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Magnetic Properties of Dilute Alloys of Cr, Mn, and Ni in 4*d* and 5*d* Transition Metals*

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Magnetic-susceptibility measurements between 4 and 300°K of dilute magnetic alloys of Cr, Mn, and Ni in various 4*d* and 5*d* transition metals are reported. Mn in Mo and Rh shows a localized magnetic moment with Kondo-type behavior. Below the Kondo temperature T_K, the susceptibility increases, by a term proportional to $\ln(T_K/T)$, faster than the Curie-Weiss law. This logarithmic term seems to be common to all dilute magnetic alloys of 3*d* solutes in transition metals. In addition to a localized moment, Mn causes a large change in the temperature-independent susceptibility of the order of 10^{-3} emu per mole of solute. Similar large changes are also observed for other dilute magnetic alloys. It seems to be quite general that the occurrence of localized moment in dilute alloys is accompanied by a large increase in the temperature-independent susceptibility. These increases are too large to be understood within the present theories of localized magnetic states. Large increases in the temperature-independent susceptibility are also observed for some alloys with no localized moments. For Mn in Re, this increase corresponds to about 10^{-2} emu per mole of solute.

INTRODUCTION

In recent years much effort has been spent, experimentally and theoretically, to understand the magnetic properties of dilute alloys with 3*d* transition metals as solutes. One important problem is whether the 3*d* solute forms a localized magnetic moment. Dilute alloys of 3*d* metals with nontransition metals are qualitatively understood and can be well described in the Friedel-Anderson picture.[1] More difficult to understand are alloys of 3*d* solutes in 4*d* or 5*d* transition metals. Using results of Anderson[2] and Wolff,[3] Clogston *et al.*[4] correlated their experimental results on the occurrence of a localized moment in dilute alloys of Fe in 4*d* transition metals and their alloys, with the band structure in these alloys and with the perturbation (electronic charge difference) introduced by the Fe atom. Important parameters in the above model are the electronic charge difference between solute and solvent and the intraatomic Coulomb interaction of the 3*d* solute. By using, instead of Fe, different 3*d* metals as solutes, these parameters can be easily changed and their influence on the occurrence of localized moments can be investigated. So far only Fe has been systematically studied as a solute.[4–8] Less is known about Co,[8–11] and only a little about Ni, Cr,[8] and Mn.[8] We therefore studied the magnetic susceptibility of dilute alloys of Cr, Mn, and Ni in 4*d* and 5*d* transition metals.

EXPERIMENTAL DETAILS

A. Alloy Preparation

The following alloys were investigated: Cr and Mn in Nb, Mo, Re, and Rh, Ni in Mo and Rh. With one exception (*Mo*-Mn) the alloys were prepared by arc melting under argon atmosphere in a water-cooled copper crucible. The main impurities of the starting materials quoted by the supplier were (in parts per million): Nb (5 Fe, 20 Si, 1 Mg, <0.01-at.% Ta), Mo (10 Fe, 8 Cr, 5 Si, 4 Ni), Re (3 Fe, 3 Si, 2 Al, 1 Cu), Ru (99.99% pure), Rh (20 Fe, 38 Ir, 16 Ag, 12 Ca, 20 Mg) Cr (99.997% pure), Mn (5 Fe, 40 Mg, 4 Si), Ni (<50 Fe, 0.01%C). Before alloying, the 4*d* and 5*d* transition metals were once arc melted into buttons weighing 2–5 g and their magnetic susceptibility was measured as a function of temperature. The results indicate that, at least for the solvents where Fe shows a localized moment (Mo, Rh), the Fe impurity concentrations are of the order quoted by the supplier. These buttons were used in preparing the alloys. Each alloy was melted at least four times. The alloys were then homogenized in a vacuum furnace (10^{-6} Torr) under the following conditions: the Nb, Mo, Re, and Ru alloys for 7 days at 1400°C, the Rh alloys for 4 days at 1240°C. The alloys were subsequently quenched to room temperature. The chemical composition of the alloys as given in Tables I and II is based on chemical analysis and/or on the record of weight loss during melting, assuming that the weight loss is solely caused by evaporation of the 3*d* solute.

