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Effect of Pressure on the Kondo Temperature of Cu:Fe—Existence of a Universal Resistivity Curve

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The electrical resistivity of Cu–110-ppm Fe has been measured for temperatures from 1.3–40 K at pressures to 82 kbar. The Kondo temperature T_K is observed to increase with pressure, having doubled in value by 82 kbar. When plotted versus T/T_K, the spin-resistivity curves are found to accurately overlap for all measured pressures, thus confirming the existence of a universal resistivity curve $\rho = \rho(T/T_K)$ for Cu:Fe. Within the experimental uncertainty of 1.7%, the saturation value of the resistivity $\rho(T = 0\text{ K})$ does not change with pressure. This indicates that to this accuracy the spin S and the potential scattering at the magnetic impurity remain constant. From the pressure dependence of T_K one obtains the volume dependence of the effective exchange parameter J_{eff}. Approximately the same volume dependence is found for a series of CuAu:Fe alloys using their known average atomic volume. The Cu:Fe high pressure and the CuAu:Fe-alloy data are discussed within the context of a simple Fermi-gas model based on the Schrieffer-Wolff transformation. The pressure dependence of the resistivity of pure copper, ρ_{phonon}, has also been studied and can be understood using the Bloch-Grüneisen formula with known values of the compressibility and the Grüneisen parameter. A method for experimentally determining deviations Δ from Matthiessen's rule in Kondo alloys is also presented. Such deviations can be very large; in fact, for $T < 30$ K, we find that $\Delta = 1.3\rho_{\text{phonon}}$.

I. INTRODUCTION

Because of its importance to the general problem of magnetism in metals, the problem of localized magnetic states in a metallic environment has for many years been an object of considerable study. Investigations on dilute magnetic alloys have been pursued with particular intensity following Kondo's success in 1964 in explaining the resistance minimum anomaly.[1] Kondo postulated an s-d exchange interaction $\mathcal{H}_{s\text{-}d} = -J_{\text{eff}}\,\vec{s}\cdot\vec{S}$ between conduction electrons with spin $\vec{s}$ and magnetic impurities with spin $\vec{S}$ and calculated the scattering to third order in perturbation theory. For antiferromagnetic (negative) values of the effective exchange parameter J_{eff}, the resultant resistivity is found to increase logarithmically with decreasing temperature. This is in approximate agreement with measurements on a large number of dilute magnetic alloys.[2,3] Such a logarithmic divergence is, of course, not possible for arbitrarily low temperatures, as the resistivity at $T = 0$ K cannot exceed the unitary limit.[4] This saturation effect at low temperatures is clearly visible, for example, in the resistivity of Cu:Fe and Y:Ce. That such deviations from a logarithmic dependence are to be expected is seen from the fact that for temperatures below a characteristic temperature T_K, the Kondo temperature, the perturbation series expansion for the resistivity diverges $\{T_K \simeq T_F \exp[1/J_{\text{eff}} n(E_F)]$, where T_F is the Fermi temperature and $n(E_F)$ is the density of states per atom at the Fermi surface$\}$. At temperatures below T_K, strong spin correlations mediated by the impurity spin are set up between conduction electrons near the impurity and it becomes necessary to treat the above problem using more sophisticated theoretical techniques. Fortunately, a considerable mathematical apparatus was available from many-body theory which facilitated further work on the Kondo problem.[3,5] Whereas some of the ensuing theories were only applicable for $T < T_K$ or $T > T_K$, Hamann[6] and Suhl and Wong[7] were able to derive approximate expressions for the resistivity valid at all temperatures. They find that the resistivity decreases from its unitary limit value at $T = 0$ K, proceeding with a logarithmic dependence through T_K, to a high-temperature plateau for $T \gg T_K$. The measured resistivity is indeed found to have this general behavior. However, a careful comparison between experiment and theory over a wide temperature range reveals only qualitative agreement. Despite enormous efforts, it has to date not been possible, starting with the deceptively innocuous looking *s-d* Hamiltonian, to derive an exact expression for the temperature dependence of any experimentally accessible quantity. All available expressions involve approximations whose exact implications are unclear. In short, although qualitative agreement between experiment and theory does exist in the Kondo problem, it has not as yet been possible to obtain the kind of agreement which would enable one to reliably extract the sort of detailed information on dilute alloy systems which is needed.

Under these circumstances it would be desirable to establish a set of fixed points or theorems which the exact solution to the *s-d* Hamiltonian must satisfy. This would be useful not only in helping to decide which approximations should be made in solving the Kondo problem, but also it would serve as a check on the validity of existing theories. One such fixed point was supplied by Mattis[8] who proved formally that for antiferromagnetic *s-d* exchange, the ground state ($T = 0$ K) of the magnetic impurity problem for $S = \frac{1}{2}$ is a singlet. As was pointed out by Heeger,[3] this theorem implies that the integral of the impurity-conduction-electron-spin correlation is $\int d^3r \langle \vec{S} \cdot \vec{s}(r) \rangle = -\frac{3}{4}$. Several so-called ground-state theories,[9] valid only for $T < T_K$, are found to satisfy this theorem. On the other hand, Zittartz and Müller-Hartmann[10] have demonstrated that this theorem is violated by the theories of Hamann[6] and Suhl and Wong,[7] among others. As is confirmed experimentally,[11] these latter theories are thus not expected to be reliable at temperatures much below T_K.

Another theorem which seems to be satisfied by all theoretical expressions derived[3,5] from $\mathcal{H}_{s\text{-}d}$ is that T_K merely plays the role of a scaling temperature; i.e., if f is the property in question, then $f = f(T/T_K)$. The possibility of the existence of such "universal" curves seems to have been first pointed out by Heeger.[3] Work by Anderson and Yuval[12] seems to imply that $f = f(T/T_K)$ follows rather generally from $\mathcal{H}_{s\text{-}d}$. In fact, the existence of universal curves appears to also follow from spin-fluctuation theories[13,14] based on the more general Wolff or Anderson Hamiltonians which include $\mathcal{H}_{s\text{-}d}$ as a special case. Experimentally, one could test for the existence of a universal dependence $f = f(T/T_K)$ by measuring the temperature dependence of f for different values of T_K. Resistivity measurements by Loram *et al.*[11] on a series of CuAu:Fe alloys suggest the existence of a universal dependence of ρ on T/T_K. However, it has been shown[15] that in alloys the local atomic environment of a magnetic ion can play a decisive role in determining its magnetic state; Star has also pointed out[16] that it is quite possible that CuAu is not a homogeneous host alloy to Fe. One would expect, on the other hand, that applying uniform pressure to a sample should vary its properties in a relatively well-defined way. A preliminary report on high-pressure resistivity measurements on Cu:Fe was given by Schilling *et al.*[17] and Summers *et al.*[18] High-pressure experiments on La:Ce[19,20] and Y:Ce[19,21] indicate a rapid increase of T_K with pressure in these systems, but are not able to test for a universal dependence.

