indicates a finite divacancy contribution and $\Delta K = 0.95$. This value of ΔK would change f_i(ISO) in Table III to 0.43.

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Effect of Interactions on the Kondo Resistivity of Dilute *Au*Fe Alloys*

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Measurements are presented of the resistivity of a number of *Au*Fe alloys with concentrations varying from 0.002- to 2- at.% Fe in the temperature range 0.45–40 °K. Very dilute *Au*Fe alloys have already been found to obey an equation due to Hamann. The more concentrated alloys studied here show departures from Hamann's equation due to interactions between the impurities, and these have been taken into account by assuming that they modify the Hamann equation by a factor $(1-\gamma'/T^2+\delta'/T^4)$. It is found that for concentrations greater than 0.01-at.% Fe, the spin resistivity no longer increases linearly with concentration; it is suggested that this is due to correlations between partially formed spin-compensated states. It is also found that the internal field distribution in *Au*Fe alloys can be approximated by a Gaussian. In the most concentrated alloys, the low-temperature resistivity varies linearly with temperature with a slope which is independent of concentration, in agreement with theoretical prediction.

I. INTRODUCTION

In a previous paper,[1] hereafter referred to as I, measurements were presented of the electrical resistivity of some *Au*Fe, *Cu*Fe, and *CuAu*Fe alloys between 0.45 and 300 °K. From these measurements, it was possible to determine Kondo temperatures and find temperature and concentration dependences of the resistivity, so that the theoretical predictions of various authors could be tested.[2] In this paper the resistivity of the *Au*Fe system is studied to higher concentrations, and the results are analyzed by taking into account the effect of interactions between impurities.

Most of the theoretical work on dilute magnetic alloys assumes that the alloys are sufficiently dilute to neglect interactions between the impurities. However, in practice, a long-range magnetic ordering occurs through the Ruderman-Kittel-Yosida (RKY) interaction and this suppresses the spin scattering and can give rise to a resistance maximum. The problem of interactions between impurities in a random alloy has been treated by Marshall[3] and by Klein and Brout[4] in terms of a probability distribution of internal fields. The impurities are assumed to be subjected to effective fields; the value of these fields can be described by a continuous probability distribution function $P(H)$. They showed that the $P(H)$ curve is symmetrical about $H=0$ with a finite probability $P(0)$ of having a zero field, which is inversely proportional to the concentration of impurities.

An early pre-Kondo attempt to account for the resistance maximum is due to Yosida,[5] who calculated the resistivity to second order in perturbation theory. At very high temperatures the resistivity is proportional to $J^2S(S+1)$, of which two-thirds comes from spin-flip scattering. At absolute zero the resistivity is proportional to J^2S^2, the spin-flip scattering freezing out and making no contribution.

More recently, Silverstein[6] put forward a theory in which he showed that the Kondo expression for the spin resistivity is modified by a factor due to the internal field distribution. In the low-concentration limit he made a moment expansion in even powers of H/T and showed that the concentration dependence of the temperature of the resistance maximum depended on the form used for the distribution of internal fields.

Abrikosov[7] calculated the resistivity due to transition-metal impurities by developing a special quantum-field-theory technique for the spin system. He extended his calculations to consider the effect of a unique internal field[8] and found that his original expression was modified by a factor due to the field, this factor having a form similar to that derived by Yosida.[5] Abrikosov predicted that close to absolute zero the resistivity should be varying exponentially with temperature, a result due to his use of a unique internal field rather than a distribution of internal fields.

A detailed calculation of the effect of internal fields on the spin resistivity was made by Harrison and Klein.[9] A similar calculation was made by Béal-Monod and Weiner[10] in their study of the effect of an applied field on the resistivity. In both papers the resistivity was calculated to third order in perturbation theory. Well below the resistance maximum the resistivity is predicted to vary linearly with temperature with a slope which is independent of the impurity concentration, assuming a finite $P(0)$ which is inversely proportional to the concentration. The temperature of the resistance maximum was found to increase linearly with the concentration but, in contrast to the conclusions of Silverstein,[6] it did not depend on the form of the internal field distribution.

