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Magnetic susceptibility of α-phase CuAu and dilute magnetic CuAu(Fe) alloys*

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The magnetic susceptibilities of a series of α-phase CuAu and dilute magnetic CuAu(Fe) alloys with Fe ($\leq$ 760 ppm) have been measured at six different magnetic fields in the range from 3.95 to 12.75 kOe and over the temperature range from 1.5 to 300 K. The excess susceptibility attributed to Fe in the Fe-bearing CuAu samples was evaluated by comparison with the results of measurement on similar CuAu samples. We separate the single-impurity from impurity-impurity interaction effects in the part of the total susceptibility attributable to the Fe additions. In this treatment, the high-temperature excess susceptibility data are fitted to the expression $\chi(H,T) = \chi_0 + C/(T+\Theta) + C_2[JB_J(H/T)/H]$ that consists of a constant χ_0, a Curie-Weiss term, and a single Brillouin-function term. The low-temperature single-impurity susceptibility χ_s is obtained by the use of the coefficients χ_0 and C_2 in the expression $\chi_s = \chi(H,T) - \chi_0 - C_2[JB_J(H/T)/H]$. The single-impurity term χ_s is field independent. The interaction effect of parallel-coupled-spin impurities which contribute to the field dependence of the measured susceptibility is explained by a single Brillouin function with $J = 3$. The effective Bohr magneton μ_{eff} was found to be $(3.4 \pm 0.1)\mu_B$ for all Fe-bearing samples. The pair concentration C_{pair} deduced from the coefficient C_2 appears to be independent of Au concentration and has a magnitude of $(68 \pm 14) C_{\text{Fe}}$. Indications are that the characteristic temperature T_c, obtained from the relation, $\Theta = 1.25 T_c$, depends on both Au and Fe concentration. The existence of a universal relation for χ_s has been found in which T_c acts as a scaling temperature with the values of 13.6, 12.2, 10.6, 9.6, and 9.6 K for the alloys with Au concentrations of 0, 2.4, 4.8, 10.0, and 10,8 at.% and Fe concentrations of 750, 450, 440, 350, and 760 ppm, respectively.

I. INTRODUCTION

In recent years, significant progress has been made in understanding certain aspects of dilute magnetic alloys. Most theoretical investigations[1] have concentrated mainly on the Kondo problem, which is essentially a single-impurity effect. Many experimental results[2-4] show other effects that can be attributed to the correlation between impurities acting as magnetic pairs and influencing the properties of dilute alloys. These magnetic pairs are assumed to be responsible, in part, for the observed impurity-impurity interaction effects in the magnetic susceptibility, NMR linewidth in Cu, magnetization, resistivity, and specific-heat experiments conducted in magnetic fields.[5] Such magnetic pairs appear to have a small or zero characteristic temperature and they produce significant effects at low temperature ($T < T_c$). There is general agreement that the anomalous susceptibility of a dilute magnetic alloy could be due in part to magnetic pairs. The separation of the single-impurity susceptibility from that due to pairs or other entities has presented a problem. Some susceptibility measurements have been analyzed by means of fitting the data to the sum of several Curie-Weiss expressions. This is an empirical method for the analysis of the data that fails to adequately describe the single-impurity susceptibility at low temperatures ($T < T_c$). Recently a theory for the temperature- and field-dependent isolated-impurity susceptibility has been proposed by Götze and Schlottmann.[6-9] This theory is in general agreement with the results of an experiment on a dilute Cu(Fe) system.[10] A comparison can be made of this theoretical result and the single-impurity susceptibility obtained by our analysis.

We have measured the magnetic susceptibility of four dilute Fe in α-phase CuAu alloys in the temperature interval from 1.5 to 300 K and at six magnetic fields from 3.95 to 12.75 kOe. Measurements were also made on three CuAu host samples with similar Au concentrations but without Fe impurities. The latter measurements were subtracted from the former to evaluate the excess susceptibility contributed by Fe in the corresponding Fe-bearing sample. The results have been analyzed by a simple procedure in which the single-impurity susceptibility can be separated from the susceptibility attributed to pairs. The present result strongly indicates that CuAu alloys can be interpreted in terms of an average of electronic properties of the constituents as observed in both low-temperature-resistivity[11] and specific-heat[12] experiments.

The samples, apparatus, and measuring technique are described in Sec. II. A summary of the theoretical treatment bv Götze and Schlottmann for the susceptibility is presented in Sec. III. Results and discussion of the measurements on the set of seven samples are given in Sec. IV.

II. EXPERIMENTAL

A. Samples

The samples used in this susceptibility experiment are polycrystalline α-phase CuAu host alloys and dilute CuAu(Fe) alloys. These were obtained from the same alloys used in the calorimetry experiment by Delinger *et al.*[12] and were cut from the section of the same alloy bar which adjoined his samples. These alloys were prepared from Cu and Au of 99.999% purity by Material Research Corp., Orangeberg, N.Y. The starting materials were combined in a spectrographically pure high-density graphite crucible[9] and were induction melted for over 8 hs under high-vacuum conditions. Then the samples were removed from their molds. After being annealed in argon-filled quartz tubes at 900 °C for 72 hs, they were quenched immediately in ice water. The alloy bars were stored at room temperature for three years and were used in our susceptibility experiments without additional heat treatment. Except for the period of storage the susceptibility samples had the same heat treatment as the specific-heat samples.[12]

The actual concentration of Au in these alloys was determined in several different laboratories by the following methods: standard quantitative method, atomic absorption method, precision back-reflection powder pictures, and from density measurements (determined by hydrostatic weighing). The Fe concentration of impurity-doped samples was determined by atomic absorption analysis in two independent laboratories. Chemical analysis and a weak Kondo effect in the nonmagnetic samples indicated as ion content of a few ppm. A value of 3–4 ppm for the iron content was obtained by the use of the Curie law for the paramagnetic contribution to the low-temperature susceptibility of the nonmagnetic samples. The compositions of the $Cu_{1-x}Au_x$ and $Cu_{1-x}Au_x(Fe)$ alloys are shown in Table I. The quantity x is the atomic fraction of Au in the alloy. In Table I and the figures this quantity is reported as atomic percent (at.%).

