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# Magnetization of Dilute *Au*Fe Alloys*

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Magnetization measurements are presented of *Au*Fe alloys containing 0.002, 0.005, and 0.01 at.% Fe over the temperature range 1.5–120 °K and in magnetic fields up to 60 kG. From low-field susceptibility measurements we find a spin $S$ of $1.29 \pm 0.03$ and a Kondo temperature $T_K$ of $(0.10 \pm 0.01)$ °K for the *Au*Fe system. It is shown that recent perturbation calculations of the magnetization for $T \gg T_K$ can be expressed as a Brillouin function with spin $S$ and a modified $g$ factor dependent on temperature, applied field, and $T_K$. Magnetization curves computed from the values of $T_K$ and $S$ determined from low-field susceptibility measurements are found to be in good agreement with the measured magnetization over a wide range of fields and temperatures.

## I. INTRODUCTION

When a transition-metal impurity is dissolved in a metallic host, mixing of the conduction electron and localized $d$-wave functions occurs.[1] Coulomb repulsion between $d$ electrons of opposite spin on the impurity site causes the state to magnetize, and a Curie law susceptibility results,[2] similar to that observed for transition-metal impurities in a nonmetallic host. In a metallic alloy, the mixing leads to a spin on the impurity which fluctuates rapidly with time and has a mean value rather less than that predicted by Hund's rule. If the mixing is sufficiently strong, the impurity moment may average to zero over a time of the order of $\hbar/kT$, where $T$ is the temperature, and the state appears to be nonmagnetic. In the case of weak mixing, it has been shown[2,3] that the system can be described by a well-defined local spin $\vec{S}$ coupled to the conduction electrons of spin density $\vec{\sigma}$ by the $s$-$d$ interaction $\mathcal{H} = -J\vec{S} \cdot \vec{\sigma}$, and that the exchange constant $J$ will in general be negative.

When deducing the impurity moment from the measured susceptibility it has been customary to neglect the influence of the impurity on the conduction-electron polarization, and vice versa. In the *Au*Fe system the inclusion of these effects to first order in $J\rho$, where $\rho$ is the density of conduction-electron states per atom, of each spin direction, at the Fermi energy, leads to a correction of less than 4% to the calculated impurity spin. However, Kondo[4] has demonstrated that the coupling between the local spin and the conduction electrons at the Fermi surface increases logarithmically with decreasing temperature for negative $J$, when the calculation is taken to higher order in $J\rho$, diverging when the temperature falls below the Kondo temperature, $T_K \sim E_F e^{-1/J\rho}$. Thus, even above $T_K$ we may expect that the corrections to the total spin will be substantially larger than those calculated from first-order theory. At temperatures well below $T_K$ where perturbation theory is no longer valid, it is generally considered that the state ceases to be magnetic, though whether the resulting spin-compensated state should be considered to be a well-defined local spin totally compensated by an extended cloud of antiparallel polarized conduction electrons,[5] or a fluctuating local moment whose time average over a time of the order of $\hbar/kT$ tends to zero,[6] is not yet clear.

Recent measurements[7] on very dilute *Au*Fe alloys at temperatures above 0.5 °K, have shown that the resistivity is well described by a temperature dependence given by Hamann,[8] with $T_K = (0.24 \pm 0.15)$ °K and $S = 0.77 \pm 0.25$. Thus in the temperature range of the present measurements, 1.5–120 °K, perturbation theory expressions[2,8–12] for the susceptibility should be valid. In Sec. II, the first-order corrections to the susceptibility will be discussed in detail, and the predictions of higher-order perturbation theory will be reviewed. Following a discussion of experimental techniques (Sec. III), the magnetization results will be compared with theoretical predictions (Sec. IV).

