\frac{2y_0\ln 2}{\pi(1+y_0{}^2)^2}\left(\frac{kT}{\Delta(\infty)}\right) +\left(\frac{kT}{\Delta(\infty)}\right)^2\left(\frac{y_0}{1+y_0{}^2}\right)\left[\left(\frac{2\ln 2}{\pi(1+y_0{}^2)}\right)^2 \times\frac{4+3y_0{}^2}{1+y_0{}^2}-\frac{\pi^2}{12}\right]+O(T^3)\right\}. \tag{4.9}$$

Equation (4.9) shows that the magnetization per impurity near $T\to 0$ is given by $\tan^{-1}[H_{\rm ext}/\Delta(\infty)]$. Thus, it is predicted that the magnetic susceptibility per impurity near zero temperatures should have a slope of $1/\Delta(\infty)$. Physically, this result shows that the very low T magnetic susceptibility is independent of the impurity concentration for very low temperatures and concentrations.[8]

D. Possible Application to a Mössbauer Experiment

It was discussed in Sec. III that the high-temperature width of the distribution function in the absence of an external field becomes very small and the expression for the width $\Delta(T)$ is approximately given by[8]

$$\Delta(T)\approx(k_BT)(e^{\pi k_BT/\Delta(\infty)-73/45}-1)^{-1/2}. \quad (4.10)$$

However, in the presence of an external field, the approximate width $\Delta(\beta)$ is given by

$$\Delta(\beta)=\Delta(\infty)\|\bar{\mu}(\beta)\|\approx\Delta(\infty)\tanh\beta H_{\text{ext}}, \quad (4.11)$$

where Eq. (3.16) has been used in Eq. (4.11). Thus, at high temperatures

$$P(h,\beta)\approx\frac{1}{\pi}\frac{\Delta(\infty)\tanh\beta H_{\text{ext}}}{[\Delta(\infty)\tanh\beta H_{\text{ext}}]^2+(h-H_{\text{ext}})^2}. \quad (4.12)$$

In the limit as $H_{\text{ext}}\to 0$, $P(h,\beta)$ becomes a δ function about $h=0$. Therefore, if one assumes that the hyperfine field h_{hf} is given by the phenomenological equation

$$h_{\text{hf}}=b\tanh\beta(\bar{H}+H_{\text{ext}})+H_{\text{ext}}, \quad (4.13)$$

at high temperatures with $H_{\text{ext}}=0$ the Mössbauer pattern of the alloy, say Au-Fe, is predicted to be characteristic of a paramagnetic spectrum. However, upon the application of a sufficiently large external field it is predicted that the Mössbauer hyperfine pattern should reappear and should have a distribution of fields given by Eq. (4.12). Since $\Delta(\infty)$ is proportional to impurity concentration one should hope, at least in principle, to be able to obtain $\Delta(\infty)$ from the Mössbauer spectrum at relatively high temperatures in the presence of an applied field. Should this turn out to be the case, it would present an interesting comparison with the low-temperature value of $\Delta(\infty)$ predicted by this theory.

V. DISCUSSIONS AND CONCLUSIONS

An effective field theory has been presented to discuss the properties of random dilute Ising-model systems in the presence of an externally applied magnetic field. The Ising spins were assumed to interact via a long-range interaction of the Ruderman-Kittel-Yosida type. It has been shown previously[15] that the molecular-field theory is the zeroth-order term in an expansion of the inverse of the effective number of neighbors z. In the limit as $z\to\infty$ one rigorously recovers the Weiss molecular-field approximation. If one may argue that even though the impurity concentration is low, because of the long interaction range of the interaction potentional the effective number of neighbors z is large enough, the molecular-field theory used in this paper may become a good approximation to the rigorous statistical-mechanical solutions desired.

One may, therefore, ask whether the results derived here are applicable to dilute alloys in general. This question was discussed previously[8] and the comments of Ref. 8 on the use of the Ising model as well as the Kondo temperature also apply here. The fact that the validity of the Ising model to magnetic alloys is questionable makes the Ising-model predictions of the magnetic field dependence of the thermodynamic properties even more interesting. For, if the measured field dependence of the specific heat and magnetization should be in qualitative agreement with the results predicted here, it would give further motivation to examine the predictions of the Heisenberg model and find how they differ from the Ising model.

