meV and $2\Delta_2 = 2.76$ meV), nor with the recent results of Lykken *et al.*[8] on single-crystal Pb films. This perhaps means that the films of Leger and Klein[3] were not single crystalline.

The two gaps Δ_1 and Δ_2 observed in tunneling on single-crystal Pb[1,5] do not vary greatly with orientation (~5%) and Leger and Klein[3] suggested that this is experimental evidence for a rather nonselective tunneling process. However, the fact that (i) the current ratio I_2/I_1,[1,5] (ii) the resolution of the gaps in dV/dI, and (iii) the detailed shapes and amplitudes of the transverse phonon peaks in d^2V/dI^2 all vary significantly with orientation[5] would seem to suggest that the tunneling is selective.

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High-Temperature Magnetic Susceptibility of Dilute Alloys in the Mean-Random-Molecular-Field Approximation

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An examination of the high-temperature magnetic susceptibility in the mean-random-field approximation gives new information on the interaction mechanism between the impurities in a dilute-magnetic-alloy system. More specifically, the experimental "high"-temperature susceptibilities of Au-Fe, Au-Cr, and Au-Mn are analyzed and found to be in good semiquantitative agreement with theory. A theoretical connection between the high- and low-temperature properties of the alloy system agrees well with experiments on Au-Fe and Au-Cr.

Dilute magnetic impurities in nonmagnetic metals exhibit a maximum[1,2] in their temperature-dependent susceptibility $\chi(T)$, and the temperature of the maximum T_{max} is approximately proportional to the impurity concentration c for impurity concentration of the order of 1%. Whereas a reasonable amount of theoretical and experimental work has been done to understand the behavior of these alloys at temperatures[3] much below T_{max}, very little attention has been paid to their properties at temperatures greater than T_{max}.

The purpose of this note is to point out that new and useful information on the interaction mechanism between the magnetic impurities can be obtained from intermediate- and high-temperature magnetic susceptibility of the dilute-magnetic-alloy system using a mean-random-field (MRF) approximation recently developed by one of the authors.[4]

A brief summary of the theory is as follows: The magnetic impurities are assumed to be randomly and uniformly distributed throughout the solid, and the statistical model of Margenau[5] is used to derive the probability distribution $P(H, T)$ of the random internal field H at temperature T. In order to obtain an analytical expression for $P(H, T)$, an approximation is used in which, when calculating the field distribution about a particular impurity site, functions of the random fields at all other impurity sites are replaced by their mean values. This approximation is called the mean-random-field approximation. One thus obtains a self-consistent integral equation for the probability distribution $P(H, T)$. The result is[4]

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

where Δ is the width of the probability distribution

and is given by

$$\Delta(T) = \gamma c \, \|\mu\| \, , \tag{2}$$

where $\gamma = \frac{2}{3}\pi^2 n_0 |a|$, where n_0 is the number of sites per unit cell and a is the strength of the interaction at a distance of one lattice constant. $\|\mu\|$ is obtained from the integral equation

$$\|\mu\| = \int_{-\infty}^{\infty} P(H, T) |\tanh\beta H| \, dH \, . \tag{3}$$

Equations (1)–(3) define a self-consistent probability distribution which is presumably valid for all temperatures for a Ruderman-Kittel[6] potential.

The theory is believed to be valid for low impurity concentrations and when Kondo-like[7] effects are unimportant, either because the Kondo temperature is much below the temperature under consideration, or because the Kondo spin-compensated state is suppressed by impurity-impurity interaction. For a discussion of the latter the reader is referred to a recent paper by Tsay and Klein.[8]

Once the probability distribution is obtained, the thermodynamic variables are calculated by averaging over all internal fields. The expression for the magnetization M in an Ising model is thus

$$M = N_0 c g \mu_B \int_{-\infty}^{\infty} P(H, T) \tanh\beta(H + H_{\text{ext}}) \, dH \, , \tag{4}$$

where g is the gyromagnetic ratio, μ_B the Bohr magneton, and H_{ext} the external magnetic field. The expression for the magnetic susceptibility χ at temperatures around and above $T_{\max}$ is, for a general spin S, approximately given by the relationship[9]

$$\chi(T) \approx \frac{N_0 c P_{\text{eff}}^2 \mu_B^2}{3 k_B T} \left[1 - \left(\frac{2}{\pi}\right)^3 \frac{\Delta(T)}{k_B T}\right] \, . \tag{5}$$