## II. DISCUSSION

When a magnetic impurity is dissolved in a metal, the effect of the $s$-$d$ interaction is to polarize the conduction electrons around the impurity. Yosida and Okiji[9] have shown that to first order in $J\rho$ the integrated conduction-electron spin polarization around an impurity in the state $S_z$ is equal to $(J\rho/2)S_z$. When a magnetic field is applied to the system the conduction electrons gain a uniform magnetization $2\mu_B^2 \rho H$ due to their Pauli susceptibility, and this exerts an exchange field on each impurity of $H_{\mathrm{ex}} = (J\rho/2)H$ (obtained by equating the $s$-$d$ exchange energy to a magnetic energy $g\mu_B S_z \times H_{ex}$ and assuming that the $g$ values for the electrons and local spin are the same). The total magnetization per solvent atom is then

$$M = 2\mu_B^2 \rho H + cg\mu_B \frac{J\rho}{2} B_s\left(\frac{g\mu_B}{kT}(H + H_{\mathrm{ex}})\right) + cg\mu_B B_s\left(\frac{g\mu_B}{kT}(H + H_{\mathrm{ex}})\right),$$

where the terms represent, respectively, the Pauli magnetization of the host, the magnetization of the polarized conduction-electron cloud, and the impurity magnetization. $B_s(g\mu_B H/kT)$ is the Brillouin function for spin $\vec{S}$, and $c$ is the concentration of magnetic impurities. $M$ can also be expressed as

$$M = 2\mu_B^2 \rho H + cg_{\mathrm{eff}}\mu_B B_s(g_{\mathrm{eff}}\, \mu_B H/kT), \tag{1}$$

where $g_{\mathrm{eff}} = g(1 + J\rho/2)$. Thus the impurity and its cloud of polarized conduction electrons can be considered as a single entity with the degeneracy $2S+1$ of the local spin and a modified $g$. The magnetization introduced by each impurity varies between $(1+J\rho)g\mu_B B_s(gx)$ for $x \ll 1$, and $(1+J\rho/2)g\mu_B B_s(gx)$ for $x \gg 1$, where $x = \mu_B H/kT$, i.e., the exchange field can only significantly affect the local spin occupation for $x \ll 1$.

Yosida and Okiji[9] extended their perturbation calculation to higher order in $J\rho$ and found that an impurity in the well-defined state $S_z$ in the absence of the $s$-$d$ interaction has an expectation value of $S_z$ in its presence given by

$$\langle S_z \rangle = S_z \left(1 - \frac{(J^2\rho^2/2)\ln(E_F/kT)}{1 + J\rho \ln(E_F/kT)}\right) \tag{2}$$

for $T \gg T_K$. This results because $S_z$ is no longer a good quantum number in the presence of the $s$-$d$ interaction, and mixing with other $S_z$ states reduces its average value. They also find that the expectation value of the conduction-electron spin is reduced by the same factor; thus

$$\langle \sigma_z \rangle = (J\rho/2)\langle S_z \rangle\ . \tag{3}$$

This perturbation result suggests that the reduction in spin as the temperature is lowered towards the Kondo temperature is not a result of a compensating conduction-electron spin cloud ($\langle \sigma_z \rangle$ decreases as well as $\langle S_z \rangle$) but is rather a reduction of the expectation value of each local spin state. The lifetime in any $S_z$ state is reduced by spin-flip scattering with the conduction electrons, and never exceeds a value of the order of $\hbar/kT_K$ (corresponding to a susceptibility of $T=0$ of the order of $\mu_B^2/3kT_K$).

Giovannini *et al.*[10] have calculated the magnetization for $T \gg T_K$ in both high and low fields, for a local spin $S = \frac{1}{2}$, and find that this is given by

$$M = 2\mu_B^2 \rho H + cg\mu_B \left(1 + \frac{J\rho}{1 + J\rho \ln(E_F/kT)}\right) B_{1/2}(gx) \tag{4}$$

for $x \ll 1$ and

$$M = 2\mu_B^2 \rho H + cg\mu_B \left(1 + \frac{J\rho/2}{1 + J\rho \ln(E_F/g\mu_B H)}\right) B_{1/2}(gx)$$

for $x \gg 1$. This result also follows from Eqs. (2) and (3) if the exchange field acting on the local spin due to conduction electrons at the Fermi surface (the only electrons polarized by the external field) is given by

$$H_{\mathrm{ex}} = \frac{(J\rho/2)H}{1 + J\rho \ln(E_F/kT_{\mathrm{eff}})}\ , \tag{5}$$

where $kT_{\mathrm{eff}} = kT$ for $x \ll 1$ and $g\mu_B H$ for $x \gg 1$.

