*Supported in part by the Advanced Research Projects Agency.

¹J. A. Appelbaum and D. Penn, Bull. Am. Phys. Soc. <u>14</u>, 371 (1969). ²J. A. Appelbaum and D. Penn, Phys. Rev. <u>188</u>, 874

²J. A. Appelbaum and D. Penn, Phys. Rev. <u>188</u>, 874 (1969).

³J. A. Appelbaum, D. Penn, and P. E. Bloomfield, Phys. Rev. B 1, 4490 (1970).

⁴A. Theumann, Phys. Rev. 178, 978 (1969).

⁵P. A. Wolff, Phys. Rev. <u>124</u>, 1030 (1961).

⁶P. W. Anderson, Phys. Rev. <u>124</u>, 41 (1961).

⁷D. N. Zubarev, Usp. Fiz. Nauk <u>71</u>, 71 (1960) [Soviet Phys. Usp. <u>3</u>, 320 (1960)].

⁸The trial integrands were approximate forms of f_r in which the logarithmic terms were expanded about the maximum value of the integrand. This resulted in test integrals which were within a few percent of the actual integrals.

⁹It is easily shown that $T_K \simeq 1.1 (nD/\pi) \exp(V/d)$.

¹⁰See, for example, P. E. Bloomfield and D. R. Hamann, Phys. Rev. <u>164</u>, 856 (1967).

¹¹The role of Fermi-surface singularities due to the Kondo effect in generating the Curie law was first emphasized by Hamann. See Ref. 12.

PHYSICAL REVIEW B

¹²D. R. Hamann, Phys. Rev. Letters <u>17</u>, 145 (1966).

¹³J. Zittartz, Z. Physik <u>217</u>, 155 (1968).

¹⁴Y. Nagaoka, Phys. Rev. <u>138</u>, A1112 (1965).

¹⁵D. R. Hamann, Phys. Rev. <u>158</u>, 570 (1967).

¹⁶G. Bayam, Phys. Rev. <u>127</u>, 1391 (1962).

¹⁷See Refs. 5 and 6 as well as P. Lederer and D. L.

Mills, Solid State Commun. 5, 131 (1967); R. Rivier and

M. J. Zuckermann, Phys. Rev. Letters <u>21</u>, 904 (1968).
 ¹⁸D. R. Hamann, Phys. Rev. <u>186</u>, 549 (1969).

¹⁹S. Q. Wang, W. E. Evenson, and J. R. Schrieffer,

Phys. Rev. Letters 23, 92 (1969); W. E. Evenson, J. R.

Schrieffer, and S. Q. Wang, J. Appl. Phys. <u>141</u>, 1199 (1970).

²⁰D. R. Hamann, Phys. Rev. Letters <u>23</u>, 95 (1969).

²¹H. Mamada and F. Takano (unpublished).

²² P. W. Anderson and G. Yuval, Phys. Rev. Letters
 23, 89 (1969).
 ²³ P. W. Anderson, G. Yuval, and D. R. Hamann, Phys.

²³P. W. Anderson, G. Yuval, and D. R. Hamann, Phys. Rev. B (to be published).

 $^{\rm 24}{\rm Even}$ at that, you cannot avoid the problem entirely; see Sec. IV.

²⁵J. A. Appelbaum and J. Kondo, Phys. Rev. Letters **19**, 906 (1967).

VOLUME 3, NUMBER 3

1 FEBRUARY 1971

Determination of the Kondo Temperature and Internal Field Distribution in Dilute AuMn Alloys*

J. W. Loram, T. E. Whall, and P. J. Ford University of Sussex, Brighton, United Kingdom (Received 26 June 1970)

The resistivities of several AuMn alloys, containing between 0.005 and 1.5 at. % Mn, have been measured in the temperature range 0.5-40 °K. The magnitude of the spin resistivity is determined from the decrease in resistance due to ordering in internal fields. This, combined with a measurement of the coefficient of the logarithmic term in the resistance at temperatures well above the resistance maximum, allows the Kondo temperature to be determined, the value so obtained being of the order of 10^{-13} °K. The predicted resistivity at T=0, in the absence of interactions, is shown to be comparable with the unitarity limit for *d*-wave scattering. Information is also obtained concerning the magnitude and distribution of internal fields within the alloys. The distribution is found to approximate to a Gaussian, but with a dip in the region of very low fields.

I. INTRODUCTION

Previous investigations, ^{1,2} and the present results, demonstrate that the temperature dependence of the resistivity of dilute AuMn alloys is qualitatively the same as that observed in many other dilute-magnetic-alloy systems. In very low concentration alloys, the resistivity increases logarithmically with decreasing temperature in the liquid-helium temperature range. At rather higher concentrations the logarithmic increase is terminated and a broad maximum is observed, associated with the onset of magnetic ordering, and the resistance falls as the temperature is lowered further. The main quantitative difference is the rather small magnitude of the logarithmic term, which is approximately 15 times smaller in AuMn alloys² than in AuFe alloys³ of comparable concentration. As the spin value of Mn in Au is significantly larger than that of Fe in Au, it may be concluded that the Kondo temperature in AuMn is extremely low.

