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Effect of pressure on impurity-impurity interactions in dilute Au:Mn, Cu:Mn, and Au:Fe spin-glass alloys*

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The electrical resistivity of the dilute spin-glass alloys Au–0.10-at.% Mn, 0.15-at.% Mn, 0.10-at.% Fe, 0.13-at.% Fe, and Cu–0.15-at.% Mn has been measured from 1.2 to 40 K at pressures to 100 kbar. In these alloys the cooperative "locking-in" of the impurity spins at a temperature T_0 leads to a resistivity maximum at $T_{\max}$. Application of pressure is found to shift $T_{\max}$ in a manner which is strongly system dependent: $dT_{\max}/dP > 0$ for Au:Mn, $dT_{\max}/dP \approx 0$ for Cu:Mn, and $dT_{\max}/dP < 0$ for Au:Fe. These results are shown to be clearly incompatible with the widely held belief that $kT_{\max} \approx \Delta_{\mathrm{RKKY}}$, where $\Delta_{\mathrm{RKKY}} \approx cJ^2S(S+1)/E_F$ is the average strength of the Ruderman-Kittel-Kasuya-Yosida interaction, and indicate that $T_{\max}$ is a function of *both* Δ_{RKKY} *and* T_K, the Kondo temperature. This expectation is confirmed in a recent theory of Larsen who introduces an impurity-impurity interaction strength Δ_c into the parquet approximation of the Kondo resistivity and obtains an explicit expression for $T_{\max} = T_{\max}(\Delta_c, T_K)$. It is shown that both the sign and the magnitude of $dT_{\max}/dP$ for the systems studied here are a natural consequence of both increasing T_K and Δ_c in *all* systems and depend on the relative magnitudes of Δ_c and T_K. In particular, one would expect to find $dT_{\max}/dP < 0$ in systems such as Au:Fe with relatively high Kondo temperatures. A further result of the analysis is that $\Delta_c(P) \approx \Delta_{\mathrm{RKKY}}(P)$, lending support to the view that in these systems the long-range RKKY oscillations represent the dominant impurity-impurity interaction mechanism.

I. INTRODUCTION

A prerequisite to the successful application of the local-moment approach for a description of magnetism in magnetically concentrated metallic alloys is an understanding of the nature of the various interaction mechanisms between local moments. In addition to the relatively-short-range direct-exchange interactions, due to the overlap of neighboring local-moment orbitals, there exists an indirect long-range interaction, mediated by the conduction electrons, which is usually described in terms of the well-known Ruderman-Kittel-Kasuya-Yosida (RKKY) spin-density oscillations. In a given magnetic alloy these interactions lead to magnetic ordering at a temperature $kT_0 \approx \Delta_c$, where the average interaction strength at a local moment site, Δ_c, is just balanced by the thermal fluctuations kT_0. The type of the magnetic ordering which occurs (ferromagnetic, antiferromagnetic, spin glass, etc.) depends upon both the spatial distribution of the spins and the exact nature of the interaction mechanisms involved.

The interaction between the local-moment spin $\vec{S}$ and the conduction-electron spin $\vec{s}$, which leads to the indirect RKKY spin-spin interaction, is usually described in terms of the s-d exchange Hamiltonian $\mathcal{H} = -2J\vec{S}\cdot\vec{s}$, where J is the effective exchange parameter. Information about this interaction is obtained most directly by measurements on alloys where the magnetic-impurity concentration c is sufficiently small so that interactions between impurities are negligible in the temperature range of interest. Negative values of J lead to a spin-scattering contribution to the resistivity ρ_{Kondo} which increases with decreasing temperature and leads, if the phonon-scattering resistivity of the host is included, to the well-known resistivity minimum.[1] This is illustrated in Fig. 1. The inflection point of ρ_{Kondo} determines the characteristic Kondo fluctuation temperature T_K, defined by

$$kT_K = De^{-1/n|J|}, \tag{1}$$

where D is the bandwidth and n the density of states at the Fermi level. Although no exact solution for ρ_{Kondo} has yet been found, present theories predict the existence of a "universal" resistivity law $\rho = \rho(T/T_K)$, a confirmation of which was demonstrated by resistivity measurements on a Cu-110-ppm-Fe alloy where T_K was found to increase rapidly with pressure.[2] Subsequent work on very dilute alloys combining Cu and Au hosts with Fe,

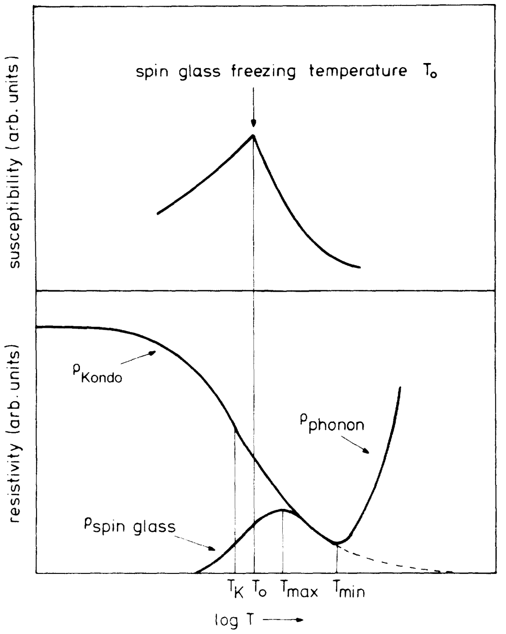

FIG. 1. Schematic diagram of the electrical resistivity of isolated impurities ρ_{Kondo} and interacting impurities $\rho_{\mathrm{spin\ glass}}$ as a function of $\log T$. ρ_{Kondo} has the characteristic temperature T_K. Interactions between impurities give rise to a resistivity maximum at T_{max}. The onset of the phonon resistivity ρ_{phonon} is also illustrated and gives rise to the well-known resistivity minimum at T_{min}. The sharp peak in the initial susceptibility, which occurs at the freezing temperature T_0, is also shown (see Refs. 6–8).

Mn, and V impurities[3,4] has shown that the increase of T_K with pressure is a result of general validity.[5] From the measured pressure dependence of T_K it can be estimated from Eq. (1) that for these systems $|J|$ increases by roughly 15% between 0 and 100 kbar pressure.

As the concentration of such very dilute magnetic alloys is increased, the indirect RKKY interaction between impurities becomes important and leads to "spin-glass" behavior[6] which is characterized by a "freezing out" of the impurity spins in random directions below a characteristic temperature T_0.[7] This so-called spin-glass freezing temperature is marked by a sharp peak in the initial susceptibility,[6,8] which is illustrated in Fig. 1, and also by an abrupt hyperfine splitting of the Mössbauer line.[9] In this so-called spin-glass regime, where the indirect RKKY interactions are believed to represent the dominant interaction mechanism, it is expected, and will be confirmed by comparison between theory and experiment, that the interaction parameter Δ_c is approximately equal to the mean RKKY-interaction energy Δ_{RKKY} given by the expression[10]

$$\Delta_{\mathrm{RKKY}} \simeq cJ^2S(S+1)/E_F . \tag{2}$$

In the limit $\Delta_{\mathrm{RKKY}} \gg T_K$, where the interaction strength is far greater than the Kondo fluctuation temperature, one would expect that $kT_0 \simeq \Delta_{\mathrm{RKKY}} \propto cJ^2$. Indeed, within the so-called spin-glass scaling regime,[11] T_0 is observed to be approximately proportional to concentration; in addition, an increase of T_0 with pressure would be expected from the results on the very dilute alloys which show that $|J|$ increases with pressure (the pressure dependence of E_F is comparatively small[2] and S is assumed constant).