Since no Mn could be dissolved in Mo by the usual alloying techniques, the *Mo*-Mn alloy was prepared by a diffusion method: A $\frac{1}{4}$-in.-diam Mo rod was immersed in molten Mn for 1 week. The surface of the rod was then ground to a depth of 1.1 mm. This

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TABLE I. Susceptibility results for alloys with no localized moments: $\Delta x/x$ is the relative accuracy of the solute concentration, (a) estimated from melting record assuming all weight loss is due to solute, (b) from chemical analysis. The solvent susceptibility χ_s is given at 80°K. $d\chi/dx$ gives the change and $(1/\chi_s)d\chi/dx$ gives the relative change of the susceptibility caused by the solute.

Solvent	x, atomic fraction of solute	$\Delta x/x$ (%)	$10^4\chi_s$(80°K) (emu mol^{-1})	$10^4 d\chi/dx$ (emu mol^{-1})	$(1/\chi_s)d\chi/dx$
Nb	0.0045 Cr	10^a	2.23	−0.9	−0.4
	0.0014 Mn	10^b		−39	−17.6
Mo	0.0194 Cr	5^b	0.833	+0.72	0.87
	0.0070 Ni	4^b		−0.71	−0.86
Re	0.0017 Cr	5^b	0.639	21	33
	0.0004 Mn	15^b		110	172
Ru	0.0165 Cr	10^a	0.398	2.0	5.0
	0.0035 Mn	10^a		2.7	6.8
Rh	0.0133 Cr	3^a	0.969	−0.5	−0.5
	0.0097 Ni	3^a		5.0	5.1
	0.0206 Co	3^a		27	28

specimen was annealed for 18 days at 1400°C in a vacuum of 10^{-6} Torr and quenched to room temperature. Electron microprobe analysis showed that the Mn had diffused for a depth of 0.5 mm. This corresponds to about 20 volume percent of the specimen. The Mn content was determined by chemical analysis, using the total specimen. From these two analyses the effective Mn concentration was 4 at.% or higher.

TABLE II. Parameters of the Curie-Weiss law [Eq. (1)] for various alloys: $\Delta x/x$ is the relative accuracy of the solute atomic fraction (a) estimated from melting record assuming all weight loss is due to solute, (b) from chemical analysis. χ_0/x is the increase in the temperature-independent susceptibility per mole of solute. p is the effective moment per solute atom calculated from the Curie constant C of Eq. (1) ($C=Np^2/3k$). T_K is the Kondo temperature from Eq. (1).

Solvent	x, atomic fraction of solute	$\Delta x/x$ (%)	$10^4\chi_0/x$ (emu mol^{-1})	p (μ_B)	T_K (°K)
Mo	0.0166 Mn	5^a	18	2.0	15
Rh	0.0022 Mn	10^b	18	2.27	14.5
	0.0045 Mn	5^b	17	2.23	14.6
	0.0084 Mn	3^b	15	2.10	14.2
$Mo_{0.7}Nb_{0.3}$	0.0071 Fe	5^b	14	1.25	4.2
	0.01^c Fe			1.3^c	4^c

B. Apparatus

The magnetic susceptibility was measured by a standard Faraday method using a recording microbalance with an accuracy of ±0.01 mg, and a magnetic field with constant gradient, $(dH/dz)/H=0.007\ \text{cm}^{-1}$. The magnetic field was varied between 0 and 12.6 kOe. The temperature was varied between room temperature and 4.2°K by means of a continuous-flow cryostat: Liquid He is drawn from a storage Dewar into a heat exchanger surrounding the specimen. The specimen, suspended by a nylon thread from the balance, is in