In the present investigation, the resistances of AuMn alloys containing between 0.005 and 1.5% Mn have been measured in the temperature range 0.5-40 °K. The decrease in resistance below the ordering temperature is combined with the magnitude of the logarithmic term at higher temperatures to provide an estimate of the Kondo temperature. The deviation of the resistance from a logarithmic temperature dependence is used to provide information on the magnitude and distribution of internal fields within the alloy.

II. RESULTS

The alloys were prepared by induction melting appropriate quantities of Johnson Matthey "Spec Pure" Au, and 99.99% pure Mn (Koch Light Industries). After an homogenizing anneal at 950 °C for six hours, resistance specimens were prepared in the form of tapes approximately $8 \times 0.3 \times 0.01$ cm in size. This was followed by a strain-relieving anneal at 500 °C for two hours. The alloys were analyzed chemically for Mn and Fe content, and the Mn concentrations are given in Table I. The Fe impurity concentration was found to be typically 5-10 ppm in both the AuMn and pure Au samples. Simultaneous resistance measurements of five alloys and one pure Au specimen were made in a conventional He₃ cryostat operating in the temperature range 0.5-300 °K, using a standard fourterminal potentiometric technique. Details of the apparatus and method of measurement have been described previously.³

The excess resistivity per atomic percent of Mn, ρ/c , where $\rho = \rho(\text{Alloy}) - \rho(\text{Au})$, and c is the Mn concentration in atomic percent, is plotted against temperature in Fig. 1. The logarithmic-temperature-dependence characteristic of conduction-electron scattering from noninteracting magnetic impurities⁴ is evident in the most dilute alloys. In the more concentrated alloys this is modified by interaction effects which lead to a decrease in resistance at low temperatures. The temperature T_m of the resistance maximum increases as the concentration increases.

At higher temperatures (10-15 °K) the excess resistance ρ passes through a minimum and increases as the temperature is raised further. This results from positive deviations from Matthiessen's rule.⁵ This deviation (which is observed in both dilute magnetic^{1,3} and nonmagnetic alloys⁶) varies initially as $\gamma \rho_p$, where ρ_p is the phonon resistivity and γ is a constant of the order of 1. Significant effects are therefore only expected (and observed) in the phonon region, and for this reason the analysis of the results is limited to temperatures below 7 °K where the deviation is



FIG. 1. The excess resistivity per atomic percent of Mn, ρ/c plotted against temperature. The nominal Mn concentrations are as shown in the figure. The scale refers to the 0.1% alloy. Small vertical displacements (not exceeding 0.2 $\mu\Omega$ cm/at.%) have been applied to the other curves for the sake of clarity.

sufficiently small to be ignored.

III. ANALYSIS OF RESULTS

There have been several calculations⁷⁻¹² of the resistivity of a dilute magnetic alloy in a magnetic field (either external, internal, or both). Yosida's⁷ calculation to second-order perturbation theory reveals several important results. At high temperatures, $T \gg H$, ¹³ the resistivity can be written

TABLE I. Data relating to the resistivities of the AuMn alloys.

Mn conc. $(c \text{ at. }\%)$	Mn conc. $(c \text{ at. }\%)$	<i>T_m</i> (°K)	$\Delta \rho_m$	A'	Β'	C'	D'	$\left(\frac{d\rho}{dT}\right)_{0.5^{\circ}\mathrm{K}}$
(Nominal)	(Analyzed)	(11)	()()	()	()	()	(,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	$(\mu\Omega \text{ cm }^{\circ}\text{K}^{-1})$
0.005	0.0025			0.0129	0.00031			
0.01	0.004			0.0242	0.00083	0.00007		
0.02	0.018	0.68		0.0472	0.00144	0.00058	0.00007	
0.05	0.036	1.4	0.0088	0.124	0.00295	0.0045	0.0022	
0.1	0.11	2.4	0.0185	0.246	0.0053	0.022	0.026	0.016
0.2	0.18	3.9	0.0355	0.476	0.0091	0.089	0.24	0.020
0.5	0.54	8	0.095					0.027
1.5	1.58	20	0.24					0.028

$\rho = AV^2 + AJ^2S(S+1),$

where $A = 3\pi m^* cv/2e^2 \hbar E_F$, c is the impurity concentration, E_F is the Fermi energy and v the atomic volume of the host, and m^* is the conduction-electron effective mass. J and V are defined by the *s*-*d* Hamiltonian $\mathcal{K} = V + 2JS \cdot s$, ¹⁴ where *S* is the impurity spin, s the conduction-electron spin density at the impurity site, and V is the screened coulomb potential of the impurity (we assume V $\gg J$). In the temperature range $T \gg H$, one-third of the spin-dependent term is contributed by elastic non-spin-flip scattering and two-thirds by spin-flip scattering. As the temperature falls below H, the populations of the allowed spin components M_{\star} of the impurities are modified (the local field is directed parallel to the z axis) and. at T=0, spin-flip scattering falls to zero. Nonspin-flip scattering contributes two terms, one proportional to $J^2 \overline{M_z^2}$, ¹⁵ which increases to $A J^2 S^2$ at T=0. The second term $-4AJ^2 \overline{M}_{g}^2$ arises from an interference between spin and potential scattering, and this dominates the variation of the resistance at low temperatures in an applied magnetic field or for ferromagnetic ordering. For an alloy of the kind considered here, which exhibits antiferromagnetic ordering, \overline{M}_{z} is zero, and the interference term therefore makes no contribution to the zero-field resistance. Thus, for an alloy undergoing antiferromagnetic ordering, Yosida⁷ shows that to second order in J, and in zero applied field, the spin resistivity decreases by an amount $A J^2 S$ [from $A J^2 S (S+1)$ to $A J^2 S^2$] as the temperature falls to zero.