Chemical etching was used to remove any ferromagnetic impurities on the surface and work damage done in shaping the samples. First, the samples were cleaned in aqua regia. A black film of Au resulted which was removed by a mixture of 10% solution of potassium cyanide and ammonium persulfate. Final cleaning was in several changes of deionized water and in methanol. After the above process the mass of each sample was about 800 mg.

TABLE I. Composition of the CuAu alloys investigated.

Sample	Au (at.%)	Fe (at.%)
CuAu	2.4 ± 0.1	a
CuAu	4.8 ± 0.1	a
CuAu	10.0 ± 0.1	a
CuAu(Fe)-1	2.4 ± 0.1	0.045 ± 0.005
CuAu(Fe)-2	4.8 ± 0.1	0.044 ± 0.005
CuAu(Fe)-3	10.8 ± 0.1	0.035 ± 0.005
CuAu(Fe)-4	10.0 ± 0.1	0.076 ± 0.005

[a] Less than a few ppm Fe as determined by low-temperature-resistivity and susceptibility measurements.

B. Apparatus and measuring technique

A Faraday susceptibility apparatus was used for this magnetic susceptibility measurement. The apparatus operates in the temperature range of 1.5–300 K with a precision of $\pm 4\times 10^{-10}$ cgs emu/g. This corresponds to a minimum detectable change in force on the sample of 3×10^{-3} dyn. The balance was calibrated at room temperature with 99.999%-purity zone-refined ASARCO copper as the primary standard. The standard had been annealed for seven days at 650 °C. The measurements were all compared to the mass susceptibility of pure ASARCO copper at 295 K, which value was assumed to be 0.0858×10^{-6} cgs emu/g. In order to extend the calibration to include forces corresponding in magnitude to those obtained for the largest susceptibility measurement, samples of very-high-purity annealed Al, Nb, and Pt were used as secondary standards.

At each temperature in this experiment the susceptibility was measured at six different magnetic fields. The measurement of each sample was taken in two stages. In the temperature range from 77 to 300 K, the ^{4}He exchange gas in the research chamber was maintained at 15 Torr. In the range from 1.5 to 77 K it was kept at 25 mTorr. These particular pressures were used to avoid the undesirable effects due to thermomolecular flow. The stabilization of temperatures above 4.2 K was obtained with the aid of a temperature controller and below 4.2 K was maintained by pumping on liquid helium in a small helium chamber surrounding the sample chamber. Temperatures between 1.5 and 40 K were measured by a calibrated germanium thermometer; in the temperature interval from 15 to 300 K a calibrated platinum thermometer was used. Both thermometers were calibrated to produce temperature scales uncertain to less than 0.1 K. The details of the cryostat design and operation have been given by Huck *et al.*[13]

III. THEORY OF SINGLE-IMPURITY MAGNETIC SUSCEPTIBILITY

Recently a new treatment of the *s*-*d* exchange model has been proposed by Götze and Schlottmann.[9] For the Tomonaga[14] model, they have calculated the impurity spin polarization, static susceptibility, and longitudinal impurity spin relaxation rate as a function of temperature and magnetic field. This was done using a self-consistent perturbation approach for both ferromagnetic and an-

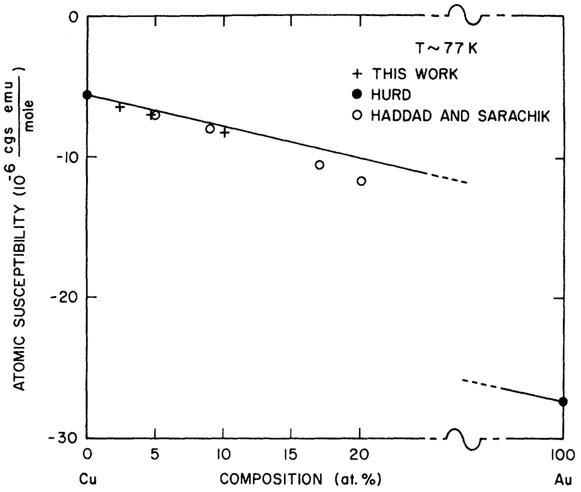

FIG. 1. Atomic susceptibility (at 77 K) as a function of Au concentration for CuAu solid solution. The solid line is drawn from the additive law.

tiferromagnetic coupling between impurity spin and the conduction electrons. For the antiferromagnetic coupling, the zero-field susceptibility $\chi_s(T)$ obeys a Curie-Weiss law for high and intermediate temperatures and approaches a finite constant at zero temperature:

$$\frac{1}{\chi_s(T)}=\begin{cases}4.6(T+\Theta)\ , & T>T_c\\ 3.8\Theta\ , & T=0\ ,\end{cases} \tag{1}$$

where Θ is a parameter with a value of $1.25T_c$. The characteristic temperature T_c has been defined by Emery and Luther.[15] The analytic expression of $\chi_s(T)$ vs T/T_c obtained by Götze and Schlottmann has been compared with a Monte Carlo calculation for a long-time approximation by Schotte and Schotte,[16] and also compared with the accurate Mössbauer hyperfine-field measurements for a very dilute Cu(Fe) alloy.[10] The agreement appears to be within the experimental uncertainties.

