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Magnetic Susceptibility and Low-Temperature Specific Heat of Dilute Transition-Metal Alloys*

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Magnetic-susceptibility measurements between 4 and 800 °K and specific-heat measurements between 1.5 and 4.2 °K are reported for a series of dilute *Rh*-Mn, *Mo*-Fe, *Mo*-Co, and *Au*-Fe alloys in the concentration range 0.02–0.7 at.%. The solute susceptibility $\Delta\chi$ of all alloys shows local-moment behavior and scales with the concentration over a wide concentration range, but large deviations from simple Curie-Weiss behavior, qualitatively the same for all alloys, are observed. This deviation consists of a rapid increase of $\Delta\chi$ at low temperatures. A pronounced field dependence of $\Delta\chi$ and large specific-heat anomalies at these temperatures strongly suggest that this rapid increase in $\Delta\chi$ arises from solute-solute interactions. This anomalous part of $\Delta\chi$ can readily be separated from the part due to isolated solute atoms. The conclusion is drawn that scaling of the solute susceptibility with concentration does *not* necessarily signify that the alloys are "dilute" in the conventional sense.

I. INTRODUCTION

In recent publications[1,2] we reported that dilute alloys of Mn in Rh exhibit a local moment but that the magnetic susceptibility does not show simple Curie-Weiss behavior. It was found that the solute susceptibility contains a very substantial temperature-independent term and at low temperatures increases much faster than predicted by the Curie-Weiss equation. Since these deviations from Curie-Weiss behavior scale with the Mn concentration, they were attributed to single-impurity effects. Large temperature-independent terms in the solute magnetic susceptibility of alloys exhibiting local moments have also been reported for dilute alloys of Mn in Mo,[2] Co in Mo,[3] and V in Au.[4] As in the *Rh*-Mn alloys, the magnetic susceptibility of *Rh*-Fe alloys increases at low temperatures much faster than a simple Curie-Weiss equation would predict.[5,6] Generally, deviations from Curie-Weiss behavior of the magnetic susceptibility are observed for almost all dilute alloys exhibiting local moments.[7–9] This is not surprising since the Curie-Weiss equation can only be considered as a convenient interpolation formula between a high-temperature Curie law and a finite zero-temperature solute susceptibility as predicted by both Kondo-type and fluctuation theories for the single-impurity limit.[9–11] In the absence of general theoretical predictions for the detailed temperature dependence of the magnetic susceptibility for dilute magnetic alloys, the observed systematic deviation from Curie-Weiss behavior should give valuable information about the electronic structure of a single magnetic impurity.

Quite recently, it has been demonstrated that even in very dilute alloys, interactions between solute atoms can contribute significantly to the magnetic susceptibility.[12,13] The question then arises as to whether the deviations from Curie-Weiss behavior observed earlier do indeed reflect properties of noninteracting impurity states. To investigate this question, we have made a magnetic study of a variety of dilute alloy systems over a wide temperature range. In addition, low-temperature specific-heat measurements were performed on the same alloys.

II. EXPERIMENTAL DETAILS

A. Alloy preparation

The following new alloys were prepared: Mo with 0.02-, 0.1-, 0.2-, 0.5-, and 0.7-at.% Fe, Mo with 0.1- and 0.4-at.% Co, Rh with 0.1- and 0.6-at.% Mn, and Au with 0.02- and 0.1-at.% Fe. The solvent materials Mo, Rh, and Au were 99.999% pure as quoted by the supplier. The solute materials were 99.99% pure. Before alloying, the magnetic susceptibility and the low-temperature specific heat of the solvent materials were determined. No significant amounts of magnetic impurities could be detected. For each of the four alloy systems investigated, a master alloy with about 3-at.% solute
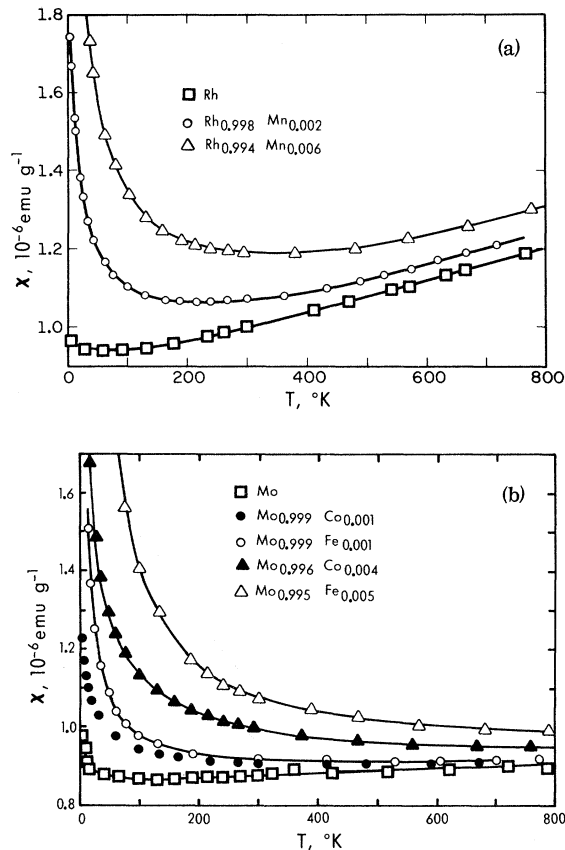
FIG. 1. Magnetic susceptibility of typical alloys vs temperature. (a) Rh alloys; (b) Mo alloys. Concentration x is given in atomic fraction.

was prepared. This master alloy was used to prepare the more dilute alloys, parts of which in turn were used to prepare the even more dilute alloys. This procedure allows a fairly good control of the alloy composition. The relative accuracy of the alloy concentration is estimated to be better than 10%. Buttons of about 10 g in weight were prepared for each alloy by arc melting under 1 atm of argon in a water-cooled copper crucible. For the Au-Fe alloys, the argon atmosphere was purified by striking the arc first on a vanadium getter.