A further result common to calculations based on both $\mathcal{H}_{s\text{-}d}$[6,7] and the Anderson[14,22] and Wolff[13] Hamiltonians is that at $T = 0$ K the exchange scattering resistivity has reached its unitary limit value $\rho_{\text{spin}} = 2S\rho_0$, where ρ_0 is the resistivity for *s*-wave resonant impurity scattering.[4] If the ordinary non-spin-flip potential scattering (phase shift δ_V) at each impurity is also included, the unitary limit value is found[23] to be reduced by the factor $\cos^2\delta_V$. This would imply that if T_K could be shifted, without changing ρ_0, S, or δ_V, the resistivity at $T = 0$ K would remain constant.

Dilute Cu:Fe would seem to be a suitable alloy on which to study the questions of the existence of a universal resistivity curve and the change of S or δ_V with pressure. The resistivity of Cu:Fe has been extensively studied over a wide concentration range[11,16] to very low temperatures. In addition, its spin-scattering resistivity exhibits a transition from an approximate logarithmic temperature dependence above 10 K to a T^2 dependence below about 2 K, thus allowing an accurate estimate of the value of the resistivity at $T = 0$ K.

II. EXPERIMENTAL

The alloys used in these experiments were prepared from 99.999+%-pure Cu and 99.998%-pure Fe.[24] The concentration of magnetic impurities in pure Cu as estimated by the manufacturer is quoted as being typically Fe < 0.7 ppm and Cr < 1 ppm. The resistivity ratio of pure Cu samples prepared in the same manner as the alloy samples was found to be $\rho_{300\,\mathrm{K}}/\rho_{4\,\mathrm{K}} \simeq 950$, which is compatible with a total impurity content less than 5 ppm. A very weak Kondo effect was also observed on this sample, indicating an Fe or Cr impurity content of less than 0.2 ppm.

The alloys were prepared by induction melting the components on a water-cooled silver boat in high vacuum. The sample was held in the molten state for a total period of 15 min, being rotated several times and jostled continually. Initially a 0.5-at.% Fe alloy was prepared and then further diluted to obtain a nominal Fe concentration of 114 ppm. Chemical analysis of this sample indicated (110 ± 5)-ppm Fe. After being cold rolled into 50-μ-thick foils, the samples were cut to size and tempered for 15 min by Joule-heating the sample to 700 °C in a water-cooled chamber under high vacuum. The homogeneity of these samples was confirmed to 1 μ by electron-microprobe examination. More detailed information on the homogeneity of Cu : Fe alloys is provided by a careful study of their magnetization by Hirschkoff *et al.*[25] They conclude that a 110-ppm alloy might be expected to contain 1 ppm of Fe-Fe pairs. Such effects can be very important at extremely low temperatures (~ 10 mK), where the main temperature dependence of the resistivity is due to scattering from atom pairs, the single-ion scattering having saturated out. In the temperature range of our experiments (above 1.3 K), the temperature dependence of the resistivity from single-ion scattering is large and one would expect the effect of Fe pairs on our measurements to be very small indeed. This expectation is confirmed by the low-temperature resistivity measurements of Star.[16] To test for possible clustering in our samples we also carried out resistivity measurements on 55- and 27-ppm Cu : Fe alloys which revealed a linear dependence of the Kondo effect on Fe concentration. In light of the above, it seems reasonable to assume that the resistivity measured in these experiments is due to scattering from isolated Fe ions.

High pressures are applied to the samples using the quasihydrostatic Bridgeman technique of opposing anvils. The high-pressure cell and anvil assembly is mounted inside a massive Cu-Be clamp which retains the applied force after removal from a hydraulic press. The applied pressure is changed at room temperature after each low-temperature resistivity measurement. The B_4C anvils used are semiconductors and thus serve as insulators at He temperatures, thereby simplifying electrical insulation problems. The samples are mounted between two epoxy disks each 4 mm in diam and 0.3 mm thick. These disks in turn are surrounded by an annular Pyrophyllite ring 10 mm in diameter and 0.6 mm thick, which is itself supported by a belt made of a special hardened steel.

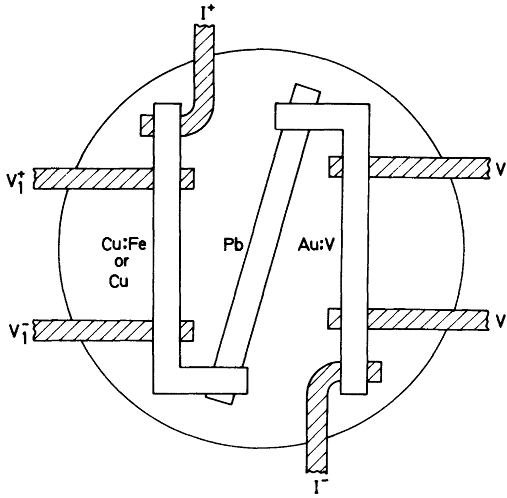

FIG. 1. Arrangement of samples and electrical leads on epoxy disk (4-mm diam) mounted in pressure cell. Cu : Fe and Cu are the Kondo and pure copper samples, respectively; Pb serves as internal manometer; Au-0.5% V detects pressure-cell deformation. *I* and *V* label current and potential leads, respectively.

The arrangement of the samples on the epoxy disk is shown in Fig. 1. The three samples (Cu : Fe or Cu; Pb and Au:V) are arranged in series. Essential for an accurate pressure determination at low temperatures is the presence of an internal manometer in the high-pressure cell itself. The Pb sample serves this function well because the pressure dependence of its superconducting transition temperature T_c is known to 10% accuracy.[26] In addition, the width of the superconducting transition ΔT_c gives a measure of the pressure inhomogeneity in the pressure cell. Typical values of the pressure inhomogeneity over the area of the Cu : Fe sample as estimated from ΔT_c lie between 2% and 7% of the measured pressure.

Applying or removing pressure from the pressure cell was found to not only change the amount of defect scattering in the sample but also to alter the sample's dimensions, the magnitude of this change depending on the pressures involved and the past history of the pressure cell. Due to the nature of these investigations, it was essential to be able to estimate any changes in the length/area factor α of the Cu: Fe sample. One way to estimate changes in α is to include within the pressure cell an additional sample whose resistivity has a very low intrinsic pressure dependence. This is the case for the alloy Au + 0.5-at.% V where at room temperature we estimate that $-(\Delta\rho/\rho\Delta P) < 0.04\%/$kbar. In order to minimize both changes in α and variation in defect scattering in measurements at different pressures, the pressure cell was initially preloaded to 90 kbar and then unloaded to zero pressure before commencing with high-pressure measurements. The success of this procedure will be shown in the Sec. III. Complete details of the high-pressure techniques employed in these experiments will be described elsewhere.[27]

The cryostat used in the high-pressure measurements was of standard design. Temperatures between 1.3 and 60 K were determined by a calibrated Germanium resistor supplemented by a copper-constantan thermocouple for higher temperatures. Both temperature sensors are located directly next to the high-pressure anvils. The accuracy of the temperature determination is always better than 0.5%.