We have calculated the temperature- and field-dependent susceptibility $\chi_s(H, T)$ by adopting this theoretical model. The single-impurity susceptibility which can be separated from the total measured susceptibility by our procedure may then be compared with the theoretical result. In our data analysis we have assumed that impurity-impurity interaction effects are superimposed on the single-impurity effect. The Fe concentrations in our alloys are not small enough that the Fe-Fe interaction can be neglected. At low temperatures, the interaction effects possibly contribute the major portion of the total susceptibility. The field-dependent susceptibility observed in our data could be attributed to interaction effects which can be described by a single Brillouin function. We found that the single-impurity susceptibility is field independent within our range of fields and temperatures. The success of the separation of the single-impurity susceptibility from that due to Fe-Fe interaction effects is supported by the existence of a universal relation for the single-impurity magnetic susceptibility of CuAu(Fe) alloys.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. CuAu nonmagnetic binary alloys

The results of our measurements on binary host alloys are shown in Fig. 1 and 2. The mean susceptibility used in Fig. 1 is obtained by extrapolating six different sets of field susceptibility data by the Honda-Owen method.[17] Figure 1 shows the atomic susceptibility concentration curve (at 77 °K), which demonstrates the variation of atomic susceptibility with Au concentration. In this figure, some previous experimental data of Haddad and Sarachik[18] are also presented for comparison. The solid line is drawn as if a simple additive relation of the atomic susceptibilities are valid for this binary alloy. The straight line connects the susceptibilities obtained by Hurd[19] for pure Cu and Au. The degree to which the susceptibility of our samples deviated from a straight line agrees with other experiments.

In Fig. 2 the temperature-dependent property of the susceptibility of our CuAu alloys is shown. The diamagnetic susceptibility is reduced slightly with increasing temperature, and at low temperatures there is a strong tendency toward paramagnetism. These two interesting features are found in many other pure metals and metallic alloys. We agree with Hurd's[20] interpretation of a similar observation of the low-temperature trend toward paramagnetism for Cu and attribute the effect to a small amount of magnetic impurity, probably Fe, in the specimens. Such impurities in our samples, estimated from the temperature dependence, are about 3–4 ppm, which is consistent with the

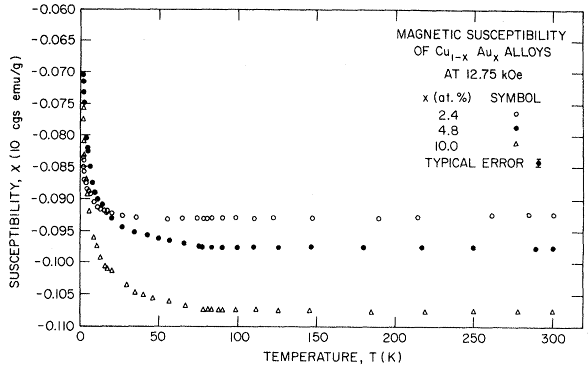

FIG. 2. Susceptibility of the Fe-free specimens measured at 12.75 kOe in the temperature interval from 1.5 to 300 K.

TABLE II. Coefficients of Eq. (2), which approximately describes the temperature dependence in the range 77–300 K of the susceptibility of the Fe-free hosts.

Sample	Gold concentration (at.%)	Atomic susceptibility (300 K) (10^{-6} cgs emu/g)	Coefficients in Eq. (2) after least-squares fitting		
			$x(0)$ (10^{-8} cgs emu/g)	$C(1)$ (10^{-11} cgs emu/g K)	$C(2)$ (10^{-14} cgs emu/g K^2)
CuAu-1	2.4 ± 0.1	−0.0924	−9.32	−1.08	2.60
CuAu-2	4.8 ± 0.1	−0.0974	−9.73	−1.07	4.78
CuAu-3	10.0 ± 0.1	−0.1080	−10.67	−1.05	3.15

result of the electrical resistivity measurement.[21]

The temperature-dependent characteristic of nonmagnetic alloys has been found and discussed in several papers. Both the Pauli paramagnetism and the Landau diamagnetism are predicted by theory to be temperature independent. With the reference to the observed temperature dependence of the susceptibility of alkali metals, Stoner[22] first suggested that the temperature-dependent property arises mainly from the change in the density of states at the Fermi level brought about by the thermal expansion. This suggestion has been supported by Hurd[23] in his experiments on group-IB metals, Cu, Ag, and Au. In the temperature range of 6–300 K, Hurd suggests an empirical equation that can be used for representing the susceptibility of pure metals such as Cu and Au:

$$\chi(T)=\chi(0)+C(1)T+C(2)T^2 \quad , \tag{2}$$

where T is temperature in kelvin, $\chi(T)$ is the measured gram susceptibility, and $\chi(0)$, $C(1)$, and $C(2)$ are parameters to be determined by a weighted least-squares method. Since the alloys used in our experiment are formed from group-IB metals, we use this equation for the host susceptibility. The constants for our alloys are presented in Table II. Also, In Fig. 2 it shows that the temperature dependence is less pronounced as more Au is added to the Cu. This phenomenon also seems to exist in CuSn alloy systems.[24] The accuracy of the measurement and the lack of data on the thermal expansion coefficients of such alloys restrict further detailed analysis in this section.

B. CuAu(Fe) magnetic alloys

The total Fe-impurity contribution to the susceptibility of the magnetic alloys was obtained by subtracting the appropriate diamagnetic portion of the host susceptibility of CuAu from the measured susceptibility of CuAu(Fe) over the whole temperature range. In Fig. 3, the reciprocal gram susceptibility at high temperatures from 20 to 300 K has been plotted versus absolute temperature at a field of 12.75 kOe. In order to distinguish curve B from A, we have shifted the vertical coordinate for curve B in this figure. In Figs. 4–7 the χ curve in each figure shows more clearly the low-temperature characteristics of the data.