Each alloy was melted at least four times. The alloys were then homogenized under the following conditions: the Mo-Fe and Mo-Co alloys for 4 days at 1400 °C in a vacuum furnace (10^{-6} Torr) and oil quenched; the Rh-Mn alloys for 4 days at 1250 °C in a vacuum furnace (10^{-6} Torr) and oil quenched; the Au-Fe alloys for 5 days at 1020 °C in sealed quartz capsules under argon pressure and water quenched. To protect the alloys during homogenizing they were wrapped in Mo foil.

After annealing, each alloy specimen was cleaned by grinding off the surface and subsequent chemical

etching. The flat part of the arc-melt buttons was then polished for specific-heat measurements Both measurements, specific heat, and magnetic susceptibility, were performed on the same alloy specimen.

B. Apparatus

The low-temperature specific heat was measured between 1. 5 and 4. 2 °K using a He-exchange-gas calorimeter previously described.[14,15] The susceptibility was measured by a Faraday method using a continuous-flow cryostat for temperatures between 4. 2 and 300 °K.[2] For temperatures between room temperature and 800 °K, an inert-gas furnace was used. The susceptibility was measured in fields up to 13 kOe.

III. RESULTS AND DISCUSSION

A. Susceptibility

Figures 1(a) and 1(b) compare the magnetic susceptibility of the unalloyed solvent materials to that of some typical alloys. The low-temperature increase in the susceptibility of the unalloyed solvents Rh [Fig. 1(a)] and Mo [Fig. 1(b)] is probably due to Fe impurities in these materials. These low-temperature increases have been included in the solvent susceptibility χ_s when determining the solute susceptibility $\Delta\chi = \chi - \chi_s$ from the susceptibility of the alloy χ. Figures 1(a) and 1(b) clearly demonstrate that at higher temperatures the temperature dependence of the Pauli susceptibility of Rh and Mo cannot be neglected when determining $\Delta\chi(T)$ as a function of temperature. We assume here that this temperature dependence of the Pauli susceptibility is not affected by small additions of solutes. This is justified by measurements on alloys of Rh and Mo with nonmagnetic solutes, where the susceptibility of the alloy has essentially the same temperature dependence as that of the solvent.[2,16]

Figures 2(a) and 2(b) show the solute susceptibilities $\Delta\chi$ divided by the concentration x:

$$\Delta\chi/x = [\chi - (1 - x)\chi_s]/x \simeq (\chi - \chi_s)/x \ ,$$

where χ is the susceptibility of the alloy and χ_s is the susceptibility of the solvent as shown in Figs. 1(a) and 1(b). The logarithmic temperature scale has been chosen to enhance the low-temperature part of the curves. As can be seen in Figs. 2(a) and 2(b), the solute susceptibility for the dilute alloys scales reasonably well with the solute concentration for all four alloy systems. The strong temperature dependence of the solute susceptibility reflects the local-moment behavior of the solute atoms.

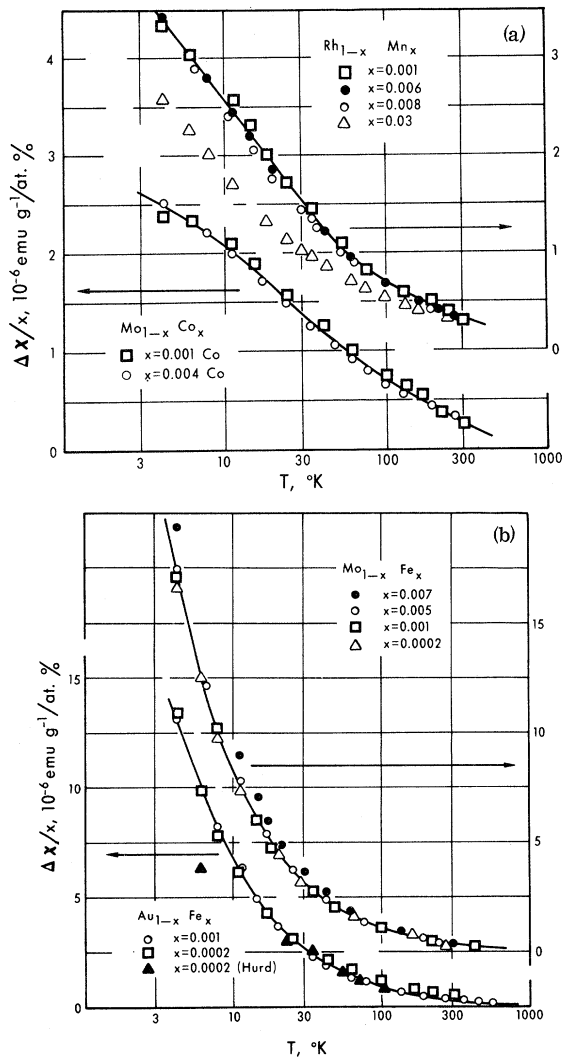As has been mentioned above, large temperature-