In deriving the expression for the $\chi(T)$ given in Eq. (5) we have neglected the part of $\chi(T)$ which arises from the term involving $\partial P(H, T)/\partial H_{\text{ext}}$ under the integral sign. The reasons for this omission are the following: (a) We expect $P(H)$ to be slowly varying with H_{ext} near $H_{\text{ext}} \to 0$, and (b) we do not have the analytical expression for the probability distribution as a function of the external field for high temperatures. One can qualitatively argue on how the $\partial P(H, T)/\partial H_{\text{ext}}$ term will modify Eq. (5), by considering an analogy with the magnetic susceptibility of an Ising-model ferromagnet at high temperatures in the molecular field theory. The magnetization R of the ferromagnet is given by[10]

$$R = \tanh\{\beta[H_{\text{ext}} + V(0) R]\} \, , \tag{6}$$

where $v(0)$ is the mean value of the exchange potential. The very-high-temperature magnetic susceptibility for the ferromagnet is

$$\chi(T) \propto (T - \theta)^{-1} \, , \tag{7}$$

where $\theta = v(0)$. θ in the expression for the ferromagnetic susceptibility arises from the differentiation of R in $\tanh\beta[H_{\text{ext}} + v(0)R]$. Were we to neglect this differentiation, we would obtain the incorrect Curie law ($\chi \propto T^{-1}$) rather than the Curie-Weiss law observed.

In examining the derivation for $P(H, T)$ it becomes clear that in neglecting $dP(H, T)/dH_{\text{ext}}$ we have neglected a term analogous to dR/dH_{ext} in $\tanh\beta[H_{\text{ext}} + v(0)R]$. Therefore we argue that the expression for $\chi(T)$ given in Eq. (5) should be modified to be

$$\chi(T) = \frac{N_0 c P_{\text{eff}}^2 \mu_B^2}{3 k_B T^*} \left[1 - \left(\frac{2}{\pi}\right)^3 \frac{\Delta(T)}{T}\right] , \tag{8}$$

where $T^* = T - \theta$ and $P_{\text{eff}} = g[S(S+1)]^{1/2}$ is the effective moment of the system.

For temperatures below $2T_{\max}$, Δ is approximately temperature independent and $\Delta \approx \Delta(T = 0) \equiv \Delta^0$. The theory predicts that $\Delta^0 \approx 2.5\, T_{\max}$. For higher temperatures, Δ is approximately given by

$$\Delta(T) \approx \beta^{-1}\{\exp[\pi/(\beta\gamma c) - \tfrac{73}{45}] - 1\}^{-1/2} \, . \tag{9}$$

Note that for very high temperatures $\Delta \to 0$ and Eq. (8) reduces to the expression for the high-T susceptibility of a set of interacting spins. For later use we also give the expression for the magnetic susceptibility at $T \to 0$, $\chi(0)$. This is obtained from Eqs. (4)–(6) of Ref. 4 and is modified for a general spin S; thus we have

$$\lim_{T \to 0} \chi(T) = \frac{2 N_0 c P_{\text{eff}}^2 \mu_B^2}{3\pi\Delta^0} \, . \tag{10}$$

Examining Eq. (8) one can make the following remarks: (i) In the temperature range $T_{\max} < T < 2T_{\max}$, where Eq. (8) is valid and Δ is only weakly temperature dependent, a plot of $[T^*\chi(T)]/c$ vs $1/T$ should give approximately a straight line. (ii) The slope of the line should be approximately proportional to Δ^0 and therefore proportional to the impurity concentration c. (iii) The value of Δ^0 obtained from the slope should be approximately $\Delta^0 \approx 2.5\, k_B T_{\max}$ as was tabulated in Table I of Ref. 4. (iv) At sufficiently high temperature Δ approaches zero according to Eq. (9) and therefore $[T^*\chi(T)]/c$ should become a constant. Let T_0 be the temperature where the straight line with slope Δ^0 intercepts the line $T^*\chi(T)/c = \text{const}$. Then, T_0 should be proportional to the impurity concentration c. (v) Since the theory was derived for all temperatures one would hope that the value of Δ^0 obtained from the high-temperature experiments would correlate with the very-low-temperature susceptibility as obtained from Eq. (10).

