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EFFECT OF PRESSURE ON THE KONDO TEMPERATURES OF Au(Fe) AND Au(Mn)

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The electrical resistivity of four Kondo systems, Au–(5 p.p.m. Fe), Au–(39 p.p.m. Fe), Au–(17 p.p.m. Mn) and Au–(50 p.p.m. Mn) has been measured in the temperature range 1.3–20 K at pressures up to 80 kbar. The Kondo temperature T_K is found to increase initially with pressure at the rate of 1.1%/kbar for Au(Fe) and 6%/kbar for Au(Mn). The volume dependence of the effective exchange constant J_{eff} is derived.

THE HIGH PRESSURE technique has distinct advantages compared with alloying when it is essential to vary the electronic and magnetic properties of a solid in a well defined manner. In recent measurements^{1,2} of the electrical resistivity of the classic Kondo alloy Cu–(110 p.p.m. Fe) from 1.3–40 K, it is found that application of pressures as high as 82 kbar causes the Kondo temperature T_K ($T_K = T_f \exp(1/n(E_f)J_{\text{eff}})$, where T_f is the Fermi temperature, $n(E_f)$ is the density of states at the Fermi surface and J_{eff} is the effective exchange constant) to increase initially at the rate of +1.1%/kbar without a detectable change in the spin S or the potential scattering at the magnetic impurity site. The existence of a “universal” resistivity law $\rho = \rho(T/T_K)$ for Cu(Fe) was also confirmed. It would be of interest to investigate to what extent the above results for Cu(Fe) can be generalized to other very dilute magnetic alloys with different combinations of magnetic impurity and host metal. Of special importance would be alloys possessing appreciably different Kondo temperatures from Cu(Fe), thus allowing in the temperature range 1.3–20 K a study of the effect of pressure

on an entirely different section of the “universal” resistivity curve. The aim of such studies is not only to check the various theoretical descriptions of such dilute spin systems but also to contribute to the understanding of the more general question of the nature of the interaction of an isolated magnetic impurity with its surroundings.

In this paper we present high pressure resistivity measurements on four further Kondo alloys [Au–(5 p.p.m. Fe), Au–(39 p.p.m. Fe), Au–(17 p.p.m. Mn) and Au–(50 p.p.m. Mn) where the nominal impurity concentrations are given]. Comparison of our measurements with the results of Ford³ and Loram^{4,5} would imply concentrations of 7 p.p.m. Fe, 32 p.p.m. Fe, 34 p.p.m. Mn and 68 p.p.m. Mn, respectively. The Kondo temperatures of these alloys for zero pressure lie far below that of Cu(Fe) [Au(Fe), $T_{K0} \approx 0.24$ K;³ Au(Mn), $T_{K0} \approx 10^{-4}$ K;⁶ Cu(Fe), $T_{K0} \approx 24$ K].⁵ The concentrations of the above alloys have been chosen so low that interimpurity interaction effects are unimportant in our temperature range. The low temperature resistivity of the Au used in preparing the alloys (99.9999% pure from Cominco Gardner) showed a slight increase after being treated in the same way as the above dilute alloys, indicating a magnetic impurity concentration less than 0.8 p.p.m. Fe. This effect is negligible in Au(Fe) alloys but must be taken into consideration in Au(Mn) due to the fact that Fe

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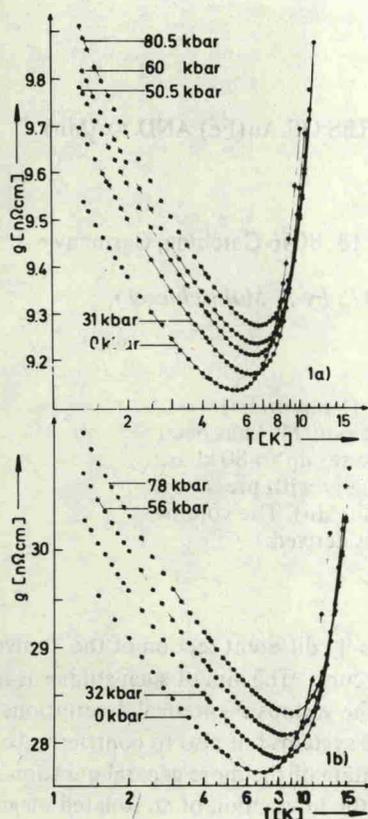


FIG. 1. Measured resistivity vs temperature. Left is spin-scattering resistivity, right is phonon resistivity. The solid lines connecting dots are to distinguish data at different pressures. (a) Au-(5 p.p.m. Fe), (b) Au-(39 p.p.m. Fe).

shows a 12 times stronger increase of the Kondo resistivity in the temperature range reported. The sample preparation and high pressure techniques have been discussed in detail in a previous publication.²

The results of our resistivity measurements at high pressures on the Au(Fe) alloys are shown in Fig. 1. At a given pressure the competition between the spin-scattering resistivity from the magnetic impurity and the phonon-scattering resistivity from the Au-lattice gives rise to a resistivity minimum at T_{\min} . Application of pressure is seen for both alloys to shift the spin-scattering curve to higher temperatures and to increase T_{\min} , the step height $\rho(T = 1.5 \text{ K}) - \rho(T_{\min})$, and the slope $|d\rho_{\text{spin}}/d \log T|$.