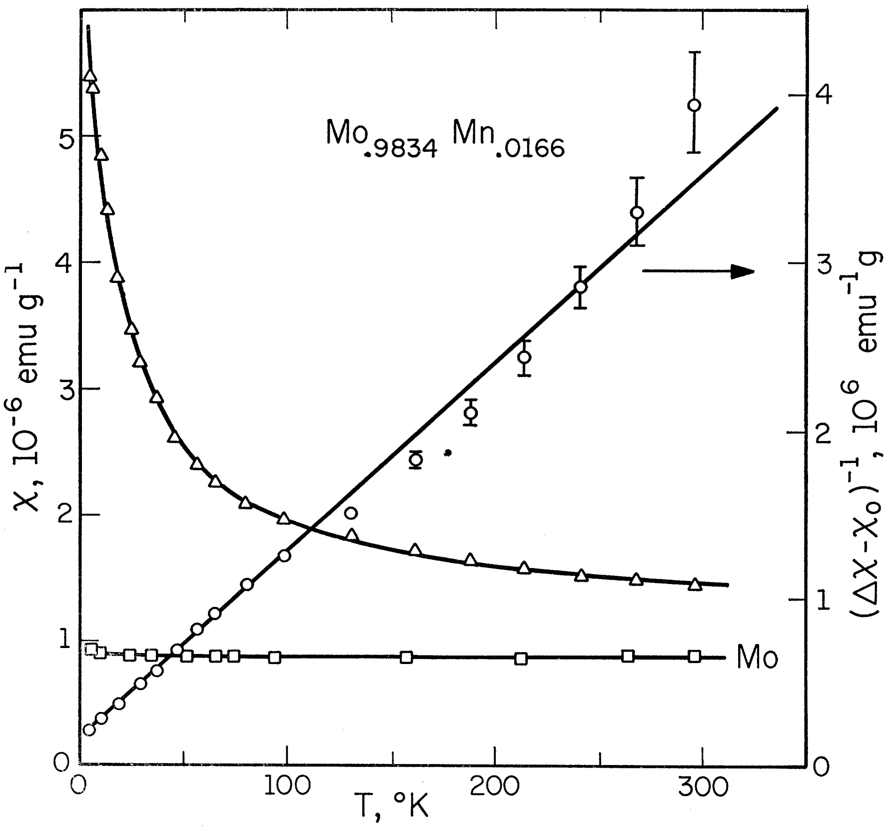

FIG. 1. Magnetic-susceptibility results for the alloy $Mo_{0.9834}\,Mn_{0.166}$: triangle represents susceptibility of the alloy, χ; square represents susceptibility of the unalloyed solvent, χ_s; circle represents reciprocal of susceptibility difference, $(\chi-\chi_s-\chi_0)^{-1}=[C/(T+T_K)]^{-1}$ versus temperature, where χ_0 is determined by Eq. (1).

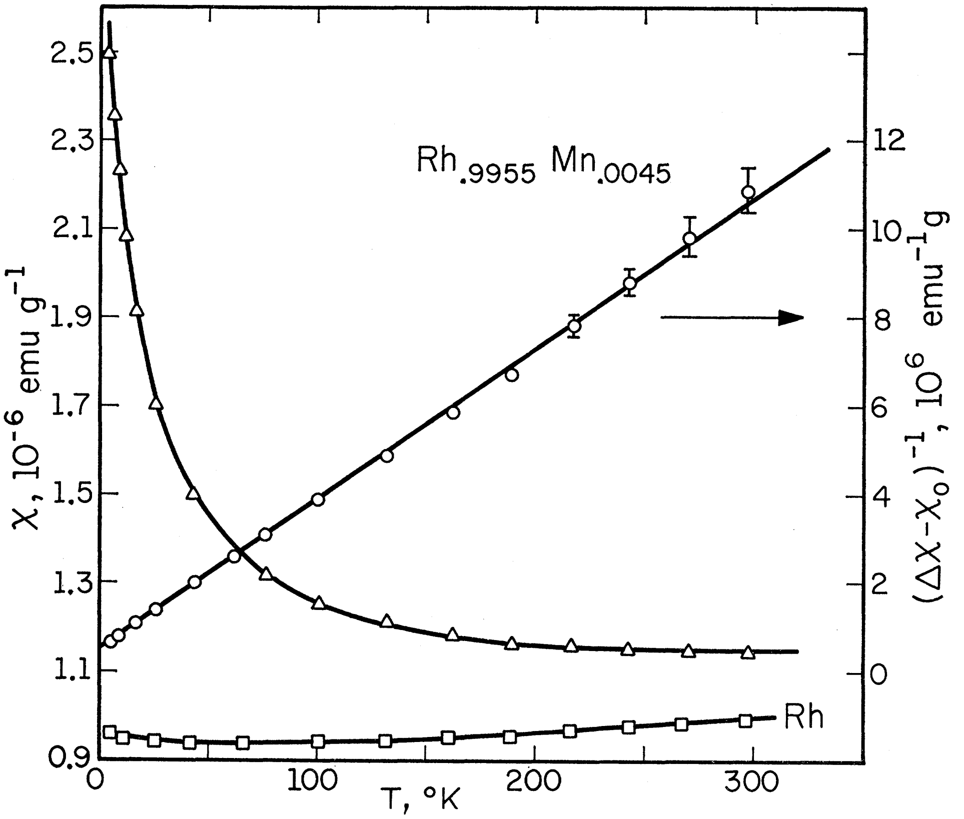

FIG. 2. Magnetic-susceptibility results for the alloy $Rh_{0.9955}$ $Mn_{0.0045}$: triangle represents susceptibility of alloy, χ; square represents susceptibility of the unalloyed solvent, χ_s; circle represents reciprocal of susceptibility difference $(\chi-\chi_s-\chi_0)^{-1}=[C/(T+T_K)]^{-1}$ versus temperature, where χ_0 is determined by Eq. (1).