Using Eqs. (2), (3), and (5), Eq. (4) may be generalized for a spin $\vec{S}$ and for all $x$ to

$$M = 2\mu_B^2 \rho H + cg_{\mathrm{eff}}\, \mu_B B_s(g_{\mathrm{eff}}\, x), \tag{6}$$

where

$$g_{\mathrm{eff}} = g\left(1 + \frac{J\rho/2}{1 + J\rho \ln(E_F/kT_{\mathrm{eff}})}\right). \tag{7}$$

It may be concluded that the $s$-$d$ interaction modifies the energy splitting of the spin states of the local moment plus polarization cloud by a factor which is temperature and field dependent, but that the system retains the degeneracy of the local spin[13] (for $T \gg T_K$).

For the purposes of analyzing the low-field susceptibility data, we require the limit of Eq. (6) for $x \ll 1$,

$$\chi = 2\mu_B^2 \rho + c\, \frac{g^2\mu_B^2 S(S+1)}{3kT} \left(1 + \frac{J\rho}{1 + J\rho \ln(E_F/kT)}\right).$$

For negative $J$, and $T \gg T_K$, this may be written

$$\chi = 2\mu_B^2 \rho + c\, \frac{g^2\mu_B^2 S(S+1)}{3kT} \left(1 - \frac{1}{\ln(T/T_K)}\right) \tag{8}$$

in agreement with the result obtained by Nagaoka.[11] When comparing Eq. (6) with the measured magnetization, we require an expression for $kT_{\mathrm{eff}}$ which is valid for all $x$. Following the suggestion by Suhl,[14] we will assume that this may be approximated by

$$kT_{\mathrm{eff}} = [\,(kT)^2 + (g\mu_B H)^2]^{1/2}. \tag{9}$$

## III. EXPERIMENTAL DETAILS

The magnetization of each specimen was measured by integrating the flux change in a pick-up coil as the specimen was removed from the center of the coil, and is a modification of a technique first developed by Shoenberg.[15] The main difference in the present apparatus is the use of a 60-kG superconducting solenoid operating in the persistent mode. The high stability of the magnetic field permits the use of a single pick-up coil (15 000 turns of 48-SWG copper wire), and obviates the need for a carefully balanced double coil system. The shape and size of the pick-up coil were chosen so that the induced emf depends only on the magnetic moment of the specimen, and is independent of its shape, demagnetization effects being negligible in the present alloys. The sample, whose mass is typically of the order of 1 g, is held in a small aluminium bucket (0.8 cm diam × 0.5 cm in length), sheathed in sufficient gold foil to reduce the susceptibility of the structure nearly to zero. The small unbalanced susceptibility remaining was measured as a function of field and temperature and corrections have been made for this in the final analysis.

The signal from the pick-up coil, resulting from the extraction of the specimen, was amplified by a Tinsley galvanometer amplifier (which also acts as a 50-cps filter) followed by a Keithley 149 millimicrovoltmeter, integrated, and displayed on a chart recorder. Care was taken to ensure that the integrated signal was independent of the rate of specimen extraction.

A small back-off coil (100 turns of 48-SWG copper wire) was wound around the specimen holder. The current passing through the back-off coil could be adjusted to give zero deflection on the chart recorder (and hence zero flux change) on removal of the specimen. The magnetic moments of the specimen and back-off coil are then equal. From the known dimensions of the back-off coil, and the current required for a null deflection, the magnetization of the specimen could be determined with an accuracy of the order of $10^{-5}$ emu, without recourse to calibration with a sample of known susceptibility.