We now compare the theory with experiment. For convenience we use the tabulated results of Lutes and Schmit[1] (LS) for Au-Fe, Au-Cr, and Au-Mn. Figure 1 shows a plot of $T^*\chi/c \equiv A(T)$

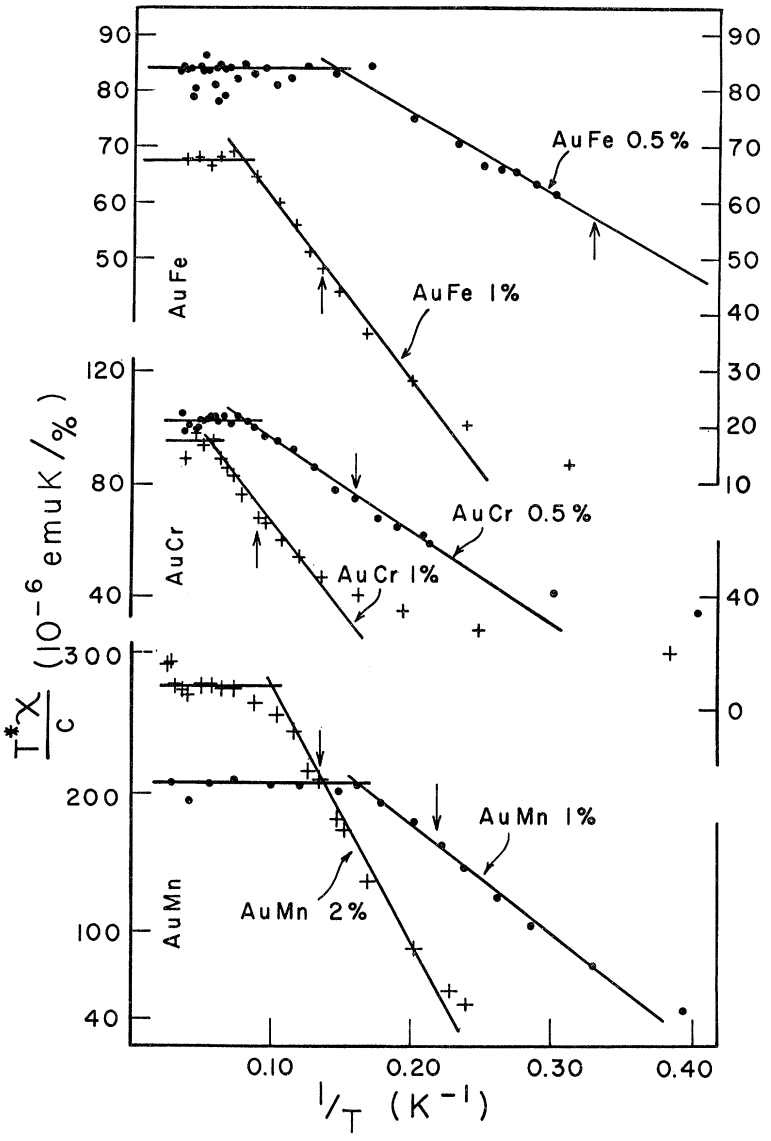

FIG. 1. $T^*\chi/c$-vs-$1/T$ plot for AuFe, AuCr, and AuMn. T_{max} is indicated by arrows.

vs $1/T$ of the data using the experimentally determined values of θ. We find that there is an approximately linear relationship between $A(T)$ and $1/T$ in the temperature range of $T_{max} \lesssim T \lesssim 2T_{max}$ for all samples in agreement with remark (i). The T_{max} values are indicated by arrows on the figure and also listed in Table I. At higher temperatures $A(T)$ becomes temperature independent as one expects, since the system obeys a Curie-Weiss law and since Δ goes to zero for high temperatures [remark (iv)]. The different high-temperature values of $A(T)$ reflect the different P_{eff} values, which are also listed in Table I.

The values of Δ^0 are calculated from the slope of the straight line and are listed in Table I. In Table I, we also list the values of T^0 and the ratios r_c, r_Δ, and r_{T^0}, where r_c is the ratio of the nominal impurity concentrations given by LS, r_Δ is the ratio of the values of Δ^0, and r_{T^0} is the ratio of the T^0 for the different concentrations. The value of $\Delta^0/k_B T_{max}$ is also given for each impurity concentration.

