test of the Γ model of ferroelectrics. (The usual isotopic effect on phonons predicts a decrease of 1 part in 138 of the phonon frequency in the deuterated case.)

The T^4 law for the order parameter could possibly be experimentally checked by doing nuclear magnetic resonance on the protons, looking for the NMR lines resulting from protons in the "wrong" sites.[7] These lines should grow as T^4 at low temperatures.

Low-temperature measurement of the spontaneous polarization in the ferroelectric would yield less direct information, as we have shown that both anharmonicity and the order parameter influence $\langle \mathbf{p} \rangle$ with the same T^4 dependence. However, our prediction is that the T^4-dependent decrease of $\langle \mathbf{p} \rangle$ will be greater in undeuterated ferroelectrics because the order parameter has no T^4 dependence in the dueterated case.

ACKNOWLEDGMENT

The author is indebted to V. H. Schmidt for several valuable discussions on experimental aspects of KH_2PO_4-type ferroelectrics.

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[7] Techniques for narrowing proton NMR lines will probably be needed for such an experiment; see S. Clough and I. R. McDonald, Proc. Phys. Soc. (London) **90**, 1019 (1962).

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Temperature-Dependent Internal Field Distribution and Magnetic Susceptibility of a Dilute Ising Spin System*

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The temperature-dependent probability distribution of internal exchange fields, $\bar{H}$, is obtained for a set of randomly distributed Ising-model spins using a modified form of the statistical model of Margenau. The impurities are assumed to interact via a convergent long-range potential which alternates in sign as a function of position. When spin correlations between the magnetic impurities are neglected and a *mean-random-field* (MRF) approximation is used, the probability distribution $P(\bar{H})$ is given by a nonlinear integral equation. For a $1/r^3$ potential, the self-consistent probability distribution is, in the MRF approximation, a Lorentzian with a temperature- and concentration-dependent width $\Delta(\beta)$, where $\beta = 1/(k_BT)$ and T is the temperature. The function $\Delta(\beta)$ is also given by a nonlinear integral equation which is solved for very high and very low temperatures. Using $P(\bar{H})$ derived for a $1/r^3$ potential, the magnetic susceptibility $\chi(\beta)$ and the specific heat $C_v(\beta)$ are obtained for all temperatures. The model gives a magnetic susceptibility which exhibits a maximum as a function of temperature for all nonzero (but sufficiently small) impurity concentrations. The temperature of the maximum is proportional to the impurity concentration. Possible applications of the model to the temperature and concentration dependence of $\chi(\beta)$ and $C_v(\beta)$ of dilute magnetic alloys are discussed.

1. INTRODUCTION

THE method of random molecular fields has been used to discuss the thermodynamic properties of various magnetic impurities interacting via a long-range potential. Marshall[1] has introduced this concept to obtain the low-temperature specific heat of magnetic impurities interacting via a Ruderman-Kittel-Yosida[2] (RKY) interaction. Later Klein and Brout[3] obtained the probability distribution of the random Ising-model internal field for a system interacting via the RKY interaction in the limit as the temperature approaches 0°K. An attempt to describe the variation of the probability distribution of the internal field with temperature as one departs from $T=0$ has been made by the author.[4,4a] However, the validity of this approach has not been satisfactorily shown. The purpose of this paper is to use a modified form of the statistical model of Margenau[5] to derive the temperature-dependent internal field distribution for a set of randomly distributed Ising-model impurity spins which interact via a convergent long-range potential which alternates in sign as a function of position. In the model used the formal expression for the probability distribution of the internal field, $P(\bar{H}_0)$, at a particular site 0 is given in terms of the effective internal fields at all other impurity sites in the solid. To evaluate the expression for $P(\bar{H}_0)$, a *mean-random-field* (MRF) *approximation* is used in which, when calculating the probability distribution of the internal field at site 0, all functions of the internal field at sites other than 0 are replaced by their mean

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[5] H. Margenau, Phys. Rev. **48**, 755 (1935).

values. Neglecting spin-spin correlations between the magnetic impurities and using the MRF approximation and imposing a self-consistency condition gives an integral equation for the probability distribution. For a $1/r^3$ potential the self-consistent $P(\bar{H})$ is a Lorentzian with a temperature- and concentration-dependent width $\Delta(\beta)$ which is monotonically decreasing with increasing temperatures. $\Delta(\beta)$ is given in a simple power series of the temperature for low temperatures and in terms of an expansion in the inverse temperature for high temperatures. Using the expression for $P(\bar{H})$, the temperature-dependent magnetic susceptibility $\chi(\beta)$ is obtained. It is found that $\chi(\infty)$ is independent of the impurity concentration and that $\partial\chi(\beta)/\partial T$ is positive and is inversely proportional to the impurity concentration for small T. As a consequence of this, the predicted magnetic susceptibility has a maximum as a function of temperature for all nonzero (but small) concentrations, where $T_{\max}$ is proportional to the impurity concentration. The model also predicts a maximum in the low-temperature specific heat $C_v(T)$ as well as a maximum in $\partial C_v(T)/\partial T$ as a function of the temperature, where both of these maxima are proportional to the impurity concentration.

In Secs. 4–7 a possible application of the theory to the very recently obtained magnetic susceptibility measurement of Cu–Mn, Au–Fe, and Cu–Fe is discussed. It is argued that the very-low-temperature magnetic susceptibility is internal field dominated for sufficiently large concentrations c_0 such that the average effective internal field is of the order of or greater than the Kondo temperature divided by the Bohr magneton. For Cu–Mn $c_0<0.01\%$, for Au–Fe $c_0\gtrsim0.05\%$, and for Cu–Fe $c_0\gtrsim0.1\%$. It is also shown that the details of the very-low-temperature specific heat of Cu–Fe obtained by Frank, Manchester, and Martin[6] *cannot* be explained by an internal-field model.

2. DERIVATION OF THE PROBABILITY DISTRIBUTION

Consider a system of interacting Ising-model spins randomly distributed in a nonmagnetic host. Let the interaction Hamiltonian $\mathcal{H}$ between the magnetic impurities be of the form

$$\mathcal{H}=\sum_{i<j} v_{ij}\mu_i\mu_j, \tag{2.1}$$

where v_{ij} is the interaction potential between the particles located at positions i and j, r_{ij} is their separation, and μ_i and μ_j are the respective Ising-model spin variables. μ_i may take values of ±1. Let the interaction potential v_{ij} be of the form

$$v_{ij}=a(\cos kr_{ij}/|\cos kr_{ij}|)r_{ij}^{-n}, \tag{2.2a}$$

with the requirements that $\int_V v_{ij}d^3r_{ij}<\infty$, where V is the volume of the solid. Let there be N_0 sites in the solid, N of which are occupied by magnetic impurities, and let $N\to\infty$ such that $(N/N_0)=c$, where c is the fractional impurity concentration. This paper considers the limiting case as the concentration $c\to0$, such that terms in c^2 may be neglected. This condition is imposed by the use of the statistical model where multiple occupancy of the sites is permitted, introducing an error of order c^2. Let $\bar{R}$ be the average distance between the magnetic impurities. It is required that the wave vector k in Eq. (2.2a) be such that $k\bar{R}\gg1$, i.e., the periodicity of the potential varies sufficiently rapidly that an impurity j is, *on the average*, just as likely to experience a positive as a negative potential from impurity i. For this reason, in subsequent calculations the potential given in Eq. (2.2a) will be approximated by one of the form

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

The potential in Eq. (2.2b) is chosen as an approximation to the Ruderman-Kittel interaction in dilute alloys to be discussed in Sec. 6. The replacement of Eq. (2.2a) by Eq. (2.2b) will give physically incorrect results when long-range magnetic order exists in the solid.

