Magnetization and susceptibility of dilute CuAu(Fe)†*

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The magnetization M and susceptibility χ of a series of dilute CuAu(Fe) alloys with varying host compositions and Fe concentrations have been measured as a function of magnetic field to 50 kG at temperatures between 1.38 and 120 °K. The results for M and χ for the Cu-rich alloys can be separated into linear and quadratic terms in the concentration c. The term proportional to c^2 has a spin value near $S = 3$, and is attributed to the presence of pairs of ferromagnetically coupled Fe atoms. The single-impurity susceptibility yields values of the Weiss temperature Θ_1 which decrease rapidly as the Au content of the host is increased. Further, all the single-impurity data fit on a single universal curve, and yield values of T_K for the various CuAu hosts which are in good agreement with those deduced from other measurements. These results provide further support for the use of a one-parameter model to describe the CuAu(Fe) system.

I. INTRODUCTION

Extensive measurements[1] on Cu(Fe) have shown that its Kondo temperature T_K occurs in the vicinity of 30 °K. Careful magnetization and susceptibility measurements by Tholence and Tournier[2] have indicated that, in addition to the single-impurity (Kondo) contribution, which is linear in concentration, there exists also a term proportional to c^2, which has been attributed to the presence of pairs of Fe atoms. Other measurements of susceptibility and magnetization[3–5] and Mössbauer,[6] magnetoresistance,[7] and specific-heat[8] results also indicate the presence of this additional term.

Resistivity[9] and specific-heat[10] studies have shown that the addition of Au to Cu(Fe) rapidly depresses the Kondo temperature T_K. The impurity contribution was found to fit a universal curve when plotted as a function of T/T_K, and thus the behavior of each alloy is adequately described by a single parameter T_K. Magnetoresistance[11] and Hall-coefficient[12] data support the conclusion that T_K decreases rapidly with Au content. On the other hand, the susceptibility of Fe in $Cu_{0.9}Au_{0.1}$ was measured by Ekström and Myers,[3] and their results indicate that T_K for this alloy is unchanged from its value in Cu(Fe).

In order to shed further light on the applicability of a single-parameter description for the susceptibility of CuAu(Fe) and to investigate the behavior of T_K, a systematic study[13] was undertaken of the magnetization and susceptibility of a series of CuAu hosts containing various Fe concentrations. In addition, such a study would also yield information concerning the existence and behavior in CuAu of a contribution due to Fe atom pairs of the kind found in Cu.

II. EXPERIMENTAL PROCEDURE

The samples were in the form of 1-g buttons prepared from 99.999%-purity starting materials by melting in an argon-atmosphere arc furnace on a water-cooled hearth. For each $Cu_{1-x}Au_x$ host composition, master alloys without Fe and with 600 ppm Fe were prepared, and appropriate amounts of these two masters were then melted together to obtain a series of samples with Fe concentrations between 0 and 600 ppm. The samples were then annealed in vacuum at 830 ° C for 8 days and quenched in iced brine. Before melting, annealing, or measuring, all materials were thoroughly cleaned using appropriate etching and electropolishing techniques. All samples were stored in liquid nitrogen when not in use.

The magnetization was measured using the Faraday method. A uniform magnetic field of up to 50 kG was supplied by a superconducting solenoid, and a field gradient of 485 G/cm was provided by a separate set of superconducting coils. A Cahn electrobalance with a sensitivity of 2 μg was used to measure the change in sample weight when the polarity of the gradient field was reversed. This method eliminates all (non-time-varying) forces that are not odd powers of the gradient field. The magnetic field produced by the gradient coil is zero at its center, and thus the gradient field coil alone will not produce a force on a sample positioned at its center. This provides the further advantage that the center of the coil, which corresponds to the ideal sample position, can be easily and accurately located. Measurements of the magnetization of a nominally pure Au sample on several different occasions indicate that the uncertainty due to errors in sample repositioning is less than 0.1%. The largest difference in any of these measurements was 0.11%.

The estimated over-all error in relative values of the magnetization is less than 0.3%. Measurements on the nominally pure Au samples were used to calibrate the instrument by comparison with Hurd's[14] data to determine the proportionality factor relating the microbalance output to magnetization. There is approximately a 1% uncertainty in this procedure, stemming mainly from the presence of some Fe impurities in the nominally pure samples.