The "locking in" of the impurity spins below T_0 destroys the growth of the delicately balanced Kondo resonance state and reduces the spin-flip scattering between impurity and conduction-electron spins. In ferromagnetic alloys the removal of the spin-flip scattering results in a rapid drop in the spin disorder resistivity near the Curie temperature.[12] Similarly, in spin-glass alloys, ρ_{spin} is forced to fall off at temperatures near or below T_0, resulting in a resistivity maximum at T_{max},[13] as illustrated in Fig. 1. For the alloys considered here, T_{max} is typically about twice as large as T_0, although in general the exact relationship between these two temperatures is unknown. Whereas some calculations of the resistivity maximum[14] have supported the conjecture that $T_{\mathrm{max}} \simeq \Delta_{\mathrm{RKKY}} \propto cJ^2$, a calculation by Matho and Béal Monod[15] based on the interacting pair model indicates that in general no exact proportionality between T_{max} and c exists. It is difficult to satisfactorily resolve this question using present experimental results. One of the present authors (U.L.) has recently developed a theory of the resistivity maximum[16] (see preceding paper) which treats the Kondo effect in interacting spin systems in more detail than previous attempts. T_{max} is shown to be a composite function of both Δ_c and T_K, and for $\Delta_c \gg T_K$ it is found that

$$T_{\mathrm{max}} \simeq \tfrac{1}{2}\Delta_c \ln(\Delta_c/T_K) . \tag{3}$$

Deviations from $T_{\mathrm{max}} \propto c$ are, therefore, *always* expected to be present to some extent and should be especially pronounced as c is decreased or T_K increased such that $\Delta_{\mathrm{RKKY}} \simeq T_K$. Such effects have been reported by Laborde and Radhakrishna[17] in measurements of the concentration dependence of T_{max} for a series of dilute Au:Fe alloys, and by Star[18] who found that, although T_K (and therefore $|J|$) *increases* with increasing Cu content in the $\mathrm{Au}_{1-x}\mathrm{Cu}_x$–0.15-at.%-Fe alloy series, T_{max} *decreases*. In the latter measurements it is not clear whether the suppression of T_{max} is due to the intrinsic T_K increase or to the weakening of the RKKY interaction by the increasing number of Cu

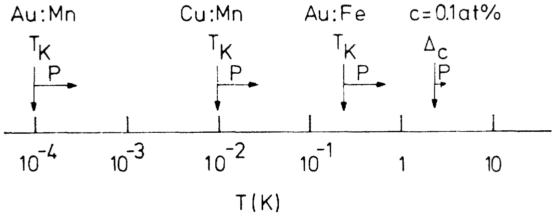

FIG. 2. Temperature scale showing relative values of T_K and a typical value of Δ_c for the alloys studied. The effect of pressure on T_K and Δ_c is also indicated.

defect atoms. Star also proposes that similar mechanisms are responsible for the behavior of $T_{\max}$ observed by Sarachik *et al.*[19] in the $Mo_{1-x}Re_x$(Fe) alloy series.

In a previous paper[20] we reported on high-pressure resistivity measurements on a Au-0.13-at.%-Fe alloy, where $T_{\max}$ is found to decrease as T_K (and $|J|$) increases with pressure, in analogy with the results of Star. This behavior is in marked opposition to $T_{\max} \propto J^2$ and is related, we believe, both to the rather close proximity of T_K and $T_{\max}$ and to the rapid increase of T_K with pressure. It would be of interest to test these ideas by carrying out measurements on systems which have much lower values of T_K. In the present paper we present further measurements[21] on Au:Fe, Cu:Mn, and Au:Mn alloys which, as shown in Fig. 2, have successively smaller T_K values.[1] Indeed, for Au-0.10-at.% Mn, where $T_K \leq 10^{-4}$ K $\ll T_{\max} = 2.5$ K, $T_{\max}$ is observed to *increase* with pressure. This is in agreement with the relation $T_{\max} \propto |J|$ which can be derived from Eq. (3) in the limit of *very* large values of Δ_c/T_K. For Cu:Mn, where T_K is intermediate between those of Au:Fe and Au:Mn, no pressure dependence of $T_{\max}$ is measured. A detailed comparison of the above theory with experiment reveals that, although for the above systems the observed pressure dependence of $T_{\max}$ is radically different, the interaction parameter Δ_c increases with pressure in all cases. In fact, the analysis shows that $\Delta_c \approx \Delta_{RKKY}$ and thus that it is possible to derive from measurements on very dilute magnetic alloys information about impurity-impurity interactions in more concentrated systems.

II. EXPERIMENTAL

The alloys used in these experiments were prepared from 99.9999% pure Au and Cu and from 99.99% pure Fe and Mn.[22] Melting was done in a standard arc furnace under approximately $\frac{1}{2}$ atm of ultra-pure argon, the samples being turned over many times and remelted. After rolling the samples between plastic sheets to a thickness of approximately 80 μm, the resulting foils were strain annealed in quartz ampules under high vacuum for several hours at 800 °C and quenched into iced water. The composition of the alloys determined by chemical analysis agrees to within 10% of the nominal value; the temperatures of the resistivity maxima, which increase with the concentration, correspond in general closely with those of previous measurements.[13,23,24]

The tiny samples used in the high-pressure studies are taken from a much larger foil sample whose resistivity had been previously measured in a conventional zero-pressure sample holder. The absolute accuracy is limited to about 1% by uncertainties in the length-to-area ratio. Although a direct determination of the exact dimensions of the samples used in the high-pressure cell is not possible, a comparison of the absolute zero-pressure and high-pressure data allows a definition of the absolute resistivity scale for all data.[2]

High pressures are applied to the samples using the quasihydrostatic Bridgeman technique of opposing anvils. The high-pressure cell and the anvil assembly are mounted inside a massive Cu-Be clamp which retains the applied force of up to 50 tons after removal from a hydraulic press. Measurements are carried out at fixed pressure and variable temperature and the applied pressure is changed at room temperature after each low-temperature run. In order to minimize sample dimension changes due to pressure-cell deformation upon initial loading, the cell is preloaded to about 80 kbar and then fully unloaded before commencing with measurements. The changes in the length-to-area ratio of the sample upon reloading to 100 kbar are estimated[2] to be less than a few percent. The pressure cell contains, in addition to two spin-glass specimens, a lead manometer whose superconducting transition temperature T_c is a sensitive function of pressure. The T_c-versus-pressure scale used here is somewhat revised from that of Eichler and Wittig[25] and includes recently redetermined pressure fixed points of Bi and Pb,[26] 7.20 K-0 kbar, 6.21 K-25 kbar, 4.70 K-74 kbar, 3.60 K-130 kbar. Further details of the high-pressure technique are described in previous publications.[2,27]

Resistivity measurements are carried out using the standard four-terminal technique. The current source from North Hills Electronics is constant to better than $1:10^5$ during the 6 h needed to carry out a temperature run. The potential drop across a sample is first amplified by a temperature stabilized Amplispot galvanometric photocell amplifier and then displayed on a Solatron LM 1490 $5\frac{1}{2}$ digit digitalvoltmeter using 2-sec integration time. The resolution of the measuring system is better than 1 nV and the time stability of the amplifier is such that after a 4-h temperature run from 4.2 to 1.2 K the resistance at 4.2 K returns without any measurable shift to its original value. The scatter in the data is less than $1{:}10^4$.

The sample temperature is measured using a calibrated Ge thermometer located directly next to the high-pressure anvils. The accuracy of the temperature measurement below 4.2 K is at least 0.01 K and better than 0.5% at any temperature. Near $T_{\max}$ data are taken in intervals of 0.1 K with the temperature stabilized to within ± 0.002 K. The small shifts of the temperature of the resistivity maximum with pressure (a few tenths of a degree) are thus well within the accuracy of the measuring system.