Most recent theoretical predictions and experimental measurements on the susceptibility of dilute magnetic alloys have confirmed that the single-impurity susceptibility should saturate to a constant value at $T=0$, and it should obey a Curie-Weiss law at temperatures large with respect to the characteristic temperature of the dilute-alloy system. Furthermore, Steiner *et al.*, from their recent experimental results on very dilute Cu(Fe), Ag(Fe), and Au(Fe) alloys,[25] indicated that the single-impurity susceptibility follows a T^2 behavior rather than a Curie-Weiss law at low temperatures. Therefore, the assumption that the single-impurity susceptibility obeys the Curie-Weiss law at low temperatures may not be appropriate.

In the analysis of the data, a new two-step procedure has been employed. At first, we have made

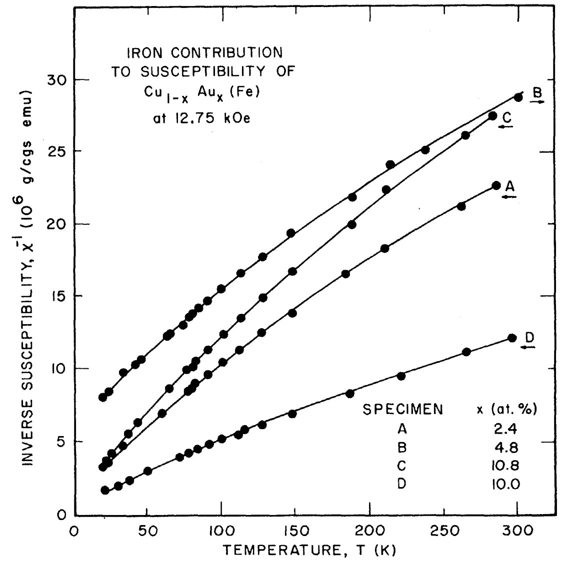

FIG. 3. Fe contribution to the susceptibility of each Fe-bearing specimen. The data at 12.75 kOe are shown for the temperature interval from 20 to 300 K. The solid lines are the best-fitting curves resulting from a least-squares analysis of the data by using the expression in Eq. (3).

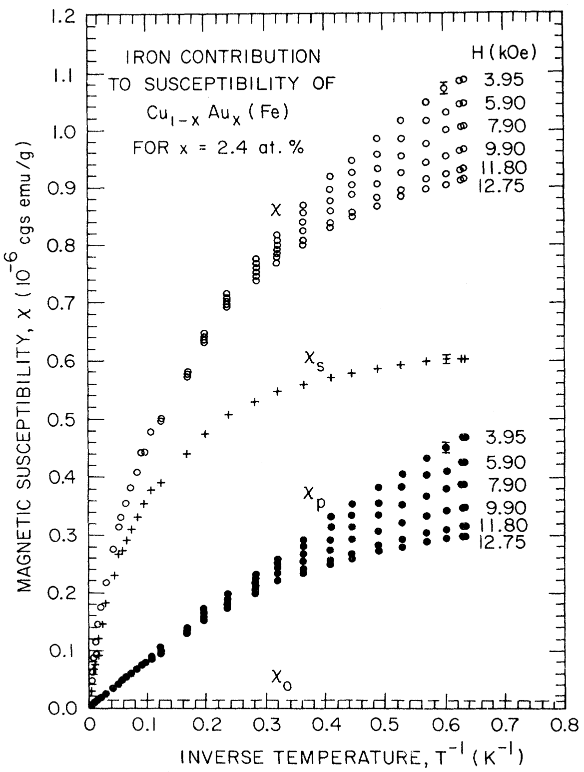

FIG. 4. Fe contribution of the susceptibility of the 2.4-at.%-Au alloy (0.045-at.% Fe). The figure also shows the χ_0, χ_s, and χ_p curves resulting from an analysis of data by using the expression in Eq. (4).

two assumptions: (i) The impurity-impurity interaction effect between two isolated Fe impurities is considered as a parallel-spin-coupled pair, with an effective spin just twice that of the isolated impurity ($S_{\mathrm{Fe}}=\frac{3}{2}$ for an isolated Fe atom in Cu). The susceptibility arising from this effect can be expressed by $\chi_p=C_2[JB_{J=3}(H/T)/H]$, where $B_{J=3}(H/T)$ is the Brillouin function and H is the applied magnetic field. The pairs of Fe impurities are regarded as isolated and noninteracting. (ii) The single-impurity susceptibility is assumed to be nearly field independent for the analysis of our measurements. The high-temperature portion of the excess susceptibility can be described by a Curie-Weiss term and a constant. The low-temperature portion cannot be described by such a simple expression.

From the high-temperature behavior of the single-impurity susceptibility and the assumptions made about the pairs, the following expression was used to fit the measured impurity susceptibility data at high temperatures (20–300 K):

$$\chi(H,T)=\chi_0+\frac{C}{T+\Theta}+\chi_p\ , \qquad (3)$$

where χ_0 and C are constants and Θ is a parameter with positive value. The resulting values of χ_0 and χ_p, obtained from Eq. (3) by a weighted least-squares fitting method, were used to find the single-impurity susceptibility $\chi_s(H,T)$ at low temperatures (1.5–20 K) by the equation

$$\chi_s(H,T)=\chi(H,T)-\chi_0-\chi_p\ . \qquad (4)$$

The single-impurity susceptibility $\chi_s(H,T)$ can be separated from the field-dependent susceptibility attributed to pairs over the range of the experiment by an extension of Eq. (4) to higher temperatures.