Temperatures between 1.38 and 122° K were obtained in an insert Dewar, surrounded by an insulated He^4 space; standard methods involving exchange gas and heaters were used. Thermometry was provided by an Allen-Bradley carbon resistor up to 20° K and by a platinum resistance thermometer at higher temperatures. The thermometers were mounted on a copper block which was thermally connected to the sample by exchange gas. These thermometers were calibrated by measuring the susceptibility of chrome potassium alum, and by comparison at zero field with a calibrated germanium thermometer hung in place of the sample. Temperatures were reproducible to 0.2%, and their absolute values were determined to 1%.

The magnetization was measured in fields of 0, 1, 2, 5, 10, 20, 30, 40, 45, and 50 kG in liquid helium at atmospheric pressure (4.2° K), and at each of the temperatures 1.38, 9.5, 44, 55, 66, 77, 89, 100, 111, and 122° K. These temperatures were controlled by using a feedback circuit to maintain the thermometer resistance constant to 0.1%. No corrections were made for the magnetoresistance of the thermometers. As the field is raised from 0 to 50 kG, this causes a 3% increase in the temperature at 1.38° K and an increase of about 1% at 9.5° K. This will have an effect on the results presented for the magnetization as a function of field, where the plots shown will be only approximately isothermal (Figs. 1, 4, and 5). As will be shown in Secs. III and IV, however, the important results concern the magnetization of samples with various concentrations at fixed field (Figs. 2 and 3) and the temperature dependence of the low-field susceptibility (Figs. 6 and 7); these are unaffected by the magnetoresistance error.

III. DATA ANALYSIS

Figure 1 presents data for magnetization as a function of magnetic field at several temperatures for a sample of $Cu_{0.91}Au_{0.09}$(Fe), and is typical of the data obtained for all 33 alloys containing Cu. We attribute the curvature at low fields (below ~5 kG) to the presence of superparamagnetic clusters similar to those previously observed in Cu(Fe).[3,15] For the curves above 40° K, extrapolation of the linear high-field behavior to $H=0$ yields values of the intercept M_c which are roughly independent of temperature. The magnetization curves at lower temperatures, when fitted to a polynomial function of H, yield similar values of M_c, the saturation magnetization of the clusters. Assuming that each Fe atom in a cluster contributes $2.2\mu_B$ to M_c, we estimate that less than 0.5% of the nominal Fe concentration is involved in the clusters. We have corrected the data for the presence of these clusters by subtracting M_c from all data for 5 kG and above.

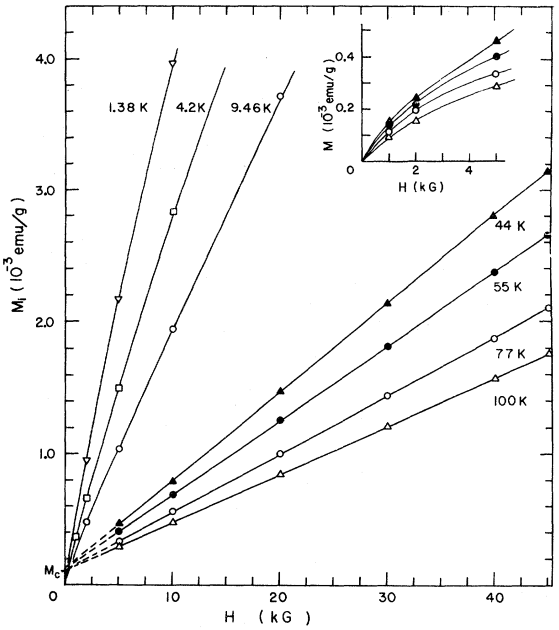

FIG. 1. Impurity magnetization as a function of magnetic field for 197 ppm Fe in $Cu_{0.91}Au_{0.09}$.

The impurity magnetization divided by the nominal Fe concentration c is plotted in Fig. 2 as a function of c. The impurity magnetization has been obtained by subtracting the magnetization of a nominally pure sample from the magnetization of the samples containing Fe. The data presented are for 1.4 °K and 40 kG, and are typical of plots for other fields and temperatures. The Cu(Fe) data of Tholence and Tournier[2] at nearly the same temperature and field are shown for comparison. As in the case of Cu(Fe), the magnetization of the Cu-rich alloys ($Cu_{0.95}$ and $Cu_{0.91}$) contains linear and quadratic terms in concentration, which can be associated with the presence of single Fe impurities and pairs of ferromagnetically coupled Fe atoms. At these low Fe concentrations any higher-order terms in concentration are expected to be small, and our data are adequately described within experimental error using only linear and quadratic terms.