Resistivity measurements were carried out using the standard four-terminal technique. The current source is constant to better than 10^{-5} during the 6 hr needed to make a temperature run. The sample potentials are first amplified by a galvanometric photocell amplifier and then displayed on an integrating $5\frac{1}{2}$ digit digitalvoltmeter. The relative accuracy of the measurement is better than 10^{-4}.

The resistivity measurements at zero pressure were carried out in a separate cryostat on a Cu: Fe sample with known dimensions. The relative accuracy of the measurement is better than 10^{-4}, the absolute accuracy being limited to about 1% by uncertainties in the length/area determination. The splinters of Cu: Fe alloy used in the high-pressure measurements are taken from this sample. A comparison of the zero-pressure and high-pressure data allows a determination of the absolute resistivity scale for all data. The experimental details of the zero-pressure measurement will be described in a future publication.[28]

III. RESULTS OF THEORY

Before presenting the results of the high-pressure resistivity measurements on Cu: Fe, let us consider the following approximate expression for the resistivity derived from $\mathcal{H}_{s\text{-}d}$ by Hamann[6] with the unitary limit value given by Schrieffer[4] and generalized by Fischer[23] to include potential scattering:

$$\rho_{\text{spin}} = S\rho_0 \left(1 - \cos 2\delta_V \frac{\ln T/T_K}{[\ln^2 T/T_K + \pi^2 S(S+1)]^{1/2}}\right), \qquad (1)$$

where the Kondo temperature including potential scattering is now given by

$$T_K \simeq T_F \exp[1/n(E_F) J_{\text{eff}} \cos^2\delta_V] \; . \qquad (2)$$

It is noted that this expression is of the form $\rho = \rho(T/T_K)$ expected for a universal resistivity curve, assuming S, δ_V, and ρ_0 are constants. The resistivity given by Eq. (1) is shown schematically in Fig. 2 [29] for several values of T_K. Although it is well known that this expression agrees well with experiment only for $T > T_K$, its qualitative features are probably correct at all temperatures. From Fig. 2 it is seen that as T_K increases (S, δ_V, and ρ_0 being held constant), the spin-scattering curve is shifted bodily to higher temperatures. A change in either S, δ_V, or ρ_0 as T_K increases would have the effect of changing the shape of the spin-scattering curve as it is shifted.

From Eq. (1) it is seen that the saturation value of the resistivity is given by

$$\rho_{\text{spin}}(T = 0 \text{ K}) = 2S\rho_0 \cos^2\delta_V . \qquad (3)$$

This saturation value is thus a sensitive function of S, ρ_0, and δ_V.

IV. RESULTS OF EXPERIMENT

A. Temperature Dependence of Resistivity

In Figs. 3(a) and 3(b) are shown the results of resistivity measurements on Cu–110-ppm Fe at pressures to 82 kbar and temperatures from 1.3 to 40 K. From the data it is immediately seen that increasing the pressure shifts the spin-scattering curve to successively higher temperatures. Applying pressure to Cu: Fe therefore increases the Kondo temperature T_K. The rapidly rising resis-

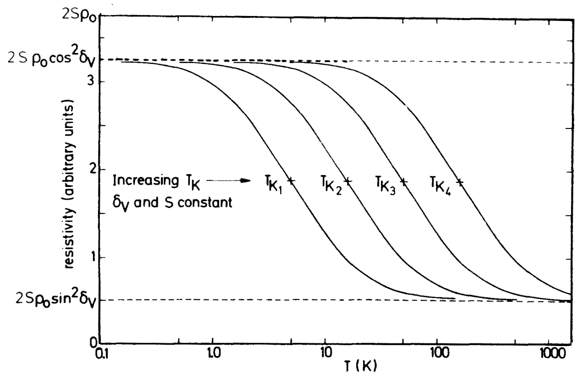

FIG. 2. Spin-resistivity curves from Hamann's expression [Eq. (1)] for increasing values of the Kondo temperature T_K. Because Eq. (1) is of the form $\rho = \rho(T/T_K)$, increasing T_K shifts the spin-resistivity curve bodily along the temperature scale.

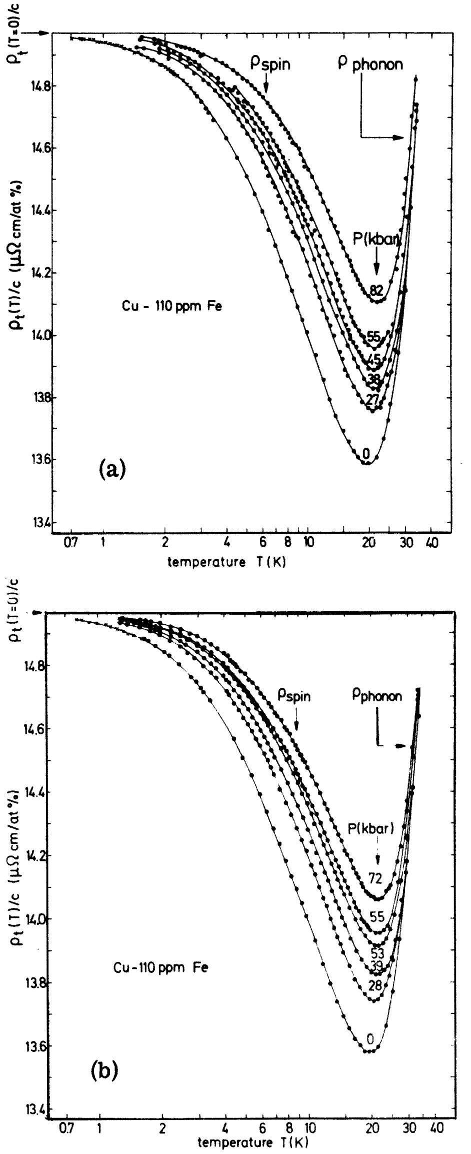

FIG. 3. Total measured resistivity of Cu : Fe versus temperature for different pressures. Dots are our data, crosses are from Loram *et al.* (Ref. 11). The solid lines connecting dots are to distinguish data at different pressures. *c* is Fe concentration (110 ppm). (a) Relative accuracy is 10^{-3}. Pressure cell was not preloaded. (b) Relative accuracy is 10^{-4}. Pressure cell was preloaded.

tivity above 20 K is due to phonon scattering. It is also seen that with increasing pressure the resistivity minimum fills up and that the temperature of this minimum, $T(\rho_{\min})$, increases slightly. It will be shown that the former effect is due to the shifting of the spin-scattering curve to higher temperatures whereas the latter follows from the reduction of the phonon scattering with increasing pressure.