Abrikosov⁸ summed the perturbation series to infinite order in J/E_F and determined the resistivity in the presence of an external magnetic field, or for ferromagnetic ordering. We will assume that for an antiferromagnetically-ordering system the interference term in Eq. (15) of Ref. 8 averages to zero for the reasons just described, and this equation may then be written

$$\rho = AV^{2} + A \frac{J^{2}}{[1 - 2JN(0)\ln(D/kT_{eff})]^{2}} \times \left(S(S+1) - \overline{M}_{z} \frac{\sinh x - x}{\cosh x - 1}\right) , \qquad (1)$$

where x = H/T, and $T_{eff} = T$ for $x \ll 1$ and is of the order of H for $x \gg 1$. D is an energy of the order of the bandwidth, and N(0) is the density of conduction-electron states at the Fermi level. The term in large parentheses results from the modification of the equilibrium populations of spin states in a finite field and is similar, though not identical, to the result found by Yosida. The scattering of electrons from impurities in each M_z state has now been modified by many-body effects, and this has led to the replacement of the exchange potential J by a

renormalized potential $J_{\rm eff}$ for the scattering of electrons within an energy kT of the Fermi surface, given by

$$J_{\rm eff} = \frac{J}{1 - 2JN(0)\ln(D/kT_{\rm eff})}$$

The temperature dependence of J_{eff} leads to the approximately logarithmic variation of resistance with temperature for $x \ll 1$, and its field dependence leads to a logarithmic field dependence of the resistivity in the limit $x \gg 1$. The third-order perturbation calculations of Harrison and Klein,⁹ and of Béal-Monod and Weiner,¹⁰ demonstrate that the simple separation of the resistivity into the product of two terms (a modified J and a term in S) is incorrect, though their results are identical to that of Abrikosov⁸ (to third order in J) in the limits of high and low temperatures if T_{eff} is written

$$T_{\text{eff}} = (T^2 + \beta^2 H^2)^{1/2}$$

where $\beta = 0.32$ for $x \ll 1$ and $\beta = 0.77$ for $x \gg 1$. (A T_{eff} of this form, but with a temperature-independent β , has been suggested by Suhl.¹⁶)

In the analysis which follows, the spin-dependent resistivity is estimated from the decrease in resistivity due to ordering in internal fields. This, combined with a measurement of the coefficient of the logarithmic term in the resistivity for $x \ll 1$. allows the Kondo temperature to be determined [the Kondo temperature is defined by⁸ $T_{\kappa} = (D/k)$ $\times e^{-1/2JN(0)}$]. Thus, from Eq. (1), the resistivity change $\Delta \rho_m = \rho(T_m) - \rho(0)$ [where $\rho(T)_m$ is the excess resistivity at temperature T_m of the resistance maximum and ρ (0) is the excess resistivity of the alloy, extrapolated to T = 0] is given approximately by $AJ_{eff}^2 S$ (for very low T_K) and the coefficient of the logarithmic term by $-4AJ_{eff}^3 N(0) S(S+1)$. The third-order perturbation result is similar, but with $J_{\rm eff}$ replaced by J. For $T_{\rm K} = 10^{-10} \, {}^{\circ}{\rm K}$, $\Delta \rho_m$ and $d\rho/$ $d\ln T$ are, respectively, 2.2 and 3.3 times larger than the values predicted by third-order theory, and values of J and T_K , deduced assuming the validity of this theory, will be in considerable error. We will therefore assume Eq. (1) to be correct in the limit $T \gg T_K$ and obtain

$$\frac{\Delta \rho_m}{d\rho/d\ln T} \sim \frac{\ln T/T_K}{2(S+1)}$$

From this ratio, and a knowledge of S, a value for the Kondo temperature can be obtained without assuming values for the rather uncertain parameters A and V. It will be assumed that the internal fields can be described by a temperature-independent probability distribution curve P(H) of the kind described in detail by Klein and Brout.¹⁷