The magnetization can be expressed as

$$M = M_h + M_s n_s + M_p n_p, \tag{1}$$

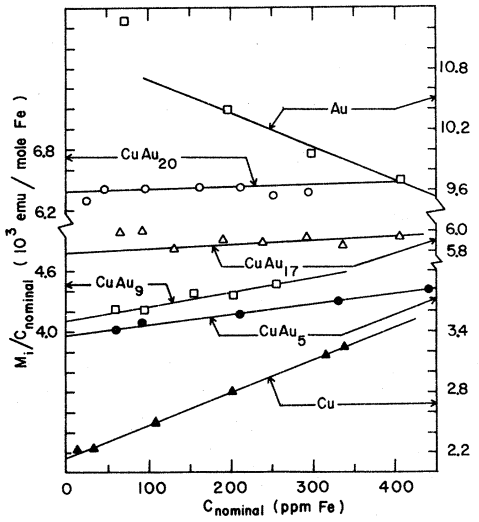

FIG. 2. Impurity magnetization per mole of Fe vs nominal Fe concentration in CuAu hosts at 40 kG and 1.4 °K. Data for Cu(Fe) are taken from Ref. 2.

where M_h is the host magnetization, M_s and M_p are, respectively, the magnetization per single Fe impurity and the magnetization per pair, n_s and n_p are the corresponding concentrations, and $c = n_s + 2n_p$. The concentration of pairs[16] is proportional to c^2, so that $n_s = c - 2kc^2$, and the magnetization is

$$M = M_h + M_s c + (M_p - 2M_s)kc^2. \tag{2}$$

Solving $n_s = c - 2kc^2$ for c, the magnetization can also be written in terms of n_s

$$M = M_h + M_s n_s + M_p k n_s^2 + O(n_s^3). \tag{3}$$

We have chosen to analyze our data in terms of the latter expression, dropping terms of order n_s^3 and higher. The advantage of this choice is that the quadratic term involves the pair magnetization M_p only, rather than the combination M_p and M_s which enters in Eq. (2). Further, the single-impurity concentration n_s can be deduced from high-field susceptibilities and exhibits less scatter than the nominal concentrations c.

We assume that the pair magnetization is nearly saturated at our lowest temperature (1.4 ° K) and our highest field (50 kG). This is similar to the procedure invoked by Tholence and Tournier for Cu(Fe), although they had a somewhat higher field.[2] A detailed analysis[17] shows that if this assumption is not fully satisfied, the coefficient of the quadratic term in Eq. (3) will have a small admixture of the single-impurity magnetization M_s(thus perhaps introducing an error into values later deduced for M_p). Relative values of the single-impurity concentrations n_s were thus deduced from the high-field slopes of the magnetization curves, and their absolute values were chosen by setting $n_s = c$ at one particular concentration. This is a reasonable normalization procedure, since $c = n_s + 2n_p$, and the size of $2n_p$ is of the order of, or smaller than, the uncertainties in the values of nominal concentration c.

Figure 3 shows the total impurity magnetization M_i divided by n_s, as a function of n_s. This is analogous to Fig. 2, with the total nominal concentration c replaced by the single-impurity concentration n_s. The scatter of the data in Fig. 3 is smaller and shows that the data points for the 9-, 17-, and 20-at.%-Au samples lie on several separate straight lines, correponding to groups of samples which were arc melted in separate furnace charges. Further, the 17- and 20-at.%-Au curves appear to have small positive slopes, indicating the presence of Fe pairs in these hosts. This was not as apparent in Fig. 2, partly owing to scatter of data points. A further reason is that the slope of the curves in Fig. 2 is not a direct measure of the pair contribution, since it involves the term $(M_p - 2M_s)$ of Eq. (2), whereas the slope of the curves in Fig. 3 is given by Eq. (3) and involves M_p only. The decrease in slope for increasing Au content suggests a decrease in the number of Fe pairs. However, this may not be the case for the following reason. As mentioned earlier, if the pair magnetization is not fully saturated at our highest field and lowest temperature (as is likely), there will be a systematic error in the values deduced for n_s, with a consequent small admixture of the single-impurity magnetization M_s in the quadratic term of Eq. (3). It will be shown in Sec. IV that, as the Au content of the host is increased, M_s at a given temperature and field becomes larger. Any admixture of the single-impurity term thus becomes more serious.