II. EXPERIMENTAL DETAILS

The *Au*Fe system is a very suitable one to examine experimentally. The solubility of iron in gold is around 1 at.% at room temperature, rising to 11 at.% at 300 °C.[11] There are therefore few metallurgical problems with this alloy system. In addition, while iron contamination during the preparation of the specimens can seriously affect the behavior of many other dilute alloy systems, it is obviously unimportant in the *Au*Fe system. The concentration of the alloys varied from a few at. ppm to 2-at.% Fe. The specimens were in the form of wires several cm long and 0.025 cm in diameter. Their preparation is identical to that described in I. Resistivity measurements were made between 0.45 and 40 °K using the normal four-probe technique. Six specimens, five alloys and one pure gold, were measured during each experimental run. On several occasions different samples of alloys with the same nominal concentration were measured; these have shown that the reproducibility of the measurements was very satisfactory. The nominal and analyzed concentrations of the alloys are given in Table I.

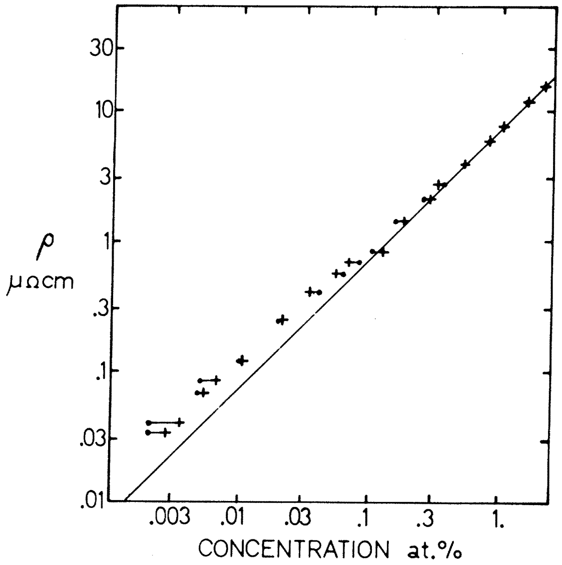

FIG. 1. ρ(μΩ cm) versus iron concentration in at.%. The closed circles indicate nominal concentrations, estimated from the initial weights of the materials, and the crosses indicate the concentration as determined by chemical analysis. The 45° line passes through the most concentrated alloys and shows that the resistivity tends to a value of 7.6 μΩ cm/at.%.

III. RESULTS

Figure 1 is a plot of the resistivity at 1 °K as a function of the concentration of impurities. The resistivity of the more concentrated alloys varies linearly with concentration and tends to a value of

TABLE I. Nominal and analyzed iron concentrations in at.% and the coefficients A, B, C, and D in μΩ cm evaluated from the analysis.

Nominal Fe conc. (at.%)	Analyzed Fe conc. (at.%)	A (μΩ cm)	B (10^{-2} μΩ cm)	C (10^{-4} μΩ cm)	D (10^{-4} μΩ cm)
0.002	0.0028	0.029	1.6 ± 0.1	...	...
0.0025	0.0037	0.032	2.1 ± 0.1	...	...
0.005	0.0053	0.048	3.1 ± 0.2	...	...
0.005	0.0070	0.060	4.3 ± 0.2	2.5 ± 0.1	...
0.01	0.011	0.089	6.1 ± 0.2	9.2 ± 0.7	0.8 ± 0.2
0.02	0.025	0.200	12.2 ± 0.4	108 ± 12	37 ± 6
0.04	0.035	0.341	18.4 ± 0.6	386 ± 45	218 ± 50
0.06	0.056	0.493	23.2 ± 1.0	910 ± 110	909 ± 240
0.08	0.071	0.647	27.2 ± 1.4	1510 ± 200	1820 ± 570
0.10	0.13	0.782	30.4 ± 2.0	2870 ± 600	5830 ± 2600

7.6 μΩ cm (at.%), which is in good agreement with the value quoted by Gerritsen.[12] Deviations away from linearity are observed for the most dilute alloys. This is partly due to uncertainties in the concentration and to the presence of nonmagnetic impurities. More important is the larger spin resistivity contribution in the dilute alloys, which becomes progressively suppressed by magnetic ordering as the concentration increases.