In the process of fitting the high-temperature data to Eq. (3) by the method of least squares, a reduced-residual-squared test was used as the "goodness-of-fit" criterion. Note that Eq. (3) is nonlinear in the parameter Θ. The remaining three parameters, χ_0, C, and C_2, appear in the equation in a linear way. An initial range was estimated for the nonlinear parameter Θ. Then, for each value of Θ the linear least-squares method was used to find values of χ_0, C, and C_2 that gave a minimum value for the goodness-of-fit criterion. Since the minimum in the sum of squares of the residuals is not a sharp function of Θ, an addition criterion was used to locate the "best-fit" value. The value of Θ was chosen such that the Brillouin

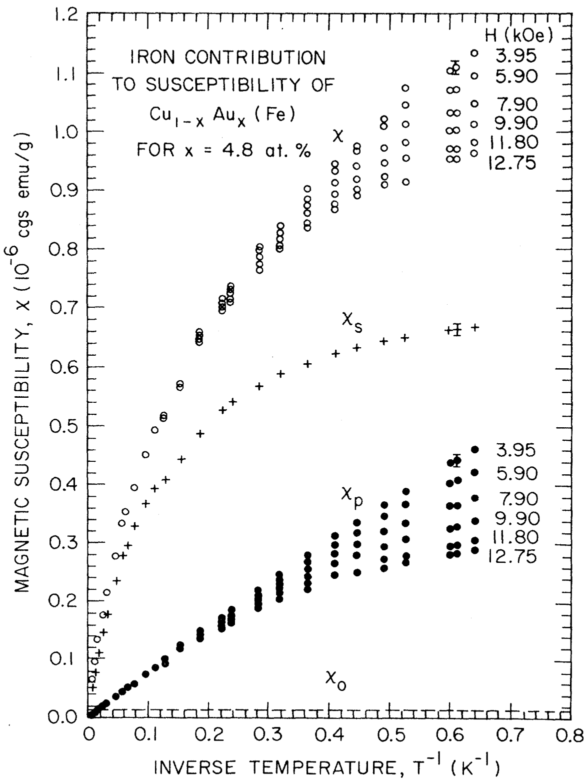

FIG. 5. Fe contribution to the susceptibility of the 4.8-at.%-Au alloy (0.044-at.% Fe). The figure also shows the χ_0, χ_s, and χ_p curves resulting from an analysis of data by using the expression in Eq. (4).

function gave a good description of the field dependence. The assignment of uncertainties to our results includes the effect of our fitting procedure. The parameter Θ is used to obtain a characteristic temperature T_c by the relation $\Theta = 1.25T_c$.

Before we present the final analysis on CuAu(Fe) alloys, it is important to apply this procedure to Cu(Fe) alloys. The magnetic-susceptibility data on Cu(Fe) (750 ppm) obtained by Huck *et al.*[13] in this laboratory has been reanalyzed by our procedure. The resulting χ_s term is shown in Fig. 8, where the inverse normalized susceptibility $\chi_s(0)/\chi_s(T)$ is plotted against the reduced temperature T/T_c. This figure also shows the theoretical relation of Götze and Schlottmann along with a plot of the results of a Mössbauer experiment on a very dilute Cu(Fe) (10 ppm) alloy by Steiner *et al.* The agreement among the experimental data in Fig. 8 indicates the existence of a universal susceptibility relationship in such alloys. The experimental data agree within the random fluctuations but do not follow the theoretical relation exactly. Götze and Schlottmann[9] also note that their expression does not agree exactly with the data on Cu(Fe).

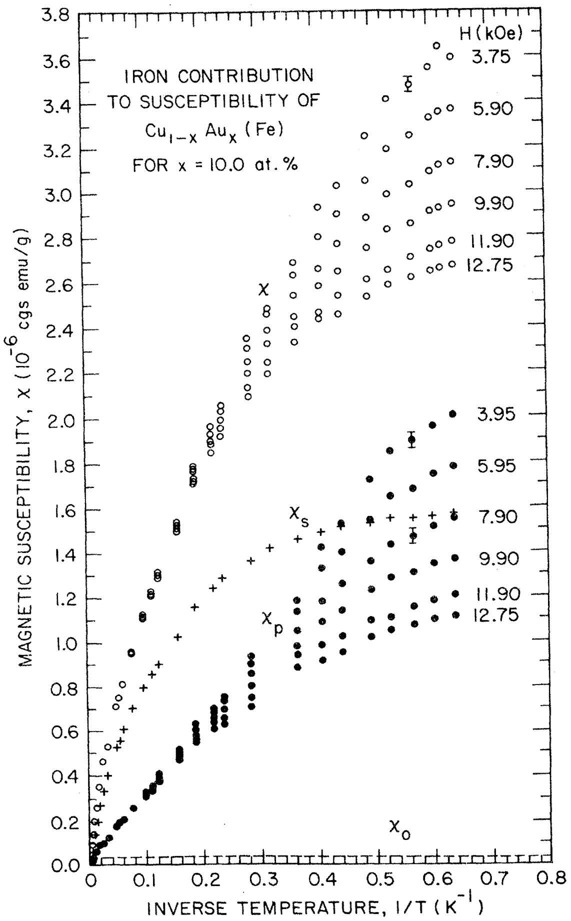

FIG. 6. Fe contribution to the susceptibility of the 10.0-at.%-Au alloy (0.076-at.% Fe). The figure also shows the χ_0, χ_s, and χ_p curves resulting from an analysis of data by using the expression in Eq. (4).