Let the effective field $\bar{H}_0$ at an arbitrary impurity site r_0 be defined by the expression

$$\bar{H}_0=\sum_j v_{0j}\bar{\mu}_j. \tag{2.3}$$

Since r_{0j} and hence $v(r_{0j})\equiv v_{0j}$ are random variables, $\bar{H}_0$ is also a random variable. The bars over $\bar{H}$ and $\bar{\mu}$ indicate that they are thermally averaged quantities. The definition of Eq. (2.3) is thus consistent with the usual molecular-field approximation.

The purpose of this paper is to obtain the temperature-dependent probability distribution $P(\bar{H}_0)$ of the random variable $\bar{H}_0$. The thermodynamic functions for the system are then obtained by integrating the thermodynamic variables for a single spin in fixed internal field $\bar{H}$ over the probability distribution of all fields. The quantity $\bar{\mu}_j$ in Eq. (2.3) is

$$\bar{\mu}_j=\mathrm{tr}\exp(-\beta\mathcal{H})\mu_j/\mathrm{tr}\exp(-\beta\mathcal{H}), \tag{2.4}$$

where tr represents the trace over all spin operators, i.e.,

$$\mathrm{tr}\exp(-\beta\mathcal{H})=\sum_{\mu_1=\pm1}\sum_{\mu_2=\pm1}\cdots\sum_{\mu_N\pm1}\exp(-\beta\mathcal{H}).$$

The spin average in Eqs. (2.3) and (2.4) is performed for a fixed set of position coordinates. It should be remarked that, in order to obtain the thermodynamic variables for the system, one has to average over the position coordinates only *after* the thermodynamic average over the spins is performed. The specific order of this averaging is dictated by the physical requirement that the spins in the random system are in thermal

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[6] J. P. Franck, F. D. Manchester, and D. L. Martin, Proc. Roy. Soc. (London) **A263**, 494 (1961).

equilibrium whereas the spatial coordinates of the impurities are frozen in the solid in a definite *nonequilibrium* fashion. This requirement has been discussed previously in connection with the dilute alloy problem.[3]

Using the standard molecular-field approximation, the Hamiltonian given in Eq. (2.1) is expressed in terms of the effective fields at the individual sites. Then

$$\mathcal{H}=-\sum_i \mu_i\left(\sum v_{ij}\bar{\mu}_j\right),$$

$$\mathcal{H}=-\sum_i \bar{H}_i\mu_i, \tag{2.5}$$

where $\bar{H}_i$ is defined in Eq. (2.3). Thus Eq. (2.4) becomes

$$\bar{\mu}_j=\frac{\operatorname{tr}\exp(\beta\sum_i\bar{H}_i\mu_i)\mu_j}{\operatorname{tr}\exp(\beta\sum_i\bar{H}_i\mu_i)}=\tanh\beta\bar{H}_j. \tag{2.6}$$

For a fixed set of spatial coordinates, $\bar{H}_0$ as well as the $\bar{H}_j$'s are constants. The distribution of $\bar{H}_0$ arises from the requirement that the positions of the particles are random variables. The probability distribution of $\bar{H}_0$ is calculated as follows. Fix an impurity μ_0 at the origin of coordinates, site 0, and average over an ensemble of systems, each containing N spins, where the impurities in each member of the ensemble have their position coordinates fixed, but where different members of the ensemble have the coordinates fixed in different positions. The density of the ensembles is determined by assuming a random distribution of the N impurities on the N_0 sites. The consequence of this procedure will be that average thermodynamic quantities (like specific heat, magnetic susceptibility, etc.) are those obtained from calculating the thermodynamic quantities of a single spin in an internal field arising from a fixed spatial configuration of the surrounding spins, and then averaging over all possible spatial configurations of all spins except that at the origin of coordinates.

To find a formal expression for the probability distribution $P(\bar{H}_0)$, the method of Margenau[5] is used and each of the $3N$-dimensional configurations which contribute to $\bar{H}_0$ is multiplied by unity, whereas configurations which do not contribute are multiplied by zero. For this purpose the well-known δ-function trick[5] to express $P(\bar{H}_0)$ is used. Thus

$$P(\bar{H}_0)=\sum_{v_N}\int_{r_N}\delta\left(\bar{H}_0-\sum v_{0j}\bar{\mu}_j\right)\times P(r_{01},r_{02},\cdots,r_{0N})\,d^3r_{0N}, \tag{2.7}$$

where $\sum_{v_N}$ is the sum over each of the potentials $v_{0j}=\pm a/r_{0j}{}^n$ as given in Eq. (2.2b), giving a total of n such sums, $\int_{r_N}$ indicates an integral over a $3N$-dimensional volume, and $P(r_{01},r_{02},\cdots,r_{0N})\,d^3r_{0N}$ is the joint probability for particle 1 to be in the volume of d^3r_1 at r_1, particle 2 to be in d^3r_2 at r_2, and particle N to be in d^3r_N at r_N. Rather than summing over discrete sites in the lattice, integration is used. This is expected to be valid for the very dilute case, for the model is restricted to sufficiently low concentrations such that the average interparticle distance is much greater than a near-neighbor distance.

It is now assumed in Eq. (2.7) that the positions of each of the particles are *independent* random variables which are uniformly distributed over the volume V of the solid with probability $1/V$. Thus the probabilities factorize and

$$P(r_N)=(1/V)^N. \tag{2.8}$$

Rewriting Eq. (2.7) using Eq. (2.8) gives

$$P(\bar{H}_0)=(2\pi)^{-1}\int d\rho\,\exp(i\rho\bar{H}_0)\times\int_{r_N}\prod_{j=1}^{N-1}\left\{\sum_{v_{0j}=\pm a/r_{0j}n}\left[(2V)^{-1}\exp(-i\rho v_{0j}\mu_j)\,d^3r_{0j}\right]\right\}. \tag{2.9a}$$

Using Eq. (2.2b) in (2.9a) thus gives

$$P(\bar{H}_0)=\int d\rho\,\exp(i\rho\bar{H}_0)\prod_{j=1}^{N-1}\int_{r_{0j}}(2V)^{-1}\times\left[\exp(i\rho a\bar{\mu}_j/r_{0j}{}^n)+\exp(-i\rho a\bar{\mu}_j/r_{0j}{}^n)\right]d^3r_{0j}. \tag{2.9b}$$

Equation (2.9) cannot be evaluated because $\bar{\mu}_j=\tanh\beta\bar{H}_j=\tanh\beta(\sum_k v_{jk}\bar{\mu}_k)$ is explicitly a function of all the position coordinates separating the impurities at r_j and r_k, and implicitly a function of the distances between all other impurities via the $\bar{\mu}_k$'s. Therefore, the evaluation of Eq. (2.9) will have to be done using an approximation, the details of which are described below.