[15] R. Brout, Phys. Rev. 118, 1009 (1960).

APPENDIX A: EVALUATION OF $d\Delta/d\beta$

In this Appendix, the derivative of the width of the distribution function $\Delta(\beta,c,H_{\text{ext}})$ with respect to β is evaluated. The arguments of Δ will be suppressed in what follows:

$$\frac{\Delta'}{\Delta(\infty)}=\frac{d\Delta}{d\alpha}=\frac{d\|\mu\|}{d\beta}, \quad (A1)$$

where α is defined in Eq. (3.1). Using Eq. (3.3) gives

$$\frac{d\|\bar{\mu}\|}{d\beta}=\frac{d\Delta}{d\alpha}=\frac{d}{d\beta}\int_0^\infty \tanh\alpha y[P^+(y)+P^-(y)]dy. \quad (A2)$$

Differentiating the right-hand side of Eq. (A2) and transposing the terms $d\|\mu\|/d\beta$ arising from $dP^\pm(y)/d\beta$ and combining it with the left-hand side gives

$$d\|\bar{\mu}\|/d\beta=\Delta(\infty)\|\bar{\mu}\| \times\int_0^\infty \frac{y\,\text{sech}^2y[P^+(y)+P^-(y)]dy}{D}\equiv\frac{N}{D}, \quad (A3)$$

where D in Eq. (A3) is

$$D=1-4\pi\|\bar{\mu}\|\int_0^\infty\frac{e^{-2\alpha y}}{1+e^{-2\alpha y}} \times\{[P^+(y)]^2+[P^-(y)]^2\}dy. \quad (A4)$$

The very low temperatures, the expansion for $P^+(y)$ given in Eq. (3.5) is used to evaluate Eq. (A4):

$$D=1-\frac{4\|\bar{\mu}\|^3\ln 2}{\pi(\|\mu\|^2+y_0^2)^2}\frac{1}{\alpha}+O(\alpha^{-3}). \quad (A5)$$

Evaluating the numerator N of Eq. (A3) using Eq. (3.7) gives for very low temperatures

$$N=\frac{2}{\pi\alpha}\frac{\|\bar{\mu}\|^2}{\|\bar{\mu}\|^2+y_0^2}\left(\ln 2+\frac{1}{\alpha^3}\frac{3y_0^2-\|\bar{\mu}\|^2}{\|\bar{\mu}\|^2+y_0^2} \times\int_0^\infty x^3\,\text{sech}^2x dx\right). \quad (A5')$$

Using Eqs. (A4) and (A5) and the power-series expansion for $\|\bar{\mu}\|$ as given in Eq. (3.10) in Eq. (A1) gives

$$\beta\frac{d\|\bar{\mu}\|}{\beta}=\frac{2\ln 2}{\pi\beta\Delta(\infty)(1+y_0^2)}+\left(\frac{2\ln 2}{\pi\beta\Delta(\infty)(1+y_0^2)}\right)^2\left(\frac{2}{1+y_0^2}\right). \quad \text{(A5'')}$$

Equation (A5) will be used to evaluate the low-temperature specific heat.

APPENDIX B: DERIVATION OF LOW-TEMPERATURE SPECIFIC HEAT

Let C_v be the specific heat and S the entropy for the system. Then

$$C_v=\frac{dQ}{dT}=T\frac{\partial S}{\partial T}=-\beta\frac{\partial S}{\partial\beta}, \quad \text{(B1)}$$

where Q is the heat applied and S the entropy associated with N randomly distributed impurities; each impurity experiences an effective field which is a random variable. Let S_1 be the entropy of a *single* impurity in a fixed field $\bar{H}$, let U_1 and Z_1 be the corresponding internal energy and partition function, respectively. Then, $S_1=k_B[\beta U_1+\ln Z_1]$, where in an Ising model with the Bohr magneton μ_B set to unity, $Z_1=\sum_{\mu=\pm 1}\exp\mu\bar{H}=2\cosh\beta\bar{H}$, and $U_1=\bar{H}\tanh\beta\bar{H}$. Thus,[16]

$$\begin{aligned}S_1&=-k_B[\beta\bar{H}\tanh\beta\bar{H}-\ln(2\cosh\beta\bar{H})]\\&=k_B\{\beta\bar{H}(1-\tanh\beta\bar{H})+\ln[1+\exp(-2\beta\bar{H})]\}.\end{aligned} \quad \text{(B2)}$$