thermal contact with the heat exchanger by means of He exchange gas. The temperature can be controlled to an accuracy of better than 0.01°K by varying the flow rate of the liquid He. The temperature is measured by a GaAs diode, attached to the heat exchanger. The diode has been calibrated by comparison with a calibrated Pt thermometer and by measuring the susceptibility of $CuSO_4 \times 5H_2O$. The accuracy of the temperature measurement is estimated to be about ±0.1°K at the lowest temperatures and it decreases to about ±0.5°K at higher temperatures.

The susceptibility for all alloys was measured in a field of 12.6 kOe on cooling down from room temperature to 4.2°K, usually at about 20 different temperatures. At 4.2°K the susceptibility was then measured as a function of the applied magnetic field. For all investigated alloys the susceptibility was independent of the field.

RESULTS AND DISCUSSION

A. Alloys without Localized Moments

For all solvent materials we found a small temperature dependence of the susceptibility caused either by the Fe impurities or by changes in the Pauli susceptibility. The largest change occurs in Re where the susceptibility changes as much as 10% between 300 and 4°K: from 0.37×10^{-6} emu g^{-1} at 300°K to 0.342 $\times10^{-6}$ at 40°K to 0.346×10^{-6} at 4°K. The susceptibility of Nb increases from 2.338×10^{-6} emu g^{-1} at 300°K to 2.417×10^{-6} emu g^{-1} at 4.2°K; for Ru the change is from 0.412×10^{-6} at room temperature to 0.392×10^{-6} at 50°K to 0.397×10^{-6} emu g^{-1} at 4°K. The temperature dependence of the susceptibility for Mo and Rh is shown in Figs. 1 and 2, respectively. For most of the alloys of Table I, the temperature dependence of the susceptibility of the solvents remains unchanged on alloying; that is, within the accuracy of the measurements the susceptibility-versus-temperature graphs become parallel displaced towards lower or higher susceptibilities. Since for most alloys and elements of Table I, $d\chi/dT$ is zero near $T=80$°K, Table I gives susceptibility data at this temperature, but the change of the susceptibility with concentration $d\chi/dx$ is essentially independent of temperature.

There is a surprisingly large change of the Nb susceptibility on alloying Mn to Nb. This effect is more than ten times larger than that predicted by a rigid-band theory, using susceptibility data for Nb-Mo

TABLE III. Change $d\gamma/dx$ and relative change $(1/\gamma)d\gamma/dx$ of the low-temperature electronic specific-heat coefficient γ, and change $d\chi/dx$ and relative change $(1/\chi)d\chi/dx$ of the Pauli susceptibility with concentration x for various isoelectronic alloy systems.

	Mo-Cr	*Re*-Mn	*Ru*-Fe	*Rh*-Co	*Pd*-Ni
$d\gamma/dx$, 10^{-4} cal mol^{-1} deg^{-2}	2.54[a]		27[b]		415[c]
$(1/\gamma)d\gamma/dx$	0.58		3.8		15[c]
$d\chi/dx$, 10^{-4} emu mol^{-1}	0.72 1.0[d]	110	6.0[d]	28 20[e]	90[c]
$(1/\chi)d\chi/dx$	0.87	172	15	29	87[c]
$\xi=\dfrac{\chi d\gamma/dx}{\gamma d\chi/dx}$	0.67		0.25		0.2[c]

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[d] Reference 17.

[e] References 10 and 11.

alloys.[12] Since, so far, we did not succeed in increasing the Mn content, the concentration dependence of the susceptibility decrease could not be studied. At present we do not have an explanation for this effect.