Using the method of least squares, the measured magnetization for the 5-, 9-, and 17-at.%-Au alloys was fitted to Eq. (3) at a given temperature and

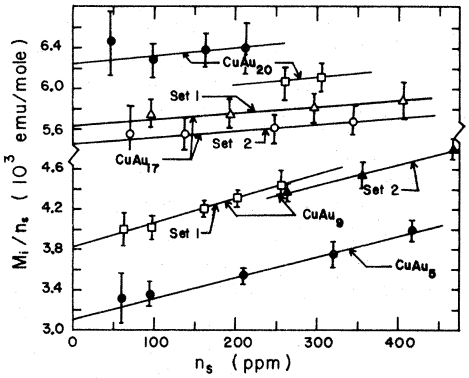

FIG. 3. M_i/n_s as a function of single-impurity concentration n_s in CuAu hosts at 40 kG and 1.4 °K.

magnetic field, and values were determined for M_h, M_s, and kM_p which will be presented in Sec. IV. The low-field susceptibilities, and values of the Curie-Weiss temperature Θ_1 and Θ_2 associated with single impurities and pairs, were deduced and will also be presented and dicussed later. The data for the 20-at.%-Au alloy were not fitted to Eq. (3), since for the reasons detailed above, the values deduced for M_p are increasingly less reliable as the Au content of the host is increased.

In order to check the validity of the above procedure, we performed a different analysis which does not rely on a determination of the single-impurity concentration n_s. We determined the total (low-field) impurity susceptibility directly from our data, and for each sample fitted it to the expression

$$\chi_i = \chi_0 + [C_s/(T+\Theta_s)] + [C_p/(T+\Theta_p)] , \quad (4)$$

using the method of least squares. The first two terms describe the single-impurity behavior[18] and the last term refers to pairs. When Θ_s and Θ_p are held fixed at the values Θ_1 and Θ_2 mentioned above, the other parameters χ_0, C_s and C_p were found to be in good agreement with those deduced from our first analysis. When all parameters were allowed to vary, Θ_p was found to depend on concentration in the case of $Cu_{0.95}Au_{0.05}$. This behavior is very similar to that found by Ekström and Myers in Cu(Fe), and is not well understood. Very good agreement was obtained for $Cu_{0.91}Au_{0.09}$. The single-impurity term in both the 5- and 9-at.%-Au cases was largely unaffected. For higher Au content (17 and 20 at.%), a reliable separation becomes difficult, because Θ_s and Θ_p become comparable (as will be shown later), and thus the temperature dependence of the last two terms in Eq. (4) becomes very similar.

IV. RESULTS AND DISCUSSION

Data for the host alloys containing 5-, 9-, and 17- at.% Au were analyzed as described in Sec. III by subtracting a contribution to the magnetization due to clusters and fitting the remainder to Eq. (3) using the method of least squares. This method of separation becomes more difficult as the Au content of the host is increased, and was not carried out for $Cu_{0.80}Au_{0.20}$ (Fe) and Au(Fe).

The values derived for the host magnetization using Eq. (3) are found to be linear functions of field at all measured temperatures. There is a slight increase (the largest is 2%) in the susceptibility as the temperature is decreased to 1.38 °K, which we attribute to small uncertainties in the concentrations used in Eq. (3), or to the presence of a small amount (1 ppm or less) of another magnetic impurity. The values found for the host susceptibility at 77 °K are -1.005×10^{-7}, -1.081×10^{-7}, -1.227×10^{-7}, and -1.30×10^{-7} emu/g for the 5-, 9-, 17-, and 20-at.%-Au hosts, respectively. These values lie, as expected, between the susceptibilities of pure Cu and pure Au, and the host diamagnetism increases as the Au content of the alloys is increased.

In Secs. IV A and IV B we shall present and discuss the contributions to the magnetization and susceptibility due to pairs and to single impurities.