Figures 2 and 3 show the impurity resistivity $\Delta\rho$, the difference between the alloy resistivity and the pure-gold resistivity, as a function of temperature up to 15 °K. A resistance maximum is first observed above 0.5 °K in a 0.04-at.% Fe alloy. As the concentration of impurities increases, the average interaction energy becomes stronger and the temperature of the resistance maximum increases approximately linearly with the impurity concentration, in agreement with theoretical prediction.[9] Above 0.2-at.% Fe a separate resistance maximum and minimum are no longer observed. These alloys are sufficiently concentrated that the decrease in the resistivity due to Kondo scattering takes place in a temperature region where phonon scattering has become important. The Kondo resistance is overshadowed by the positive-resistance contribution resulting from deviations from Matthiessen's rule.[13]

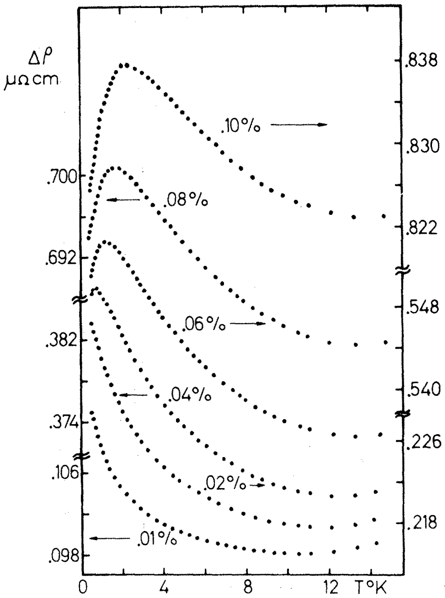

FIG. 2. $\Delta\rho$(μΩ cm) against T for *Au*Fe alloys. The nominal iron concentration in at. % is indicated for each alloy.

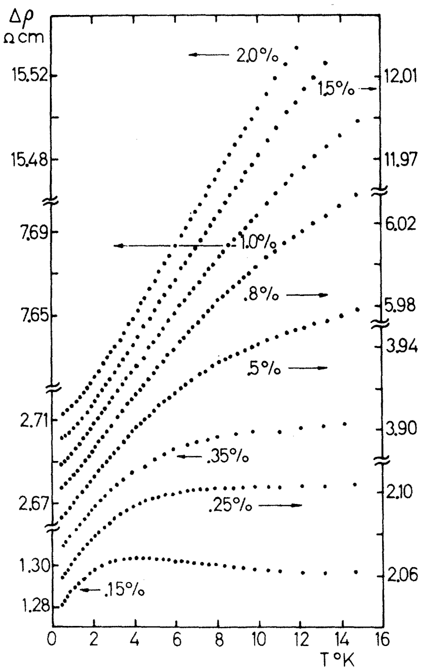

FIG. 3. $\Delta\rho$(μΩ cm) against T for *Au*Fe alloys. The nominal iron concentration in at. % is indicated for each alloy.

IV. ANALYSIS OF RESULTS

In I resistivity measurements were presented of some very dilute *Au*Fe alloys, with concentrations less than 25-at. ppm Fe. It was observed that the resistivity of the dilute alloys showed a positive upward curvature when plotted against $\ln T$, instead of the linear variation in $\ln T$ as predicted by Kondo.[14] An analysis was made on the 25-at. ppm Fe alloy and it was found that the resistivity could be fitted to an expression due to Hamann[15] with an impurity spin S of 0.77 and a Kondo temperature T_K of 0.24 °K. It was demonstrated that the dilute alloys obeyed the equation

$$\rho = A + \tfrac{1}{2} B F(T), \qquad (1)$$

where

$$F(T) = 1 - \left(1 + \frac{13.5}{[\ln(T/0.24)]^2}\right)^{-1/2} \qquad (2)$$

is the temperature-dependent part of Hamann's[15] equation with $S = 0.77$ and $T_K = 0.24$ °K, and A is the temperature-independent contribution due to potential scattering from the iron and from nonmagnetic impurities. A plot of ρ against $F(T)$ was linear for all the dilute alloys.