III. RESULTS OF EXPERIMENT

In Figs. 3–5 are shown the results of resistivity measurements on the three spin-glass alloys Au-0.10-at.% Mn, Cu-0.15-at.% Mn, and Au-0.10-at.% Fe at pressures up to 96 kbar and temperatures from 1.2 to 40 K. All three systems have resistivity maxima located at approximately 2.5 K. The resistivity curves for each pressure are displaced vertically for clarity. Additional data were taken on the same systems at different concentrations and will be included in the following discussion. The data reveal that for Au:Mn a pressure of 100 kbar causes a shift of $T_{\max}$ to higher temperatures. On the other hand, for Cu:Mn no pressure shift of $T_{\max}$ is observed, and for Au:Fe, in agreement with previous measurements on a Au–0.13-at.% Fe alloy,[20] pressure even shifts $T_{\max}$ to lower temperatures. To bring the pressure shifts out clearly, in Fig. 6 the data near the maxima are shown on an expanded scale for the lowest and highest pressures measured on a given sample. The values of $T_{\max}$ are determined to ± 0.01 K by carrying out a linear regression on the slopes of consecutive data points near the maximum and are displayed in Fig. 7 as a function of the relative volume V/V_0. A linear least-squares fit to the data is also shown. Data on the same system at different concentrations retain the same sign but not the same magnitude of $dT_{\max}/dP$.

The major portion of the spin-scattering resistivity is lost in the phonon resistivity for temperatures greater than $T_{\min}$. The presence of the phonon resistivity and large deviations from Matthiessen's rule prevent us from extracting precise information about the spin resistivity in this upper temperature region. The question arises whether such disturbing influences have any effect on the temperature of the resistivity maximum itself.

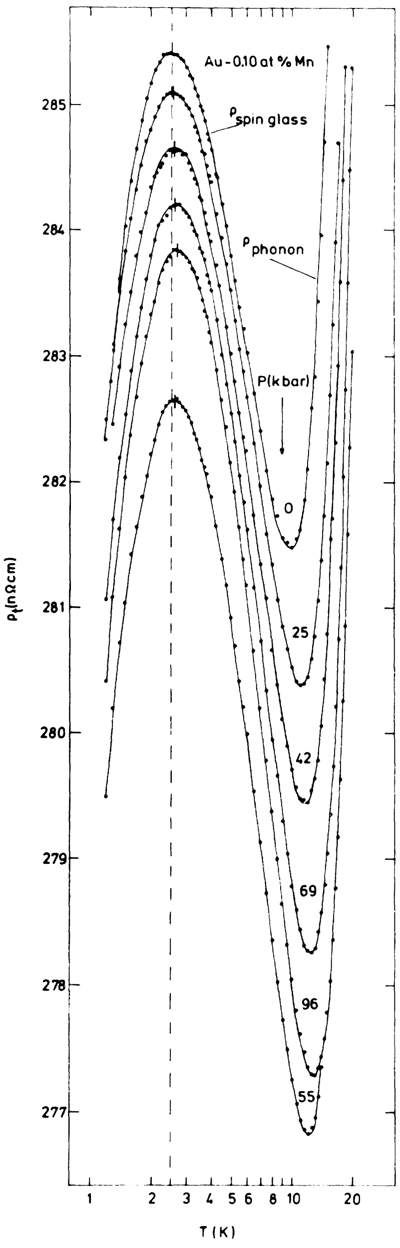

FIG. 3. Total measured resistivity ρ_t of Au–0.10-at.% Mn as a function of $\log T$ for pressures of 25, 42, 69, 96, and 55 kbar measured in that sequence on the same sample. The solid lines connecting the points are drawn for clarity. The absolute scale applies to the $P = 0$ data. The high-pressure data are shifted vertically, although the relative scale is the same for all data. The values of ρ_t at $T_{\max}$ are: 285.4, 304.0, 314.7, 331.5, 345.3, and 343.1 nΩ cm at pressures of 0, 25, 42, 69, 96, and 55 kbar, respectively. The high-pressure data contain ≈4 nΩ cm more defect scattering than at $P = 0$. The vertical dashed line through 2.53 K defines $T_{\max}$ for $P = 0$. Pressure shifts $T_{\max}$ to higher temperatures.

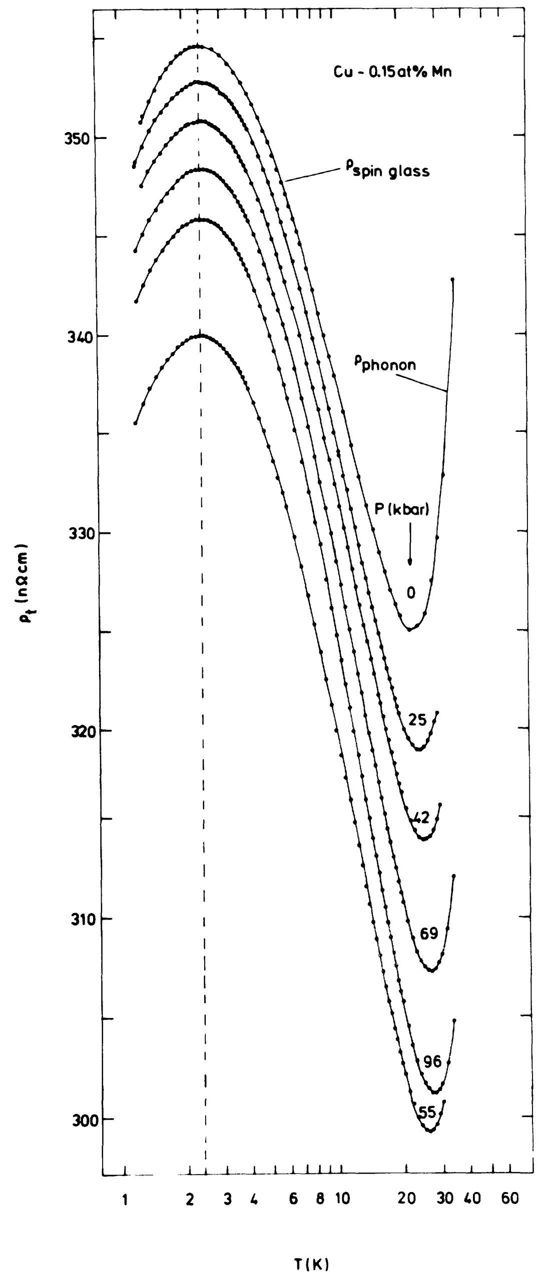

FIG. 4. Total measured resistivity ρ_t of Cu–0.15-at.% Mn as a function of log$T$ for pressures of 25, 42, 69, 96, and 55 kbar measured in that sequence on the same sample. The solid lines connecting the points are drawn for clarity. The absolute scale applies to the $P=0$ data. The high-pressure data are shifted vertically, although the relative scale is the same for all data. The values of ρ_t at $T_{\max}$ are: 354.6, 412.2, 433.2, 467.4, 495.1, and 482.1 nΩ cm at pressures of 0, 25, 42, 69, 96, and 55 kbar, respectively. The high-pressure data contain ≈ 24 nΩ cm more defect scattering than at $P=0$. The vertical dashed line through 2.41 K defines $T_{\max}$ for $P=0$. Pressure causes no measurable shift in $T_{\max}$.