The function $\exp(-i\rho v_{0j}\bar{\mu}_j)$ for a fixed v_{0j} depends upon the field $\bar{H}_j$. Now, consistent with the initial postulate of a probability distribution of the field $P(\bar{H}_0)$ at site 0, a probability distribution of $\bar{H}_j$, $P(\bar{H}_j)$, is also postulated. (This is equivalent to evaluating $P(\bar{H}_j)$ over an ensemble of systems in which an impurity is fixed at site j and other impurities are randomly distributed.) Using this $P(\bar{H}_j)$, $\exp(\pm i\rho v_{0j}\bar{\mu}_j)$ for a fixed v_{0j} is approximated by replacing it by its mean value over the distribution of fields at site j, $P(\bar{H}_j)$. Thus

$$\exp(\pm i\rho a\bar{\mu}_j/r_{0j}{}^n)\xrightarrow{\text{MRFA}}\int P(\bar{H}_j)\,\exp(\pm i\rho a\bar{\mu}_j/r_{0j}{}^n)\,d\bar{H}_j. \tag{2.10}$$

This approximation will be denoted by the *mean-random-field approximation* (MRFA) and is indicated by MRFA over the arrow in Eq. (2.10). At the moment no assumption is made about $P(\bar{H}_j)$ except that it exists. $P(\bar{H}_j)$ may, for example, be a conditional probability distribution with the condition that H_0 is specified. However, later on, $P(H_j)$ will be required to be the same for all j.

It is appropriate to draw a comparison between the MRF approximation and the usual Weiss molecular-field approximation. In the Weiss molecular-field approximation all fields are replaced by a mean field which is the same at each site in the solid. Here, in order to calculate the distribution of fields at site 0, functions of the internal field at sites other than 0 are replaced by their mean values, obtained by averaging over a distribution of fields. Thus in a sense the MRF approximation goes one step beyond the Weiss molecular-field approximation.

With the approximation in Eq. (2.10), Eq. (2.9) becomes

$$P(\bar{H}_0)=(2\pi)^{-1}\int d\rho\,\exp(i\rho\bar{H}_0)$$

$$\times\prod_{j=1}^{N-1}\left(\int d\bar{H}_j\int_{-\infty}^{\infty}P(\bar{H}_j)\,V^{-1}\cos\frac{\rho a\bar{\mu}_j}{r_{0j}{}^{n}}\,d^3r_{0j}\right).\quad(2.11)$$

It should be noted that, since each $\bar{H}_j$ and $\bar{\mu}_j$ was assumed to be independent of all other $\bar{H}$'s, the MRF approximation neglects spin-spin correlations between the impurities. Such an approximation would be physically incorrect if a long-range correlation between magnetic impurities existed. It is only because, on the average, the spins are only correlated over a short region[3] of space that the model presented may have applicability to experiments performed on dilute magnetic alloy systems. It should also be remarked that even though the method goes beyond the usual Weiss field approximation, in that it permits the internal fields to have a probability distribution, only upon repeated comparison with experiment can one gain confidence that the model may have applicability to real physical systems.

Equation (2.11) gives $P(\bar{H}_0)$ in terms of the $P(\bar{H}_j)$ at all other sites. In principle, each random variable $\bar{H}_j$ may have a different probability distribution. At this point, however, as a *self-consistency condition* it is *imposed* that each $P(\bar{H}_j)$ has the same functional form. Therefore the subscripts on $\bar{H}_0$ and $\bar{H}_j$ are dropped in Eq. (2.11), and Eq. (2.11) becomes

$$P(\bar{H})=(2\pi)^{-1}\int_{-\infty}^{\infty}d\rho\,\exp(i\rho\bar{H})$$

$$\times\left(V^{-1}\int_{-\infty}^{\infty}P(\bar{H})\,d\bar{H}\int_{-\infty}^{\infty}\cos\frac{\rho a\bar{\mu}}{r^n}\,d^3r\right)^{N-1}.\quad(2.12)$$

Let

$$V'=\int_V\int_{-\infty}^{\infty}P(\bar{H})\,d\bar{H}\left(1-\cos\frac{\rho a\bar{\mu}}{r^n}\right)d^3r.\quad(2.13)$$

Then Eq. (2.12) becomes

$$P(\bar{H})=(2\pi)^{-1}\int_{-\infty}^{\infty}d\rho\,\exp(i\rho\bar{H})\left(1-\frac{V'}{V}\right)^N$$

$$=(2\pi)^{-1}\int_{-\infty}^{\infty}d\rho\,\exp[(i\rho\bar{H})-(N/V)\,V'],\quad(2.14)$$

where V' is given in Eq. (2.13) and $N-1$ was replaced by N.

Equation (2.14) is a nonlinear integral equation for $P(\bar{H})$ and is the central result of this section.

Next V' is evaluated:

$$V'=4\pi\int_{-\infty}^{\infty}P(\bar{H})\,d\bar{H}\int_0^{\infty}\left(1-\cos\frac{\rho a\bar{\mu}}{r^n}\right)r^2dr.$$

Let

$$Z=\mid a\rho\bar{\mu}\mid/r^n,\quad(2.15)$$

where the vertical bars indicate absolute values. Then Eq. (2.15) becomes

$$V'=\frac{4\pi}{n}\mid\rho a\mid^{3/n}\int_{-\infty}^{\infty}P(\bar{H})\,d\bar{H}\mid\bar{\mu}\mid^{3/n}\int_0^{\infty}\frac{1-\cos Z}{Z^{(3+n)/n}}\,dZ\quad(2.16)$$

and Eq. (2.16) is to be used in Eq. (2.14).

3. EVALUATION OF $P(\bar{H})$ FOR A $1/r^3$ POTENTIAL

Next Eq. (2.16) is evaluated for a $1/r^3$ potential. Equation (2.16) becomes

$$V'=\tfrac{4}{3}\pi\mid\rho\mid\int_{-\infty}^{\infty}P(\bar{H})\mid\bar{\mu}\mid d\bar{H}\int_0^{\infty}\frac{1-\cos Z}{Z^2}\,dZ$$

$$=\tfrac{2}{3}\pi^2\mid\rho a\mid\,\|\bar{\mu}\|,\quad(3.1)$$

where

$$\|\bar{\mu}\|=\int_{-\infty}^{\infty}P(H)\mid\bar{\mu}\mid d\bar{H},\quad(3.2)$$

with $\bar{\mu}$ given in Eq. (2.6). Substituting Eq. (3.1) into Eq. (2.14) gives

$$P(\bar{H})=(1/\pi)\{\Delta(\beta)/[\Delta(\beta)^2+\bar{H}^2]\},\quad(3.3)$$

where

$$\Delta(\beta)=\tfrac{2}{3}\pi^2\mid a\mid n_0c\,\|\bar{\mu}\|\equiv\gamma c\,\|\bar{\mu}\|,\quad(3.4)$$

where $\gamma=\tfrac{2}{3}\pi^2\mid a\mid n_0$ and depends only upon the strength of the interaction at a distance of a lattice constant and the number of sites per unit cell n_0. It is expected that this model gives too large a probability for obtaining high fields from what one would expect in an actual physical situation. The reason for this is that in a real solid the maximum field is limited by the nearest-neighbor distance between the impurities, whereas here no restrictions were placed on the distance of closest approach. Equation (3.3) is an integral equation defining $P(\bar{H})$ via Eqs. (3.4) and (3.2).