It is readily shown that for very low T_K (i.e., $|\ln T_K| \gg 1$), for temperatures in the region of 1 °K, and in the absence of interactions, Eq. (1) can be

written

$$\rho = AV^{2} + B(1 - 2\ln T / |\ln T_{K}|), \qquad (2)$$

where

$$B = AJ^{2}S(S+1)[\frac{\ln(D_{F}/kT_{K})]^{2}}{|\ln T_{K}|^{2}}$$

is the spin resistivity in the absence of internal or external fields at T=1 °K. In the presence of a temperature-independent distribution of internal fields (the validity of this assumption will be discussed below), the resistivity can be written, for $x \ll 1$,

$$\rho = A' - B' \ln T - C'/T^2 + D'/T^4 - O(1/T^6), \qquad (3)$$

where

$$\begin{aligned} A' &= AV^2 + B, \\ B' &= 2B / \left| \ln T_K \right|, \\ C' &= \overline{H^2} \frac{1}{9} B(1 - 2 \ln T / \left| \ln T_K \right| + 9\beta^2 / \left| \ln T_K \right|), \\ D' &= \overline{H^4} \frac{B}{135} \left(S^2 + S + 1 \right) \\ &\times \left(1 - \frac{2\ln T}{\left| \ln T_K \right|} + \frac{15}{4} \frac{(2 + 9\beta^2)}{(S^2 + S + 1)} \frac{\beta^2}{\left| \ln T_K \right|} \right). \end{aligned}$$



FIG. 2. The excess resistivity ρ plotted against $\log_{10} T$. The $(\mathcal{P})^{\circ}$ indicate experimental values of ρ . Crosses represent the quantity $\rho + C'/T^2 - D'/T^4$, which is the expected variation in resistivity in the absence of interactions.



FIG. 3. A plot of A' (open circles) and B' (crosses) against nominal concentration c at. %, where A' is the resistivity at T=1 °K in the absence of interactions and B' is the coefficient of the logarithmic term in the resistivity.

 $\overline{H^2}$ and $\overline{H^4}$ represent the second and fourth moments of the internal field distribution. For $|\ln T_K| \gg 1$ and for temperatures in the range 0.5 < T < 10 °K, the coefficients C' and D' are approximately independent of temperature. The terms in β [which arise from the term T_{eff} in Eq. (1)] give a negligible contribution to C' and D' and will be neglected.

From Eq. (3), the slope $d\rho/d\ln T$ is given by

$$d\rho/d\ln T = -B' + 2C'/T^2 - 4D'/T^4 \tag{4}$$

(assuming that B', C', and D' are independent of temperature and that terms of order $1/T^6$ and higher can be neglected). To determine the coefficients B', C', and D', the measured slopes are analyzed as a power series in $1/T^2$. An expression of the form of Eq. (4) was found to be appropriate for all alloys at temperatures above T_m , and the values of the coefficients so obtained are listed in Table I. The rejection of terms in $1/T^6$ can lead to a substantial underestimate of the coefficient D' of the $1/T^4$ term, and we estimate that this term may be in error by as much as a factor of 2. The coefficient C' can be obtained with more confidence and has a probable uncertainity of $\pm 20\%$. The uncertainty in the coefficient B' of the logarithmic term is $\pm 10\%$ in the 0.2% alloy, and $\pm 5\%$ in the more dilute allovs.

In Fig. 2, the excess resistivity ρ of each of the alloys is plotted against $\ln T$. The crosses represent the values of the quantity $\rho + C'/T^2 - D'/T^4$, which is the expected variation of the resistance

956

in the absence of interactions. The $1/T^4$ term is approximately one-sixth of the $1/T^2$ term at $T = T_m$. The crosses are linear in $\ln T$ (and can be written $A' - B' \ln T$), thus demonstrating the internal consistency of the analysis. Values of A' (the resistivity at $T = 1^{\circ}$ K in the absence of interactions) are also listed in Table I and a plot of A' and B' against nominal concentration is shown in Fig. 3. Both A'and B' are linear in concentration, with slopes

$$A'/c = 2.40 \pm 0.05 \ \mu\Omega \ \mathrm{cm}/\%$$

and

$$B'/c = 0.048 \pm 0.008 \ \mu\Omega \ \mathrm{cm}/\%$$
.

The small intercept in B' at c=0 could be accounted for by the presence of approximately 5 ppm of Fe in solution.

From Eq. (1), the resistivity at T=0 is given approximately by

$$\rho(0) = AV^2 + B\frac{S}{S+1}\left(1 - \frac{2\ln\beta|\overline{H}|}{|\ln T_K|}\right), \qquad (5)$$

where $\overline{|H|}$ is the mean field, and from Eq. (2), the resistivity at $T = T_m$ in the absence of interactions is

$$\rho'(T_m) = A V^2 + B (1 - 2 \ln T_m / |\ln T_K|).$$
(6)

Thus from (3), (5), and (6)

$$\left|\ln T_{K}\right| = 2(S+1)(\Delta \rho_{m}^{\prime}/B^{\prime}) + 2S\ln(T_{m}/\beta \overline{|H|}) + 2\ln T_{m},$$
(7)

where $\Delta \rho'_m = \rho'(T_m) - \rho(0)$. It is shown in the discussion below on internal fields that $T_m/\overline{|H|} \sim 1.5$. Thus, the second term is independent of concentration and is approximately equal to 1.3S.