FIG. 2. Solute susceptibility per atomic percent of solute vs log$T$. (a) Rh-Mn and Mo-Co alloys; (b) Mo-Fe and Au-Fe alloys. Concentration x is given in atomic fraction. The data by Hurd of one of the Au 0.0002 at.fraction Fe alloys are from Ref. 22.

independent terms in the solute susceptibility have previously been reported for Rh-Mn[1,2] and Mo-Co[3] alloys. This temperature-independent term χ_0 was originally used as a parameter in a least-squares-fit analysis of the solute susceptibility to an equation of the form

$$\Delta\chi = \chi_0 + C/(T+\theta) \ . \tag{1}$$

The evaluation of the parameter χ_0 can, of course, be affected by deviation of the temperature dependence of $\Delta\chi - \chi_0$ from simple Curie-Weiss behavior. Since any kind of local-moment behavior of the susceptibility should disappear as $1/T$ when $T \to \infty$, χ_0 can be determined directly from high-temperature measurements. The results for typical alloys are

shown in Fig. 3 where the solute susceptibility is plotted vs $1/T$. As can be seen (Fig. 3), only for the Rh-Mn alloys does $\Delta\chi$ *not* extrapolate to zero for $1/T \to 0$. The value of χ_0 for the Rh-Mn alloys obtained from the intercept at $1/T = 0$ is proportional to the Mn concentration. But its value of $\chi_0 = 8 \times 10^{-4}$ emu/mol of Mn is only about one-half of the value determined earlier by a least-squares fit to Eq. (1) of data below room temperature.[1,2] This difference in χ_0 for the Rh-Mn alloys and also the apparent large χ_0 value reported earlier for Mo-Co alloys[3] is due, as shown below, to signifi-cant deviations of the solute susceptibility from Curie-Weiss behavior at low temperatures. Van Dam[17] recently reported that when fitting suscepti-bility data of Au-V alloys to Eq. (1), the χ_0 value depends largely on the temperature range of the data in question. It is therefore possible that the χ_0 value originally reported for these alloys[4] is a result of curve fitting and not a real temperature-independent term in the susceptibility.

The value of χ_0 for the Rh-Mn alloys as deter-mined from Fig. 3 is still very large being of the order of magnitude of a large exchange-enhanced Pauli susceptibility.[2] This large temperature-in-dependent term in the susceptibility in addition to a local-moment behavior now seems to occur only in alloys with Mn as a solute, and it has been found that in all cases this term is of the same order of magnitude.[18] Orbital paramagnetism is a possible explanation, but it is not clear why of all $3d$ solutes only Mn should exhibit this term.[19]

In order to study the temperature dependence of the solute susceptibility it is instructive to plot
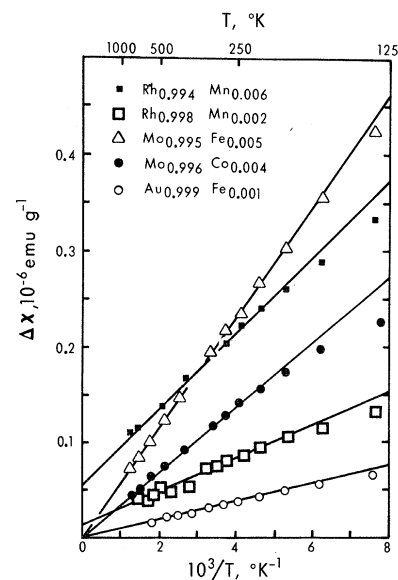

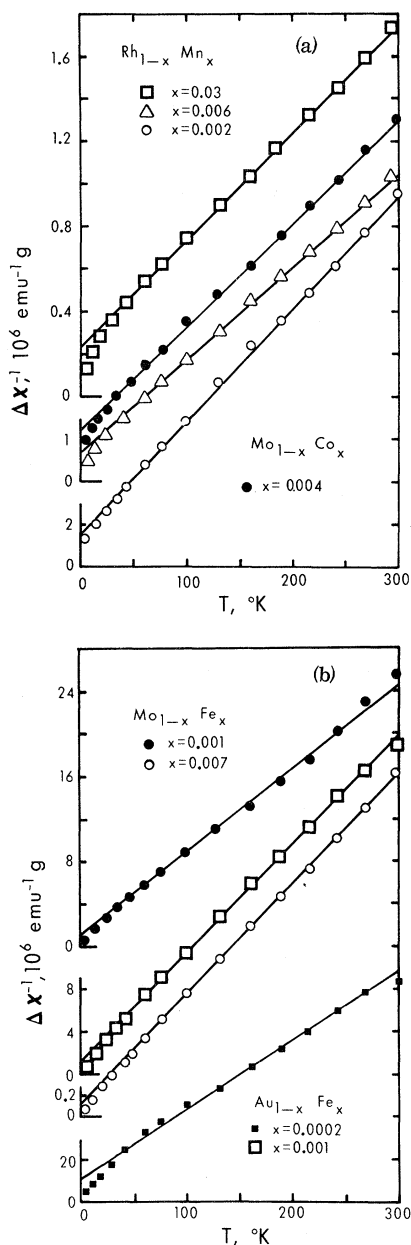FIG. 3. High-temperature solute susceptibility vs reciprocal temperature.