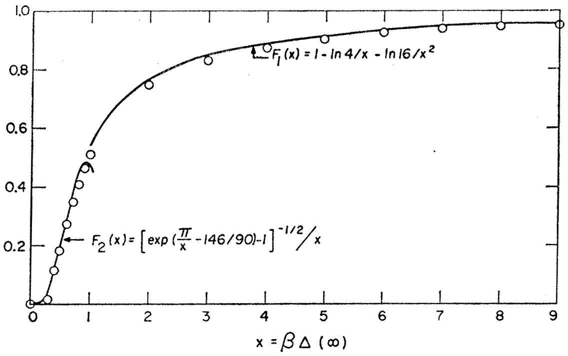

FIG. 1. Values of $\|\mu(x)\|$ as a function of $\beta\Delta(\infty)$, where $\beta=1/(k_BT)$, T is the temperature, and $\Delta(\infty)$ is the width of the probability distribution function at $T=0$. The circles show the machine-calculated values, whereas the solid lines give the low-temperature approximation $F_1(x)$ and the high-temperature approximation $F_2(x)$ given in Eqs. (4.5) and (5.2), respectively.

A. Solution of Δ(β) for Low Temperatures

The temperature-dependent solution for $\Delta(\beta)$ may be obtained by finding approximations to the integral equation

$$\|\bar{\mu}\| = 2\int_0^\infty P(\bar{H}) \tanh\beta\bar{H}d\bar{H}. \tag{3.5}$$

Expanding Eq. (3.5) in terms of $\bar{H}/\Delta(\beta)$ gives

$$\begin{aligned}\|\bar{\mu}\| &= 1-[4/(\pi\Delta(\beta))] \\ &\quad\times\int_0^\infty \frac{\exp(-2\beta\bar{H})}{1+\exp(-2\beta\bar{H})} \\ &\quad\times\left[1-\left(\frac{\bar{H}}{\Delta(\beta)}\right)^2+\left(\frac{\bar{H}}{\Delta(\beta)}\right)^4+\cdots\right]d\bar{H} \\ &= 1-\{2[\pi\beta\Delta(\beta)]^{-1}\}\{\ln 2+\tfrac{1}{2}[\beta\Delta(\beta)]^{-2} \\ &\quad\times\sum_{n=1}^{\infty}\frac{(-1)^n}{n^3}-\tfrac{3}{2}[\beta\Delta(\beta)]^{-4}\sum_{n=1}^{\infty}\frac{(-1)^n}{n^5} \\ &\quad+O([\beta\Delta(\beta)]^{-6})\}.\end{aligned} \tag{3.6}$$

Equation (3.6) gives

$$\lim_{\beta\to\infty}\|\bar{\mu}\| = 1. \tag{3.7}$$

In Eq. (3.4) let

$$\Delta(\beta) = \Delta(\infty)\,\|\bar{\mu}(\beta)\|, \tag{3.8}$$

where $\Delta(\infty)$ is the value of $\Delta(\beta)$ at $\beta\to\infty$. Using Eq. (3.6) in Eq. (3.5) gives

$$\|\bar{\mu}(\beta)\| = 1-(q/\beta)-(q/\beta)^2-O(q/\beta)^3, \tag{3.9}$$

where

$$q = \ln 4[\pi\Delta(\infty)]^{-1}. \tag{3.10}$$

It is found that in the MRF approximation, for a set of spins for which spin-spin correlations between impurities are neglected, the temperature-dependent probability distribution is a Lorentzian with a width $\Delta(\beta)$ which is proven in the Appendix to be monotonically decreasing with temperature. The expression for the width for low temperatures is given by Eqs. (3.8) and (3.9).

B. Evaluation of Δ(β) for High Temperatures

For high temperatures and low internal fields let $\tanh\beta\bar{H}\approx\beta\bar{H}-\frac{1}{3}(\beta\bar{H})^3+(2/15)(\beta\bar{H})^5$ and separate the integral in Eq. (3.5) into two parts:

$$\begin{aligned}\|\bar{\mu}\| &\approx \frac{2}{\pi}\int_0^{1/x}\frac{\|\bar{\mu}\|}{\|\bar{\mu}\|^2+y^2}[xy-\tfrac{1}{3}(xy)^3+(2/15)(xy)^5]dy \\ &\quad+\frac{2}{\pi}\int_{1/x}^{\infty}\frac{\|\bar{\mu}\|}{\|\bar{\mu}\|^2+y^2}[1-2\exp(-2xy)]dy,\end{aligned} \tag{3.11}$$

where $x=\beta\Delta(\infty)$. For a very small value of x the above equation becomes

$$1\approx(x/\pi)\ln[1+(x\|\bar{\mu}\|)^{-2}]+(73/45\pi)x+O(x^2),$$

which gives a solution

$$\|\bar{\mu}\|\approx[\exp(\pi/x-73/45)-1]^{-1/2}/x. \tag{3.12}$$

Equation (3.12) shows that the width of the Lorentzian decreases proportionally to $\exp[-\pi kT/2\Delta(\infty)]$ for high temperatures.

Solutions to the integral equation for $\|\mu\|$ were obtained by solving Eq. (3.5) using a computer. In Fig. 1 the values of $\|\bar{\mu}\|$ are plotted as a function of the temperature divided by the width of the Lorentzian at $\beta\to\infty$, $\Delta(\infty)$. In the same figure the low-temperature solution $F_1(x)=\|\bar{\mu}\|$, as given by Eq. (3.9), and the high-temperature solution $F_2(x)=\|\bar{\mu}\|$, as given by Eq. (3.12), are also shown. It is found that $F_1(x)$ and $F_2(x)$ approximate quite well the solutions for $\|\bar{\mu}\|$ obtained by machine calculations.

4. MAGNETIC SUSCEPTIBILITY

In this section the magnetic susceptibility at low temperatures is calculated. The single-impurity partition function Z_1, in the presence of an internal field $\bar{H}$ and an external field $\bar{H}_{\text{ext}}$, is

$$\begin{aligned}Z_1(\beta,\bar{H},H_{\text{ext}}) &= \sum_{\mu=\pm1}\exp[-\beta(\bar{H}+H_{\text{ext}})\mu] \\ &= 2\cosh\beta(\bar{H}+H_{\text{ext}}).\end{aligned} \tag{4.1}$$

The magnetization of the single impurity $M_1(\bar{H},\beta)$ in an effective field $\bar{H}$ is

$$M_1(\bar{H},\beta) = \lim_{H_{\text{ext}}\to0}(\partial\ln Z_1/\partial\beta H_{\text{ext}}) = \mu_B\tanh\beta\bar{H}, \tag{4.2}$$

where μ_B is the Bohr magneton.