Magnetic field measurements were made with a Siemens SB RHY 18 Hall probe mounted in close proximity to the pick-up coil. The Hall probe was calibrated during a subsidiary calibration of the magnetic field with a pick-up coil of known dimensions. The cryostat consisted of a high conductivity inner copper can surrounding the specimen, the inner can being enclosed by, and thermally isolated from, an outer can which was immersed in liquid helium. The pick-up coil was attached to the outside of the outer can, and was located at the center of the solenoid. Helium exchange gas surrounding the specimen ensured good thermal contact with the inner copper can, to which was attached a constant volume gas thermometer. Temperatures below 4.2 °K were obtained by condensing in, and pumping on, a small volume of liquid helium in the inner can. From the vapor pressure of this liquid, the temperature of the specimen could be determined to within a few millidegrees. Temperatures above 4.2 °K were determined to within 0.5% using the gas thermometer.

The specimens used for magnetization measurements and those used for the resistivity measurements described in Refs. 7 and 16 were taken from the same ingots, and details of their preparation have been given in Ref. 7. The nominal concentrations of the alloys were 0.002, 0.005, and 0.01 at.%Fe and the Fe concentrations determined by resistance ratio measurements were 0.0029, 0.0054, and 0.0099 at.%. These compare favorably with the concentrations deduced by chemical analysis, which were 0.0028, 0.0053, and 0.01 at.% for the three alloys. In analyzing the results, we have taken, somewhat arbitrarily, the concentrations to be those determined by resistance-ratio measurements.

## IV. RESULTS

In Fig. 1 the susceptibility of a pure Au sample is plotted against $T$ between 1.5–100 °K. The absence of any detectable temperature dependence at low temperatures indicates that this sample contains less than 2 ppm of paramagnetic impurities. The measured value of the susceptibility of Au, $(2.56 \pm 0.05)\times 10^{-6}$ emu/cm$^3$, is in reasonable agreement with the value of $2.74\times 10^{-6}$ emu/cm$^3$ found by Henry and Rogers.[17] Also shown in the same figure is the susceptibility of the Au 0.0054 at.% Fe sample, measured in a field of 59 kG, and this indicates the magnitude of the effects under investigation.

In Fig. 2, the inverse susceptibility $1/\chi - \chi_{\mathrm{Au}}$ is plotted against $T$ in the temperature range 1.5–4.2 °K for the three *Au*Fe alloys. It is evident that the results fit an expression of the Curie-Weiss form

$$\chi = \chi_{\mathrm{Au}} + \frac{cN_A}{V}\,\frac{\mu_{\mathrm{eff}}^2}{3k(T-\theta)} ,$$

where $N_A$ is Avogadro's Number, $V$ is the molar volume of Au, and $\mu_{\mathrm{eff}} = g\mu_B[S(S+1)]^{1/2}$ is the effective moment per Fe atom. Values of $\mu_{\mathrm{eff}}$ and $\theta$ for the three alloys are compared with values obtained in previous investigations of the *Au*Fe system in Table I.[18–20]

In more concentrated alloys $\theta$ is concentration dependent, resulting from interactions between impurities. In the present alloys, $\theta$ is independent of concentration, and cannot therefore result from such interactions. (This is consistent with the measurements of Dreyfus *et al.*[21] who found that a concentration dependence is observed only for concentrations greater than 0.01 at.% Fe.) We can therefore conclude that the finite Curie temperature is a property of isolated Fe atoms and is associated with their interaction with the conduction electrons.