The total contribution to the entropy of a random system with N impurities is

$$S=\frac{N_0ck_B}{2}\int_{-\infty}^{\infty}P(\bar{H})\{\beta(\bar{H}+H_{\text{ext}})[1-\tanh\beta(\bar{H}+H_{\text{ext}})]+\ln[1+\exp(-2\beta(\bar{H}+H_{\text{ext}}))]\}d\bar{H}, \quad \text{(B3)}$$

where $P(\bar{H})$ is given in Eq. (2.19) and H_{ext} is the applied magnetic field. A factor of 2 is introduced in the denominator of Eq. (B3) in order not to count each interaction twice. Using Eqs. (3.1) and (3.2) in Eq. (B3) gives

$$S=\frac{N_0ck_B}{2}\int_{-\infty}^{\infty}P(y)\{\alpha(y+y_0)[1-\tanh\alpha(y+y_0)]+\ln(1+e^{-2\alpha|y+y_0|})\}dy, \quad \text{(B4)}$$

where $P(y)$ is the value of $P^+(y)$ with $y_0=0$. Let

$$F(x)=x(1-\tanh x)+\ln(1+e^{-2x}); \quad \text{(B5)}$$

then Eq. (B4) becomes

$$S=\frac{N_0ck_B}{2}\int_0^{\infty}F(\alpha y)[P^+(y)+P^-(y)]dy, \quad \text{(B6)}$$

where $P^\pm(y)$ and α are defined in Eqs. (3.1) and (3.2). Using Eq. (B1) gives

$$C_v=-\beta\frac{N_0ck_B}{2}\left(\int_0^{\infty}\frac{\partial F(\alpha y)}{\partial\beta}[P^+(y)+P^-(y)]dy+\int_0^{\infty}F(\alpha y)\frac{\partial}{\partial\beta}[P^+(y)+P^-(y)]dy\right). \quad \text{(B7)}$$

Using the identity

$$I^\pm=\int_0^{\infty}F(\alpha y)\frac{\partial P^\pm(y)}{d\beta}dy=\frac{\Delta'}{\Delta}\left(\int_0^{\infty}F(\alpha y)P^\pm(y)dy-2\pi\int_0^{\infty}F(\alpha y)[P^\pm(y)]^2dy\right), \quad \text{(B8)}$$

where $\Delta'=d\Delta/d\beta$, and Δ is given in Eq. (2.17). Integrating the second term on the right-hand side of Eq. (B8) by parts gives

$$I^\pm=\frac{\Delta'}{\Delta}\left[\pm\frac{y_0}{\pi}\frac{\ln 2}{1+y_0^2}+\int_0^{\infty}(y\pm y_0)P^\pm(y)\left(\frac{\partial F}{\partial y}\right)dy\right]. \quad \text{(B9)}$$

Using Eq. (B9) in Eq. (B7) gives

$$C_v=\frac{N_0ck_B}{2}\left[\left(1+\frac{\beta\Delta'}{\Delta}\right)\int_0^{\infty}(\alpha y)^2\,\text{sech}^2(\alpha y)\times[P^+(y)+P^-(y)]dy+(\alpha y_0)\frac{\beta\Delta'}{\Delta}\times\int_0^{\infty}(\alpha y)\,\text{sech}^2(\alpha y)[P^+(y)-P^-(y)]dy\right]. \quad \text{(B10)}$$

Equation (B10) gives the final expression for the specific heat, with

$$\frac{\beta\Delta'}{\Delta(\infty)}=\beta\frac{d\|\bar{\mu}\|}{d\beta}$$

given in Eq. (A6).

[16] Alternatively, the entropy may be obtained by using $\sum_i P_i\ln P_i$, where the summation is over the possible spin orientations.