The magnetization is, on the average,

$$M(\beta) = N_0c\mu_B\int_{-\infty}^{\infty}P(\bar{H})\tanh\beta\bar{H}d\bar{H}, \tag{4.3}$$

where N_0 is the number of sites per unit volume. The magnetic susceptibility $\chi(\beta)$ is

$$\chi(\beta)=\lim_{H_{\mathrm{ext}}\to 0} N_0c\mu_B{}^2\beta\int P(\bar{H})\times\frac{\partial^2}{\partial(\beta H_{\mathrm{ext}})^2}\ln Z_1(\beta,\bar{H},H_{\mathrm{ext}})\,d\bar{H} \tag{4.4}$$

$$=N_0c\beta\mu_B{}^2\int_{-\infty}^{\infty}P(\bar{H})\,\mathrm{sech}^2\beta\bar{H}d\bar{H}. \tag{4.5}$$

Next the magnetic susceptibility, Eq. (4.5), is evaluated for low temperatures, neglecting the change of $P(\bar{H})$ with external magnetic field,[6a]

$$\chi(\beta)=\frac{2N_0c\mu_B{}^2}{\pi\Delta}\left[1-\frac{\pi^2}{12}\left(\frac{1}{\beta\Delta}\right)^2+O\left(\frac{1}{\beta\Delta}\right)^4\right], \tag{4.6}$$

$$\lim_{\beta\to\infty}\chi(\beta)=2N_0c\mu_B{}^2/\pi\Delta(\infty)=2N_0cP(0)\mu_B{}^2,$$

where $P(0)$ is the probability density at $\bar{H}=0$. Since Δ as given by Eq. (3.5) is proportional to the impurity concentration, Eq. (4.6) shows that $\chi(\infty)$ is independent of the impurity concentration. This result has already been obtained previously.[7] The slope of the magnetic susceptibility near $T=0$ is given by

$$\lim_{\beta\to\infty}(\partial\chi/\partial T)=-(1/k_BT^2)(\partial\chi/\partial\beta)$$

$$=\frac{2N_0ck_B\mu_B{}^2}{\pi\Delta(\infty)}q\left[1+\frac{4q}{\beta}-\frac{\pi^2}{6\Delta(\infty)^2\beta q}+O(\beta^{-2})\right], \tag{4.7}$$

where q is given in Eq. (3.10). Substituting for q gives, for very low temperatures,

$$\frac{\partial\chi}{\partial T}=\frac{4N_0c\mu_B{}^2k_B\ln 2}{[\pi\Delta(\infty)]^2}\left[1-\left(\frac{\pi^3}{12\ln 2}-\frac{4\ln 4}{\pi}\right)\frac{k_BT}{\Delta(\infty)}+O(T^2)\right]. \tag{4.8}$$

Equation (4.8) shows that the initial slope of $\chi(T)$ versus T is inversely proportional to the impurity concentration and is positive for small temperatures. A necessary consequence of this result is that the model gives a maximum in the magnetic susceptibility for all nonzero concentrations.[8] To determine the temperature of the maximum, one has to solve the equation

$$\frac{\partial\chi}{\partial\beta}\propto\frac{2}{\pi}\int_0^{\infty}\frac{H^2(\beta\Delta'+2\Delta)-\Delta^2(\beta\Delta')}{(H^2+\Delta^2)^2}\,\mathrm{sech}^2\beta HdH\equiv 0, \tag{4.9}$$

where $\Delta'=\partial\Delta/\partial\beta$. It should be noted that for temperature $kT\simeq\Delta$, Eq. (3.9) is no longer valid, making the analytic solution of Eq. (4.9) difficult.

A. High-Temperature Magnetic Susceptibility

Equation (4.5) is conveniently rewritten in the form

$$\chi(\beta)=\frac{N_0x}{\pi[\Delta(\infty)/c]}\int_{-\infty}^{\infty}\frac{\|\bar{\mu}\|^2}{\|\bar{\mu}\|^2+y^2}\,\mathrm{sech}^2xydy, \tag{4.10}$$

where $x=\beta\Delta(\infty)$. Equation (4.10) can be evaluated by using a contour integration in the complex plane. Closing the contour in the upper half of the complex plane with an infinite radius R, the integrand has a first-order pole at $y=i\|\bar{\mu}\|$ and a set of second-order poles at $y=(2n+1)i\pi/2x$, where n is any positive integer. The residue at $y=i\|\bar{\mu}\|$ is $[2i\cos^2(\|\mu\|x)]^{-1}$, and the residue at $(2n+1)i\pi/2x$ is

$$\frac{(2n+1)i\pi/2x^3}{[\|\mu\|^2-((2n+1)\pi/2x)^2]^2}.$$

Evaluating the contour integral, Eq. (4.10) becomes

$$\chi(\beta)=\frac{N_0\mu_B{}^2}{\Delta(\infty)/c}\left\{\frac{x}{\cos^2(\|\bar{\mu}\|x)}-\sum_{n=0}^{\infty}x^{-1}\frac{[(2n+1)/2x]\pi}{(\|\bar{\mu}\|^2-\{[(2n+1)/2x]\pi\}^2)^2}\right\}. \tag{4.11}$$

For $k_BT\gg\Delta(\infty)$, Eq. (3.12) is used to find the approximate value of $\chi(\beta)$:

$$\chi(\beta)=\frac{N_0c\mu_B{}^2}{k_BT}\left\{1-\left(\frac{2}{\pi}\right)^3\frac{\Delta(\infty)}{k_BT}\sum_{n=0}^{\infty}(2n+1)^{-3}+O\left[\exp\left(-\frac{\pi k_BT}{2\Delta(\infty)}\right)\right]\right\}. \tag{4.12}$$

Equation (4.12) gives a leading term for high-temperature magnetic susceptibility which is the same as for a paramagnetic Ising system, with corrections proportional to $1/T^2$.

The magnetic susceptibility, Eq. (4.5), was calculated using a computer and the results are shown in Fig. 2. There is a maximum in the susceptibility at $1/[\beta_{\max}\Delta(\infty)]\simeq 0.4$, or $k_BT_{\max}\simeq 0.4\Delta(\infty)=0.4\gamma c$, where γ is given in Eq. (3.4). Thus the maximum in $\chi(\beta)$ is proportional to the impurity concentration. Also note from Eq. (4.10) that the susceptibility is only a function of $x=\beta\Delta(\infty)$, and the temperature at which the maximum in $\chi(\beta)$ occurs is given by $x\simeq 0.4$,

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[6a] *Note added in proof:* We have since shown that in the MRF approximation the change of $P(\bar{H})$ with external field gives no contribution to the magnetic susceptibility.

[7] J. Friedel, J. Phys. Radium **23**, 962 (1962); M. W. Klein, Phys. Rev. **141**, 489 (1966).

[8] See the qualitative arguments of S. Liu [Phys. Rev. **157**, 411 (1967)].

TABLE I. Summary of the results for the width of the probability distribution Δ, the magnetic susceptibility χ, and the magnetic specific heat C_v. $\gamma=(8\pi^2/3)|a|n_0$, where a is the strength of the interaction between two impurities at a distance of one lattice constant and n_0 = the number of sites per unit cell, $n_0=4$ for an fcc lattice. c is the impurity concentration, N_0 is Avogadro's number, k_B is the Boltzmann constant.