In Fig. 4 the curves labeled 1 are similar plots of $\Delta\rho$ against $F(T)$ for two more concentrated alloys containing 0.02- and 0.06-at.% Fe. It can be seen that, in contrast to the very dilute alloys, these more concentrated alloys show strong deviations away from linearity at the lowest tempera-

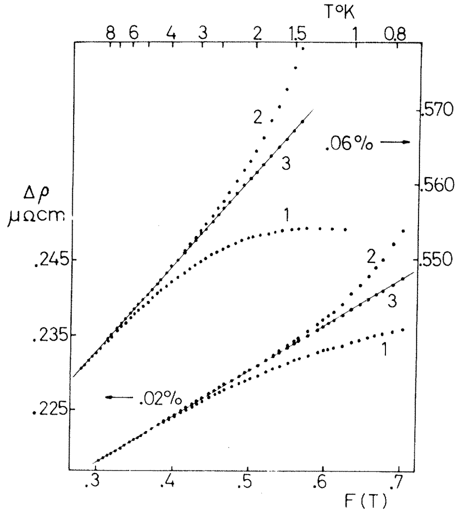

FIG. 4. $\Delta\rho$($\mu\Omega$ cm) against $F(T)$ [defined in Eq. (2)] for *Au*Fe alloys containing nominal iron concentrations of 0.02 and 0.06 at.%. Curve 1 is the plot with both interaction terms present. Curve 2 shows the effect of adding the interaction term $CF(T)/T^2$, and curve 3 shows the effect of adding the terms $CF(T)/T^2 - DF(T)/T^4$.

tures, which can be attributed to interactions between the impurities.

In order to account for the presence of internal fields we have assumed that the resistivity can be written as a function of H/T; in addition, only even functions of H/T are considered. We have also assumed that the distribution of internal fields is not temperature dependent and that all the cross terms involving interference between potential and spin scattering average to zero, as is the case for an antiferromagnetic system.[5] We postulate that in the presence of internal fields Eq. (1) takes the form

$$\Delta\rho = A + \tfrac{1}{2}BF(T_{\text{eff}})[1 - \gamma/T^2 + \delta/T^4], \qquad (3)$$

where γ and δ are proportional to the second and fourth moments, respectively, of the internal field distribution. Equation (3) is similar to expressions derived by Silverstein[6] and by Abrikosov[8] in that the resistivity has been written as a product of two temperature-dependent terms. The term $[1 - \gamma/T^2 + \delta/T^4]$ results from a redistribution of the population of the levels of the spin system due to the internal fields. To account for the effect of a magnetic field on the scattering of electrons from impurities in a *given* spin state, we have replaced the temperature by an effective temperature T_{eff} in the expression for $F(T)$, the temperature-dependent part of Hamann's equation. Following the suggestion by Suhl[16] we assume T_{eff} is equal to $(T^2 + H^2)^{1/2}$, and using this expression it can easily be shown that Eq. (3) reduces to

$$\Delta\rho = A + \tfrac{1}{2}BF(T)(1 - \gamma'/T^2 + \delta'/T^4), \qquad (4)$$

where γ' and δ' are functions which are only slowly varying with temperature and will be assumed to be temperature independent. Equation (4) would be expected to be a good approximation at temperatures far above the Kondo temperature where the effects due to spin compensation can be neglected.[17]