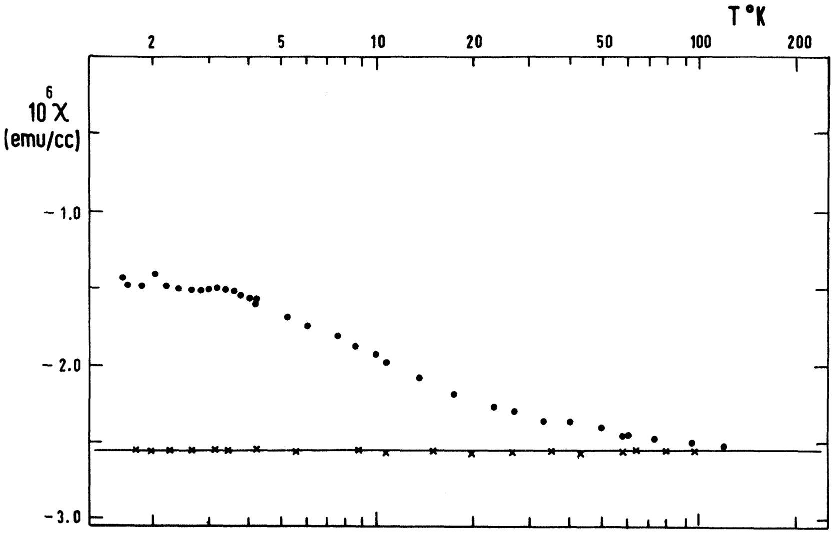

FIG. 1. Susceptibility of pure Au, and a Au 0.0054 at. % Fe alloy (in a field of 59 kG) as a function of temperature.

As described in Sec. II, perturbation theory predicts that the susceptibility has a temperature dependence of the form given by Eq. (8) for $T \gg T_K$ and $x \ll 1$. This dependence closely approximates, within the temperature range $5 < T/T_K < 100$, to an expression of the form

$$\chi = \chi_{\mathrm{Au}} + \frac{cN_A}{V}\,\frac{\mu_{\mathrm{eff}}^2}{3k\times 1.24(T+4.5T_K)}\,, \qquad (10)$$

which also describes the results in the temperature range 1.5–4.2 °K (Fig. 2). From this expression we find $\mu_{\mathrm{eff}}/\mu_B = 3.44$, 3.41, and 3.46, and $T_K = (0.10 \pm 0.01)$ °K for the 29-, 54-, and 99-ppm alloys, respectively. Assuming $g = 2$, this yields values of $S = 1.29$, 1.28, and 1.30 for the three alloys. The close agreement between the three values is probably fortuitous, considering the uncertainties in concentration.

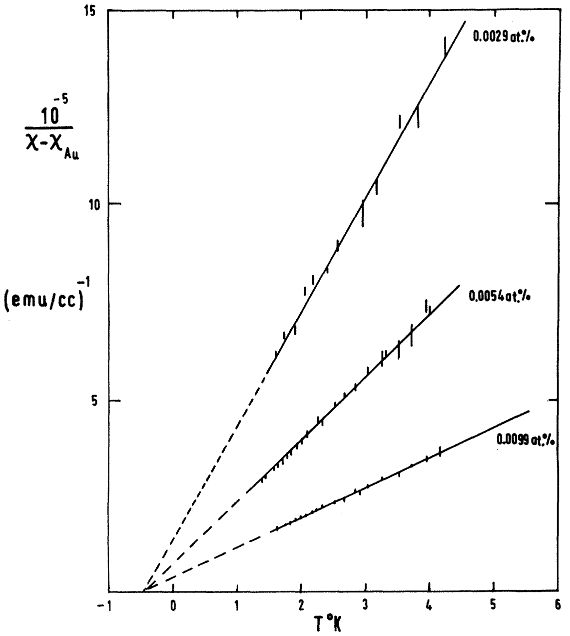

FIG. 2. Inverse susceptibility $1/(\chi - \chi_{\mathrm{Au}})$ of three *Au*Fe alloys containing 0.0029, 0.0054, and 0.0099 at.% Fe as a function of temperature.

The magnetization has also been measured as a function of field for fields up to 59 kG and at fixed temperatures of 2.02 and 4.18 °K. These results are shown in Fig. 3, where the Fe impurity magnetization $M_{\mathrm{Fe}} = M - M_{\mathrm{Au}}$ is plotted against $x$. A Brillouin function calculated with $g = 2$ and $S = 1.28$ [the value found from a fit of Eq. (10) to the low-field susceptibility] is shown by curve I. Two aspects of these results are of particular interest. Firstly the progressive reduction of the low-field magnetization (at a given value of $x$) with falling temperature, the initial slope being 33% lower than the bare Brillouin function at a temperature of 2.02 °K ($T/T_K \sim 20$). Secondly the magnetization reaches a value approximately 12% lower than the Brillouin function at the highest fields available in the present apparatus ($g\mu_B H/kT \sim 4$, $g\mu_B H/kT_K \sim 40$). Assuming the values $S = 1.28$ and $T_K = 0.1$ °K, the magnetization can be calculated using Eqs. (6) and (9) for all $x$. Curves computed in this way for $T = 2.02$–4.18 °K are compared with the results in Fig. 3, and agreement is seen to be satisfactory within the limits of experimental error.