FIG. 4. Reciprocal solute susceptibility $(\Delta\chi)^{-1}$ vs temperature. (a) Rh-Mn and Mo-Co alloys; for Rh-Mn alloys $(\Delta\chi - \chi_0)^{-1}$ is shown, χ_0 being determined from the high-temperature data of Fig. 3. (b) Mo-Fe and Au-Fe alloys.

$(\Delta\chi)^{-1}$ as a function of the temperature. This is shown in Figs. 4(a) and 4(b). For the Rh-Mn alloys, $(\Delta\chi - \chi_0)^{-1}$ is plotted where χ_0 is determined from the high-temperature data in Fig. 3. It is clear in all cases that there is Curie-Weiss behavior (straight line) down to a temperature of about 50° K, but that at lower temperatures $\Delta\chi$ increases significantly faster than that predicted by the high-tem-

perature Curie-Weiss equation of the form

$$\Delta\chi = C/(T+\theta) . \qquad (2)$$

The parameters C and θ for this high-temperature Curie-Weiss equation [straight line in Figs. 4(a) and 4(b)] are listed in Table I for typical alloys.

The deviation at low temperatures from this high-temperature Curie-Weiss equation (Curie-Weiss plus χ_0 for Rh-Mn alloys) is shown in Fig. 5 vs $\log T$. The logarithmic form of this deviation reported earlier for the Rh-Mn alloys[1,2] was based on a too-high χ_0 value, and in view of the present results must now be considered as coincidental. Indeed, the deviation curves of Fig. 5 appear to have temperature dependence of a Curie-Weiss form but with a much lower characteristic temperature θ. This is demonstrated in Fig. 6, where, for typical alloys, the low-temperature part of the solute susceptibility is shown in the form of $(\Delta\chi - \chi_0)^{-1}$ vs temperature graphs, χ_0 here being determined by a least-squares fit of these low-temperature data to Eq. (1). The parameters of Eq. (1) thus obtained are listed in Table I. Obviously, χ_0 for the low-temperature fit reflects the high-temperature Curie-Weiss term which, at low temperatures, is approximately temperature-independent due to its relative high θ value (Table I). The two sets of parameters in Table I, representing the low- and high-temperature data, respectively, are very different. This result reflects the fact that the magnetic susceptibility for the investigated alloys cannot be represented in simple Curie-Weiss form. Any fitting of the data to Eq. (1) [or Eq. (2)] yields parameters which only are characteristic of the temperature range in question and cannot give a complete description of the solute susceptibility.[20-22] The two distinct sets of parameters of Table I suggest that the temperature dependence of the solute susceptibility of the investigated alloys is caused by two different physical effects, one predominating at high temperatures the other at low temperatures, each of them characterized by a distinctive value of θ. It is tempting to attribute the higher θ value in Table I to isolated solute atoms and thus relate it to a Kondo temperature, and to attribute the lower θ value to interacting groups of solute atoms with θ characterizing the average interaction.

In the alloys investigated, solute-solute interaction is indeed important. This is indicated, at low temperatures, by an increasing dependence of the susceptibility on the magnetic field in a form which is characteristic of large magnetic dipoles (super-paramagnetism). In the presently available range of the magnetic field and temperature, this field dependence is significant only for the Mo-Fe and Au-Fe alloys. At 4.2 °K these alloys show a 10–25% decrease in the differential susceptibility when increasing the magnetic field to 13 kOe. A

TABLE I. Parameters of Eq. (1) for typical alloys determined from the high- and low-temperature data, respectively. χ_0/x and C/x are given in units per mole of solute.

Solvent	Alloy x (at. fraction solute)	χ_0/x (10^4 emu mol^{-1}) High T	χ_0/x (10^4 emu mol^{-1}) Low T	C/x (10^2 emu deg mol^{-1}) High T	C/x (10^2 emu deg mol^{-1}) Low T	θ (deg) High T	θ (deg) Low T
Rh	0.006 Mn	9.5	28	80	49	35	12
Mo	0.005 Fe	<1	23	118	91	13	1.3
Au	0.001 Fe	<3	20	193	152	12	1.5
Mo	0.004 Co	<1	26	99	49	52	19

significant decrease (30% at 4.2 °K) in the differential susceptibility of Rh 3-at.% Mn indicates the presence of large magnetic dipoles in this concentrated alloy whereas at the same time the average susceptibility per solute atom has decreased compared to the dilute alloys (Fig. 2). In those cases where the magnetic susceptibility depends on the magnetic field, the data in Figs. 1–6 represent the low-field susceptibility. At higher fields and/or lower temperatures strong curvature in the magnetization–vs–magnetic-field curves is observed for all investigated alloy systems (Au-Fe, see Ref. 23; Mo-Co, see Ref. 24; preliminary measurements at 4.2 °K of the alloy Rh-0.6-at.% Mn show a 30% decrease in the differential susceptibility when increasing the magnetic field to 50 kOe).