There is an appreciable increase in the susceptibility of Re with small additions of Mn (Table I). In this case, $\Delta\chi$ is temperature-dependent, decreasing approximately linearly with temperature, by 30% from 4 to 300°K. Large changes in the Pauli susceptibility on alloying have also been observed for other isoelectronic alloys. Walstedt *et al.*[10,11] report a susceptibility change on adding Co to Rh corresponding to $d\chi/dx\approx 2000\times 10^{-6}$ emu mol^{-1}. We remeasured one *Rh*-Co alloy to find the temperature dependence of $d\chi/dx$. Our $d\chi/dx$ value is somewhat higher than that of Ref. 10 (Table III) and the temperature dependence is small: $d\chi/dx$ decreases essentially linearly with increasing temperature by 20% between 4 and 300°K.

Since there is no electronic charge difference between solvent and solute in isoelectronic alloys, no localized electronic state in the Friedel sense can arise at the solute sites. The essential electronic difference between the 4*d* or 5*d* solvents and the isoelectronic 3*d* solute is the spatial extension of the *d*-wave functions. This can be expressed as a difference in the intraatomic Coulomb interaction[13–15] which is significantly larger for the 3*d* solutes than their 4*d* or 5*d* hosts. This intraatomic Coulomb interaction leads to a strong locally enhanced susceptibility[10–16] (local paramagnons). Table III compares the present results with results with other isoelectronic alloys. Since the intraatomic Coulomb interaction increases from Cr to Ni, one expects the locally enhanced susceptibility to do the same. In fact, $d\chi/dx$ increases in Table III towards the right, but for Mn in Re $d\chi/dx$ is much too high to fit into this sequence. Re is the only 5*d* solvent in Table III; possibly the increase in the 5*d* bandwidth over that of the 4*d* band has a large effect on the local enhancement at the Mn site. On the other hand, Caroli *et al.*[15] pointed out that Hund's rule coupling at the 3*d* site can considerably enhance the amplitude of the localized spin fluctuation and that this effect can account for the large $d\chi/dx$ for dilute alloys with Mn as solute. Where sufficient data are available, Table III also gives the parameter $\xi=(\chi/\gamma)(d\gamma/dx)/(d\chi/dx)$, which is used in the literature[15] to describe the effect of the Coulomb interaction.

Despite the relatively small increase in the susceptibility with the addition of Cr to Mo (Table III), it could well be that here also intraatomic Coulomb interaction is important. This could explain the fast decrease of the superconducting transition temperature in these alloys, which Andres *et al.*[17] could not explain by an increased average Coulomb interaction.

B. Alloys with Curie-Weiss Susceptibility

Figures 1 and 2 show the susceptibility results for the alloys where Mn has a localized moment. The measured susceptibility has been analyzed in terms of the following equation:

$$\Delta\chi\equiv\chi-\chi_s=\chi_0+C/(T+T_K)\,, \qquad (1)$$

where χ is the measured susceptibility of the alloy and χ_s is that of the solvent. The constant χ_0 allows for changes in the temperature-independent susceptibility on alloying, C is the Curie constant ($C=Np^2/3k$), and T_K is a characteristic temperature (Kondo temperature). The parameters of Eq. (1) were determined by least-squares analysis and they are listed in Table II.

For the *Mo*-Mn alloy there are considerable deviations from the Curie-Weiss law. This can be seen in Fig. 1, where $(\Delta\chi-\chi_0)^{-1}=[C/(T+T_K)]^{-1}$ is plotted versus temperature. Since the alloy is not homogeneous (see above), this deviation may be caused by Mn-Mn interaction in parts of the alloy where the Mn concentration is relatively high. Despite this difficulty, it is quite clear that Mn has a localized magnetic moment in Mo, but the parameters given in Table II may differ somewhat from those which would have been obtained in the dilute limit with no solute-solute interaction.

The magnetic moment of Mn in Mo is similar in magnitude to that of[4] Fe ($p=2.9\mu_B$) and of[9] Co ($p=2.4\mu_B$) in Mo. This indicates that for the formation of a localized magnetic state, the magnitude of the elec-

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tronic charge difference between solvent and solute is not nearly as important as the electronic structure of Mo itself. This situation is very similar to the occurrence of localized moments in nontransition metals where the appearance of such moments is a function mainly of the solvent metal.[1,2] The importance of the electronic structure of Mo in the formation of a localized magnetic moment was also demonstrated by Jaccarino and Walker[18] who have shown that the moment of Fe in Nb-Mo alloys can be directly related to the number of Mo nearest neighbors.