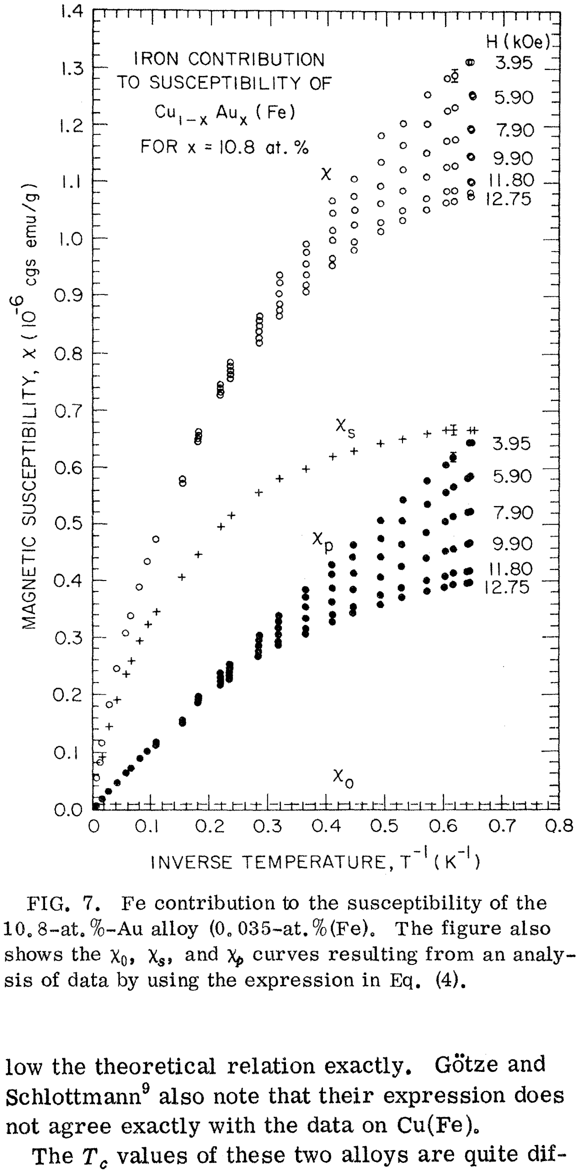

FIG. 7. Fe contribution to the susceptibility of the 10.8-at.%-Au alloy (0.035-at.%(Fe). The figure also shows the χ_0, χ_s, and χ_p curves resulting from an analysis of data by using the expression in Eq. (4).

The T_c values of these two alloys are quite different, with $T_c = 22.4$ K for Cu(Fe) (10 ppm) and $T_c = 13.6$ K for Cu(Fe) (750 ppm). This difference in T_c for these two alloys with different Fe concentrations is not surprising. Nagaoka[26] predicted in 1966 that the characteristic temperature of a Kondo alloy system would decrease with increasing magnetic impurity concentration. Star[27] first found indications of a possible concentration dependence of Kondo temperature from low-temperature-resistivity experiments on a series of Cu(Fe) alloys. Tsay and Klein[28] have also concluded theoretically that the effective Kondo temperature decreases as the impurity-impurity interaction increases. In order to obtain more experimental evidence for investigating this dependence on concentration of T_c, we applied our method of analysis to some published Cu(Fe) results: data (obtained by Ekström and Myers[29]) on a series of Cu(Fe) alloys with Fe

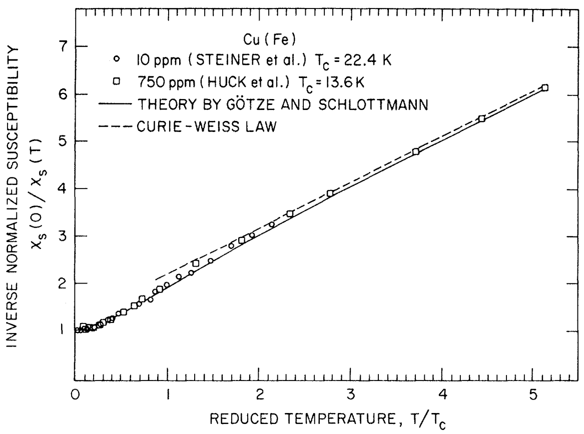

FIG. 8. Inverse normalized susceptibility $\chi_s(0)/\chi_s(T)$ vs reduced temperature T/T_C, for the Cu(Fe) alloys. Theoretical preditction with $\rho J = 0.2$ by Götze and Schlottmann and Curie-Weiss expression are shown by the solid and dashed line, respectively.

concentrations ranging from 92 to 304 ppm. The resulting T_c values associated with each single-impurity susceptibility are presented in Table IV. The concentration-dependent behavior of T_c in these Cu(Fe) alloys is shown in Fig. 9, where the dashed line indicates the trend in the behavior of this concentration dependence. In this figure, one can note that T_c decreases more rapidly with Fe concentration in the low-concentration range than a linear relationship. When the Fe concentration increases up to concentrations in excess of 100 ppm, T_c appears to decrease less rapidly with increasing Fe concentration.

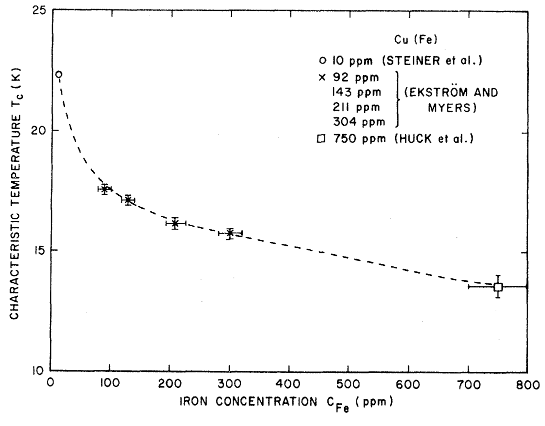

FIG. 9. Characteristic temperature T_c for copper host as a function of Fe concentration C_{Fe}.

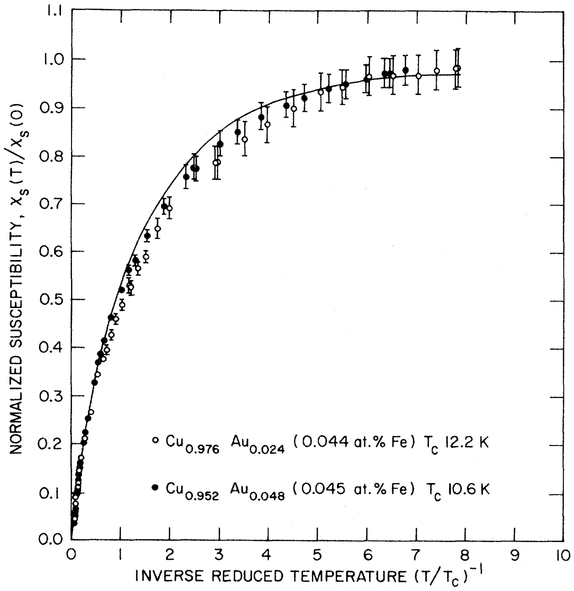

FIG. 10. Normalized susceptibility $\chi_s(T)/\chi_s(0)$ vs inverse reduced temperature $(T/T_c)^{-1}$. Solid curve is from theory of Götze and Scholttmann.