In Figs. 4(a) and 4(b) some of the data (the resistivity of the Cu host has been subtracted out) are plotted as a function of the reduced temperature T/T_K and are seen to overlap within the experimental accuracy over the entire range $T < T(\rho_{\min})$. The overlap for the other pressures is similarly accurate. Thus, over the pressure and temperature range considered here, the present experiments confirm the existence of a universal resistivity curve $\rho = \rho(T/T_K)$ for Cu : Fe.

It should be pointed out that the fact that the resistivity data shown in Figs. 4(a) and 4(b) still have minima, even though the host resistivity has been

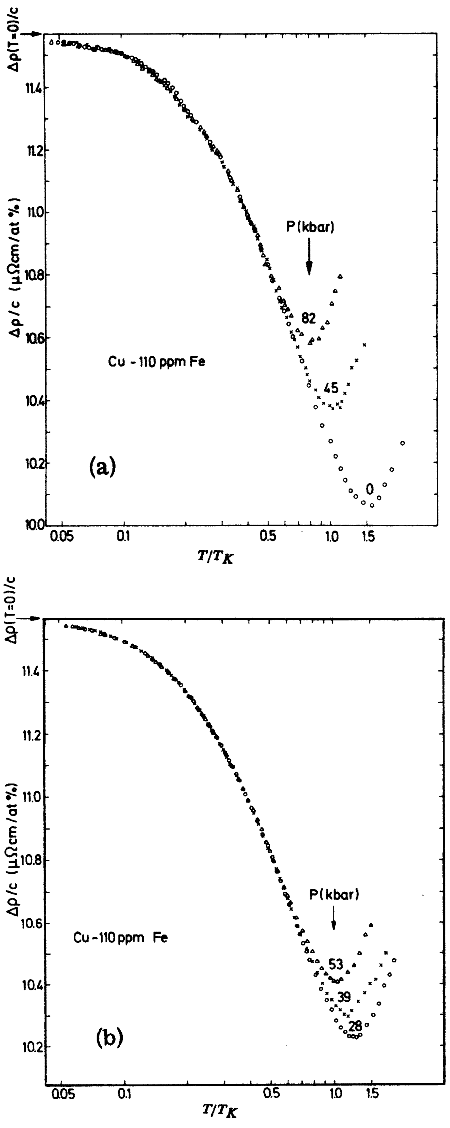

FIG. 4. Reduced resistivity of Cu : Fe versus reduced temperature for three pressures. $\Delta\rho = \rho_t - \rho_{\text{phonon}}$. ρ_t and ρ_{phonon} are the resistivities of Cu : Fe and Cu, respectively. The data shown in (a) and (b) are taken from Figs. 3(a) and 3(b). The values of T_K used for the given pressures are listed in Table I.

TABLE I. Shown for Cu: Fe as a function of pressure P are the Kondo temperature T_K, the volume change $\Delta V/V$, the density of states at the Fermi surface $n(E_F)$, and the effective exchange parameter J_{eff}. Data taken on the same sample are arranged in groups.

P(kbar)	T_K (K)	$-\Delta V/V$(%)[a]	$n(E_F)$ (states/eV atom)[b]	J_{eff} (eV)
0	24.0	0	0.294	−0.418
27 ± 1	31.6	1.75	0.292	−0.435
45 ± 3.5	37.7	2.82	0.290	−0.448
55 ± 4	40.3	3.39	0.290	−0.451
82 ± 5.7	50.6	4.79	0.288	−0.470
38 ± 1	35.2	2.42	0.291	−0.443
28 ± 0.8	31.2	1.80	0.292	−0.435
39 ± 1	34.6	2.48	0.291	−0.443
53 ± 0.8	39.0	3.27	0.290	−0.451
55 ± 3.9	41.0	3.38	0.290	−0.454
72 ± 6.5	47.5	4.30	0.289	−0.465

[a] See Ref. 35. [b] See Ref. 30.

subtracted out, is due to large deviations from Matthiessen's rule, as will be shown shortly. For this reason it is not possible for the curves to overlap for $T > T(\rho_{min})$.

In Table I are listed for each pressure the values of T_K which give the best overlap. Since our zero-pressure data agree well with the resistivity data (0.3–300 K) of Loram *et al.*[11] in our temperature range (1.3–40 K), we use their value of $T_K = 24$ K for Cu: Fe at zero pressure determined by the slope of a T^2 plot at the lowest temperatures. The Kondo temperature at high pressures is determined by the temperature shift necessary for the curves to overlap. Using Eq. (2) and the known pressure dependence of $n(E_F)$,[30] one can calculate the pressure dependence of the effective exchange parameter J_{eff}. From Table I we see that a pressure of 82 kbar is sufficient to approximately double T_K, implying that J_{eff} increases in magnitude by about 12%.

B. Unitary Limit

In Fig. 3(a) the data at 27, 45, 55, and 82 kbar were taken, in the given order, on the same sample without having preloaded the pressure cell. The saturation value of the resistance, $R(T=0$ K$)$, can be determined for each pressure by using the known T^2 dependence of the resistivity at low temperatures.[11,16] For the data shown in Fig. 3 the saturation value of the resistance (not resistivity) is found to increase by 14% between 27 and 82 kbar. The most likely explanation for this shift is that the initial application of pressure causes irreversible deformation of the pressure cell (and the sample) leading to an increased amount of temperature-independent defect scattering. The data shown in Fig. 3(a) are thus shifted vertically to have the same value of $R(T=0$ K$)$ for all pressures. Changes in the length/area factor of the Cu: Fe sample would also cause $R(T=0$ K$)$ to shift. This possibility will be discussed below.

In an effort to minimize changes in defect scattering for different pressures, the pressure cell for the data shown in Fig. 3(b) was initially preloaded to 90 kbar and then unloaded completely before proceeding with measurements at successively higher pressures. The data at 28, 39, and 53 kbar were taken on a different sample than the data at 55 and 72 kbar. The success of the preloading procedure is demonstrated by the fact that between 28 and 53 kbar, $R(T=0$ K$)$ increases by only 0.14% and between 55 and 72 kbar by 0.063%.

A Au: V sample is included in the pressure cell to supply information on changes in the length/area factor α of the Cu: Fe sample, as was discussed in Sec. II. These measurements indicate that for the Au: V sample between 28 and 53 kbar α decreases by 1.4% and between 55 and 72 kbar increases by only 0.2%. From the compressibility alone one would expect an increase in α of 0.5% and 0.3%, respectively. The measured values of α give a rough indication of the deformation of the pressure cell as the pressure is increased and they therefore represent the accuracy to which one can estimate changes in the length/area factor for the Cu: Fe sample. The actual changes in $R(T=0$ K$)$ with pressure for Cu: Fe are much less than the changes in α. Therefore, to the accuracy of our measurement (1.4% between 28 and 53 kbar and 0.2% between 55 and 72 kbar), the saturation value of the resistivity is independent of pressure.