Mn shows a localized moment also in Rh. Here the magnetic susceptibility has been investigated for different Mn concentrations. Figure 2 shows the results for one *Rh*-Mn alloy. The susceptibility is again analyzed using Eq. (1). As can be seen in Table II, the magnetic moment is essentially independent of the Mn concentration. The same is true for χ_0/x, indicating that the change in the temperature-independent susceptibility χ_0 is proportional to the concentration x. Again, the moment and the Kondo temperature T_K are similar to those of Fe in Rh ($p=3.0\mu_B$, $T_K=14$°K).[4]

In addition to the appearance of a localized moment in *Rh*-Mn and *Mo*-Mn, there is also a considerable change in the temperature-independent susceptibility (Table II). It is of the same order of magnitude as that for the isoelectronic alloys of Table III. These large changes in the temperature-independent susceptibility in addition to a localized moment formation cannot be accounted for by the usual theories of a localized magnetic state.[2] Unfortunately, we cannot compare these results with measurements on other dilute magnetic alloys, since in most previous investigations no attention was given to this effect and usually no χ_0 values were determined. To check whether this large temperature-independent susceptibility occurs also in alloys with Fe as solute, we made the alloy $(\mathrm{Nb}_{0.3}\mathrm{Mo}_{0.7})_{0.9929}\mathrm{Fe}_{0.0071}$ and measured its susceptibility. After subtracting the susceptibility of the matrix $\mathrm{Nb}_{0.3}\mathrm{Mo}_{0.7}$, the susceptibility was fitted to a Curie-Weiss equation [Eq. (1)]. The parameters are given in Table II. The magnetic moment and the Kondo temperature agree with the results of Clogston *et al.*[4] However, there is a very large temperature-independent susceptibility connected

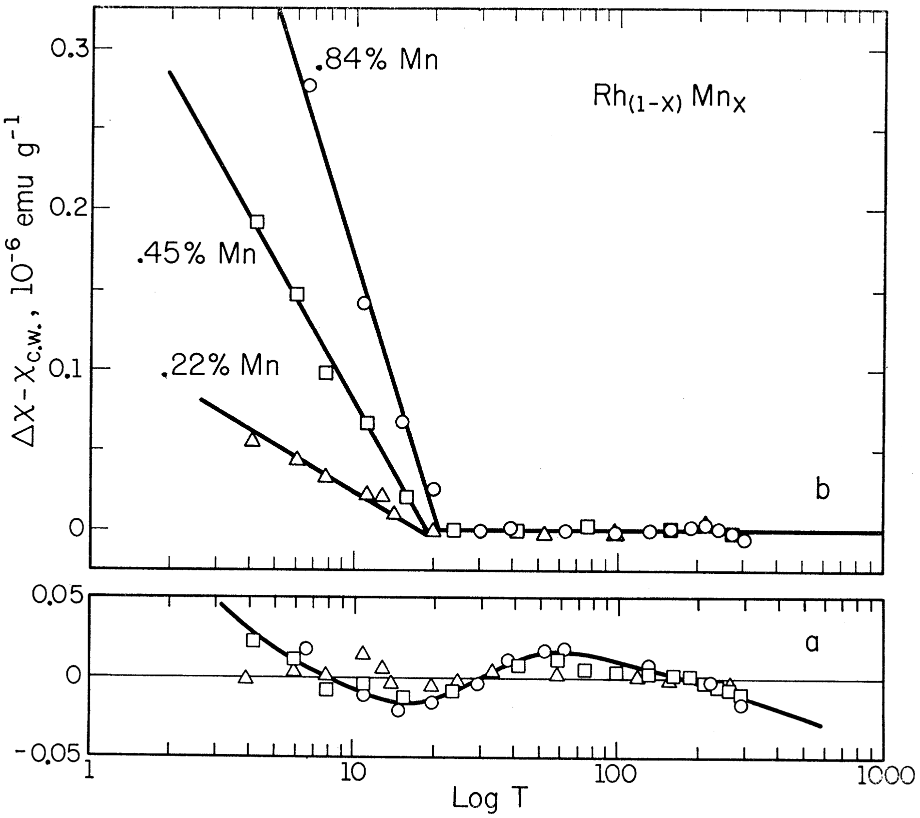

FIG. 3. Analysis of the susceptibility data for the *Rh*-Mn alloys: deviation of the experimental data from the least-squares Curie-Weiss equation 1, $\Delta\chi-\chi_{\mathrm{C.W.}}\equiv(\chi-\chi_s)-[\chi_0+C/(T+T_K)]$ is plotted versus $\ln T$. The parameters χ_0, C, and T_K are determined by least-squares analysis of the data in the temperature range $T_L\leq T\leq 300$°K, where $T_L=4$°K for (a) and $T_L=20$°K for (b).