Referring to curves 1 of Fig. 4, the gradients $\rho' = d(\Delta\rho)/dF(T)$ were evaluated for various values of $F(T)$. By comparison with Eq. (4) these gradients are given by

$$\rho' = \frac{d(\Delta\rho)}{dF(T)} = \frac{B}{2} - \frac{C}{T^2}\left(1 - \frac{2F(T)}{F'(T)}\right) + \frac{D}{T^4}\left(1 - \frac{4F(T)}{F'(T)}\right), \qquad (5)$$

where $C = \frac{1}{2}B\gamma'$, $D = \frac{1}{2}B\delta'$, and $F'(T) = dF(T)/d\ln T$. From Eq. (4) it can be seen that a plot of ρ' as a function of $(1/T^2)[1 - 2F(T)/F'(T)]$ should be linear when the term in D/T^4 is negligible and will deviate away from linearity when this term begins to become important. Such a plot is given in Fig. 5 for the 0.06-at.% alloy. The dots represent calculated values of $(1/T^2)[1 - 2F(T)/F'(T)]$, and the crosses represent the calculated values of the additional term $(D/T^4)[1 - 4F(T)/F'(T)]$. The value of the coefficient D and the position of the straight line were adjusted in a self-consistent manner so as to obtain the best possible fit to linearity. The line has intercepts on the axes of $\frac{1}{2}B$ and $B/2C$ enabling these two coefficients to be determined. Curves 2 and 3 of Fig. 4 show the plots of $\Delta\rho$ against $F(T)$ where interactions between the

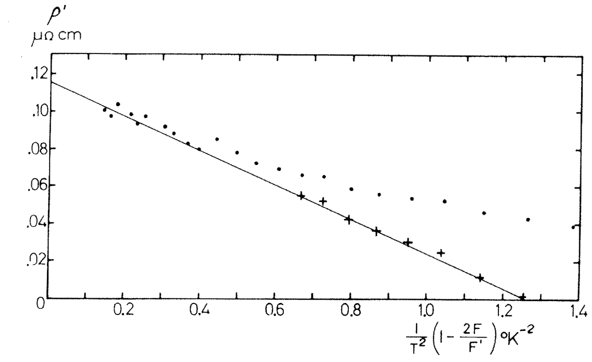

FIG. 5. $\rho' = [d\Delta\rho/dF(T)]$($\mu\Omega$ cm) against $(1/T^2)$ $[1 - 2F(T)/F'(T)]$(°K^{-2}). The closed circles are values of $(1/T^2)[1 - 2F(T)/F'(T)]$ for different values of ρ'. The crosses indicate the addition of the term $(D/T^4)[1 - 4F(T)/F'(T)]$.

impurities have been taken into consideration. Curves 2 represent the effect of adding the interaction term $CF(T)/T^2$, and curves 3 represent the effect of adding $CF(T)/T^2 - DF(T)/T^4$. Curves 3 show that when interactions between the impurities have been taken into account, $\Delta\rho$ varies linearly in $F(T)$ in a manner similar to the very dilute alloys. The resistivity at $F(T)$ equals zero, i.e., $T=\infty$ gives the value of A, which is the resistivity due to potential scattering from the iron and from nonmagnetic impurities.

V. DISCUSSION

The analysis described in Sec. IV was made on several alloys with concentrations varying between 0.005- and 0.1-at.% Fe. It was not possible to make the analysis on alloys with a higher concentration, because then it would have to be made in a temperature region where phonon scattering is important and the Kondo resistivity is overshadowed by the resistivity arising from deviations from Matthiessen's rule. Because of this deviation, the analysis is limited to the temperature region below 10 °K. For each alloy the coefficients A, B, C, and D were evaluated, which are shown in Table I.

A plot of A, the potential scattering from the iron and from nonmagnetic impurities, is linear in concentration to within the errors of both the nominal and analyzed concentrations. Because of these errors we have taken A to be the best measure of the iron concentration, the concentration in at.% being given by $A/7.6$, where A is expressed in $\mu\Omega$ cm. Figure 6 is a plot of B, the coefficient of the temperature-dependent part of Hamann's equation, against A. According to Hamann, and to all the theories of the spin resistivity due to isolated impurities, B should be proportional to concentration, whereas from Fig. 6 it can be seen that B deviates away from linearity at concentrations greater than about 0.01-at.% Fe. The rather detailed set of measurements and analysis presented here confirms our preliminary observations.[18]