At much higher temperatures, the impurity susceptibility is very small compared with the Pauli susceptibility of the Au, and small uncertainties in the latter can lead to large errors in the estimated Fe susceptibility, making a comparison with theory difficult. For this reason, to obtain an accurate

TABLE I. Effective moments and Curie temperatures of *Au*Fe alloys as determined by various authors.

| Author | Temperature range (°K) | Concentration (at. %) | $\mu_{eff}/\mu_B$ | $\theta$ (°K) |
|---|---|---|---|---|
| Kaufman *et al.* (Ref. 18) | 14–300 | 0.63–6.6 | 3.4–4.9 | −23 to +25 |
| Lutes *et al.* (Ref. 19) | 0.5–30 | 0.5 and 1.0 | 3.6 and 3.3 | −3 and +1 |
| Hurd (Ref. 20) | 6–300 | 0.0006–0.022 | 3.5–3.8 | −10 (mean) |
| Present results | 1.5–4.2 | 0.0029 | 3.10 ± 0.15 | −0.5 ± 0.15 |
| | | 0.0054 | 3.07 ± 0.08 | −0.4 ± 0.1 |
| | | 0.0099 | 3.12 ± 0.05 | −0.50 ± 0.05 |

temperature dependence at high temperatures, the field was held constant at 59 kG, and the temperature varied between 1.5–120 °K. In such a measurement, the Au magnetization, although large, remains constant, and can be estimated by extrapolating the measured magnetization to $1/T = 0$. Results obtained in this way are shown in Fig. 1, and the value of the Au susceptibility so determined is $(2.58 \pm 0.05) \times 10^{-6}$ emu/cm$^3$, in excellent agreement with the value found for the pure Au specimen. The Fe magnetization $M_{Fe} = M - M_{Au}$ is plotted against $x$ in Fig. 4, at the fixed field of 59 kG. The Brillouin function calculated with $g = 2$ and $S = 1.28$ to shown by curve I. Also included for comparison is the magnetization at a fixed temperature of 2.02 °K (shown previously in Fig. 3). Particular values of $x$ correspond to a lower value of $T_{eff}$ in the fixed temperature curve, compared with that in a fixed field of 59 kG, hence the reduction of the moment is greater in the former case. Both sets of results are compared with the predictions of Eq. (6) assuming that $T_{eff}$ is of form given in Eq. (9). As, according to Eq. (4), the curves only differ through the difference in $T_{eff}$ at a given $x$, the precise form of $T_{eff}$ can in principle be determined from these results. The measurements in the present investigation are not, however, sufficiently precise to permit such a detailed analysis.

A similar fixed field ($H = 59$ kG) magnetization curve is shown in Fig. 5 for the 99-ppm alloy. A Brillouin function with $g = 2$ and $S = 1.30$ (as estimated from the low-field susceptibility data for this alloy) is shown by curve I. The magnetization as predicted by Eq. (6) is also shown, and is in good agreement with the experimental points.