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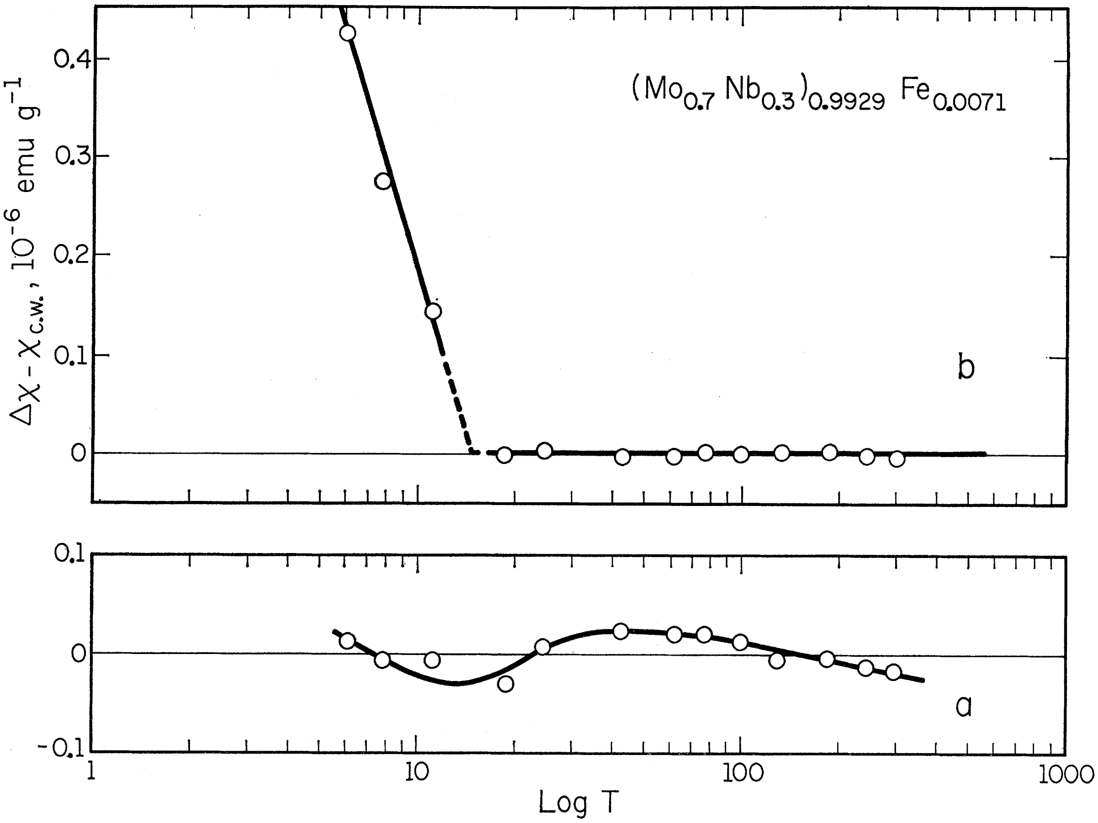

FIG. 4. Analysis of the susceptibility data for the alloy $(Mo_{0.7}Nb_{0.3})_{0.9929}Fe_{0.0071}$ (see caption of Fig. 3).

with the Fe solute. Creveling and Luo[19] found a similar change in the temperature-independent susceptibility on adding V to Au ($\chi_0/x = 15\times10^{-4}$ emu mol^{-1}). It may be, therefore, quite generally true that the occurrence of a localized magnetic moment is accompanied by a large increase in the temperature-independent susceptibility. The magnitude of the increase suggests that it is closely related to the local enhancement effect as discussed for the isoelectronic alloys of Table III, but it is not clear how this fits into the present theory of localized moments in dilute alloys. Possibly, the observed temperature dependence of the susceptibility corresponds to a situation between a localized moment and a temperature-independent susceptibility, as recently discussed by Wang, Evenson, and Schrieffer.[20]