This field dependence of the magnetic susceptibility at low temperatures in dilute magnetic alloys is quite common and recently has been generally accepted as being due to solute-solute interactions (magnetic clusters) in these alloys.[12,13,23] However, scaling of the magnetic susceptibility with solute concentration, such as we have observed (Fig. 2), is usually considered to be a criterion for noninteracting solute atoms. It would therefore appear questionable that the observed field dependence of $\Delta\chi$ indeed reflects interaction effects for the alloys
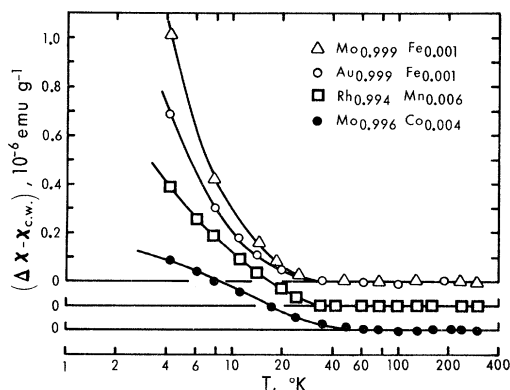
represented in Fig. 2. Very direct evidence for magnetic interactions in certain dilute alloys can be obtained from NMR measurements.[25-27] Also low-temperature specific-heat measurements should be sensitive to this type of interaction.[28] We therefore decided to measure the low-temperature specific-heat of typical alloys to further investigate the question of solute-solute interactions.

B. Specific Heat

The results of the low-temperature specific-heat measurements are shown in Fig. 7, where the specific heat C of the investigated alloys is plotted vs temperature. Also shown is the specific heat for the unalloyed solvents. As can be seen, large anomalies are observed for all alloys. The C vs T curves for the alloys do *not* extrapolate to zero for $T \to 0$, indicating a temperature-independent term in the specific heat of the alloys within the investigated temperature range (1.5–4.2 °K). These temperature-independent terms show up in form of
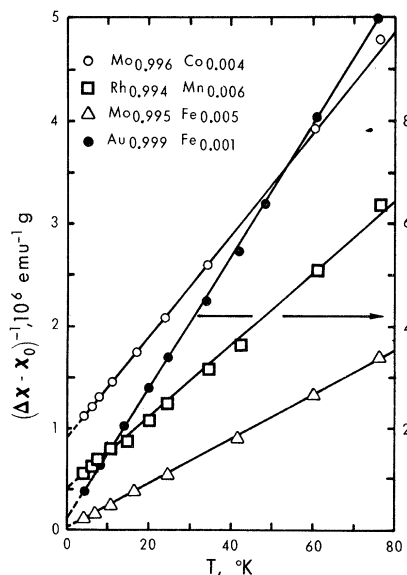

FIG. 5. Deviation of the solute susceptibility $\Delta\chi$ from the high-temperature Curie-Weiss equation χ_{CW} vs log$T$. χ_{CW} is determined from the linear part of the curves in Fig. 4.

FIG. 6. Reciprocal solute susceptibility for typical alloys vs temperature. χ_0 is determined by least-squares fit of $\Delta\chi$ to Eq. (1).
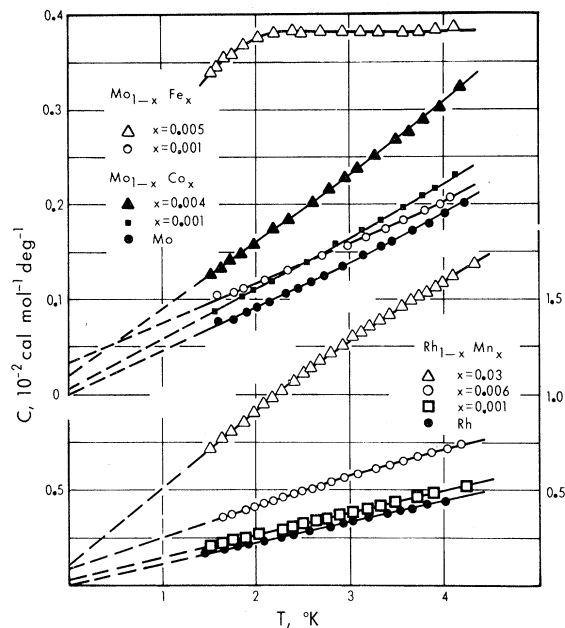
FIG. 7. Low-temperature specific heat vs temperature.