As was pointed out in Sec. II, a zero-pressure measurement on a Cu: Fe sample with known dimensions is used to determine the absolute resistivity scale of the high-pressure data. For pressures below about 20 kbar, rather large irreversible deformations of the sample take place whether the pressure is being increased or decreased. It is thus not possible to carry out a measurement near $P=0$ on a sample with the same dimensions and amount of defect scattering as the high-pressure data. The $P=0$ measurement was actually carried out on a tempered Cu: Fe sample whose length/area factor was known to about 1%. In order to compare this measurement to the high-pressure data, different amounts of temperature-independent defect scattering ρ_d are added to the $P=0$ data until these data, when shifted in temperature, overlap best with the $P=28$-kbar data. The amount of defect scattering added, $\rho_d = 35$ $\mu\Omega$ cm, agrees within 40% with that measured on a pure Cu sample before and after pressure application. The confirmation of the existence of a universal resistivity curve for Cu: Fe at pressures below 28 kbar is thus not as complete as for the other pressures. However, the accuracy of the overlap of the $P=0$ and 28-kbar data and the fact that the necessary temperature shift (T_K increase) between these two pressures is proportionately the same as for the other pressures (see Table I), supports the existence of a universal resistivity curve for pressures below 28 kbar.

The saturation value of the resistivity is determined from the $P=0$ data to be $\rho_{\text{spin}}(T=0\text{ K})=11.6 \pm 0.6$ $\mu\Omega$ cm/at.%, where the accuracy is limited by uncertainties in the Fe-concentration determination. This value of the saturation resistivity is in reasonable agreement with estimates from other sources.[11,25] Using Eq. (3) it is found that for a Cu host $\rho_{\text{spin}}(T=0\text{ K})=3.8(2S)\cos^2\delta_V\,\mu\Omega$ cm/at.%. Furthermore, using our value of the saturation resistivity and $S=\frac{3}{2}$ from susceptibility measurements,[31] one obtains $\delta_V \simeq 0$. Negligible potential scattering, however, seems to be inconsistent with the fact that Cu: Fe exhibits a sizable peak in the thermoelectric power.[3,23] EPR investigations[32] imply $S=2$ which results in the more reasonable value $\delta_V = 29°$.

C. Phonon Scattering

The temperature dependence of the resistance of pure Cu was measured on a single sample for pressures 31 ± 2.4 kbar and 57 ± 4 kbar at temperatures between 4 and 300 K. In a separate experiment, a zero-pressure measurement was carried out on a pure Cu sample with known dimensions. The results of these measurements are shown in Fig. 5 (note that the residual defect scattering has been subtracted out). It is seen that for temperatures below approximately 40 K, a $\rho \propto T^5$ law fits the data quite well. This result is in agreement with the low-temperature ($T \ll \Theta_D = 343$ K for Cu) dependence predicted by the Bloch-Grüneisen expression[33] $\rho_{\text{phonon}} = bT^5$, where $b \propto 1/(r_s^2\,\Theta_D^6)$, r_s is the radius of the Wigner-Seitz sphere, and Θ_D is the Debye temperature. At temperatures above 40 K, the data are seen to bend over, heading toward the approximate $\rho \propto T$ dependence expected for $T \gg \Theta_D$.[33]

Inspection of the pressure dependence of the data reveals that for temperatures below 40 K, the resistance curve for 53 kbar is shifted downward from that at 32 kbar by roughly 15%. This decrease is due mainly to an increase in the Debye temperature with pressure, as will be seen below. Using the above low-temperature expression for the resistivity, one finds

$$\Delta b/b = \Delta V/V(6\gamma_D - \tfrac{2}{3}), \qquad (4)$$

where $\gamma_D = -(\Delta\Theta_D/\Theta_D)/(\Delta V/V)$ is the (Debye) Grüneisen parameter. Typical values of the Grüneisen parameter for Cu determined by thermal-expansion and specific-heat measurements vary from about 1.5 to 2.0 at temperatures from 4 to 300 K.[34] In the following we set $\gamma = 1.7$. For the compressibility of Cu we use the expression[35] $\Delta V/V = -6.9 \times 10^{-4}P\text{ (kbar)} + 1.3\times 10^{-6}P^2\text{ (kbar)}$. One finds that between 31 and 57 kbar $\Delta b/b \simeq 14\%$, in good agreement with our measurements. It is interesting to note that at temperatures near 300 K, the 57-kbar curve in Fig. 5 is displaced downward by only about 5% from the 31-kbar curve. This agrees well with the downward shift of 4% expected for the high-temperature limit ($\rho \propto T$).

The value of b at all pressures is determined absolutely by extrapolating the high-pressure data to $P=0$ and comparing this with the zero-pressure

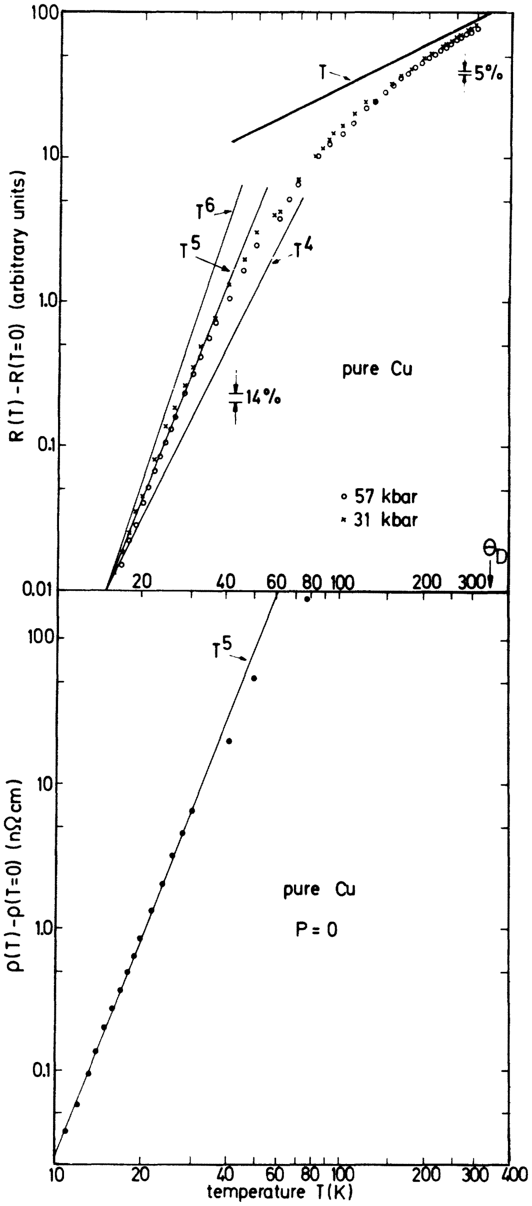

FIG. 5. Resistivity of pure Cu sample versus temperature for three pressures. The residual defect resistivity $\rho(T=0\text{ K})$ has been subtracted out. The high-pressure data can be normalized by comparing them to the $P=0$ data which were measured absolutely.