In these measurements it was not possible to

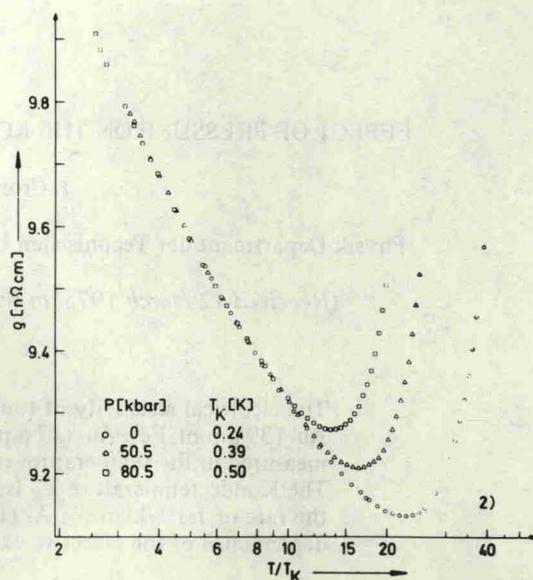


FIG. 2. Resistivity of Au-(5 p.p.m. Fe) vs reduced temperature T/T_K for several pressures.

check for the existence of a "universal" resistivity law $\rho = \rho(T/T_K)$ as was done for Cu(Fe). Rather, it was necessary to assume the existence of this law and the pressure independence of S and the potential scattering in order to be able to determine the shifts of T_K with pressure. The relative vertical position of the curves in Fig. 1 to each other was determined by requiring that all spin-scattering curves overlap accurately when shifted in temperature on a $\log T$ plot.⁷ Using this procedure one derives a relative initial increase of T_K with pressure of $\delta = (\Delta T_K/T_{K0})/\Delta p \approx +1.1\%/kbar$ for the Au(Fe) alloys. In Fig. 2 plots of the resistivity of Au-(5 p.p.m. Fe) are shown as a function of the reduced temperature T/T_K and are seen to overlap within the experimental accuracy over the entire range $T < T_{\min}$. For $T \geq T_{\min}$, the phonon-scattering and deviations from Matthiessen's rule become large and prevent any possibility of overlap. The determination of δ using the above procedure is accurate to roughly $\pm 20\%$.

An independent method of estimating the change of T_K consists in measuring the pressure dependence of the slope of the spin-resistivity curve. Since for $T \geq T_K$, $|d\rho_{\text{spin}}/d \log T| \propto |J_{\text{eff}}^3|$,⁸ the observed increase in slope seen in Fig. 1 with pressure implies

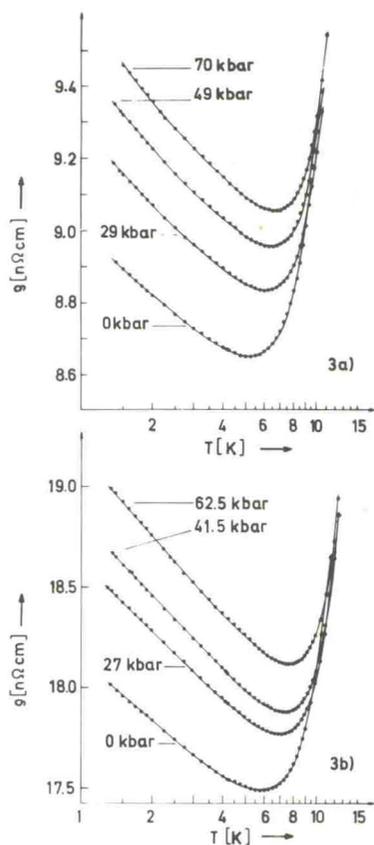


FIG. 3. Measured resistivity vs temperature. Left is spin-scattering resistivity, right is phonon resistivity. The solid lines connecting dots are to distinguish data at different pressures. (a) Au-(17 p.p.m. Mn), (b) Au-(50 p.p.m. Mn).

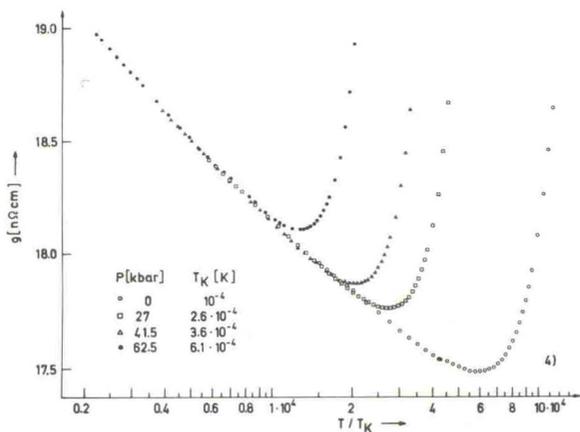


FIG. 4. Resistivity of Au-(50 p.p.m. Mn) vs reduced temperature T/T_K for several pressures.

that $|J_{\text{eff}}|$ and, therefore, T_K also increase by an amount of $\delta \approx +1.1\%/kbar$ in good agreement with the value determined above. The observed increase in slope with pressure in these measurements was found to be reversible in pressure and is thus not a spurious effect but clearly due to an increase in T_K .