so-called "upturns" when C/T is plotted vs T^2, the usual form of presenting low-temperature specific data. This is shown in Fig. 8 for the most dilute alloys. The "upturn" is much larger for the more concentrated alloys. Again the data for the un-alloyed solvent is shown (Fig. 7) yielding an electronic specific-heat coefficient (intercept at $T^2 = 0$) of $\gamma = 11.05 \times 10^{-4}$ cal deg^{-2} mol^{-1} for Rh and $\gamma = 4.46 \times 10^{-4}$ cal deg^{-2} mol^{-1} for Mo and a coefficient for the lattice specific heat (slope of C/T vs T^2) of $\beta = 0.6 \times 10^{-6}$ cal deg^{-4} mol^{-1} for Rh and $\beta = 1.7 \times 10^{-6}$ cal deg^{-4} mol^{-1} for Mo in reasonable agreement with earlier measurements.[29] Also shown in Fig. 8 are specific-heat data for the alloy $Rh_{0.98} Co_{0.02}$. Cobalt does not form a localized moment in dilute Rh-Co alloys but exhibits a very large local-exchange-enhanced Pauli susceptibility.[2,30] The observed large increase in the electronic specific-heat coefficient when adding Co to Rh corresponds to a value of $\gamma = 0.8 \times 10^{-2}$ cal deg^{-2} mol^{-1} of Co and is probably due to large local spin fluctuation on the Co sites.[2,30] The increase in the specific heat when adding Mn to Rh is more than a factor of 10 larger than that due to adding the same amount of Co (0.1-at.% Mn nearly has same effect as 2-at.% Co, Fig. 7). This strongly suggests that the origin for the specific-heat anomalies in Rh-Mn must be sought in magnetic solute-solute interactions. The same type of anomalies in the low-temperature specific heat (upturns) are very common in many concentrated para- and also ferromagnetic alloys with an alloy concentration where the ferromagnetic Curie temperature as function of alloy composition extrapo-

lates to 0 °K. In most of these alloys the appearance of these anomalies is attributed to spatially nonuniform magnetizations, i.e., magnetic clusters. In some alloys these clusters have been directly observed via the superparamagnetic properties which they exhibit. Also neutron-scattering experiments show evidence of these clusters (for example, Ni-Cu[31-33] and V-Fe[34-36] alloys). It is of interest in the present connection to note that these concentrated alloys on the paramagnetic side of the critical concentration can be considered as magnetically dilute[31,32] with only a relatively small concentration of magnetic dipoles.

In a simple approximation it has been shown[37-39] that, in a limited temperature range, the magnetic clusters contribute a constant term to the specific heat of the alloy,

$$C = A + \gamma T + \beta T^3 , \qquad (3)$$

where A is the "cluster" specific heat, γ the electronic specific-heat coefficient, and β the lattice specific-heat coefficient. A temperature-independent specific heat can, of course, only be considered a first approximation to the cluster specific heat, and one has to realize when fitting specific-heat data to Eq. (3) that the cluster specific heat can also affect the linear and cubic term in Eq. 3.[38,39]

In Fig. 9 the solute specific heat $\Delta C = C$(alloy) $- C$(solvent) is plotted vs temperature. Also shown is ΔC for a Au 0.1-at.% Fe alloy as measured by Bethoux et al.[23] The dilute Rh-Mn and Mo-Co alloys can be described by a simple equation of the form of Eq. (3), the solute specific heat ΔC consisting of a temperature-independent term and a term linear in temperature. The parameters of Eq. (3) for these alloys are listed in Table II. Like the magnetic susceptibility, the solute specific heat for these dilute alloys scales roughly with concentration (Table II). The solute specific heat for the Mo-Fe and Au-Fe alloys is of a more complicated
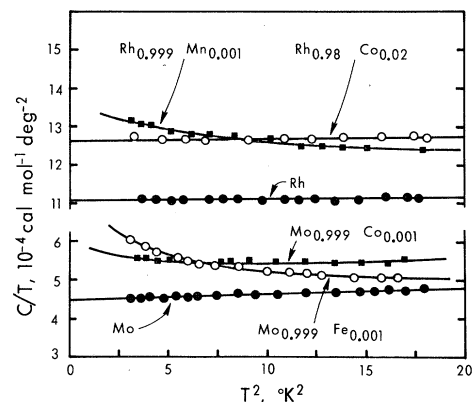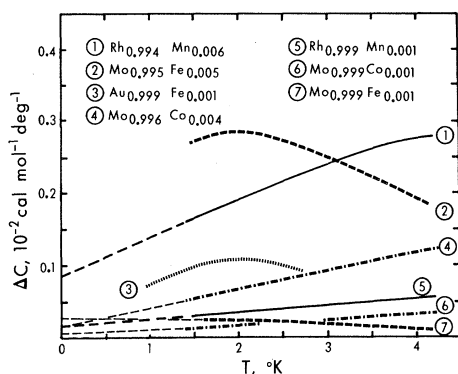

FIG. 8. C/T vs T^2 for dilute alloys.

FIG. 9. Solute specific heat $\Delta C = C_{(alloy)} - C_{(solvent)}$ vs temperature. The data for the Au-Fe alloy are from Ref. 23.

concentrated alloys near the critical concentration for ferromagnetism is small. Both the dilute alloys and the concentrated alloys are therefore magnetically dilute, the difference being the chemical composition at which localized moments first occur. This strongly suggests that as in concentrated alloys the specific-heat anomalies in the dilute alloys are caused by strong solute-solute interactions.

C. Interpretation of $\Delta\chi$

The specific-heat results discussed above indicate that despite the observed scaling of the low-field magnetic susceptibility with solute concentration, solute-solute interaction is significant for all investigated alloys. The following interpretation is suggested: The solute susceptibility can be considered to consist of the two terms

$$\Delta\chi = \chi_K - \chi_M = \frac{C_K}{T + T_K} + \frac{C_M}{T + T_M} , \qquad (4)$$

where χ_K represents the susceptibility of isolated solute atoms with a Kondo temperature T_K[41] and χ_M describes the susceptibility of interacting solute atoms with T_M being characteristic of the interaction. This separation of the susceptibility into two Curie-Weiss terms is suggested by the data as discussed above. It can only be considered as a very approximate description of the susceptibility, since in the case of solute-solute interaction one has to expect a distribution of different T_M values corresponding to solute atoms with different chemical and magnetic environments. Also, it is not justifiable to assume that the Kondo susceptibility follows a simple Curie-Weiss equation down to temperatures below the Kondo temperature T_K.