data which were measured absolutely. Using Eq. (4) with $\gamma = 1.7$ gives the following expression for the resistivity due to phonon scattering:

$$\rho_{\text{phonon}} = 2.60\times 10^{-10}(1+\Delta V/V)^{9.5}T^5\ \mu\Omega\,\text{cm}. \qquad (5)$$

In Figs. 3(a) and 3(b) it was seen that with increasing pressure the resistivity minimum fills up and that the temperature at this minimum $T(\rho_{\text{min}})$ increases by approximately 2 K between 0 and 82 kbar. It is now possible to understand the origin of these two effects. The shift of $T(\rho_{\text{min}})$ is due to a decrease in the phonon scattering with increasing pressure. Assuming that near the minimum $\rho_{\text{spin}} \propto \ln T$ and $\rho_{\text{phonon}} = bT^5$, it can be shown that $\Delta T_{\text{min}}/T_{\text{min}} = \Delta b/5b$. The former assumption is seen [Figs. 3(a) and 3(b)] to hold even for $P = 82$ kbar and the latter was shown in Fig. 5 to be valid. Thus by using Eq. (4) we find that between 0 and 82 kbar $\Delta T_{\text{min}} = 1.8$ K, in good agreement with experiment. The increase of $\rho(T_{\text{min}})$ with increasing pressure is due to the shifting of the spin-scattering curve to higher temperatures. Near the resistivity minimum, the spin scattering dominates the phonon scattering ($\rho_{\text{spin}} \simeq 15\,\rho_{\text{phonon}}$) and the spin-scattering resistivity is essentially *linear* in $\ln T$. Since the minimum itself moves only slightly with pressure, it would be expected that as ρ_{spin} is shifted to higher temperatures, $\rho(T_{\text{min}})$ should increase by a proportional amount. Our measurements confirm this expectation.

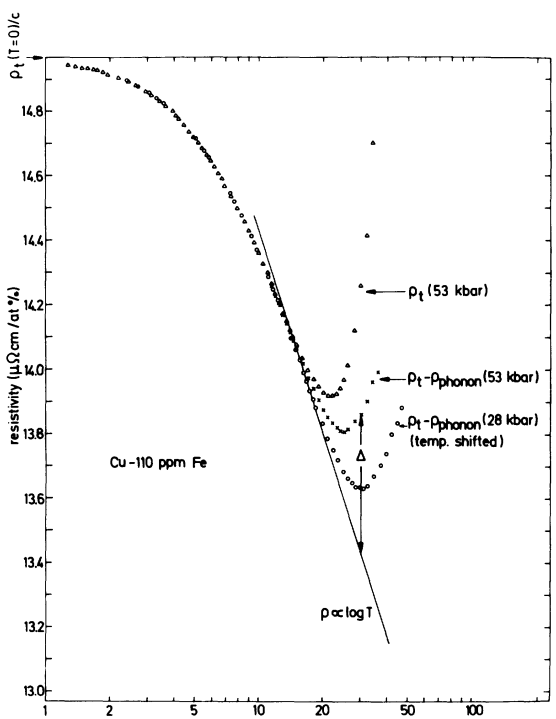

FIG. 6. The total resistivity of Cu : Fe at $P = 53$ kbar is shown before and after subtraction of the Cu host resistivity ρ_{phonon}. The universal resistivity curve for $T < 20$ K is estimated by the temperature-shifted reduced resistivity at $P = 28$ kbar and for higher temperatures by a straight line, thus allowing an estimate of the deviation Δ from Matthiessen's rule.

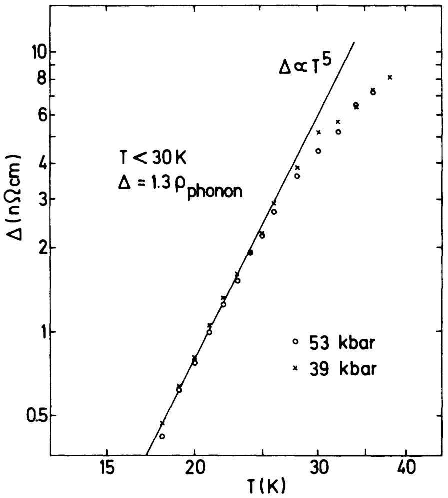

FIG. 7. Deviation from Matthiessen's rule versus temperature estimated by method shown in Fig. 6. For $T < 30$ K, Δ has approximately the same temperature dependence as ρ_{phonon}.

In Fig. 6 the Cu : Fe data at 53 kbar are shown before and after ρ_{phonon} is subtracted off. As discussed by Loram *et al.*,[11] the fact that the reduced resistivity $\Delta\rho$ still has a prominent minimum is due to large deviations from Matthiessen's rule. To estimate the magnitude of these deviations, we also show in Fig. 6 the reduced resistivity $\Delta\rho$ for $P = 28$ kbar which has been shifted in temperature until it overlaps the 53-kbar data. Due to the existence of a universal resistivity curve for Cu : Fe, the spin-scattering resistivity is thus quite well determined out to about 30 K. A straight line approximates the spin-scattering curve to higher temperatures allowing the deviation from Matthiessen's rule to be estimated to about 40 K. These deviations are seen to be approximately as large as the phonon scattering itself. In Fig. 7 we plot the estimated deviation Δ as a function of temperature. The deviation at lower temperatures is seen to have a T^5 dependence and is therefore proportional to the phonon resistivity. This result is in agreement with the findings of Whall *et al.*[36] on a series of gold alloys and with theoretical predictions[37,38] for temperatures low enough that the phonon scattering is small compared to the impurity scattering. The deviation of Δ from a T^5 dependence at temperatures above 30 K could be at least partially due to an incorrect estimate of the spin-scattering resistivity for these temperatures. However, it is known experimentally[11] and expected theoretically[38] that Δ levels off and has a maximum at about 60 K for 110 ppm Fe in Cu. Extension of the present measurements to higher temperatures and pressures is necessary for a thorough investigation of these questions.

V. DISCUSSION

A. Universal Resistivity Curves

In Sec. IV we have shown that, over the pressure and temperature range considered, the present experiments confirm the existence of a universal resistivity curve $\rho = \rho\,(T/T_K)$ for Cu : Fe. The existence of universal curves $f = f\,(T/T_K)$ in the Kondo problem is a result common to all approximate expressions derived from the s-d Hamiltonian. In fact, this result appears to follow quite generally from $\mathcal{H}_{s\text{-}d}$. Anderson and Yuval[12] have demonstrated an "asymptotically exact equivalence" between the Kondo problem based on $\mathcal{H}_{s\text{-}d}$ and the statistical mechanics of a certain one-dimensional gas. It is found that the renormalized partition function of the problem is a function of terms like T/T_K and J/T_K. This implies that in expressions for measurable quantities calculated from this partition function the temperature T is always scaled by T_K. It is conceivable that terms involving T_K and J could appear separately from T; however, from an inspection of the spectral function, it would seem intuitively reasonable that such terms tend to cancel out. The present work implies that such terms, if they do exist, do not enter the expression for the resistivity in any important way.