The positive resistivity contribution due to the breakdown of Matthiessen's rule is apparent in the resistivity of all of the alloys above 8 °K where phonon scattering becomes significant. This prohibits an accurate estimate of B' (the coefficient of $\ln T$) in alloys containing more than 0.2% Mn. Also $\rho(0)$ (and hence $\Delta \rho'_m$) can only be determined reliably for $c \ge 0.05\%$. Thus, an estimate of T_K is limited to the alloys containing 0.05, 0.1, and 0.2% Mn. Using the experimentally determined values of $\Delta \rho'_m$, B', and T_m and assuming a spin $S = \frac{5}{2}$ (as determined from susceptibility^{18,19} and specific-heat²⁰ measurements) we find values of $|\log_{10}T_K| = 10.9$ ± 2 , 12.6 ± 1 , 14.5 ± 2 for the three alloys, and the mean value of the logarithm of the Kondo temperature for AuMn is equal to $\log_{10}T_K = -12.7 \pm 1.5$. Using

$$T_{K} = (D/k) e^{-1/2JN(0)},$$

and assuming the free-electron value for N(0) for Au, we find $J = 0.09 \pm 0.01$ eV. It must be emphasized that the correctness of this value for T_K depends on the validity of Eq. (1) in the limit T $> T_K$, and on the value of $\frac{5}{2}$ assumed for the spin *S*. (If we take *S* = 2, then the mean value of the Kondo temperature is given by $\log_{10} T_K = -10.9 \pm 1.5$ and $J = 0.10 \pm 0.01$ eV.)

IV. INTERNAL FIELD DISTRIBUTION

From the measured coefficients B', C', and D'and using Eq. (3), values of the second and fourth moments of the internal field distribution curve can be obtained, and these are listed in Table II. $(\overline{H^2})^{1/2}$ and $(\overline{H^4})^{1/4}$ provide a measure of the breadth of the distribution curve, and these quantities, divided by the concentration, are plotted in Fig. 4. The fact that $(\overline{H^4})^{1/4}$ is systematically lower than $(\overline{H^2})^{1/2}$ is probably due to the fact that D' is underestimated in the present method of analysis (because of the neglect of terms of the order $1/T^6$).

It is apparent from Fig. 1 that the resistivity at temperatures well below T_m is linear in temperature. Measurements by Macdonald *et al.*² on a Au 0.04% Mn alloy demonstrate that this dependence is valid down to 0.1 °K. Harrison and Klein⁹ and Béal-Monod and Weiner¹⁰ have shown that such a linear dependence can be explained if the probability distribution curve P(H) is finite at H = 0. It may be shown from Eq. (1) that the coefficient of the



FIG. 4. The parameters T_m/c (crosses), $(\overline{H^2})^{1/2}/c$ (triangles), $(\overline{H^4})^{1/4}/c$ (open circles), and 1/2cP(0) (open squares) (in units of °K/at.%) which characterize the internal-field-distribution curve, plotted against nominal concentration c (at.%).

958

linear term is given approximately by

$$\left(\frac{d\rho}{dT}\right)_{T=0} \simeq 4.3 \frac{B}{S+1} P(0) \left(1 - \frac{2\ln\beta |\overline{H}|}{|\ln T_K|}\right), \tag{8}$$

where P(0) is the value of P(H) at H=0. The coefficient 4.3 is calculated assuming $S=\frac{5}{2}$, but it is only weakly dependent on S. From Eqs. (5), (6), and (8), we obtain a ratio

$$\frac{\Delta \rho_m'}{(d\rho/dT)_{T=0}} = \frac{1}{4.3P(0)} \left(1 - 2(S+1) \frac{\ln(T_m/\beta \mid H \mid)}{|\ln T_K|} \right)$$
$$\simeq \frac{0.20}{P(0)} . \tag{9}$$

Values of 1/P(0) obtained from the measured ratios are listed in Table II, and 1/2cP(0) is plotted against concentration in Fig. 4 (we have assumed that the slope measured at 0.5 °K is the same as that at T=0). T_m/c is also included in this figure, and may be compared with the parameters (1/c), $(\overline{H}^2)^{1/2}$, $(1/c)(\overline{H}^4)^{1/4}$, and 1/2cP(0) which characterize the distribution curve.

Several conclusions can be drawn from the results shown in Fig. 4.

(a) The magnitudes of the parameters relative to one another, and therefore the shape of the distribution curve, are approximately independent of concentration.

(b) The quantities $(\overline{H^2})^{1/2}$, $(\overline{H^4})^{1/4}$, 1/P(0), and T_m are not linear in concentration. Although nonlinearity might be expected for the more concentrated alloys, a linear dependence for c < 0.1%would be expected to result from an RKKY interaction between impurities.

(c) The ratio $(\overline{H^4})^{1/4}/(\overline{H^2})^{1/2} \simeq 0.9 \pm 0.2$ for the 0.05, 0.1, and 0.2% alloys. This value may be compared with a ratio of 1.0 for a unique field and 1.3 for a Gaussian distribution. A ratio close to one precludes the possibility of a cutoff Lorentzian distribution with a cutoff significantly greater than the width.