	Low-T behavior ($\gamma c \gg k_BT$)	High-T behavior ($\gamma c \ll k_BT$)	k_BT_{max}	Value at T_{max}
Δ	$\gamma c\left[1-\frac{\ln 4}{\pi}\frac{k_BT}{\gamma c}-\left(\frac{\ln 4}{\pi}\frac{k_BT}{\gamma c}\right)^2\right]$	$k_BT\{\exp[(\pi k_BT/\gamma c)-(73/45)]-1\}^{-1/2}$	$\cdots$	$\cdots$
$\frac{\chi(T)}{N_0\mu_B^2/\pi}$	$\frac{2}{\gamma}\left[1+\frac{\ln 4}{\pi}\frac{k_BT}{\gamma c}-\left(\frac{\pi^2}{12}-2\frac{\ln^2 4}{\pi^2}\right)\left(\frac{k_BT}{\gamma c}\right)^2\right]$	$c/k_BT[1-(2/\pi)^3(\gamma c/k_BT)\sum_{n=0}^{\infty}(2n+1)^{-3}]$	$\simeq 0.4\gamma c$	$\chi(T_{\mathrm{max}})\propto c^0$
$\frac{12C_{v(T)}}{N_0k_B/\pi}$	$\frac{k_BT}{\gamma}\left\{1+2\frac{\ln 4}{\pi}\frac{k_BT}{\gamma c}-\left(\frac{k_BT}{\gamma c}\right)^2\left[\frac{7\pi^2}{20}-5\left(\frac{\ln 4}{\pi}\right)^2\right]\right\}$		$\simeq 0.32\gamma c$	$C_{v(T_{\mathrm{max}})}\propto c^1$
$\frac{12C_{v(T)}}{N_0k_B^2T/\pi}$	$\left\{1+2\frac{\ln 4}{\pi}\frac{k_BT}{\gamma c}-\frac{k_BT}{\gamma c}\left[\frac{7\pi^2}{20}-5\left(\frac{\ln 4}{\pi}\right)^2\right]\right\}\gamma^{-1}$		$\simeq 0.16\gamma c$	$C_{v(T_{\mathrm{max}})}/T_{\mathrm{max}}\propto c^0$

thus $\chi(\beta_{\mathrm{max}})$ is independent of the impurity concentration. The behavior of the high- and low-temperature magnetic susceptibility is summarized in Table I.

5. SPECIFIC HEAT

The low-temperature specific heat for the system is now obtained. The entropy S_1 of a single impurity in an effective field H is[9]

$$S_1=k_B(\beta U+\ln Z), \tag{5.1}$$

where

$$\ln Z=\ln\sum_{\mu\pm 1}\exp(-\beta H\mu)=\ln 2\cosh\beta H$$

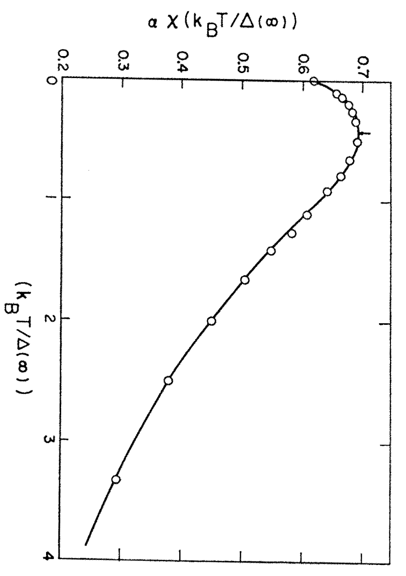

FIG. 2. Calculated results for the magnetic susceptibility as a function of the temperature divided by the width of the probability distribution $\Delta(\infty)$ at $\beta=\infty$. The maximum in $\chi(\beta)$ occurs at $k_BT\approx 0.4\Delta(\infty)$. $\alpha=[N_0\mu_B^2c/\Delta(\infty)]^{-1}$, where μ_B is the Bohr magneton, N_0 is Avogadro's number, and c is the impurity concentration.

and

$$U=\frac{\sum_{\mu\pm 1}\mu H\exp(-\beta\mu H)}{\sum_{\mu=\pm 1}\exp(-\beta\mu H)}=-H\tanh\beta H.$$

Thus the entropy for a single impurity is

$$S_1=k_B(-H\tanh\beta H+\ln 2\cosh\beta H). \tag{5.2}$$

The total entropy for the N impurities is therefore

$$S=\tfrac{1}{2}(N_0ck_B)\int_{-\infty}^{\infty}\{-\beta H\tanh\beta H+\ln[1+\exp(-2\beta|H|)]+\beta|H|\}P(H,\beta)dH, \tag{5.3}$$

where the factor of $\frac{1}{2}$ in Eq. (5.3) is introduced in

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[9] If one tries to evaluate the specific heat C_v from the energy, as was done in Refs. 1, 3, and 4, one finds that C_v/T diverges, thus violating the third law of thermodynamics. The difficulty is traced back to the fact that the probability distribution, which is required to be normalized to unity, introduces a constraint between the internal fields, and hence the energy is not a proper extensive quantity. Thus one has to use the entropy to evaluate the specific heat as is done here.

order not to count each interaction in Eq. (2.1) twice. $P(H,\beta)$ is given by Eq. (3.3). It is seen from Eq. (5.3) that the entropy is a function of T only. Thus the specific heat C_v is

$$C_v = dQ/dT = T(\partial S/\partial T) = (-1/k_BT)(\partial S/\partial\beta). \quad (5.4)$$

Let

$$F(H,\beta) = \beta H(1-\tanh\beta H) + \ln[1+\exp(-2\beta H)].$$

Then

$$\left(\frac{\partial S}{\partial\beta}\right)_H = N_0c\int_0^\infty \left(P(H,\beta)\frac{\partial F(H,\beta)}{\partial\beta} + F(H,\beta)\frac{\partial P(H,\beta)}{\partial\beta}\right)dH. \quad (5.5)$$

Integrating the second term of Eq. (5.5) by parts gives

$$\int_0^\infty F(H,\beta)\frac{\partial P(H)}{\partial\beta}dH = \frac{\Delta'}{\Delta} \times \int P(H,\beta)H\frac{F(H,\beta)}{\partial H}dH, \quad (5.6)$$

where Δ is given in Eq. (3.4) and $\Delta' = \partial\Delta/\partial\beta$. Substituting Eqs. (5.5) and (5.6) into Eq. (5.4) gives

$$C_v = N_0ck_BT\int_0^\infty P(x/\beta)x^2\,\text{sech}^2x[1+\beta\Delta'/\Delta]dx, \quad (5.7)$$

where $x=\beta H$. For very low temperatures

$$\beta\Delta'/\Delta = (q/\beta) + 2(q/\beta)^2 + O(q/\beta)^3,$$

where $q = \ln 4/\pi\Delta(\infty)$. Using this result in Eq. (5.7), the very-low-temperature specific heat becomes

$$C_v = \frac{N_0\pi k_B{}^2T}{12[\Delta(\infty)/c]}\left\{1+\frac{2\ln 4}{\pi}\frac{k_BT}{\Delta(\infty)} - \left[\frac{k_BT}{\Delta(\infty)}\right]^2 \times\left[\frac{7\pi^2}{20} - \frac{5(\ln 4)^2}{\pi^2}\right] + O[k_BT/\Delta(\infty)]^3\right\}. \quad (5.8)$$