It should also be pointed out that the existence of universal curves is a prediction which is not restricted to expressions derived from the s-d Hamiltonian but also applies to spin-fluctuation theories[13,14] based on the more general Wolff or Anderson Hamiltonians which include $\mathcal{H}_{s\text{-}d}$ as a special case.

In Sec. III it was shown using Eq. (1) (Fig. 2) that the shape of the spin-scattering curve remains unchanged as T_K increases only if ρ_0, δ_V, and S remain constant. The present work supports this result. As is seen from Eq. (3), the constancy of ρ_0, δ_V, and S in these experiments is indicated even more directly by the fact that the saturation value of the resistivity $\rho_{\text{spin}}(T = 0\text{ K})$ does not change as T_K increases. These considerations are expected to hold on general grounds since ρ_0, δ_V, and S must enter any expression for the spin-scattering resistivity.

From Eq. (1) it is clear that other Kondo alloys need not exhibit the same "universal" resistivity curve as Cu: Fe because different values of the parameters ρ_0, δ_V, and S might be appropriate. All curves, however, should have the same qualitative dependence as shown in Fig. 2. To investigate this question we are currently carrying out resistivity measurements on Au: V[39] and other alloys over a wide range of temperature and pressure.

On the basis of the above results, it would seem reasonable to expect that the universal resistivity curve for Cu: Fe exists over a much wider range of pressure and temperature than was accessible in the present experiments. If this is true for other alloys as well, then for those alloys where T_K increases with pressure, the application of very high pressures would in fact enable one to determine a portion of the universal curve which for $P = 0$ would lie at lower temperatures. This method of mapping out $\rho_{\text{spin}}(T/T_K)$ would be especially advantageous for rare-earth alloys such as Y: Ce and La: Ce, where the increase in T_K with pressure is extremely rapid.

B. Volume Dependence of T_K

In these experiments it is found that a pressure of 82 kbar is sufficient to approximately double the Kondo temperature of Cu: Fe. In Fig. 8 we plot the values of J_{eff} shown in Table I as a function of relative volume. Also shown in this figure are the results of the resistivity measurements of Loram

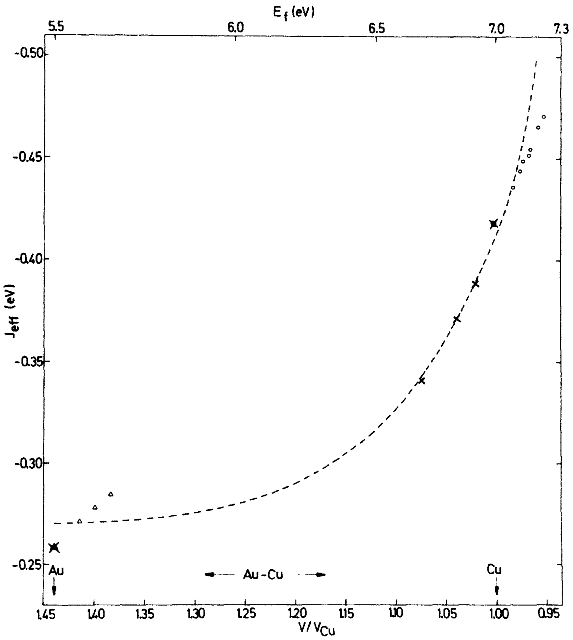

FIG. 8. Effective exchange parameter J_{eff} as a function of the average atomic volume of sample relative to that of Cu at $P = 0$. Circles are from our high-pressure data on Cu : Fe, crosses are from Cu-Au : Fe data by Loram *et al.* (Ref. 11), and triangles are preliminary results of high-pressure data on Au : Fe by Crone *et al.* (Ref. 42). The dashed line is a fit using the SW transformation [Eq. (6)]. Values of the Fermi energy E_F given are those of a free-electron gas.

et al.[11] on a series of Cu-Au: Fe alloys. It is interesting to note that (with the exception of the Au: Fe alloy) both alloy and high-pressure data show roughly the same volume dependence. This would seem to support the point of view that in these experiments changes in the value of $J_{\rm eff}$ are mainly a function of the macroscopic volume and are not dependent on details of the band structure or the immediate surroundings of the magnetic impurity. In the simplest model, therefore, we assume that changes in sample volume (whether lattice contraction under pressure or lattice expansion due to alloying Au in Cu) merely have the effect of shifting the Fermi level relative to the virtual bound states of the magnetic impurity. The value of the Fermi energy and its volume dependence are estimated from the free-electron model. The position of the virtual bound states of the magnetic impurity are assumed fixed with respect to the bottom of the conduction band.

An expression which connects $J_{\rm eff}$ with the positions of the virtual levels relative to E_F is given in the magnetic limit by the Schrieffer-Wolff (SW) transformation[4]:

$$J_- = J_{\rm eff} - J_+ = -(v^2/S)\,(1/E^+ + 1/E^-)\,, \qquad (6)$$

where J_+ is the ordinary positive exchange integral, v is the covalent admixture matrix element, S is the impurity spin, E^+ is the energy required to transfer an electron from the Fermi surface to the Fe impurity in its ground state, and E^- is the energy required to remove an electron from the impurity and place it at the Fermi level. The above expression for J_-, which was initially derived only to lowest order, was shown by Hamann[22] to retain validity in an asymptotically exact path-integral theory of magnetic alloys. Since J_+ is estimated to be small[40] [$J_+ \simeq 0.2$ eV$/(2l+1) = 0.04$ eV for transition-metal impurities] and essentially pressure independent,[19] it will be neglected in the following. For the impurity spin we use the EPR value[32] $S = 2$ for Fe in Cu. For Mn in Ag it has been determined[41] that $U = E^+ + E^- = 5$ eV; we use this value here in the absence of good spectroscopic measurements for Fe in Cu. Going from Au to Cu the value of the free-electron-gas Fermi energy increases from 5.5 to 7.0 eV. Inserting the above values in Eq. (6) we obtain the dashed curve in Fig. 8. The values of the parameters used in the fit are $v = 0.8$ eV, $E^+ = 1.0$ eV, and $E^- = 4.0$ eV for Cu: Fe. Going from Au to Cu, E^+ thus decreases from 2.5 to 1 eV.