A. Pairs

As discussed in Sec. II, it was assumed that the pair term in the magnetization was saturated at the highest field (50 kG) and lowest temperature (1.38 °K), and one expects therefore that the calculated values of the coefficient M_2 of the quadratic term of Eq. (3) will increase monotonically to a constant value at high fields. The behavior of M_2 can be seen in Fig. 4 where M_p, which is proportional to M_2, is plotted as a function of magnetic field. The magnetizations of the $Cu_{0.95}Au_{0.05}$ and $Cu_{0.91}Au_{0.09}$ hosts behave as expected, and resemble a Brillouin function. On the other hand, the $Cu_{0.83}Au_{0.17}$ host shows an unexpected maximum. As discussed in Sec. II, this may result from an admixture into M_2 of the single-impurity magnetization.[19] A value for $M_{2\ \mathrm{sat}}$ can therefore not be reliably deduced from the data. Values of $M_{2\ \mathrm{sat}}$ for the 5- and 9-at.%-Au hosts are listed in Table I. The susceptibility χ_2 associated with M_2 fits a Curie-Weiss law $C_2/(T+\Theta_2)$. Values of C_2 and Θ_2 are also listed in Table I.

The usual forms for the saturation magnetization and the Curie constant give

$$M_{2\ \mathrm{sat}} n_s^2 = g\mu_B S(N_0/M) n_p \quad (5)$$

and

$$C_2 n_s^2 = g^2\mu_B^2[S(S+1)/3k_B](N_0/M) n_p , \quad (6)$$

where N_0 is Avogadro's number, M is the gram

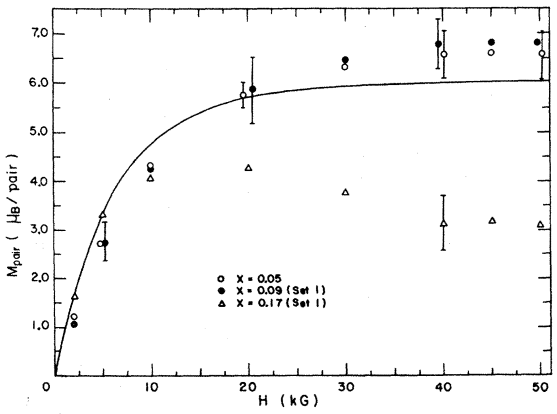

FIG. 4. Magnetization as a function of magnetic field at 1.4 °K due to pairs of Fe atoms in $Cu_{1-x}Au_x$. The solid line is a Brillouin function for $S=3$.

TABLE I. Parameters describing the pair contribution to the magnetization and susecptibility of $Cu_{1-x}Au_x$ alloys containing Fe.

x	$M_{2\,sat}$ $\{10^{-9}$ [emu/g(ppm Fe)^2]$\}$	C_2 $\{10^{-12}$ [emu °K/g(ppm Fe)^2]$\}$	k	S	Θ_2 (°K)
0.05	31 ± 2	6.2 ± 1.3	61 ± 15	3.3 ± 1.2	1.5
0.09 Set 1	43 ± 4.3	7.1 ± 1.2	86 ± 20	2.7 ± 0.9	0.7
Set 2	25 ± 2.5	5.4 ± 0.9	36 ± 15	3.8 ± 1.5	0.4
0.17 Set 1		2.5	36	assume 3	less than 1.3

molecular weight of the host, and n_s and n_p are the single-impurity and pair concentrations. Using $n_p = kn_s^2$ and $g = 2$, these two expressions can be used to find values for the spin S and the parameter k, which are listed in Table I. The parameter k listed for the 17-at.%-Au alloy was estimated from Eq. (6), using a value $S = 3$. It should be noted, however, that the single-impurity admixture also affects the value of C_2 probably resulting in an underestimate of k for this alloy. The pair magnetization $M_p = M_2/k$ at 1.38 °K is plotted as a function of magnetic field in Fig. 4. A Brillouin function for spin $S = 3$ is shown for comparison.

The calculated spin values listed in Table I are near $S = 3$, and M_p shown in Fig. 4 is quite similar to the Brillouin function for $S = 3$. This is consistent with the assumption that the quadratic term is due to pairs of ferromagnetically coupled Fe atoms. The values of Θ_2 are near 1 °K, which is similar to the value $\Theta_2 < 5$ °K found for Cu(Fe).[2,3] The concentrations of pairs, given by the parameter k, are approximately one-half the value found for Cu(Fe) by Tholence and Tournier[2] and comparable with values obtained by Franz and Sellmyer.[4]

B. Single impurities

Figure 5 shows the single-impurity magnetization per impurity as a function of magnetic field for all five host alloys studied. At higher temperatures the magnetization is a linear function of field, as expected. The single-impurity magnetization was obtained for the host alloys containing 5-, 9-, and 17-at.% Au by fitting the total magnetization to Eq. (3) and deducing the coefficient M_s of the linear term. A separation into pair and single-impurity terms was not performed for the $Cu_{0.80}Au_{0.20}$(Fe) and Au(Fe) alloys. The single-impurity magnetization presented for these two alloys was obtained by subtracting the measured host magnetization from the magnetization of the same alloy containing relatively dilute (99- and 61-ppm) Fe concentrations. Based on the results, it will be argued below that for these two alloys direct subtraction of the host magnetization yields the correct qualitative behavior of the single-impurity term, and that this behavior will not be affected in any significant way by the possible admixture of a small pair term.