The results of measurements on two Au(Mn) alloys are shown in Fig. 3. Although these results are qualitatively similar to those on Au(Fe), due to the very small value of the slope $|d\rho_{\text{spin}}/d \log T|$ for Au(Mn), the resistivity is only for $T \leq 3$ K completely free of phonon scattering, and also the scatter in the data becomes relatively more important. For these reasons, it is not possible to determine δ accurately by using the overlap method (Fig. 2). From the observed increase in slope with pressure, however, one obtains $\delta \approx +6\%/kbar$. Using this value of δ , the resistivity curves for Au-(50 p.p.m. Mn) are plotted in Fig. 4 vs T/T_K , and they are seen to overlap satisfactorily.

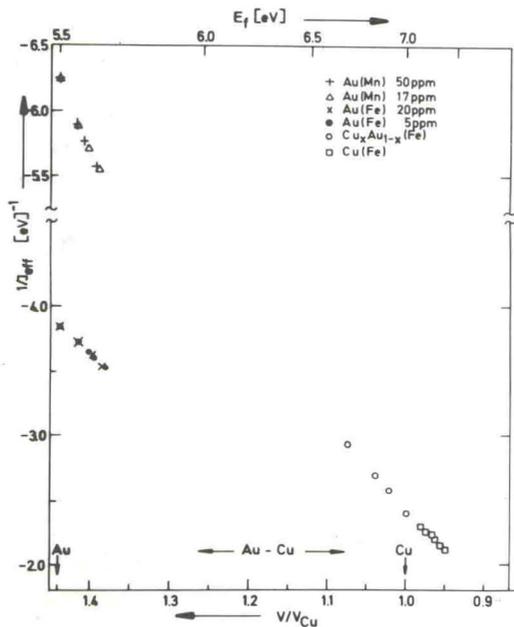


FIG. 5. Volume dependence of the inverse effective exchange constant $1/J_{\text{eff}}$ relative to the atomic volume of Cu at $p = 0$. Au(Fe) and Au(Mn) our data, open circles from Cu-Au(Fe) alloy data by Loram *et al.*⁵ squares from our Cu(Fe) high pressure data.² Values of the Fermi energy E_f are those of a free electron gas.

Since T_{\min} occurs at the temperature where the slope of the spin-scattering and phonon resistivity are equal in magnitude, T_{\min} increases with pressure due to both the observed increase in the slope of ρ_{spin} and the decrease in ρ_{phonon} with increasing pressure. The reduction in ρ_{phonon} is due to an increase in the Debye temperature of the host alloy with pressure.^{2,9} An estimate of the T_{\min} shift is in good agreement with the observed shift $(\Delta T_{\min}/T_{\min})/\Delta p \approx +0.2\%/k\text{bar}$. The $p = 0$ data shown was carried out on an annealed sample outside the pressure cell. Relative to this data the high pressure data shows a significant shift in T_{\min} to higher temperatures which is probably due to the stiffening of the lattice and raising of the Debye temperature from cold working the sample in the pressure cell.¹⁰ The increase of $\Delta\rho(T) = \rho(1.5\text{ K}) - \rho(T_{\min})$ with pressure is due essentially to the shift of ρ_{spin} to higher temperatures.

From the pressure dependence of T_K , one can extract the volume dependence of the effective exchange constant J_{eff} , as shown in Fig. 5. (The change of $n(E_f)$ with volume is small for the noble metals).² Also shown are the results of our pressure measurements on Cu(Fe) and the alloy data of Loram *et al.*⁵ on $\text{Cu}_{1-x}\text{Au}_x(\text{Fe})$. Using the Schrieffer-Wolf transformation,¹¹ one finds $\Delta(1/J_{\text{eff}})/\Delta V \approx \text{constant} \approx (S/|V_{kd}|^2)\Delta E_d/\Delta V$ where S and V_{kd} are assumed pressure independent and E_d is the energy separation of the virtual bound state from the Fermi surface,

V_{kd} being the covalent admixture matrix element. The increase of T_K with pressure found in our measurements could thus be explained in terms of a reduction of E_d with volume. However, an increase of $|V_{kd}|$ with pressure would also account for the observed T_K increase. That the results in Fig. 5 for Au(Fe) and Cu(Fe) do not lie along the same line could be due to inaccuracies in the value of T_K used for $p = 0$, or effects of local environment.

At the present time, work is being carried out on further Kondo alloys [Au(V), Au(Cr), Cu(Cr), Cu(Mn), Ag(Mn)] in an effort to ascertain to what extent the above results are of general validity. Preliminary measurements on several of these alloys indicate that also here T_K increases with pressure. Although the precise mechanism for the observed T_K increase is not known at this time, it is probable that the same mechanism is at work in all of the above systems.

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