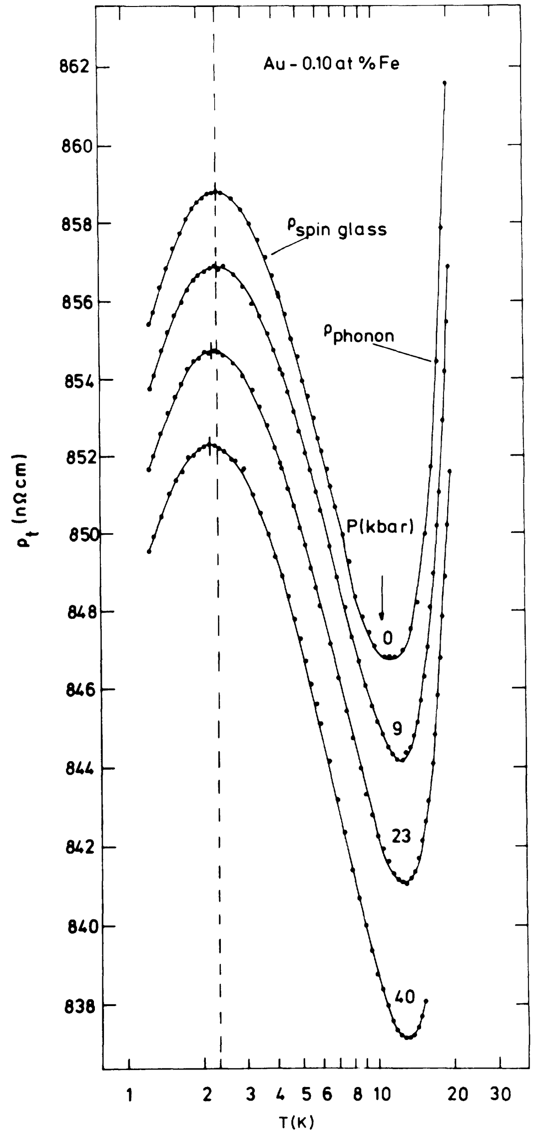

FIG. 5. Total measured resistivity ρ_t of Au–0.10-at.% Fe as a function of log$T$ for pressures of 9, 23, and 40 kbar measured in that sequence on the same sample. The solid lines connecting the points are drawn for clarity. The absolute scale applies to the $P=0$ data. The high-pressure data are shifted vertically, although the relative scale is the same for all data. The values of ρ_t at $T_{\max}$ are 858.8, 892.3, 894.8, 899.3, nΩ cm at pressures of 0, 9, 23, and 40 kbar, respectively. The high-pressure data contain ≈ 31 nΩ cm more defect scattering than at $P=0$. The vertical dashed line through 2.38 K defines $T_{\max}$ for $P=0$. Pressure shifts $T_{\max}$ to lower temperatures.

In Fig. 8 the resistivity of Au–0.10-at.% Mn at zero pressure is shown before and after subtracting the phonon resistivity of the Au host. The presence of large deviations from Matthiessen's rule is indicated by the continued presence of the

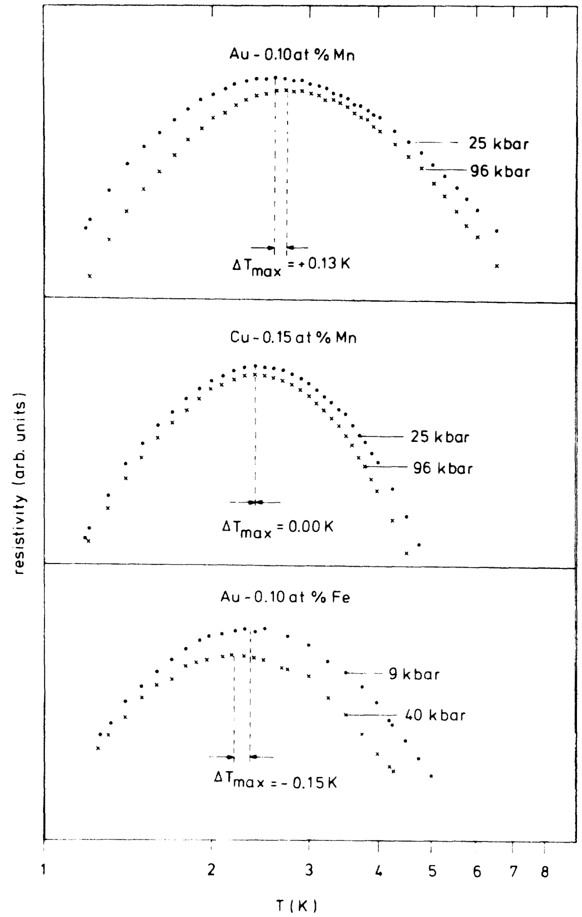

FIG. 6. Expanded plot near the resistivity maximum of the data shown in Figs. 3–5 for the lowest and highest pressures on each sample. The corresponding shifts in $T_{\max}$ with pressure are clearly visible.

resistivity minimum. This result is similar to that found on the Kondo alloy Cu–110-ppm Fe where these deviations were shown to be actually larger than the phonon resistivity itself and to have initially the same temperature dependence.[2] These and similar results found by Whall *et al.*[28] indicate that to account roughly for the phonon scattering and deviations from Matthiessen's rule one should subtract from the measured resistivity an amount equal to about 2.2 ρ_{phonon}. This subtraction is also shown in Fig. 8 and is seen to indeed affect the temperature dependence of the resistivity near $T_{\min}$ but is of negligible importance at temperatures below 6 K. This result also holds at high pressures where the phonon scattering decreases in importance. Similar results are found for the other systems investigated here. Clearly then, the values of $T_{\max}$ determined in these studies are a property solely of the spin-scattering resistivity.

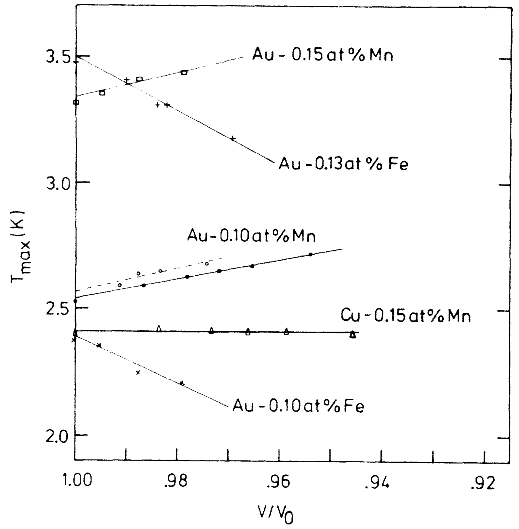

FIG. 7. Temperature of the resistivity maximum for the data shown in Figs. 3–6 as a function of the relative volume V/V_0. (●) Au–0.10-at.%Mn; (Δ) Cu–0.15-at.% Mn; (×) Au–0.10-at.% Fe. Also shown are additional results on (○) Au–0.10-at.% Mn which are shifted by about 30 mK due to a slight concentration difference; (□) Au–0.15-at.% Mn; and the (+) Au–0.13-at.% Fe data from Ref. 20.

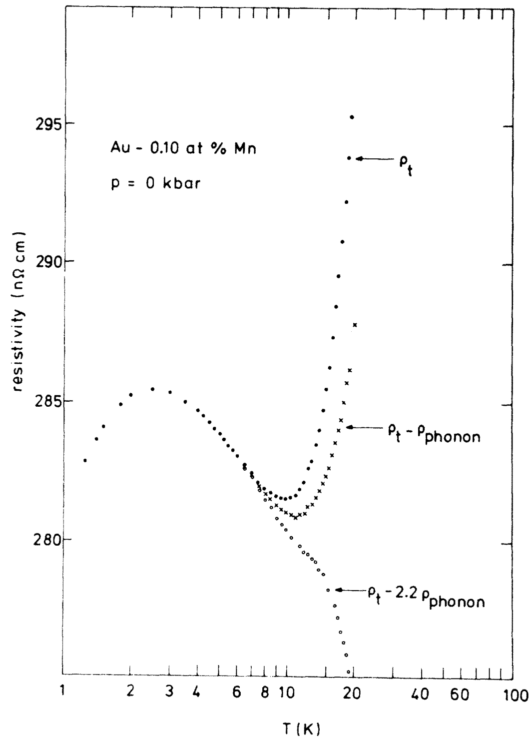

FIG. 8. Zero-pressure resistivity data for Au–0.10-at.% Mn showing the effect of subtracting out from the measured resistivity ρ_t both the simple phonon resistivity ρ_{phonon} and the quantity 2.2 ρ_{phonon}. The large contributions from deviations from Matthiessen's rule are clearly visible above 8 K.