Figure 5 shows that as the host Au content is increased there is an increase both in the magnitude of M_s and in its curvature as a function of field. Thus the impurity behaves more like a free spin, indicating a decrease in T_K as the host Au content is increased.

The inverse susceptibility χ_s^{-1} deduced from M_s is presented as a function of temperature in Fig. 6. If the low-temperature portion is fitted to a Curie-Weiss law, the intercept, which is a measure of T_K, clearly decreases as the host Au content is increased. At higher temperatures there are deviations from this simple low-temperature Curie-Weiss law. For the alloys with 5-, 9-, and 17-at.% Au, the susceptibility is adequately described over the entire range of temperature by the expression $\chi_0 + C_1/(T + \Theta_1)$, and values of χ_0, C_1, and Θ_1 obtained from least-squares fits to this expression are listed in Table II.

As the Au content of the host increases, the values of Θ_1 associated with the single-impurity term decrease rapidly and become comparable with the values of Θ_2 deduced for the pair term. Since the

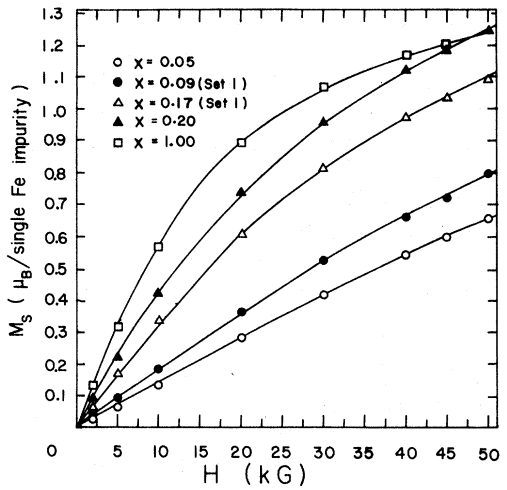

FIG. 5. Magnetization as a function of magnetic field at 1.4 °K due to single Fe impurities in $Cu_{1-x}Au_x$.

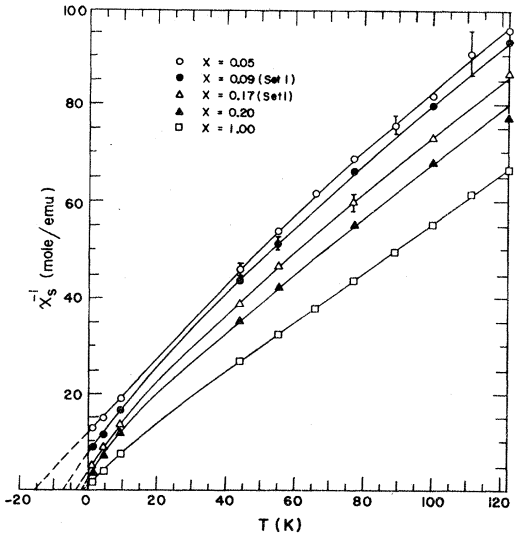

FIG. 6. Inverse susceptibility due to single Fe impurities in $Cu_{1-x}Au_x$ hosts as a function of temperature.

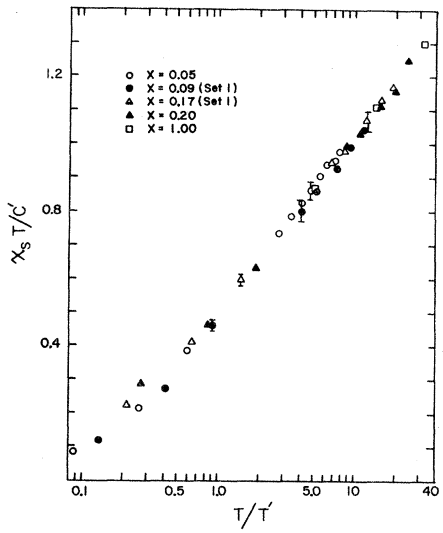

FIG. 7. Plot of $\chi_s T/C'$ vs T/T', where χ_s is the single-impurity susceptibility, T is the temperature, and C' and T' are parameters chosen so as to make all the data lie on a single universal curve.

magnetization is expected to behave roughly as a Brillouin function with argument $H/(T+\Theta)$, the pair and single-impurity terms have similar field dependences if they have comparable values of Θ. Therefore the possible admixture of a small pair term in the data presented for the $Cu_{0.80}Au_{0.20}$(Fe) and Au(Fe) alloys would not significantly affect the field dependence of the magnetization shown in Fig. 5 or of the susceptibility shown in Fig. 6.