Equation (5.8) shows that the very-low-temperature specific heat is linear in T and independent of the impurity concentration. This is in agreement with previous internal field calculations with a $1/r^3$ potential.[1,3,4] However, Eq. (5.8) also predicts that C_v/T has a positive slope for low temperatures, a result which has not been obtained before. Thus Eq. (5.8) gives a maximum in C_v as well as in C_v/T. The temperature of the maximum in both of these quantities is predicted to be proportional to the impurity concentration. The maximum in C_v is obtained by differentiating Eq. (5.8) with respect to T and setting the result equal to zero. The approximate condition for the maximum in C_v is $k_BT_{\text{max}}/\Delta(\infty) \approx 1/\pi$ or $k_BT_{\text{max}} \approx 0.32\gamma c$. The maximum in C_v/T is again found by differentiating (C_v/T) with respect to T and setting the result equal to zero. For this case $k_BT_{\text{max}} \approx \Delta(\infty)\{20\ln 4/[7\pi^3 - (100\ln 4/\pi)]\} \approx 0.16\gamma c$. Thus the temperatures of the maxima in C_v/T and C_v are, from the model, roughly $\frac{3}{4}$ and $\frac{3}{8}$, respectively, of the temperature of the maximum in the magnetic susceptibility. All three of these temperatures are proportional to the impurity concentration. The results on the low-temperature specific heat are summarized in Table I. Equation (5.8) also shows that the specific heat per impurity is a function of $\beta\Delta(\infty)$, and since $\beta\Delta(\infty)$ is a constant at $T = T_{\text{max}}$, the predicted C_v/T_{max} is proportional to the impurity concentration.

6. POSSIBLE APPLICATION TO DILUTE MAGNETIC ALLOYS

Dilute concentrations of magnetic impurities dissolved in nonmagnetic metals show various anomalies in their specific heats, their resistivities, and their magnetic susceptibilities.[10] The purpose of this section is to discuss conditions under which the model presented may have applicability to dilute alloys. The assumptions that the spins interact via an Ising-model Hamiltonian and that the magnitude of the impurity spin is fixed at a temperature-independent value impose severe restrictions on the temperature region in which the model may have applicability. Recent experimental and theoretical considerations indicate[11,12] that for antiferromagnetic J there is a Kondo temperature T_K at which the impurity conduction-electron system becomes unstable to the formation of a singlet pair state which compensates (i.e., reduces) the effective impurity spin, in which

$$k_BT_K \approx D\exp(-\tfrac{2}{3}\epsilon_F/|J|), \quad (6.1)$$

where D is an energy of the order of magnitude of the Fermi energy ϵ_F of the host metal, and J is the *s-d* exchange interaction, assumed to be a constant. In fact, experiments[11] indicate that at very low temperatures the spin compensation is close to complete. Therefore, the assumption of a rigid impurity spin S has no validity below T_K but may have limited validity above T_K. For Cu–Fe, T_K is indicated to be somewhere between[11] 6 and 15°K. The value of T_K is, however, very rapidly varying with J, as is found from Eq. (6.1). Even though T_K for dilute Cu–Fe is of the order of 10°K, for some other material T_K may be several orders of magnitude smaller. Above T_K, and for sufficiently large impurity concentrations, internal field effects may be of importance and, if so, the

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[10] G. J. van den Berg, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Co., Amsterdam, 1964), Vol. 4, and references therein.

[11] A. J. Heeger and M. A. Jensen, Phys. Rev. Letters **18**, 488 (1967); M. A. Jensen, A. J. Heeger, L. B. Welsh, and G. Gladstone, *ibid.* **18**, 997 (1967).

[12] M. D. Daybell and W. A. Steyert, Phys. Rev. Letters **18**, 398 (1967).

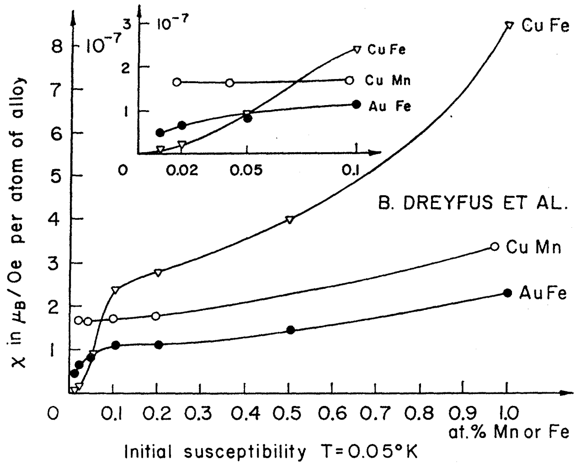

FIG. 3. Magnetic susceptibility of dilute alloys as a function of the impurity concentration. Results of Dreyfus *et al.* (Ref. 15).

magnetic susceptibility should be determined by these internal field effects. Whenever this is the case it is believed that the internal field arises from a RKY[2] interaction and the results derived in the previous section for a $1/r^3$ potential may apply. It is appropriate to remark that, at temperatures much below T_K as well as for concentrations sufficiently low that internal field effects are unimportant, the very-low-temperature magnetic susceptibility $\chi(\beta)$ per unit volume arising from a set of independent spins is expected to be proportional to the impurity concentration. This was recently found to be so by Hurd.[13] However, at very low temperatures, $\chi(\beta)$, which is dominated by internal field effects, is independent of the impurity concentration. The magnetic susceptibility and the low-temperature specific heat of Cu-Mn, Au-Fe, and Cu-Fe are next discussed.

Cu-Mn

There are very recent experiments on Cu-Mn which may have relevance to the present work. In one of these, Careage *et al.*[14] measured the low-temperature magnetic susceptibility of dilute Cu-Mn in the concentration range 0.0186 to 1.89% manganese in the temperature range from 0.1 to 4°K. They found that, for concentrations ranging from 0.0186 to 0.196%, $\chi(0.1°\text{K})$ is approximately independent of the impurity concentration, that $(\partial\chi/\partial T)_{T=0.1°K}$ increases with decreasing concentration, and that $\chi(T)$ exhibits a maximum where $T_{\max}$ increases with concentration. The approximately concentration-independent very-low-temperature susceptibility of Cu-Mn as measured by Careage *et al.*[14] may be interpreted as showing that the susceptibility is internal-field dominated. For concentrations higher than 0.9% manganese, Careaga *et al.*[14] find that $\chi(0)$ is no longer independent of the impurity concentrations but increases with concentration. This may be due to effects arising from the c^2 term, which was not considered in the statistical model. Part of these results are reproduced from a paper by Dreyfus *et al.*[15] and are shown in Fig. 3. Further possible evidence that the properties of Cu-Mn are determined by internal field effects may be found in the radically different resistivity of Cu-Mn and Cu-Fe at low temperatures, as was found by Steyert *et al.*,[12] that Cu-Fe does not exhibit a low-temperature resistivity maximum whereas Cu-Mn does.[10] This maximum has been discussed as arising from internal field effects.[16]