In Fig. 8 it is seen that the SW curve gives good qualitative agreement with the Cu: Fe–high-pressure and Cu-Au: Fe–alloy data. One obvious method of testing the above simple model would be to carry out high-pressure measurements on Au: Fe. For the SW curve shown in Fig. 8 one would expect a very weak pressure dependence of T_K. More generally, it can be inferred from Eq. (6) that if $J_{\rm eff}$ is less for Au: Fe than for Cu: Fe, as is the case, then $dJ_{\rm eff}/dV$ should also be less. This result holds independent of the exact values assumed for the parameters in Eq. (6). Preliminary high-pressure measurements on Au: Fe[42] shown in Fig. 8 confirm this expectation, although the agreement with the SW curve is rather poor. Assuming that the pressure dependence of $J_{\rm eff}$ for Au: Fe shown in Fig. 8 is correct, there could be a number of reasons for this discrepancy. The values of T_K for Au: Fe and Cu: Fe were determined[11] by fits to different theoretical expressions and are therefore not directly comparable. Whereas the value $T_K = 24$ K obtained from resistivity measurements on Cu: Fe seems reasonable, one can only estimate that for Au: Fe, $T_K < 0.5$ K[43] or equivalently $|J_{\rm eff}| < 0.27$ eV. In addition, as is seen from Eq. (2), different amounts of potential scattering for Fe in Au and Cu could also affect the determination of $J_{\rm eff}$. Shifting T_K for Au: Fe to lower temperatures and increasing U would improve the agreement substantially. Unfortunately, there is at the moment no independent information to justify this procedure.

Besides possible large errors in the values of the parameters used, it could well be that the simple model presented here is itself inadequate; there are a variety of modifications which might be necessary. Firstly, the virtual bound state of Fe is likely to be located energetically in or near the d bands of Cu or Au which lie only 2.1 and 2.5 eV, respectively, below the Fermi level. Application of pressure would tend to increase the overlap of band and virtual $3d$ wave functions and broaden the virtual bound state, thereby increasing the interband mixing. If this were the dominant effect, it would explain why in all known high-pressure experiments on Kondo alloys (Cu: Fe, Au: Fe,[42] Au: V,[39] Y: Ce,[21] and La: Ce[20]) the Kondo temperature is found to *increase* with pressure (note that in the simple model T_K could decrease with increasing pressure if $E^- < E^+$). Secondly, in the simple model we have also neglected any influence of the immediate local surroundings on T_K. The ionic radius of Au is about 40% larger than that of Cu and thus for a given lattice parameter the cage formed by the surrounding ions within which the Fe^{++} ion resides is smaller for Au than Cu. This effect could have the tendency to shift T_K for Au compared with Cu to relatively higher temperatures. Thirdly, it should also be noted that we have completely ignored numerous orbital effects such as orbit-orbit exchange, mixed spin and orbit exchange, and crystal-field splitting. As pointed out by Hirst,[32] the orbital angular momentum of $3d$ impurities in cubic metals is not necessarily quenched. In fact, EPR studies on Fe in Cu indicate that $g \neq 2$[44] where

g is the Lande g factor. The assignment to a magnetic impurity of orbital as well as spin degrees of freedom can be carried out in the context of a generalized s-d interaction model.[45] However, as pointed out by Hirst,[46] the generalized Kondo problem arising from the generalized s-d interaction model is very similar to that arising from $\mathcal{H} = -J\vec{s}\cdot\vec{S}$. It would, therefore, not be expected that consideration of the generalized model would change the main conclusions of this paper. Inclusion of orbital effects and crystal-field splittings could, however, complicate the interpretation of the pressure dependence of J_{eff}.

To investigate the importance of such effects, further experimental work is necessary. The above modifications to the simple Fermi gas model will be discussed in detail in a future publication.[42]

C. Potential Scattering

In Sec. IV it was pointed out that within the experimental accuracy of 1.7% the saturation value of the resistivity of Cu: Fe, $\rho_{\mathrm{spin}}(T=0\ \mathrm{K})$, remains constant between 28 and 72 kbar. This result is consistent with the volume dependence of the scattering from charge impurities which would give a change in resistivity over this pressure range of roughly 1.5%. Since $\rho_{\mathrm{spin}}(T=0\ \mathrm{K}) = 2S\rho_0\cos^2\delta_V$, it is therefore not possible within the accuracy of these experiments to detect any change in δ_V. This is a result which, as pointed out previously, is indicated by the existence of a universal resistivity curve.

There are several types of potential scattering which contribute to the phase shift δ_V. The simplest kind arises from the charge difference between the magnetic ion and host ions. In addition, it has been shown[47] that scattering from potential scatterers (nonmagnetic impurities, vacancies, and other crystal defects) located near the magnetic ion can also interfere significantly with the exchange scattering from the magnetic impurity. A further type of potential scattering V_d is due to resonance scattering as the virtual bound state of the magnetic impurity nears the Fermi level. From the Schrieffer-Wolff transformation one obtains $V_d = -(v^2/2)(1/E^+ - 1/E^-)$, where the phase shift δ_{V_d} for this type of potential scattering is given by $\delta_{V_d} = -\tan^{-1}[\frac{1}{2}\pi n(E_F)V_d]$. Using the value of the parameters for the fit in Fig. 8, one finds that for Cu: Fe, δ_{V_d} is very small, $\cos^2\delta_{V_d} \simeq 0.99$. Using the above simple model, the change in $\cos^2\delta_{V_d}$ upon application of 82 kbar would be less than 1% and thus not detectable in this experiment. The phase shift δ_{V_d} does not become significant until the virtual d level is very near E_F. However, it can be argued that this source of potential scattering may not enter the expression [Eq. (3)] for the saturation value of the resistivity $\rho_{\mathrm{spin}}(T=0\ \mathrm{K})$. When the virtual level is far away from E_F, $\delta_{V_d}=0$; however, at $T=0$ K the spin-scattering resistivity has reached its unitary limit, implying $\delta_J = \frac{1}{2}\pi$, where δ_J is the phase shift for spin scattering. On the other hand, when the virtual bound state is at the Fermi level, it is to be expected that $S=0$, $\delta_J=0$, and $\delta_{V_d} = \frac{1}{2}\pi$. If it still holds for the virtual level near E_F that $\delta_J + \delta_{V_d} = \frac{1}{2}\pi$, then one would expect that as the virtual level approaches E_F, the spin scattering would be wiped out from below, $\rho_{\mathrm{spin}}(T=0\ \mathrm{K})$, however, remaining constant. This model is consistent with high-pressure resistivity measurements on Y: Ce.[20] It would thus appear that δ_V in Eq. (3) does not include δ_{V_d} from this resonant scattering. This would imply that our measurements are only capable of detecting changes in the "nonresonant" types of potential scattering.

D. Deviations from Matthiessen's Rule

In Sec. IV we described an experimental procedure for determining deviations from Matthiessen's rule in Kondo alloys. This method relies on the existence of a universal resistivity curve as has been demonstrated for Cu: Fe. It is seen from Figs. 6 and 7 that below 40 K these deviations are actually larger than the phonon scattering itself. This result emphasizes the necessity, at all but the lowest temperatures, of accurately determining such deviations before attempting to extract information about the spin-scattering curves. For temperatures below 30 K it is found that these deviations are proportional to the phonon scattering in pure Cu, in agreement with experimental and theoretical results.[36–38] The departure from this rule seen at temperatures above 30 K is probably due to the presence of a maximum in Δ at about 60 K.[11] Extension of the present measurements to higher temperatures and pressures is necessary for a thorough investigation of such effects.

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