Several theoretical calculations[20] indicate that one expects universal behavior for the susceptibility of the kind

$$\chi_s = (C/T) f(T/T_K), \tag{7}$$

where χ_s, T, and C refer to the single-impurity susceptibility, the temperature, and the Curie constant, and $f(T/T_K)$ is a universal function of T/T_K. Thus plots of $\chi_s T/C$ as a function of T/T_K should superimpose for the various alloys. Figure 7 shows $\chi_s T/C'$ vs T/T', where the parameters C' and T' have been adjusted to give superposition, and the values $C' = 1.3$ emu °K/mole and $T' = 15.8$ °K were chosen for the $Cu_{0.95}Au_{0.05}$(Fe) alloy, following Table II. It is clear from Fig. 7 that values of C' and T' can be chosen to give a very good universal curve, and these parameters are listed in Table III for the various alloys. The values listed for C', which according to theory should be proportional to $N_s g \mu_B S(S+1)/3k_B$, are seen to vary only slightly; this variation may be due to small changes[21] in the spins S or, more likely, to uncertainties in n_s (i.e., the concentrations). Thus C' is essentially constant, in contrast to the values of C listed in Table II, which were found by fitting to the expression $\chi = \chi_0 + C/(T+\Theta_1)$; one should note that the parameter C in this last expression is related to the Curie constant $N_s \mu_{\text{eff}}^2$ (and Θ is related to the Kondo temperature T_K) in different ways[1] for different ranges of T/T_K.

The parameters T' deduced from the universal curve of Fig. 7 should be proportional to the Kondo temperature T_K. Table IV lists values of T'/T_0' (column 2) and Θ_1/Θ_{10} (column 3) found from the present experiments, and of T_K/T_{K0} deduced from studies of the resistivity[9,22] (column 4) and specific heat[10] (column 5), where T_0', Θ_{10}, and T_{K0} refer to

TABLE II. Parameters obtained by fitting the single-impurity susceptibility to the expression $\chi_s = \chi_0 + C_1/(T+\Theta_1)$ for $Cu_{1-x}Au_x$ alloys containing Fe.

x	χ_0 [10^{-4} (emu/mole Fe)]	C_1 (emu °K/mole Fe)	Θ_1 (°K)
0.05	6.1	1.31	15.8
0.09 Set 1	30	1.06	8.4
Set 2	28	1.04	8.4
0.17 Set 1	61	0.87	3.3
Set 2	86	0.78	2.6

TABLE III. Parameters C' and T' obtained for the universal curve of Fig. 7.

x	T' (°K)	C' (emu °K/mole Fe)
0.05	15.8	1.31
0.09	10.4	1.23
0.17	6.3	1.21
0.20	4.9	1.27
1.00	0.3	0.97

TABLE IV. Values of T', Θ_1, and T_K for $Cu_{1-x}Au_x$(Fe) alloys, normalized to their values in Cu(Fe). The parameters T' and Θ_1 listed in columns 2 and 3 are deduced from the present experiment. Values of T_K presented in columns 4 and 5 are taken, respectively, from resistivity measurements of Refs. 9 and 22 and from specific-heat measurements of Ref. 10.

x	T'/T'_0	Θ_1/Θ_{10}	T_K/T_{K0}	T_K/T_{K0}
0.05	0.54	0.54	0.54	0.75 (4.8 at.% Au)
0.09	0.36	0.32	0.36 (10 at.% Au)	0.5 (10.8 at.% Au)
0.17	0.22	0.11	0.17	
0.20	0.17			
1.00	0.01		0.01	

values for Cu(Fe). The value of Θ_1 for the 5-at.%-Au alloy was found to be 0.54 of the Θ_{10} for Cu(Fe) measured by Tholence and Tournier.[2] The ratio of T_K's for the same two alloys was also found to be 0.54 in resistivity measurements of Loram, Whall, and Ford.[9] Since no measurement of T' is available for Cu(Fe), its value was chosen by setting $T'/T'_0 = 0.54$ for $Cu_{0.95}Au_{0.05}$, and all other ratios T'/T'_0 then follow.