In addition to the $T_{\max}$ shift, a number of the other features of the data are pressure dependent. The slope of the spin-scattering resistivity on the high-temperature side of the maximum is seen to increase in all data with pressure, a result which inspection of Fig. 1 reveals should be related to the observed increase at a given temperature of the slope of ρ_{Kondo} with pressure in very dilute Kondo alloys[3] as pressure shifts T_K to higher temperatures. This relationship is confirmed by experiment where the pressure dependence of the slopes of Kondo and spin-glass alloys is the same to within 20%. The derivative of the logarithm of the slope with respect to pressure has roughly the value +0.45/100 kbar. In spin-glass alloys the slope clearly has some influence on the value of $T_{\max}$ which is, therefore, not only a function of T_0 but also of T_K. Other features common to all data are the increase with pressure of the temperature of the resistivity minimum $T_{\min}$, the peak to minimum height $R(T_{\max}) - R(T_{\min})$, and the value of the spin-scattering resistivity at the maximum $R(T_{\max})$. $T_{\min}$, which occurs at that temperature where the magnitudes of the slopes of the spin and phonon resistivity are equal, increases with pressure due to both the increase of the slope of the spin resistivity and the decrease of the phonon scattering of the host.[2] The increase of the peak to minimum height comes from the increase of $R(T_{\max})$ as well as the shifting of $T_{\min}$ to higher temperatures which reveals a greater portion of the spin resistivity curve. The quantities $R(T_{\max})$ and $d^2R/d(\ln T)^2 |_{T_{\max}}$ are independent of the host phonon scattering and are solely functions of the impurity scattering. Their pressure dependence can be understood in the context of the theory presented in Sec. IV.

The data were taken in the order presented in Figs. 3–5 from top to bottom. The reversibility of the data on the Au:Mn and Cu:Mn samples was examined by unloading the pressure cell from 96 to 55 kbar. Those features of the data which are independent of the length-to-area ratio of the sample, $T_{\max}$ and $T_{\min}$, are seen to be fully reversible. Other features which are dependent on this ratio [$R(T_{\max})$, slope at 6K, $R(T_{\max})-R(T_{\min})$] proved to be reversible to within 5%, the difference arising from a 5% increase in the length-to-area ratio due to expansion of the pressure cell upon unloading. Similar results were found in previous measurements on a Au–0.13-at.%-Fe alloy.[20]

IV. COMPARISON WITH THEORY

The strikingly different behavior of the pressure dependence of $T_{\max}$ for Au:Mn, Au:Fe, and Cu:Mn systems, which is presumably related to the proximity of T_K and Δ_{RKKY}, cannot be quantitatively understood using existing theories of the resistivity maximum and thus points to the necessity of developing a theory which goes beyond the limits of the second Born approximation in the Kondo effect. A theoretical description of resistivity maxima based on the "noise" model has recently been developed by one of us [16](U.L.) (see preceding paper) in which the single impurity s-d exchange scattering is treated in the parquet approximation and interactions between different impurities are viewed as giving rise to an additional spin-flip transition rate Δ_c. A number of the features of this theory, for example the concentration dependence of $T_{\max}$, have been shown to be in good agreement with experiment. We use this theory, which is valid for $T \gg T_K$, to analyze our data and, in particular, we wish to show that it is capable of accounting for the observed pressure dependences of $T_{\max}$ described in Sec. III.

According to the theory[16] a relation exists between $T_{\max}$, Δ_c, and T_K in the form $x_0 = x_0(d)$,

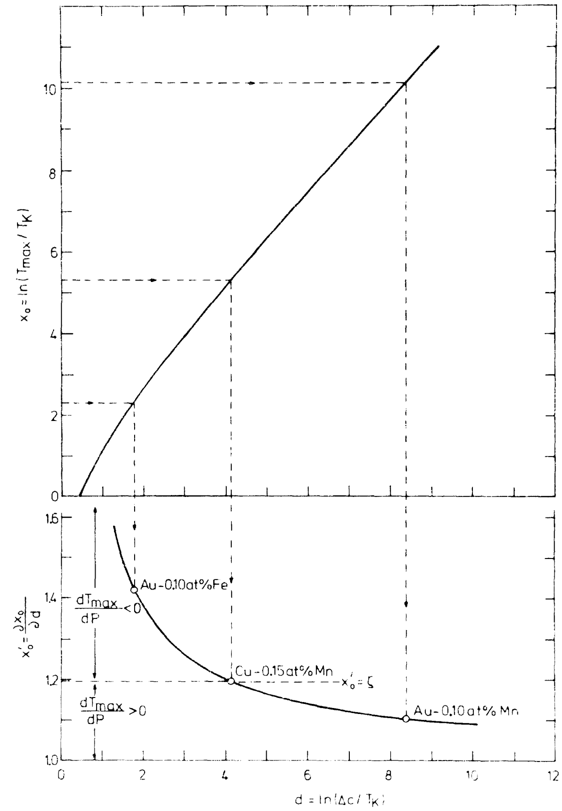

FIG. 9. Interrelationship of $T_{\max}$, Δ_c, and T_K as a function of the variables $x_0 = \ln(T_{\max}/T_K)$ and $d = \ln(\Delta_c/T_K)$. Also shown is $x_0' = \partial x_0/\partial d$. For a given system values of $T_{\max}$ and T_K define x_0 and d, and give a point on the curve $x_0'(d)$. The pressure dependence of $T_{\max}$ is determined by the relative position of this point to the line $x_0' = \zeta$, where ζ is defined by Eq. (8).

where $x_0 = \ln(T_{\max}/T_K)$ and $d = \ln(\Delta_c/T_K)$. For $d > 0.75$ or $x_0 > 0.65$ this relation is given by

$$d = x_0 + \ln\left(\frac{2(R_2 + R_1 x_0 + x_0^2)}{i_0(R_5 + R_4 x_0 + R_3 x_0^2 + x_0^3)}\right), \tag{4}$$

where $i_0 = 1.0855$, $R_1 = 9.527$, $R_2 = 20.792$, $R_3 = 10.000$, $R_4 = 25.375$, and $R_5 = 24.610$. This relation is illustrated in Fig. 9 along with the first derivative $x_0' = \partial x_0/\partial d$. An important property of this relation is that it is in general independent of the spin S of the impurity, the potential scattering at the impurity, the magnitude of the unitarity limit, and the temperature-independent defect scattering. Within the noise model, therefore, $T_{\max}$ depends only on the two "relevant" parameters of the system: the single-impurity Kondo temperature T_K and the interaction strength Δ_c. In the present experiments, the pressure dependence of the temperature of the resistivity maximum $T_{\max}(P)$ is determined. Using the above theory in the limit $\Delta_c = 0$, one may extract the pressure dependence of the Kondo temperatures $T_K(P)$ from measurements on very-low-concentration alloys. Given then $T_{\max}(P)$ and $T_K(P)$, one can use Eq. (4) to derive the pressure dependence of Δ_c.

A. Qualitative comparison

Before commencing with a quantitative comparison of experiment and theory, let us first attempt to account for the pressure dependence of $T_{\max}$ in a qualitative way. Consider small variations in T_K, $T_{\max}$, and Δ_c due to an increase in pressure:

$$\gamma = \delta \ln T_K, \quad \mu = \delta \ln T_{\max}, \quad \sigma = \delta \ln \Delta_c, \tag{5}$$

when $P \to P + \delta P$. From the relation $x_0 = x_0(d)$ one then gets

$$\delta x_0 = x_0' \delta d, \tag{6}$$

where $x_0' = \partial x_0(d)/\partial d$. This can be easily rewritten in the form

$$\mu = (\gamma - \sigma)(\xi - x_0'), \tag{7}$$

where

$$\xi = \gamma/(\gamma - \sigma). \tag{8}$$

From a comparison of Eqs. (1) and (2), it is reasonable to assume that T_K depends more strongly on the effective exchange parameter J, whose pressure variation is considered the source of the pressure effects we measure, than does Δ_c, implying $|\gamma| > |\sigma|$. This assumption will be confirmed below in the quantitative analysis. As one finds for these alloys that T_K increases with pressure,[3] one has $\gamma > 0$, and hence $\gamma - \sigma > 0$ and $\xi > 0$. From Eq. (7) it is clear that the direction of the pressure shift of $T_{\max}$, which is given by the sign of μ, is determined solely by the relative magnitudes of ξ and x_0', specifically,

$$\frac{dT_{\max}}{dP} < 0 \quad \text{for } x_0' > \xi, \qquad \frac{dT_{\max}}{dP} = 0 \quad \text{for } x_0' = \xi, \qquad \frac{dT_{\max}}{dP} > 0 \quad \text{for } x_0' < \xi. \tag{9}$$