From remarks (ii) and (iv) we find that r_Δ and r_{T^0} should each be equal to the ratio of the impurity concentration r_c. The agreement between these quantities is reasonable. (Because of the crudeness of the MRF approximation for high temperatures one expects good qualitative, but not so good quantitative, agreement.) From remark (iii), the ratio of $\Delta^0/k_B T_{max}$ should be approximately 2.5 and this is again in reasonable agreement with experiment. It is important to note that, whereas this ratio varies between 2.1 and 3.2 for the three different alloys, it varies much less for the two

TABLE I. T_{max} is obtained from the experiments of LS. P_{eff}, Δ^0, and T^0 are obtained from Fig. 1. $r_c \equiv c_{II}/c_I$, $r_\Delta \equiv \Delta^0{}_{II}/\Delta^0{}_I$, and $r_T \equiv T^0{}_{II}/T^0{}_I$. $\chi(0)_{expt}$ is the experimentally obtained $T \to 0$ susceptibility. $\chi(0)_{theor}$ is calculated from Eq. (10). We have used the LS data for $\chi(0)_{expt}$ for Au-Cr and that of Ref. 11 for Au-Fe. In both of these cases T is sufficiently low that χ is concentration independent (Ref. 4) and Eq. (10) holds.

Sample	Nominal concentrations c (%)	T_{max} (K)	$\frac{\Delta^0}{k_B}$ (K)	P_{eff}	T^0 (K)	$\frac{\Delta^0}{k_B T_{max}}$	r_c	r_Δ	r_{T^0}	$\chi(0)_{expt}$ (emu/g)	$\chi(0)_{theor}$ (emu/g)
$AuFe_I$	0.5	3	7.5	3.6	6.5	2.5					3.1×10^{-6}
							2	2.4	2.0	3.1×10^{-6}	
$AuFe_{II}$	1.0	7	17.8	3.3	13	2.5					2.5×10^{-6}
$AuCr_I$	0.5	6	12.5	4.0	12.5	2.1					2.6×10^{-6}
							2	2.1	1.6	3.0	
$AuCr_{II}$	1.0	11	26.2	3.8	20						2.2×10^{-6}
$AuMn_I$	1	4.5	14.1	5.8	6	3.1					
							2	1.7	1.9		
$AuMn_{II}$	2	7.5	23.7	6.6	11.5	3.2					

different concentrations of the same alloy. Finally we use the values of Δ^0 obtained from Fig. 1 to calculate the zero-temperature magnetic susceptibility $\chi(0)_{MRF}$ using Eq. (10), and compare this with the experimental very-low-temperature data for[11] Au-Fe and Au-Cr.[1] For these two materials there are data available in the region where T is sufficiently low such that $\chi(T)$ is concentration independent, as predicted from the very-low-T limit of the theory. The data are shown in Table I. There is a remarkably good agreement between the very-low-temperature theory and experiment when we use the value of Δ^0 obtained from the high-temperature experimental susceptibility and Eq. (8). This we consider as evidence that the theory has reasonable validity over the whole temperature range.

It is our hope that these results will motivate further and more detailed experiments on dilute magnetic systems, which will be compared with the MRF approximation. We note that the slopes of the lines in Fig. 1 are never strictly straight. This is not surprising since Δ is continuously changing with temperature (slowly in the region $T_{max} \lesssim T \lesssim 2T_{max}$) and because terms higher than $1/T$ should also be considered. For this reason one should solve the integral equation for $P(H, T)$ using a computer and compare the susceptibility point by point with experiment.

Summary: We have compared the prediction of the mean-random-field approximation with the magnetic susceptibility of gold alloys and found reasonably good agreement between experiment and theory. Our analysis also gives the width of the probability distribution Δ^0, which also enters into the very-low-temperature theory of the magnetic susceptibility, resistivity, specific heat, and the external field dependence of the thermodynamic variables of the alloy systems.[12] The value Δ^0 obtained from high T gives the low-temperature magnetic susceptibility for Au-Fe and Au-Cr in surprisingly good agreement with experiment. Thus, our results not only describe the high-temperature behavior reasonably well, but also establish an important connection between the high- and low-temperature properties of these random systems.

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