The ratios of T' shown in column 2 compare quite closely with the ratios of T_K in column 4 deduced from resistivity measurements. The specific heat values differ numerically, but show the same decrease with increasing Au content. As mentioned earlier, Θ_1 is related to T_K differently depending on the range of T/T_K considered. Since T_K varies rapidly with increasing Au content, the ratios of Θ_1 are not simply related to the ratios of T_K.

An earlier susceptibility experiment by Ekström and Myers[3] on a $Cu_{0.90}Au_{0.10}$ alloy containing Fe indicated that the Kondo temperature T_K remained essentially unchanged from its value in Cu(Fe). In contrast, the present results show a smooth monotonic decrease in T_K with increasing Au content, in agreement with results of resistivity,[9,22] specific-heat,[10] and magnetoresistance[11] measurements. Further, the susceptibility data fit a universal function of T/T_K, and the T_K values deduced agree well with the T_K's obtained from these other measurements. Thus a single-parameter T_K adequately describes the behavior of these alloys.

The present data can, in fact, be used to argue against a "local" model of the Jaccarino-Walker[23] type. The simplest form of such a model would have two terms in the susceptibility; one characteristic of the Cu(Fe) system and one characteristic of the Au(Fe) system (that is, a given Fe atom would behave as if it were in either a Cu or a Au matrix). An attempt to fit the data to the expression $\chi_0 + C_1/(T+\Theta_1) + C_2/(T+\Theta_2)$ with $\Theta_1 = 29$ °K for Cu(Fe) and $\Theta_2 = 1$ °K for Au(Fe) was clearly unsuccessful. Letting Θ_1 and Θ_2 vary freely between 1 and 30 °K gave a best fit for Θ_1 near Θ_{CuAu}, listed in Table II, and a small correction in Θ_2 with C_2 negative, which is an unphysical result.

A more sophisticated form of the local model would ascribe different behavior to an impurity with each of the 12 possible nearest-neighbor combinations. The susceptibility would then be the sum of 12 different Curie-Weiss terms, with Curie constants proportional to the probability of each combination of nearest neighbors. A detailed analysis of these probabilities and of the data shows that Θ would have to decrease from 29 °K for Fe surrounded by 12 Cu neighbors to less than 15 °K for the (11 Cu, 1 Au) configuration. Two Curie-Weiss terms with such dissimilar Θ values can be recognized as separate terms and could not have been described by a single term. If further refinements of the "local" model are made to include a larger number of neighbors, the model tends towards the continuous model.

V. CONCLUSION

The magnetization and susceptibility were measured as a function of magnetic field (1–50 kG) and temperature (1.38–120 °K) for CuAu(Fe) alloys containing 5-, 9-, 17-, and 20-at. % Au, with Fe concentrations ranging from 0 to 600 ppm. As in the case of Cu(Fe), the behavior of the Cu-rich CuAu(Fe) alloys can be separated into linear and quadratic terms in the concentration, presumably associated with single Fe impurities and with Fe atoms acting in pairs. The spin deduced for the quadratic term is near $S=3$, indicating that this term is in fact due to pairs of ferromagnetically coupled Fe atoms. The susceptibility associated with the linear term can be fitted to a Curie-Weiss law plus a constant, $C_1/(T+\Theta_1) + \chi_0$. The value of Θ_1 decreases as the host Au content is increased, in a manner similar to the decrease of T_K observed in resistivity,[9,22] magnetoresistance,[11] and specific-heat[10] measurements. Further, appropriate values of two parameters C' and T' can be found for each alloy, such that plots of all $\chi_s T/C'$ as a function of T/T' superimpose. Thus all the data lie on a single universal curve. The value of C' changes only slightly for different host compositions; according to theory $C' \propto n_s g \mu_B S(S+1)/3k_B$, and the small variation in C' may therefore be due to small changes in S, or to uncertainties in the concentrations n_s. The parameter T' is proportional to the Kondo temperature T_K, and the values of T' found for the various host CuAu compositions are in the same ratio as the T_K's found from the resistivity measurements of Loram *et al.*[9] and Star *et al.*[22] The present magnetization and susceptibility results thus provide further support for the use of a one-parameter model to describe the CuAu(Fe) system

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