The results of the analysis of the susceptibility of our CuAu(Fe) alloys are shown in Figs. 10 and 11. These are plots of normalized susceptibility $\chi_s(T)/\chi_s(0)$ as a function of reduced temperature T/T_c. The CuAu(Fe) results all agree within experimental fluctuations of the data when plotted in this reduced form and would agree with the experimental results on Cu(Fe) alloys if plotted in the same manner as in Fig. 8. The parameters obtained from Eq. (3) are listed in Table III along with the effective magneton value and characteristic temperature T_c deduced from the parameters C and Θ, respectively. The data graphed in Figs. 10 and 11 also show a universal relationship in magnetic

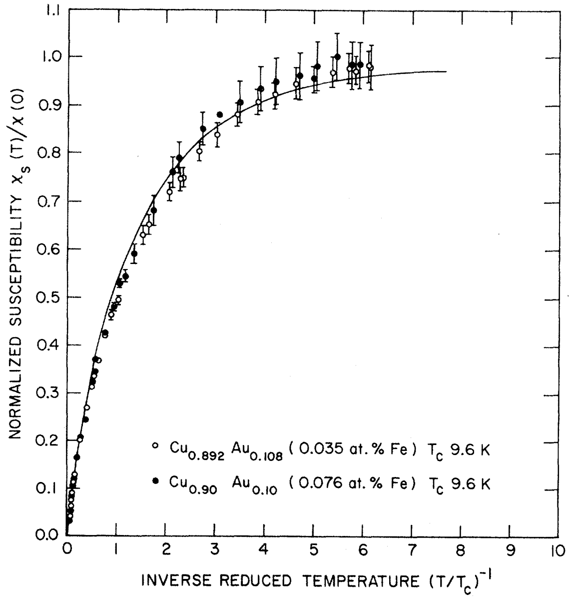

FIG. 11. Normalized susceptibility $\chi_s(T)/\chi_s(0)$ vs inverse reduced temperature $(T/T_c)^{-1}$. Solid curve is from the theory of Götze and Schlottmann.

TABLE III. High-temperature (20–300 K)-fitting result for the expression $\chi(H,T)=\chi_0+C/(T+\Theta)+C_2[JB_{J=3}(H/T)/H]$.

Sample	Fe concentration (ppm)	χ_0 (10^{-8} cgs emu/g)	C (10^{-6} cgs emu/K)	C_2 (10^{-3} erg/G g)	μ_{eff} (μ_B)	T_c (K)
Cu(Fe)	750 ± 50	1.426 ± 0.02	15.6 ± 0.2	5.7 ± 0.2	3.4 ± 0.1	13.6 ± 0.5
$CuAu_{2.4}$(Fe)-1	450 ± 50	1.151 ± 0.02	8.86 ± 0.1	1.5 ± 0.2	3.4 ± 0.1	12.2 ± 0.5
$CuAu_{4.8}$(Fe)-2	440 ± 50	1.135 ± 0.02	8.45 ± 0.2	2.0 ± 0.2	3.4 ± 0.1	10.6 ± 0.5
$CuAu_{10.8}$(Fe)-3	350 ± 50	0.874 ± 0.02	7.0 ± 0.1	1.5 ± 0.5	3.4 ± 0.1	9.6 ± 0.5
$CuAu_{10.0}$(Fe)-4	760 ± 50	1.936 ± 0.02	17.0 ± 0.2	5.5 ± 0.5	3.4 ± 0.1	9.6 ± 0.5

susceptibility. The solid line in each figure represents the graph of the theoretical expression. A similar universal relationship in low-temperature resistivity has been previously found for similar alloys by Loram *et al.*[11] The existence of this universal susceptibility curve strongly indicates that CuAu(Fe) alloys can be interpreted in terms of a single parameter T_c as observed in both resistivity[11] and specific-heat[12] experiments. A somewhat similar procedure has been used by Haddad and Sarachik[18] to obtain a universal curve for the susceptibility attributable to Fe impurities. Their procedure employs a constant and two Curie-Weiss terms and does not permit an easy comparison with our results or the theoretical description of Götze and Schlottmann. The disagreement between the experimental results and the theoretical curve at $T\sim T_c$ is consistent with the uncertainty of the determination of the T_c value. However, systematic discrepancies do exist at intermediate temperatures. This deviation of Götze and Schlottmann's calculation from the results of measurements on Cu(Fe) is shown in Fig. 8 and for our CuAu(Fe) measurements in Figs. 10 and 11. Götze and Schlottmann[9] have attributed this deviation to an imperfection of the approximation used in this method rather than a prediction of expected behavior. A characteristic temperature T_c that decreases with the addition of Au was found in both specific-heat and resistivity experiments on similar alloy systems. The amount of Fe impurity plays an important role in the concentration dependence of T_c where it decreases with increasing Fe concentration as shown in Fig. 9. The dependence of T_c upon Fe concentrations in the CuAu(Fe) system is not the same as in pure-copper host. As shown in Table III, the T_c values of those alloys with dissimilar Fe concentration and the same Au concentration are obviously not described by the relationship for Cu(Fe). The possible explanation could be that the mean free path of conduction electrons is substantially reduced in a CuAu host because of the addition of Au. Since the mean free path is the significant quantity in the impurity-impurity interaction, the formula for a CuAu host should be different from the one for a pure-Cu host. However, more theoretical and experimental investigations are needed to explore this problem. For those alloys listed in Table III with similar Fe concentration T_c appears to decrease slightly with increasing Au concentration. The exact way T_c depends on both Au and Fe concentration is still uncertain.