(d) The ratio

$$\frac{1}{P(0)} / (\overline{H^2})^{1/2}$$

TABLE II. Values for the Kondo temperature T_K , and parameters relating to the internal-field-distribution curve for the AuMn alloys.

c at. % (Nomina	1) $\log_{10}T_K$	$(\overline{H^2})^{1/2}$ °K	$(\overline{H^4})^{1/4}$ °K	<i>P</i> (0) °K ^{−1}	T _m °K
0.01		0,23			
0.02		0.51	0.47		0.68
0.05	-10.9 ± 2	0.98	0,93		1.4
0.1	-12.6 ± 1	1.62	1.48	0.172	2.4
0.2	-14.5 ± 2	2.52	2.27	0.112	3.9
0.5				0.057	8
1.5				0.031	20

= 3.7 and 3.5 for the 0.1 and 0.2% alloys, respectively. This is slightly larger than the value 2.5 for a Gaussian distribution and suggests that there is a dip in the P(H) curve at zero fields. This is consistent with the behavior of the specific heat of Au 0.08% Mn and Au 0.16% Mn as measured by du Chatenier, ²⁰ who observed a decrease in C/T(where *C* is the specific heat) below 0.7 and 1.0 °K in the two alloys. The decrease in the slope $d\rho/dT$ of the resistivity of the Au 1.5% Mn alloy (Fig. 1) below 2 °K also indicates the existence of a dip in the P(H) curve at very low fields.

(e) The RMS field $(\overline{H^2})^{1/2}$ is approximately equal to 0.65 T_m and corresponds closely to the temperature of the maximum of the specific heat of AuMn alloys of comparable concentration.²⁰

(f) The fact that the deviation from the logarithmic temperature dependence varies at high temperatures like $1/T^2$ and that the RMS field so obtained is comparable with the mean field at T $\ll T_m$ [as estimated approximately from P(0)] strongly suggests that the field distribution is not varying strongly with temperature and makes plausible the assumption of a temperature-independent field distribution in the above analysis. This is also true of other dilute alloy systems such as AuFe,²¹ CuMn,²² etc. This conclusion is, at first sight, inconsistent with the behavior of the hyperfine field as determined by Mössbauer-effect measurements in AuFe alloys.²³ The Mössbauer splitting is observed to increase rapidly from zero as the temperature falls below a Curie temperature T_{C} which is comparable to the temperature of the specific-heat maximum in AuFe alloys of a similar concentration, and this suggests that the internal fields increase cooperatively below T_c , as in a simple ferro- or antiferromagnet.

However, the resistivity responds to the mean field averaged over a time of the order of 10⁻¹⁴ sec, whereas a well-defined Mössbauer splitting is only observed if the mean field at the impurity site is nonzero when averaged over a time of the order of 10^{-8} sec. Thus, it may be speculated that at $T > T_c$, the *local field* at a given impurity site precesses in a time very much shorter than 10^{-8} sec and produces no Mössbauer splitting but affects the resistivity and specific heat. As T approaches T_c , the precession time of the local field increases rapidly and exceeds 10^{-8} sec at temperatures below T_c , and a Mössbauer splitting is observed. However, it must be emphasised that from resistivity measurements we can demonstrate that the magnitudes of the precessing fields do not change substantially from above to below T_c . (There is no sharp change in the slope of the resistance or speccific heat around T_{c} .) The comparability of the magnitudes of the local fields both in the ordered and disordered states is not surprising in a dilute

random alloy where the average coordination number of impurities is close to one. It is only in concentrated alloys, or pure ferro- or antiferromagnets, where the coordination number is large, that the local field averages to a small value at high temperatures, but increases cooperatively below a rather well-defined transition temperature.

V. DISCUSSION

Another estimate of J can be obtained if the RKKY expression²⁴ for the interaction between two impurities separated by the mean interimpurity distance is compared with the experimental value of $(\overline{H^2})^{1/2}$. Taking $(\overline{H^2})^{1/2} \sim 1.6$ °K for c = 0.1% and assuming $S = \frac{5}{2}$, we obtain J = 1.1 eV. This is more than an order of magnitude larger than the value of J = 0.09eV derived from the logarithmic term in the resistivity. Blandin²⁵ has pointed out that this discrepancy results from the assumption that J is a δ -function potential in the derivation of the RKKY interaction. If it is assumed that J is an extended potential of l = 2 symmetry, which scatters each of the ten l=2 partial waves incoherently, then we obtain a value of J = 0.22 eV from the mean interaction energy. If it is further assumed that the interaction energy is proportional to $J_{eff}^2(T)$ rather than J^2 , then, since $J_{\rm eff}(T)/J \sim 1.5$ at $T = 1 \,^{\circ} {\rm K}$ for $T_{\rm K} = 10^{-13}$ $^{\circ}$ K, we obtain J = 0.15 eV, which is in reasonable agreement with the value obtained from the logarithmic term in the resistivity.