## V. CONCLUSIONS

We have shown that the temperature dependence

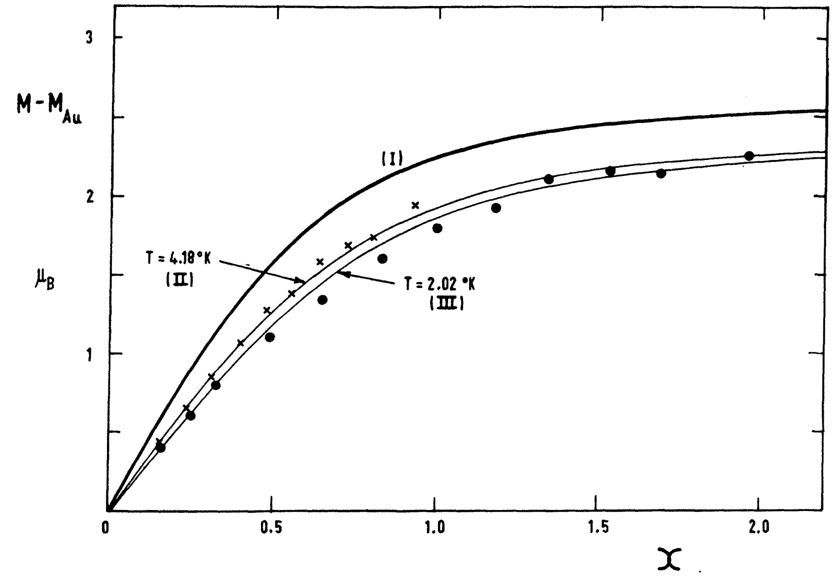

FIG. 3. Impurity magnetization $M$-$M_{Au}$ versus $x$ (where $x = \mu_B H/kT$) for the Au 0.0054 at. % Fe alloy. Crosses correspond to measurements at the fixed temperature 4.18 °K, and dots at the fixed temperature 2.02 °K. The continuous curves represent Brillouin functions with $S = 1.28$. In curve I, $g_{eff} = 2$. In curves II and III, $g_{eff}$ is given by Eq. (7) with $T = 4.18$ and 2.02 °K, respectively.

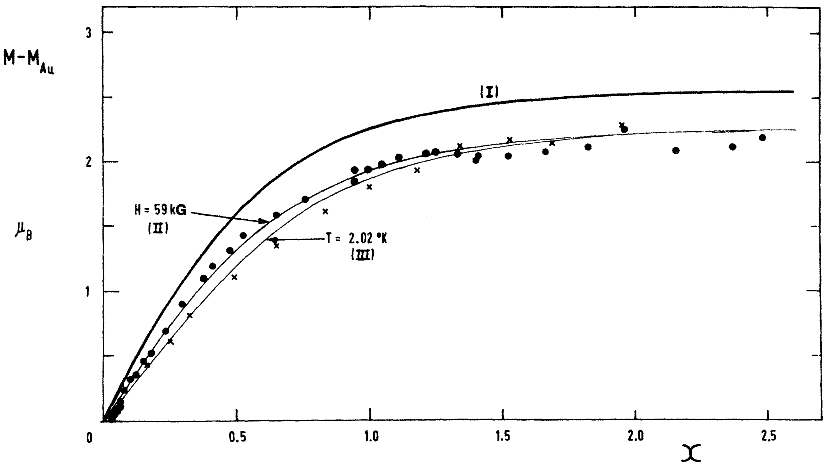

FIG. 4. Impurity magnetization $M\text{-}M_{Au}$ versus $x$ for the Au 0.0054 at.% Fe alloy. Dots correspond to measurements in a fixed field of 59 kG, and crosses measurements at a fixed temperature of 2.02 °K. The continuous curves represent Brillouin functions with $S = 1.28$. In curve I, $g_{eff} = 2$. In curves II and III $g_{eff}$ is given by Eq. (7) with $H = 59$ kG, and $T = 2.02$ °K, respectively.

of the low-field susceptibility of *Au*Fe alloys containing less than 0.01 at.% Fe is independent of concentration at temperatures above 1.5 °K. The results can be analyzed, using Eq. (10), to give a Kondo temperature $T_K = (0.10 \pm 0.01)$ °K for the *Au*Fe system. This value is in good agreement with the value of $T_K = (0.24 \pm 0.15)$ °K obtained from an analysis of the resistivity in terms of an expression given by Hamann.[8] [If the expression for the susceptibility given by Hamann is used, Eq. (4.15) of Ref. 8 instead of Eq. (10), we obtain $T_K = (0.20 \pm 0.02)$ °K, which is in even better agreement with the value obtained from the resistivity.] An estimate of the Kondo temperature can also be obtained from the thermopower, which is expected to pass through a broad maximum in the neighborhood of $T_K$. Measurements by Macdonald *et al.*[22] on very dilute *Au*Fe alloys reveal the existence of a broad maximum in the thermopower at between 1–2 °K.