Let us now apply the above considerations to the alloy systems studied in the present work. Using the values of $T_{\max}$ and T_K for zero pressure given in Table I, one can derive the initial values of x_0 and then (cf. Fig. 9) the initial value of d for each alloy. This results in a point on the curve $x_0'(d)$ vs d. Because for Cu:Mn we have from experiment that $dT_{\max}/dP = 0$, then from Eq. (9) we have that $x_0' = \xi$, which defines a horizontal line in Fig. 9. It is interesting to note that the points on the curve $x_0'(d)$ representing Au:Fe and Au:Mn lie, respectively, above and below the line $x_0' = \xi$. This would imply from Eqs. (7) and (9) that for Au:Fe, $dT_{\max}/dP < 0$ and for Au:Mn, $dT_{\max}/dP > 0$ as observed in experiment. From Fig. 9 it can also be estimated that for Au:Fe the quantity $|x_0' - \xi|$ is roughly twice as large as for Au:Mn, implying from Eqs. (5) and (7) that $T_{\max}$ shifts twice as fast with pressure for Au:Fe as for Au:Mn, a result evident in the experimental results shown in Figs. 6 and 7. Here the assumption is made that approximately the same value of ξ is appropriate for all three systems, an assumption which will be corroborated by the quantitative analysis considered below. This simple result is only possible with the given order of T_K values: $T_K(\text{Au:Fe}) > T_K(\text{Cu:Mn}) > T_K(\text{Au:Mn})$. If the reversed order of T_K values were postulated, the observed pressure dependence of $T_{\max}$ could only be explained by assigning widely different values of ξ to each alloy. Since $\xi > 1$, it can be seen from Eq. (8) that $\sigma > 0$, which implies that in these measurements Δ_c always increases with pressure. This is also a reasonable result in view of the proportionality $\Delta_c \propto J^2$ from Eq. (2) and the known increase of $|J|$ with pressure.[3] If $\sigma < 0$ for the above systems, then one would have $dT_{\max}/dP < 0$ in all cases, contrary to experiment.

From Fig. 9 it can also be seen that for a given impurity concentration, alloys having relatively high values of T_K (i.e., low x_0 and large x_0') will tend to have $\mu < 0$, and thus, $T_{\max}$ values which shift to lower temperatures with increasing pressure, as is the case with Au:Fe. Similarly, in a given alloy, as pressure increases, the increase in T_K dominates in d, which thus decreases. It then follows that $dT_{\max}/dP$, irrespective of its

TABLE I. Summary of results for the alloy systems studied here. Zero-pressure parameters are shown on the left-hand side and high-pressure parameters on the right-hand side. Symbols are defined in the text.

Alloy	$T_{\max}$ (K) [h]	T_K (K) [i]	J (eV) [j]	Δ_c (K) [k]	Δ_{RKKY} [l]	μ_1 [m]	γ_1 [n]	σ_1 [o]	β_1 [p]	ζ [q]
Au–0.10-at.% Mn [a]	2.54	10^{-4}	0.157	0.433	0.455	1.52	47.7	7.9	5.5	1.20
Au–0.10-at.% Mn [b]	2.57	10^{-4}	0.157	0.444	0.455	1.82	47.7	7.3	5.5	1.18
Au–0.15-at.% Mn [c]	3.34	10^{-4}	0.157	0.567	0.683	1.47	47.7	6.5	5.5	1.16
Cu–0.15-at.% Mn [d]	2.42	0.012	0.213	0.721	0.987	−0.15	33.3	8.6	5.5	1.35
Au–0.10-at.% Fe [e]	2.39	0.24	0.251	1.425	0.798	−3.84	17.7	3.0	3.1	1.20
Au–0.13-at.% Fe [f]	3.50	0.24	0.251	1.866	1.037	−3.05	17.7	3.2	3.1	1.22
Au–0.13-at.% Fe [g]	3.53	0.24	0.251	1.883	1.037	−2.46	17.7	3.5	3.1	1.25

[a] Data in Figs. 3 and 7 (●).
[b] Data in Fig. 7 (○).
[c] Data in Fig. 7 (□).
[d] Data in Figs. 4 and 7 (△).
[e] Data in Figs. 5 and 7 (×).
[f] Data in Fig. 7 (+), see Ref. 20.
[g] Data at 22 and 40 kbar.
[h] Linear regression intercepts of $T_{\max}(P)$ data.
[i] For Au:Mn see Ref. 15, for Cu:Mn see Ref. 1, for Au:Fe see Ref. 29.
[j] From Eq. (1) using $D = 10^5$ K and $n = 0.294$ states/atom for Cu and $n = 0.308$ states/atom for Au.
[k] From Eq. (4).
[l] From Eq. (2) using $S = \frac{5}{2}$ for Mn and $S = 2$ for Fe.
[m] Defined in Eq. (12).
[n] Defined in Eq. (10).
[o] Defined in Eq. (13).
[p] Defined in Eq. (11).
[q] $\zeta = (1 - \sigma_1/\gamma_1)^{-1}$.

sign at $P = 0$, will *decrease* with increasing pressure. This universal downward curvature in $T_{\max}(P)$ should become apparent if the present pressure range were expanded appreciably. In the same way, for a given system, one would expect that $dT_{\max}/dP$ should always *decrease* as the impurity concentration, and therefore Δ_c, decreases. This is confirmed in the present experiment where it is observed that pressure shifts $T_{\max}$ more rapidly to lower temperatures in Au–0.10-at.% Fe than in Au–0.13-at.% Fe.

In this qualitative analysis we have thus shown it to be plausible that the sign and magnitude of $dT_{\max}/dP$ in the systems studied is a natural consequence of both increasing T_K and Δ_c ($\gamma > 0, \sigma > 0$) in *all* alloys and depends on the relative magnitudes of Δ_c and T_K, i.e., on the relative strength of the spin-glass interaction and the Kondo effect.

B. Quantitative comparison

If the pressure dependence of both the temperature of the resistivity maximum $T_{\max}(P)$ and the Kondo temperature $T_K(P)$ are known, it is possible, using Eq. (4) to calculate directly $\Delta_c(P)$, the pressure dependence of the average interimpurity interaction strength. In recent measurements at pressures to 80 kbar by Crone *et al.*[3] on very dilute Au:Fe, Au:Mn, and Cu:Mn alloys, it was found that T_K always increases with pressure and that its volume dependence could be empirically represented by a linear relation

$$\ln T_K(v) \simeq \ln T_K(0) + \gamma_1 v \,, \qquad (10)$$

where $v = (V_0 - V)/V_0$ is the relative volume decrease which in the present systems is ~5% for $P \simeq 80$ kbar. A least-squares fit to these data yields the values of γ_1 tabulated in Table I which also lists the $T_K(0)$ values assumed. In addition, this table includes values of the parameter β_1 which is defined by the expression

$$J^2(v) \simeq J^2(0)(1 + \beta_1 v) \,, \qquad (11)$$

where $J(P)$ is derived from $T_K(P)$ using Eq. (1). Note that β_1 also gives the pressure dependence of Δ_{RKKY} since $cS(S+1)/E_F$ is essentially pressure independent. As seen in Fig. 7, $T_{\max}(P)$ approximately obeys a linear relationship

$$T_{\max}(v) \simeq T_{\max}(0)(1 + \mu_1 v) \,. \qquad (12)$$

Calculating values of $\Delta_c(P)$ using Eq. (4) and fitting them to a linear expression

$$\Delta_c(v) \simeq \Delta_c(0)(1 + \sigma_1 v) \qquad (13)$$

results in the values of the parameters β_1, μ_1, and σ_1 given in Table I. It is seen that the interaction parameter Δ_c increases with pressure and the parameter $\zeta = \gamma_1/(\gamma_1 - \sigma_1)$ changes little for all systems studied, in agreement with the results of the qualitative analysis.