Au-Fe

The low-temperature magnetic susceptibility of Au-Fe ($T=0.05$°K) as a function of the impurity concentration has been obtained by Dreyfus *et al.*[15] and is reproduced in Fig. 3. It is to be noted that, for concentrations between 0.08 and 0.4%, $\chi(T)$ is approximately concentration-independent. However, for $c<0.08\%$, the susceptibility drops with temperature. Qualitatively one may interpret these results as indicating that at $c=0.1\%$ or greater the internal fields are sufficiently large that at very low temperatures the energy of the magnetically (short-range) ordered state is lower than that of the Kondo compensated state. As the concentration is further lowered, those impurities which experience very low internal fields will have a Kondo spin-compensated state and will thus lower the magnetic susceptibility. Again, Au-Fe at and above 0.1% concentrations exhibits a maximum in the resistivity which can be interpreted in terms of internal field effects, as was discussed before.[16]

Cu-Fe

The very-low-temperature, very-low-concentration Cu-Fe is definitely not internal field dominated. This is seen from the magnetic susceptibility obtained by Dreyfus *et al.* shown in Fig. 3. However, at concentrations of 0.1% and above, internal field effects become important. It should also be noted that in Cu-Fe there is no concentration-independent region as in Cu-Mn and Au-Fe, possibily indicating a more complicated behavior than in Cu-Mn and Au-Fe. It should also be remarked that the specific-heat maximum in the internal field model is proportional to the impurity concentration as is exhibited in Table I. This is not observed for very-low-concentration Cu-Fe. This

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[13] C. M. Hurd, Phys. Rev. Letters **18**, 1127 (1967) and references therein.

[14] A. J. Careage, B. Dreyfus, R. Tournier, and L. Weils, in *Proceedings of the Tenth International Conference on Low-Temperature Physics, Moscow, 1966* (Proizvodstrenno-Izdatel'skii Kombinat, VINITI, Moscow, 1967).

[15] B. Dreyfus, J. Souletie, J. L. Tholence, and R. Tournier, J. Appl. Phys. **39**, 846 (1968).

[16] S. D. Silverstein, Phys. Rev. Letters **16**, 466 (1966); R. J. Harrison and M. W. Klein, Phys. Rev. **154**, 540 (1967).

shows that the low-temperature specific heat Cu–Fe *cannot* be explained by internal field effects alone. The previous specific-heat results of Cu–Fe[17] were derived from the knowledge of $P(\bar{H})$ near $T=0$, and the more complete $P(\bar{H})$ derived here shows that the conclusions drawn from the limited knowledge of the $P(\bar{H})$ curve were incorrect. Before concluding, a brief discussion of the relationship to previous work on the magnetic susceptibility[3] and the effect of correlations is in order. Previously[3], the probability distribution of the internal field near $\beta\to\infty$ was obtained and the following semiquantitative argument was used to derive the temperature of the susceptibility maximum. At any one temperature the impurities were divided into two groups, one for which $\mu_B\bar{H}>k_BT$ and another for which $\mu_B\bar{H}<k_BT$. The impurities of the latter group were assumed to contribute like a set of paramagnetic impurities, whereas the former were considered to experience large internal fields and thus give an exponentially small contribution to the magnetic susceptibility. The above physical reasoning gave the qualitative behavior of the experimentally obtained magnetic susceptibility of Cu–Mn and predicted a temperature of the maximum, $T_{\max}$, which is proportional to the impurity concentration. The present paper agrees with, and justifies within the approximation used, the previous qualitative arguments, at least as far as the maximum in the magnetic susceptibility is concerned.

It is also to be noted that the qualitative results are not expected to depend upon the appropriate consideration of spin-spin correlations (provided only short-range correlations exist). However, quantitatively the correlations will tend to reduce the probability for small internal fields and increase the probability for obtaining large internal fields. This is expected to result in a decrease in $\chi(T=0)$ and in

$$\lim_{T\to 0} C_v(T)/T$$

and an increase in $T_{\max}$ for $\chi(T)$ as well as for $C_v(T)$. A quantitative comparison between theory and experiment will be made elsewhere.

7. SUMMARY AND CONCLUSION

The properties of a set of randomly distributed Ising-model spins were examined. The probability distribution of the internal field as a function of temperature was derived in the limit of zero concentration, using a MRF approximation. The low- and high-temperature values of the magnetic susceptibility, the specific heat, and the width of the probability distribution were obtained. The magnetic susceptibility obtained has a temperature-dependent maximum where $T_{\max}\propto c$ and $\chi(T_{\max})\propto c^0$. The specific heat $C_v(T)$ as well as $C_v(T)/T$ obtained from the model have a temperature-dependent maximum where $T_{\max}$ is in both cases proportional to the impurity concentration. The results for the detailed temperature and concentration dependence of these quantities are summarized in Table I in terms of a parameter γ, where $\gamma=\frac{2}{3}\pi^2|a|n_0$, where $|a|$ is the strength of the interaction between two impurities at a distance of one lattice constant, and n_0 is the number of sites per unit cell. $n_0=4$ for a face-centered-cubic lattice.

ACKNOWLEDGMENTS

The author wishes to thank Dr. R. Tournier for permitting him to use Fig. 3 prior to publication and Dr. R. J. Harrison for some valuable discussions.

APPENDIX

The purpose of this Appendix is to prove that $\Delta(\beta)$ is a monotonically increasing function with increasing β. From Eq. (2.5), $\Delta(\beta)=A\,||\bar{\mu}||$, where $A=\frac{2}{3}\pi^2|a|n_0c$. Thus, it is sufficient to show that $d\,||\bar{\mu}||/d\beta>0$.

$$||\bar{\mu}||=\frac{2}{\pi}\int_{-0}^{\infty}\frac{A\,||\bar{\mu}||}{(A\,||\bar{\mu}||)^2+\bar{H}^2}\tanh\beta\bar{H}d\bar{H}, \tag{A1}$$

$$d\,||\mu||/d\beta=B(\beta)/[1-C(\beta)], \tag{A2}$$

where

$$B(\beta)=\frac{2}{\pi}\int_0^{\infty}\frac{A\,||\bar{\mu}||}{(A\,||\bar{\mu}||)^2+\bar{H}^2}\operatorname{sech}^2\beta HdH>0.$$

$$C(\beta)=\frac{2}{\pi}\int_0^{\infty}\frac{A\tanh\beta\bar{H}d\bar{H}}{(A\,||\bar{\mu}||)^2+\bar{H}^2}-\frac{4}{\pi}\times\int_0^{\infty}\frac{A(A\,||\bar{\mu}||)^2}{[(A\,||\bar{\mu}||)^2+\bar{H}^2]^2}\tanh\beta\bar{H}d\bar{H}. \tag{A3}$$

Using the definition of $||\bar{\mu}||$ in Eq. (A3), Eq. (A2) becomes

$$\frac{d\,||\bar{\mu}||}{d\beta}=\int_0^{\infty}P(\bar{H},\beta)\bar{H}\times\operatorname{sech}^2\beta\bar{H}d\bar{H}\bigg/2\int_0^{\infty}\pi A[P(H,\beta)]^2\tanh\beta\bar{H}d\bar{H}>0,$$

Q.E.D.

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[17] M. W. Klein, Phys. Rev. Letters **16**, 408 (1963).