For $T \gg T_K$ and $T \gg T_M$, Eq. (4) reduces to Eq. (2) with $C = C_K + C_M$ and

$$\theta = (C_K T_K + C_M T_M)/(C_K + C_M),$$

form showing a maximum in the investigated temperature range. It does *not* scale with concentration (Fig. 7 and Ref. 23). The A/x values for the Au-Fe and Mo-Fe alloys in Table II represent the average solute specific heat, averaged over the investigated temperature range. When fitting the specific heat of these alloys to Eq. (3) one obtains anomalously low or even negative β values. Again this behavior is often observed in many concentrated alloys near the onset of ferromagnetism.[34,40] Whereas the detailed temperature dependence of these specific-heat anomalies in concentrated alloys is not always well understood, it has been demonstrated that it is due to the above-mentioned spatially inhomogeneous magnetization (magnetic clusters).[31,34,35]

The specific-heat anomaly observed in the dilute alloys is similar to that observed in the concentrated paramagnetic alloys. This similarity is probably more than coincidental. As in the dilute alloys, the concentration of magnetic dipoles in the

TABLE II. Low-temperature specific-heat parameters of Eq. (3) for the investigated alloys. The A/x value for the Au-Fe alloy is determined from Ref. 23. $\Delta\gamma/x$ and A/x are given in units per mole of solute.

Alloy Solvent	x (at. fraction solute)		γ (10^{-4} cal mol^{-1} deg^{-2})	A (10^{-4} cal mol^{-1} deg^{-1})	$\Delta\gamma/x$ (10^{-2} cal mol^{-1} deg^{-2})	A/x (10^{-2} cal mol^{-1} deg^{-1})
Rh			11.05			
Rh	0.001	Mn	12.0	1.8	10	18
Rh	0.006	Mn	16.0	9.2	8.3	15
Rh	0.03	Mn	30	11	10	4
Mo			4.46			
Mo	0.001	Co	5.3	0.5	8.4	5
Mo	0.004	Co	7.0	2.0	6.3	5
Mo	0.001	Fe				25
Mo	0.005	Fe				50
Au	0.001	Fe				90
Rh	0.02	Co	12.67		0.82	

TABLE III. Parameters of Eq. (4) for the investigated alloys. The χ_0 values for Rh-Mn are determined from the high-temperature data in Fig. 3. χ_0/x, C_K/x, and C_M/x are given in units per mole of solute.

| Alloy | | | | χ_K | | χ_M | |
Solvent	x (at. fraction of solute)		χ_0/x (10^4 emu mol^{-1})	C_K/x (10^2 emu deg mol^{-1})	T_K (deg)	C_M/x (10^2 emu deg mol^{-1})	T_M (deg)
Rh	0.001	Mn	7.9	50	70	43	11.8
	0.002	Mn	8.0	48	50	41	11.3
	0.006	Mn	9.5	51	50	29	8.0
	0.008	Mn	8.5	49	50	27	7.6
	0.030	Mn	9.4	61	57	10	2.0
Mo	0.0002	Fe				(53)	(0.3)
	0.001	Fe		56	30	66	0.3
	0.002	Fe		60	29	67	0.3
	0.005	Fe		48	32	70	0.3
	0.007	Fe		44	31	86	0.7
Au	0.0002	Fe				(110)	(0.5)
	0.001	Fe		53	40	140	1.5
Mo	0.004	Co		86	60	13	8

C and θ being determined by the high-temperature Curie-Weiss equations (straight lines in Fig. 4, see also Table I).

It can be shown that at low temperatures Eq. (4) can again be approximated by a Curie-Weiss equation of the form of Eq. (1) with the parameters of Eq. (1) depending on the temperature range in question. The four parameters of Eq. (4) can be best determined by requiring that they represent the correct high-temperature parameters and at the same time give the best fit to the data at low temperatures. The parameters thus determined are listed in Table III. As expected from the behavior discussed above (Figs. 3, 4, and 6) the solute susceptibility for all investigated alloys can be very well represented by Eq. (4). In order to better demonstrate the concentration dependence of C_K and C_M, they are divided by the concentration x and given in units per mole of solute. In the absence of systematic high-field measurements, it is at present not possible to separately determine from the two Curie constants the concentration and effective magnetic moments of the isolated solute atoms and of the interacting solute atoms, respectively. The following concentration dependence of the parameters of Eq. (4) is observed.