The constant χ_0, a temperature-independent orbital contribution to the susceptibility of Fe in a Cu host,[30] shows a possible linear relationship with Fe concentration in Fig. 12 for both Cu(Fe) and CuAu(Fe) systems. This indicates the possibility of an alloying effect by Fe in Cu-host alloys. Although the data for Cu(Fe) and CuAu(Fe) fall on separated straight lines, it is certainly possible for a single relation to describe the relationship of χ_0 that would be consistent with the experimental uncertainties.

The effective magnetic moment of the single Fe ion can be deduced from the constant C in the Curie-

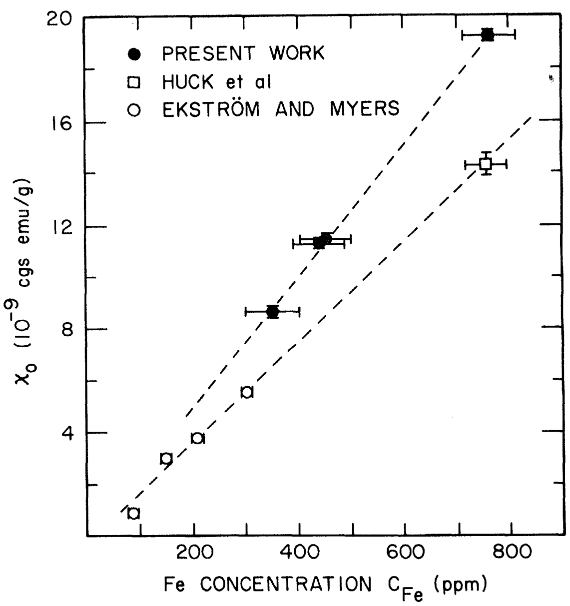

FIG. 12. Temperature- and field-independent susceptibility χ_0 vs Fe concentration.

TABLE IV. Iron-cencentration dependence in several Cu(Fe) alloys of T_c calculated by the use of Eq. (3).

Sample	Iron concentration (ppm)	Characteristic temperature T_C(K)	Source
1	10	22.4	Steiner *et al.*
2	92	17.6 ± 0.2	Ekström and Myers
3	143	17.2 ± 0.2	Ekström and Myers
4	211	16.2 ± 0.2	Ekström and Myers
5	304	15.8 ± 0.2	Ekström and Myers
6	850	13.6 ± 0.5	Huck *et al.*

Weiss term. The results from all Fe-bearing alloys show the same value of $(3.4 \pm 0.1)\mu_B$ as presented in Table IV. This number agrees with the values found from two independent experiments by Tholence and Tournier[2] and Ekström and Myers.[29]

The concentration C_{pairs} of magnetic pairs with $J = 3$ can be estimated from constant C_2 in Eq. (3). A plot of C_{pairs} versus C_{Fe}^2, which does not correct for differences in Au concentrations, is shown in Fig. 13, and shows a rough linear dependence. The ratio of the numbers of pairs to that of the singles is estimated to be about $(68 \pm 14)C_{\text{Fe}}$. This result is consistent with the value of $75C_{\text{Fe}}$ obtained by Franz and Sellmyer[3] on a Cu(Fe) magnetization experiment, and is also consistent with a Mössbauer experiment by Window[31,32] on a series of Cu(Fe) alloys.

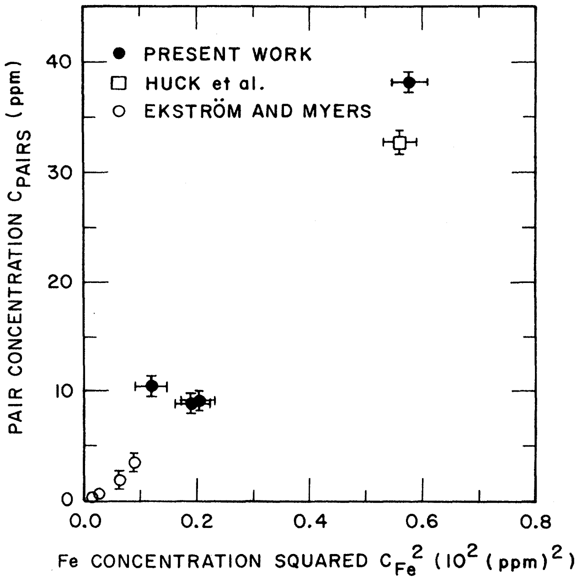

FIG. 13. Dependence of pair concentration C_{pair} on the square of the single-Fe concentration C_{Fe}.

V. CONCLUSION

By means of a simple procedure, we have successfully separated the single-impurity susceptibility χ_s from that due to magnetic pairs χ_p acting with spin equal to 3. The single-impurity susceptibility for all CuAu(Fe) alloys, χ_s, was compared with the theoretical curve based on the *s-d* exchange model calculation by Götze *et al.* The field-dependent part of the observed susceptibility for CuAu(Fe) can be described completely by a single Brillouin function with $J = 3$ for pairs. This contributed a susceptibility value which was comparable to χ_s at low temperatures. A dependence on both Au and Fe concentrations of the characteristic temperature of CuAu(Fe) alloys, T_c, was found. The characteristic temperature is dependent upon the iron concentration for both Cu(Fe) and CuAu(Fe). The estimated pair concentration and the average effective-moment value of Fe for all Fe-bearing samples are $(68 \pm 14)C_{\text{Fe}}$ and $(3.4 \pm 0.1)\mu_B$, respectively. A universal relationship of the normalized susceptibility, $\chi_s(T)/\chi_s(0) = f(T/T_c)$, was found over the entire temperature range for our experimental results.

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