The magnitude of the potential scattering AV^2/c and the spin resistivity at T=1 °K, B/c, can be estimated from the measured values of A'/c and B'/c. Using Eq. (3), we find

 $AV^2/c=1.7 \ \mu\Omega \ \mathrm{cm/at.\%}$

and $B/c = 0.71 \ \mu\Omega \ \mathrm{cm/at.\%}$.

Abrikosov's expression, Eq. (1), in zero field, is the high-temperature limit (for $V/E_F \ll 1$ and for s-wave scattering) of an expression given by Fischer,²⁶ in which the potential scattering is described by a potential phase shift δ_v . For an extended potential, scattering partial waves of l symmetry, and assuming $JN(0) \ll 1$, Fischer²⁶ finds that the resistivity may be written

$$\rho = (2l+1) \frac{1}{2} \rho_0 c \left\{ 1 - \cos 2\delta_v \ln(T/T_K) \right. \\ \left. \times \left[\left(\ln(T/T_K) \right)^2 + \pi^2 S(S+1) \right]^{-1/2} \right\},$$
(10)

where $\rho_0 = 4\pi\hbar/z' e^2 k_F$ is the *s*-wave unitarity limit, k_F is the Fermi wave vector, and *z'* is the number of conduction electrons per atom. Assuming the validity of this expression at T=0, we may estimate the resistivity of AuMn at T=0 in the absence of interactions. Comparing Eqs. (2), (3), and (10), we have

$$AV^{2}/c = (2l+1)\rho_{0}\sin^{2}\delta_{v},$$

$$B/c = (2l+1)\rho_{0}\cos 2\delta_{v} \left[\pi^{2}S(S+1)/4(\ln T_{F})^{2}\right],$$

and the predicted resistivity at T=0 (in the absence of interactions)

$$\rho'(0) = (2l+1)\rho_0 \cos^2 \delta_n$$
.

Using the measured values of A'/c and B'/c and asuming $S = \frac{5}{2}$, we find

 $\sin \delta_v = 0.23 \pm 0.02$, $\rho'(0) = 28 \pm 7 \ \mu\Omega \ cm/at.\%$, and

 $(2l+1)\rho_0 = 30 \pm 7 \ \mu\Omega \ cm/at.\%$

Although these values depend strongly on the value assumed for S and the estimated value of T_K , the predicted value of $(2l+1)\rho_0$ is in reasonable agreement with the free-electron value of the *d*-wave unitarity limit in Au alloys of 21 $\mu\Omega$ cm/at.%. [The values deduced assuming S = 2 are $\log_{10}T_K = -10.9 \pm 1.5$, $AV^2/c = 1.8\mu\Omega$ cm/at.%, $\rho'(0) = 22$ $\mu\Omega$ cm/at.%, and $(2l+1)\rho_0 = 24 \ \mu\Omega$ cm/at.%.] The expression for T_K derived by Fischer,²⁶

$$T_{K} = (D/k) \exp[-1/2JN(0)\cos^{2}\delta_{v}]$$
,

differs from that used in our previous discussion as the result of an interference between spin and potential scattering. (This higher-order interference does not average to zero in the absence of a magnetic field as does the first-order interference between J and V discussed in Sec. III.) Using this expression for T_K and the value deduced for $\cos^2 \delta_v$, we find a value of $J = 0.10 \pm 0.01$ eV from the estimated value of T_K .

The rather small value (1.7 $\mu\Omega\,cm/\%$) of the predicted resistivity at $T \sim E_F$ demonstrates that the up- and down-spin subbands of the virtual state are well separated on either side of the Fermi level, with *d*-wave phase shifts at the Fermi level for up- and down-spin electrons close to π and 0, respectively. This is, therefore, consistent with the measured spin value of $\frac{5}{2}$, which also indicates total polarization of the virtual state. A very small value of the antiferromagnetic J resulting from the covalent admixture of conduction electrons and local d orbitals^{25,27} would be expected in this case, and the resulting J may be further reduced by normal ferromagnetic atomic exchange, leading to the very small value 0.10 eV which we observe.

The predicted resistivity at T = 0 of $28 \pm 7 \ \mu\Omega$ cm/at.% follows from the assumption of resonant *spin* scattering of all ten l = 2 partial waves at that temperature. The change in phase shift from $\sim \pi$ to $\sim \pi/2$ for the up-spin electrons and from ~ 0 to $\sim \pi/2$ for the down-spin electrons takes place mostly within an energy range of the order of kT_K of the Fermi energy.

VI. CONCLUSIONS

From the behavior of the low-temperature resistivity, the internal field distribution has been shown to approximate to a Gaussian, but with a dip in the P(H) curve at low fields. The results also indicate that the strengths of the fields do not change substantially as the temperature increases from well below to well above the ordering temperature.

Use has been made of the fact that T_K (and hence J) can be derived from the *vatio* of the spin resistivity (as determined by the decrease in resistivity on ordering) and the coefficient of the logarithmic term for $x \ll 1$, while the resistivity in the absence of interactions at T = 0 and $T \sim E_F$ can be derived from the *magnitudes* of these quantities. From measurements on dilute AuMn alloys we obtain $T_K \sim 10^{-13}$ °K and a T = 0 resistivity of $28 \pm 7 \ \mu\Omega$ cm/%. This latter value is consistent with the resonant scattering of all ten l = 2 partial waves, as expected for the compensation of a Mn atom of bare spin $\frac{5}{2}$.