DEVIATION FROM CURIE-WEISS LAW

In dilute magnetic alloys with Kondo-type behavior, the Curie-Weiss law, in the absence of general theoretical calculations, is a convenient interpolation formula for the magnetic susceptibility.[1,21–23] However, it has been known for some time that there are systematic deviations from the Curie-Weiss law.[1,24–26] For the alloys listed in Table II, systematic deviations from Eq. (1) are observed. For the *Mo*-Mn alloy these deviations are large and can be easily seen in Fig. 1. As already mentioned above, these deviations are thought to be caused by Mn-Mn interaction and will not be further discussed in this section. For the *Rh*-Mn alloys, where the concentration dependence of the parameters of Eq. (1) indicates that these alloys can be considered to be dilute, with no solute-solute interaction, the deviations of the reciprocal susceptibility from a straight line are not so obvious (Fig. 2), but nevertheless they are significant. The same is true for the $(Mo_{0.7}Nb_{0.3})$-Fe alloy. This can be seen in Figs. 3(a) and 4(a), where the systematic deviation of the experimental data from the least-squares fit Curie-Weiss law [Eq. (1)] is shown. The analytical representation of

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the data can be considerably improved in the following way: To determine the parameters of Eq. (1), only the high-temperature data are used, that is, data above a temperature where the deviation curves in Figs. 3(a) and 4(a) change the sign of their curvature (at about 20°K). These data ($T>20$°K) follow very well a Curie-Weiss law with the constants of Eq. (1) given in Table IV. Figures 3(b) and 4(b) show the deviation of the experimental data from this high-temperature Curie-Weiss equation. There is no systematic deviation at higher temperatures, but the low-temperature data deviate logarithmically [linear as a function of $\ln T$, Figs. 3(b) and 4(b)]. This logarithmic deviation of the susceptibility starts at the temperature T_K [Table IV, Figs. 3(b) and 4(b)], T_K being determined by Eq. (1) using only high-temperature data. The susceptibility of the alloys can be, therefore, very well represented by the following equation:

$$\begin{aligned}\Delta\chi\equiv\chi-\chi_s=\chi_0+C/(T+T_K)&\\ +\alpha\ln(T_K/T)\,,&\quad T\leq T_K\\ +\qquad\qquad 0\,,&\quad T>T_K\,.\end{aligned}\qquad(2)$$

This equation has only one additional parameter as compared with Eq. (1). The parameters of Eq. (2) are listed in Table IV. They are only slightly different from those listed in Table II, determined by fitting the data to Eq. (1). Again, for the *Rh*-Mn alloys the parameters in Table IV are essentially concentration independent.

TABLE IV. Parameters of Eq. (2): α is the coefficient of the logarithmic term in Eq. (2), for $T>T_K$, $\alpha=0$. χ_0/x is the increase in the temperature-independent susceptibility per mole of solute. p is the effective magnetic moment calculated from the Curie constant C of Eq. (2) ($C=Np^2/3k$). T_K is the Kondo temperature from Eq. (2).

Solvent	x, atomic fraction of solute	$10^4\,\alpha/x$ (emu mol^{-1})	$10^4\,\chi_0/x$ (emu mol^{-1})	p (μ_B)	T_K (°K)
$Mo_{0.7}Nb_{0.3}$	0.0071 Fe	65	9.4	1.51	14.2
Rh	0.0022 Mn	20	14	2.39	18.1
	0.0045 Mn	28	12	2.40	20.1
	0.0084 Mn	28	11	2.30	21.1

The theoretical meaning of the observed logarithmic divergence which starts below the Kondo temperature is at present not clear. Since it is observed in two electronically very different alloys, *Rh*-Mn and ($Mo_{0.7}Nb_{0.3}$)-Fe, it seems to be generally true for transition-metal solvents. A similar behavior has been recently reported for dilute alloys of Fe in $Rh_{1-x}Pd_x$.[7,24] There the situation is more complicated due to polarization effects in the exchange-enhanced solvent susceptibility,[7] but the logarithmic behavior of the deviation from the Curie-Weiss law is shown over a wider temperature range. It may well be that the observed logarithmic divergence is caused by the interaction of the *d* electrons of the solute with the *d* electrons of the solvent, as suggested by Nagasawa.[7] If this is true, no such deviation should occur in alloys like *Cu*-Fe. As discussed by Daybell and Steyert,[1] deviations from Eq. (1) do occur in these alloys,[25,26] but it is not yet known whether or not they are of the type described above.

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