We now investigate the effect of a small concentration change on the pressure dependence of $T_{\max}$. From Eqs. (7) and (8) one readily derives that

$$\mu_1 \simeq \sigma_1 x_0' + \gamma_1 (1 - x_0') . \tag{14}$$

Making a small change in the concentration δc one obtains, assuming that $\delta\sigma_1 = 0$ and using the fact that $\delta\gamma_1 = 0$,

$$\delta\mu_1 = (\sigma_1 - \gamma_1)\delta x_0' . \tag{15}$$

Since $\gamma_1 > \sigma_1$, and Δ_c and d increase with concentration, one has $\delta x_0' < 0$ for $\delta c > 0$ (see Fig. 9 or expand $x_0' = 1 + 1/d + \cdots$) and thus $\delta\mu_1 > 0$ in all cases. It, therefore, follows that, as was discussed in the qualitative analysis, μ_1 should always *increase* in value as the concentration increases. This prediction of the theory is born out by the few data points shown in Table I. Estimating the magnitude of the effect, one has

$$\delta x_0' \simeq -\left(\frac{1}{d^2}\right)\left(\frac{\partial d}{\partial \ln c}\right)(\delta \ln c) \simeq -\frac{(\delta \ln c)}{d^2} \tag{16}$$

assuming $\Delta_c \propto c$. Using numbers for the geometric mean of the two concentrations, we find for Au:Mn (0.10 at.% → 0.15 at.%), $\delta\mu_1 \simeq +0.2$ and for Au:Fe (0.10 at.% → 0.13 at.%), $\delta\mu_1 \simeq +1.0$. These numbers are within the statistical errors in μ_1 which are the order of 0.4. For the Cu:Mn system, where nominally $c = 0.15$ at.%, no significant pressure dependence in $T_{\max}$ could be observed; from the above it is clear that, at a sufficiently high concentration, $T_{\max}$ should increase with pressure. Using Eq. (15) of the present paper and Eq. (78) of Ref. 16 it is possible to make the prediction that if the Mn concentration is increased such that $T_{\max}$ increases from 2.4 to 9.0 K, then $\delta\mu_1 \simeq +0.94$ and $T_{\max}$ should increase with pressure at the rate of approximately (0.4 K)/(100 kbar). This experiment is in progress.

The present analysis has concentrated on the quantity $T_{\max}$ because it is a function solely of the two basic system parameters Δ_c and T_K and thus allows the most unequivocal comparison between experiment and theory. An extension of this comparison to the entire temperature range over which ρ_{spin} is measured is the subject of future study. First results indicate that, for $\gamma > \sigma$ [see Eq. (5)], the quantities $R(T_{\max})$ and $d^2R/d(\ln T)^2|_{T_{\max}}$ increase with pressure for all systems, in agreement with experiment.

V. DISCUSSION

A. Resistivity maximum

The temperature of the resistivity maximum $T_{\max}$ has assumed particular importance in this work not only because it is experimentally a well-defined quantity, but also because physically it represents a balance between the two basic system parameters, T_K, the Kondo temperature, and Δ_c, the average RKKY-interaction strength. Furthermore, $T_{\max}$ is independent of the impurity spin S, the impurity potential scattering, and the temperature-independent defect scattering.[16] An expression for $T_{\max}$ based on simple relations like $kT_{\max} \simeq \Delta_c \propto c J^2$, has been shown to be unable to account for the pressure dependence of $T_{\max}$ observed in these experiments. A recent theory by one of us[16] (U.L.), correct to infinite order in perturbation theory, emphasizes the importance of taking the Kondo effect properly into account in order to understand resistivity maxima in spin glasses and shows that there is no simple relation such as the above between $T_{\max}$ and Δ_c which does not also involve T_K. Using this analysis it is possible, given the pressure dependence of T_K and $T_{\max}$, to derive values of the basic interaction parameter $\Delta_c(P)$ which are given in Table I. Within this theory the strikingly different behavior of $T_{\max}$ with pressure observed for the three systems studied here is shown to depend in a simple and natural way on the relative values of Δ_c and T_K and their monotonic increase with pressure. This is brought out clearly in Fig. 10 where $T_{\max}$ is plotted as a function of $|J|$ at fixed impurity concentration. Increasing the concentration shifts the $T_{\max}$ curve to the right. The nonproportionality of $T_{\max}$ and Δ_c is very apparent. Systems where $\Delta_c/T_K \gg 1$, such as Au-0.10-at.% Mn, will tend to have $dT_{\max}/dP > 0$, whereas for systems with smaller values of Δ_c/T_K, such as Au-0.10-at.% Fe, $dT_{\max}/dP$ can become negative in sign. This anomalous behavior is generally expected as T_K and Δ_c approach each other in value whether pressure is used to increase $|J|$ and thus move T_K towards Δ_c or concentration is decreased to lower Δ_c towards T_K. $T_{\max}(0)$ for Cu-0.15-at.% Mn lies on top of the $T_{\max}$ "hill" and should eventually decrease if pressures much higher than 100 kbar are applied. If the Mn concentration is increased, thus shifting the $T_{\max}$ curve to the right, it would be expected that pressure would shift $T_{\max}$ immediately to higher temperatures, in agreement with the prediction at the end of Sec. IV. In fact, it is possible using Fig. 10 to understand in a qualitative way all the complicated dependences of $T_{\max}$ on pressure at different concentrations found in these experiments.

Even though the present theory is no longer applicable when T_K is near Δ_c, one may speculate that the formal result of this theory, i.e., $T_{\max} \to 0$ as $\Delta_c \to T_K^+$, will continue to hold in a more sophisticated calculation. The interesting region

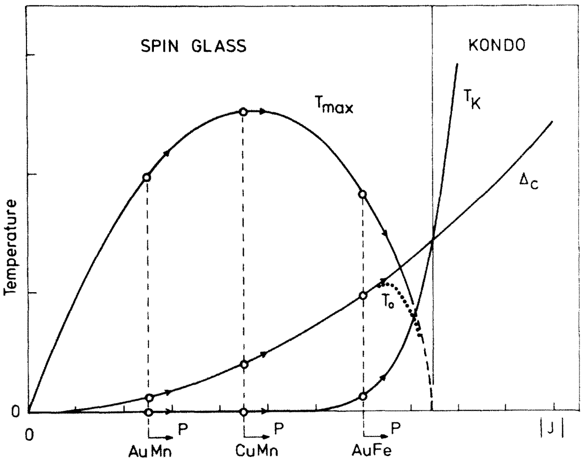

FIG. 10. Expected functional dependence of T_{max}, Δ_c, T_K, from theory and conjectured dependence of T_0 on the effective exchange parameter J at fixed impurity concentration in both the spin-glass $\Delta_c \gg T_K$ and Kondo $\Delta_c \ll T_K$ regimes. Behavior of T_{max} with pressure for Au: Mn, Cu: Mn, and Au: Fe agrees with the present experimental results. Increasing concentration increases T_{max} and Δ_c and shifts the T_{max} curve to the right. Curves shown for T_K and T_{max} are calculated from Eqs. (1) and (4), respectively, assuming $\Delta_c \propto J^2$. For $\Delta_c \simeq T_K$, the T_{max} and T_0 curves have at most qualitative significance.

$\Delta_c \simeq T_K$, where spin fluctuation and interaction effects are comparable in magnitude, requires a full self-consistent calculation and is the subject of future study.