Potton[23] has measured the specific heat of a Au 0.01 at.% Fe alloy down to 0.1 °K, and finds that this passes through a maximum at around 0.15 °K. A maximum is expected in the specific heat of a spin compensated state at around $T_K/3$,[5,24] and this result would be consistent with $T_K \sim 0.5$ °K.

The entropy associated with the magnetic ordering of the impurities can be obtained from specific-heat measurements on more concentrated alloys, and from this a spin value may be derived. From such measurements du Chatenier[25] finds $S = 0.65$ for a Au 0.092 at.% Fe alloy, and Potton[23] finds $S = 0.9 \pm 0.1$ for a Au 0.1 at.% Fe alloy. These values for the spin are substantially lower than the value of $S = 1.29 \pm 0.03$ determined from the present susceptibility measurements, and suggests that a substantial part of the entropy is removed from the

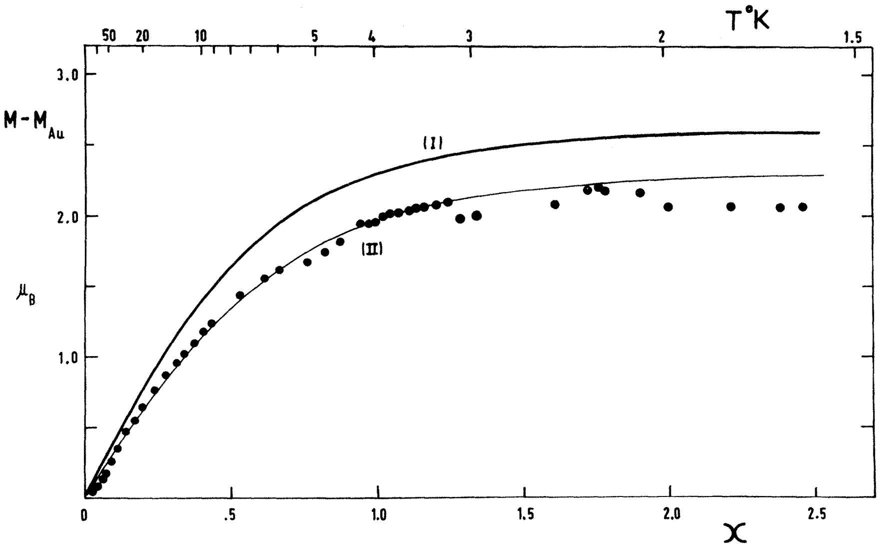

FIG. 5. Impurity magnetization $M\text{-}M_{Au}$ versus $x$ for the Au 0.0099 at. % Fe alloy, in a fixed field of 59 kG. The continuous curves represent Brillouin functions with $S = 1.30$. In curve I, $g_{eff} = 2$. In curve II, $g_{eff}$ is given by Eq. (7) with $H = 59$ kG.

spin system even at temperatures far higher than $T_K$. Further evidence for this is the rather low spin value of 0.5 determined from entropy measurements below 30 °K in the *Cu*Fe system,[26] for which $T_K$ is of the order of 20 °K, compared with the bare spin value of 1.4 determined from susceptibility measurements.[20]

It has also been demonstrated in the present investigation that the magnetization of the *Au*Fe alloys over a considerable range of fields and temperatures is well described by a Brillouin function with a spin $S$ and with a temperature- and field-dependent $g$ value given by

$$g_{\text{eff}} = 2\left(1 - \frac{0.5}{\ln T_{\text{eff}}/T_K}\right),$$

using the values of $S$ and $T_K$ derived from the low-field susceptibility measurements. It should be noted, however, that although satisfactory agreement with experiment has been obtained using a Brillouin function with a non-half-integral spin, this agreement may be fortuitous. It has not as yet been demonstrated theoretically that the magnetization of a system of fluctuating spins, of mean value $S$, is correctly described by a simple Brillouin function with nonintegral degeneracy $2S+1$.

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