The possibility of inferring the existence of a Kondo condensation so far below the lowest temperatures available experimentally is associated with the small physical scale of the isolated magnetic atoms which are being compensated. Thermal fluctuations spread the transition over many decades in temperature and produce a measureable

*Work sponsored in part by the Air Force Materials Laboratory (AFSC) through the European Office of Aerospace Research (OAR), U.S. Air Force, under contract No. F61052-68-C-0011.

- ^{1}A . N. Gerritsen and J. O. Linde, Physica <u>18</u>, 877 (1952).
- ²D. K. C. MacDonald, W. B. Pearson, and I. M.
- Templeton, Proc. Roy Soc. (London) <u>A266</u>, 161 (1962). ³J. W. Loram, T. E. Whall, and P. J. Ford, Phys.
- Rev. B <u>2</u>, 857 (1970). ⁴J. Kondo, Progr. Theoret. Phys. (Kyoto) <u>32</u>, 37

(1964).

⁵M. Z. Kohler, Physik <u>126</u>, 495 (1949); M. J. Rice, Phys. Rev. Letters <u>23</u>, 1108 (1969); H. Smith and J. W. Wilkins, *ibid*. 24, 221 (1970).

⁶J. S. Dugdale and Z. A. Baszinski, Phys. Rev. <u>157</u>, 552 (1967).

- ⁷K. Yosida, Phys. Rev. <u>107</u>, 396 (1957).
- ⁸A. A. Abrikosov, Physics <u>2</u>, 61 (1965).
- 9 R. J. Harrison and M. W. Klein, Phys. Rev. <u>154</u>, 540 (1967).

 10 M. T. Béal-Monod and R. A. Weiner, Phys. Rev. 170, 552 (1968). 11 R. More and H. Suhl, Phys. Rev. Letters 20, 500

¹¹R. More and H. Suhl, Phys. Rev. Letters <u>20</u>, 500 (1968).

¹²P. E. Bloomfield, R. Hecht, and P. R. Sievert, J. Appl. Phys. <u>40</u>, 1101 (1969).

effect at temperatures far higher than T_{K} . By contrast, a superconducting transition, which involves every site in the metal, produces little modification of the properties above the transition temperature, and its existence cannot be demonstrated if it falls below the lowest attainable temperature.

Although most of the properties of a dilute magnetic alloy are modified by the Kondo effect at temperatures far in excess of T_{κ} , few can be measured with sufficient accuracy to determine a very low T_{κ} . Thus magnetic susceptibility, nuclear orientation, Mössbauer effect, and nuclear magnetic resonance, which rely for their estimate of a very low T_{κ} on the detection of very small deviations from a Curie Law (1/T temperature depen-)dence), are extremely susceptible to uncertainties in specimen temperature, interactions between impurities, etc., which themselves lead to deviations of the kind that are sought. The effect on the impurity resistivity is to produce a deviation from a constant value and this, coupled with the very high inherent accuracy of resistivity measurements, enables a reliable estimate of a very low T_K to be made.

¹⁶H. Suhl, Phys. Rev. Letters <u>20</u>, 656 (1968).

¹⁷M. W. Klein and R. Brout, Phys. Rev. <u>132</u>, 2412 (1963).

¹⁸G. Gustafson, Ann. Physik 25, 545 (1936).

¹⁹O. S. Lutes and J. L. Schmit, Phys. Rev. <u>134</u>, A676 (1964).

 20 F. J. du Chatenier, J. de Nobel, and B. M. Boerstoel, Physica <u>32</u>, 561 (1966).

²¹An analysis of the internal field distribution, which is similar to that described here, has recently been applied to the AuFe system. P. J. Ford, T. E. Whall, and J.

W. Loram, Phys. Rev. B 2, 1547 (1970).

²²A. N. Gerritsen and J. O. Linde, Physica <u>18</u>, 877 (1952).

²³R. J. Borg, R. Booth, and C. E. Violet, Phys. Rev. Letters <u>11</u>, 464 (1963).

²⁴K. Yosida, Phys. Rev. <u>106</u>, 893 (1957).

²⁵A. Blandin, J. Appl. Phys. <u>39</u>, 1285 (1968).

²⁶K. Fischer, J. Phys. Chem. Solids <u>29</u>, 1227 (1968).
 ²⁷J. R. Schrieffer and P. A. Wolff, Phys. Rev. <u>149</u>,

491 (1966).

960

¹³We replace $g\mu_B H/k$ by H for simplicity.

 $^{^{14}{\}rm We}$ assume J positive for an antiferromagnetic exchange coupling. $_$

¹⁵In the discussion which follows, \overline{M}_z and M_z^2 are the mean and mean square values of M_z , averaged over a distance of the order of the conduction-electron mean-free path.