The results of the present investigation are similar to those of Star[18] for the alloy series $Au_{1-x}Cu_x$-0.15-at.% Fe where T_{max} was observed to shift rapidly to lower temperatures with increasing Cu concentration. In previous studies on very dilute alloys, Loram *et al.*[29] showed that T_K increases as Cu is added to Au:Fe. The present results thus indicate that the negative T_{max} shift observed by Star is a consequence of the T_K increase and not of a weakening of the RKKY interaction by Cu impurity scattering.[30] Across the above alloy series, T_K increases monotonically from 0.24 K for Au:Fe to about 24 K for Cu:Fe,[29] and thus, for the above Fe concentration where $\Delta_c \simeq 2$ K, one moves from a region where $\Delta_c > T_K$ to one where $\Delta_c < T_K$. The apparent disappearance of the resistivity maximum for $T_K > \Delta_c$ is known from other systems, e.g., Au:V, where $T_K \simeq 300$ K and for magnetic impurity concentrations near or above those used in the above systems ($c \simeq 0.1$ at.%), no resistivity maximum is observed.[31] The appearance of a resistivity maximum is indicative that magnetic ordering phenomena are dominating the dynamics of the impurity spin. However, if the concentration of the magnetic impurity is low enough that $\Delta_c < T_K$, Kondo spin fluctuations dominate the spin dynamics and magnetic ordering of the spins is prevented. The relatively weak interactions between the spins in such a system act to disturb the formation of the delicately balanced many-body Kondo resonance. This is seen in measurements on[32] Cu:Cr ($T_K \simeq 1$ K) where increasing the Cr concentration to 600 ppm reduces the unitarity limit value $R(T=0\text{ K})$, with no resistivity maximum being observed down to a temperature of 60 mK. A similar reduction of $R(T=0\text{ K})$ has been observed on a number of systems upon application of an external magnetic field,[33] in agreement with theory.[34]

B. Interaction parameter Δ_c

One of the more important results of the present analysis is the information gained about the interaction parameter Δ_c which is summarized in Table I. This parameter presumably represents the average strength of the indirect RKKY interactions which lead to a random locking in of the impurity spins at temperatures below the spin-glass freezing temperature T_0. To check this, it would be interesting to compare Δ_c, which is estimated using the present theory from the parameters T_K and T_{max}, to the mean RKKY-interaction energy Δ_{RKKY} given by Eq. (2). The values of the exchange parameter J are extracted from measurements on very dilute alloys where interaction effects are unimportant. In Table I the values of Δ_c and Δ_{RKKY} as well as their respective pressure dependences σ_1 and β_1 are seen to agree in all cases to within a factor of 2. This good agreement not only supports the correctness of the present theory and analysis but also affirms the possibility of using parameters derived from measurements on very dilute alloys to yield information about interaction phenomena in more concentrated systems.

The values of the parameters σ_1 and β_1 depend to some extent on the choice of the Kondo temperature at zero pressure $T_K(0)$. On the other hand, if we *assume* that $\Delta_c \propto \Delta_{RKKY} \propto J^2$ holds *exactly*, then $\sigma_1 \equiv \beta_1$ and $T_K(0)$ can, in fact, be estimated. This procedure yields $T_K(0) \simeq 0.2$ K for Au:Fe, in agreement with the value given in Table I. For Au:Mn one finds $T_K(0) \simeq 10^{-11}$ K, a value which differs from that in Table I, but is in rough agreement with with the low value $T_K(0) \simeq 10^{-13}$ K quoted by Loram *et al.*[29] This procedure for determining $T_K(0)$ could, in principle, be applied to other low T_K systems.

Deviations from $\Delta_c \simeq \Delta_{RKKY}$ would be expected if the present studies were extended to higher concentrations where the molecular-field scaling regime no longer applies and Eq. (2) is inappropriate as an estimate of Δ_c. From such studies one could conceivably gain information about the spatial dependence of both the RKKY oscillations and J. Chemical clustering would of course complicate such studies tremendously. A further complication is that 3-d transition-metal nearest-neighbor impurities certainly have direct d-d exchange interactions which will contribute to Δ_c and lead to deviations from the proportionality $\Delta_c \propto cJ^2$. Indeed, as is discussed in more detail below, the dependence of the freezing temperature T_0 on concentration can be quite complicated.

C. Spin-glass freezing temperature T_0

Although unfortunately no simple relationship exists between T_0 and $T_{\max}$, from the theory of Adkins and Rivier,[35] kT_0 and Δ_c would be expected for $T_k \ll \Delta_c$ to be approximately equal and to have, therefore, the same concentration and pressure dependence. It must, however, be emphasized that in that theory, as well as in any other theory of spin-glass alloys known to the authors, the effect on the freezing of Kondo spin fluctuations was not taken into account. T_0 can be looked upon as that temperature where thermal fluctuations and spin fluctuations just balance the interaction strength Δ_c. As T_K and Δ_c become comparable in magnitude, spin fluctuation effects should become important and T_0 is expected to fall rapidly to $T \simeq 0$ K. This behavior of T_0 is illustrated in Fig. 10. A suitable theory in this region is still lacking and requires a more sophisticated treatment of the Kondo effect than was given by Larsen.[16] Such spin fluctuation effects would be, considering the systems studied here, most important for Au-0.10-at.% Fe which has the highest T_K value. Whether or not the calculated increase of Δ_c with pressure in this alloy is also accompanied by a similar increase in T_0, or by a decrease as with $T_{\max}$, is an unresolved question which is currently a subject of active experimental investigation.

Consider the idealized case of a dilute spin-glass system where the uniformly distributed magnetic impurities interact solely by the indirect RKKY interaction and all impurities have the same magnitude (not necessarily same direction) of interaction with the magnetic environment. For such a homogeneous magnetic system, no magnetic ordering should be possible at temperatures below T_K. In actual alloy systems such an ideally uniform impurity distribution is not realizeable and clearly the presence of correlated pairs, triplets, etc., of impurities[36] will complicate the "intrinsic" concentration or pressure dependence of T_0 and $T_{\max}$. An impurity pair has a much lower Kondo temperature T_{Kp} than an isolated impurity and thus can take part in ordering phenomena even in the temperature range $T_{Kp} < T_0 < T_K$, where isolated impurities are "compensated" and cannot interact. In this concentration range one observes an ordering temperature $T_{op} \propto c^2$, given by a clear maximum in the susceptibility characteristic of pairs of impurities, with apparently little influence from single impurities. On the other hand, the contribution of impurity pairs to the resistivity is negligible compared to that from isolated impurities until one reaches a temperature near T_{Kp}. If the great majority of impurities are single impurities, the resistivity maxima from impurity pairs ordering will be extremely weak compared to the very large Kondo resistivity of the single impurities which reaches unitarity limit values. It is, therefore, readily understandable why susceptibility peaks are observed in some alloys where no resistivity maximum could be measured; an example is the system Cu-0.1-at.% Fe,[18,37] where the formation of pairs and even clusters is believed to be especially prominent. Whereas here the resistivity reflects the behavior of the dominant impurity species, the susceptibility focuses on the impurity type which orders, even if it be a small minority. That the majority of impurities need not take part in the so-called spin-glass freezing process is suggested from measurements of the specific heat through T_0.[38] Because the susceptibility can be quite sensitive to the spin dynamics of a minority impurity type, care must be taken not to assume the pressure and concentration dependence of T_0 to be characteristic of the entire impurity system. Furthermore, recent experimental results[7] show that there is no generally valid correlation between T_0 and some characteristic feature of the resistivity, such as the resistivity maximum or the inflection point. Although to date no theory exists which relates T_0, $T_{\max}$, T_K, and Δ_c to each other over a wide range of the parameters, the present theory allows one, within certain limits, to derive information about the important interaction quantity Δ_c from a knowledge of $T_{\max}$ and T_K.

Note added in proof. Subsequent measurements on Cu-0.35-at.% Mn show that between 0 and 67 kbar $T_{\max}$ increases from 9.1 to 9.5 K, in good agreement with the prediction at the end of Sec. IV. For recent developments see the papers by the present authors in the *Proceedings of the Second International Symposium on Amorphous Magnetism* (Plenum, New York, 1976).

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