Rh-Mn: For these alloys Table III also gives the temperature-independent term of the solute susceptibility χ_0 as determined directly from high-temperature measurements. As already mentioned above, this term is roughly proportional to the Mn concentration even up to 3-at.% Mn (Table III). The same is true for the Kondo susceptibility χ_K of Eq. (3), C_K/x and T_K being approximately constant. Contrary to this there is a tendency for both

C_M/x and T_M to systematically decrease towards the low value of both parameters for the 3-at.% alloy. As mentioned above, the slower than linear increase of χ_M with concentration is coupled with the appearance of larger magnetic dipoles giving rise to a field-dependent susceptibility. This concentration dependence can only be understood in terms of complex magnetic interactions. The large T_K value for the 0.1-at.% Mn alloy may not be very significant since the total susceptibility for this alloy is small, and any systematic errors in the susceptibility measurements could have large effects on the parameters of the weakly temperature dependent χ_K. The average effective moment per Mn atom as determined from the high-temperature Curie-Weiss equation (Table I) amounts to $p = 2.5\mu_B$ per Mn atom.

Mo-Fe: The Kondo susceptibility χ_K scales roughly with concentration, the Kondo temperature T_K being constant. Similarly, χ_M increases linearly with concentration, except for the most concentrated alloys, where both C_M and T_M have increased. [For the most dilute alloy, $x = 0.0002$ at. fraction Fe, the solute susceptibility at high temperature is not accurate enough for determination of the parameter of Eq. (4) C_M and T_M for this alloy (Table III) are estimated from the low-temperature data.] The large value for the parameters of χ_M at high concentrations is reflected in the data of Fig. 2 where the solute susceptibility of the most concentrated alloy ($x = 0.007$ at. fraction) no longer scales with concentration, but shows a faster than linear increase of $\Delta\chi$ with the concentration. This concentration dependence of χ_M is qualitatively different from that of the Rh-Mn alloys, indicating

that the Fe-Fe interactions in the Mo-Fe alloys have a larger tendency for parallel alignment of the magnetic dipoles than the Mn-Mn interactions in Rh-Mn alloys. This is in agreement with the appearance of large magnetic dipoles already in very dilute Mo-Fe alloys.

From the high-temperature Curie-Weiss equation (Table I) one obtains an average effective moment of about $p = 3.0\mu_B$ per Fe atom.

Au-Fe: These alloys behave very similarly to the Mo-Fe alloys, but with an even larger tendency for magnetic "clustering," χ_M being about three times larger than χ_K. (For reasons given above, the parameters of χ_M for the alloy with $x = 0.0002$ at. fraction are estimated from the low-temperature susceptibility data.) The average high-temperature effective moment amounts to $p = 3.9\mu_B$ per Fe atom.

Mo-Co: Except for a smaller tendency for magnetic clustering these alloys are very similar to the Rh-Mn alloys. The solute susceptibility of the Mo 0.1-at.% Co alloy (Fig. 1) is too small for a meaningful separation into the two terms of Eq. (4). The average effective moment at high temperatures is $p = 2.8\,\mu_B$ per Co atom.

IV. SUMMARY

For a number of different dilute magnetic alloys we have shown that although the low-field susceptibility scales with solute concentration there is evidence for strong solute-solute interaction. This evidence is reinforced by the similar specific-heat behavior of the dilute alloys and some concentrated alloys near the critical concentration for ferromagnetism. Since both types of alloys are magnetically dilute, the difference being the chemical composition at which local moments first start to appear, this similarity is probably more than coincidental. Nevertheless, despite the strong tendency for magnetic cluster formation, it is at present not clear why the low-field susceptibility scales with concentration. A complex magnetic interaction is suggested leading eventually at high concentrations either to a faster or slower than linear increase of the solute susceptibility with concentration. A simple model is used to separate the contribution due to isolated solute atoms from the total susceptibility. The Kondo temperatures obtained are generally higher than previously assumed.

Finally, we would like to mention that the presence of magnetically different solute atoms in dilute alloys should be directly seen in NMR or Mössbauer experiments which probe the solute hyperfine fields. Measurements of the Co^{59} Knight shift K in Mo-Co alloys as a function of temperature demonstrate that at low temperatures, K varies much less with temperature than the bulk susceptibility[42] but rather has a temperature dependence close to the form $1(T + 60\,°K)$.[42,43] This behavior can be readily explained in our simple model: The observed resonance signal comes from the isolated Co atoms, the susceptibility of which varies as $1/(T + T_K)$ with $T_K = 60\,°K$ (Table III). Due to large hyperfine fields seen by the strongly interacting Co atoms, the resonance of these atoms is not observed.

Mössbauer measurements of Fe^{57} in Mo at low temperatures indicate a strongly temperature- and field-dependent hyperfine field.[44] Since in this type of experiment the Fe atoms with the strongest hyperfine fields are most readily seen (give the largest splitting) it seems obvious to correlate the observed splitting to interacting Fe atoms. The temperature dependence at low fields, $H_{eff} \sim 1/(T + 0.8\,°K)$,[44] is indeed very similar to that of χ_M (Table III). Since the hyperfine field of the spin-compensated isolated Fe atoms is expected to be small and only weakly temperature dependent at low temperatures, it may be difficult to identify those atoms in Mössbauer measurements.

Whereas the results of the two experiments just described closely agree with our present interpretation of the solute susceptibility in dilute alloys, they do not completely justify this interpretation. They clearly indicate, however, that careful measurements of the temperature dependence of the solute hyperfine fields, when combined with measurements of the bulk susceptibility, are the most promising types of experiments to study interaction effects in dilute magnetic alloys. In addition, detailed measurements of the specific heat over an extended temperature range should give valuable information about these interaction effects.

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