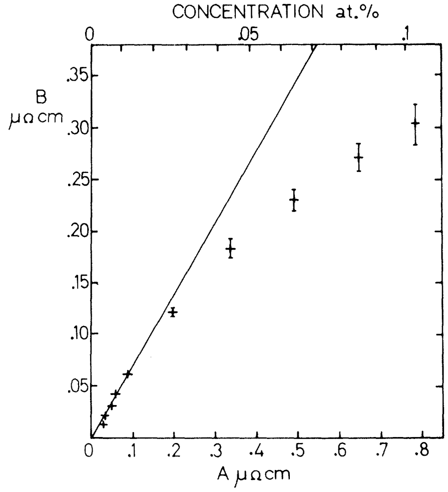

FIG. 6. $B(\mu\Omega\,\mathrm{cm})$, the coefficient of the spin resistivity term, as a function of $A(\mu\Omega$ cm). The approximate concentration, taken as equal to $A/7.6$ at.%, is also given.

The nonlinearity in the concentration dependence of the spin resistivity suggests that there are correlations between the impurities in addition to those due to the RKY interaction. Nagaoka[19] showed that, well below the Kondo temperature, the range of the conduction-electron polarization is very long and that there are correlations between the spin-compensating clouds. In I the Kondo temperature of *Au*Fe was found to be 0.24 °K and therefore the temperature range of the present analysis is only about an order of magnitude greater than the Kondo temperature. The nonlinearity of B suggests that there are still considerable correlations between spin-compensating clouds to temperatures well above the Kondo temperature. Further evidence of the large amplitude of the conduction-electron polarization comes from some recent susceptibility measurements on dilute *Au*Fe alloys[20] which indicate that at 2 °K the spin value of the iron impurity is only about 60% of the bare iron spin. In addition, some measurements on the resistivity of dilute *Au*Mn alloys,[17] which were made in the same temperature range as the *Au*Fe alloys, show that B varies much more linearly with the concentration, which is consistent with the very much lower Kondo temperature (less than 10^{-6} °K) which is observed in *Au*Mn.

In Eq. (4) the coefficient γ' is proportional to $\langle H^2\rangle$, the second moment of the internal field distribution; and the coefficient δ' is proportional to the fourth moment $\langle H^4\rangle$. It can be shown that for a cut-off Lorentzian with a half-width proportional to the concentration and a cutoff independent of concentration, both the second and the fourth moments are proportional to the concentration. For a Gaussian distribution with a half-width proportional to the concentration, $\langle H^2\rangle$ is proportional to the square of the concentration and $\langle H^4\rangle$ to the fourth power. From Fig. 7 the coefficient of γ' was evaluated to be 2.0 ± 0.3 and δ' to be 3.3 ± 0.5. This indicates that the probability distribution of internal fields in the *Au*Fe system approximates a Gaussian or, alternatively, a cut-off Lorentzian, but where the cutoff is proportional to the concentration.

It has already been mentioned that deviations from Matthiessen's rule prevented an analysis on

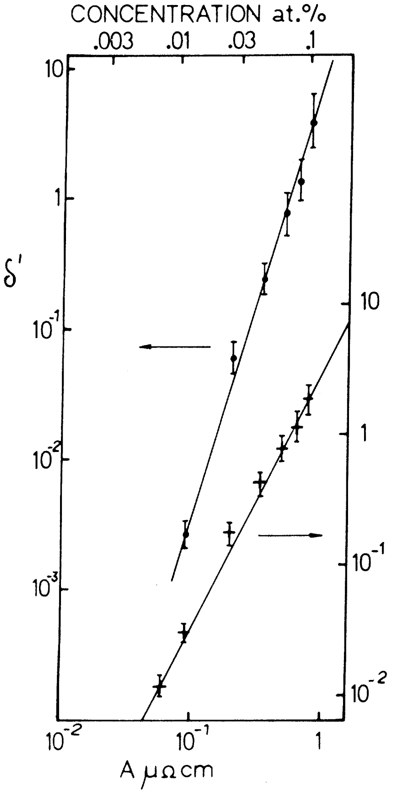

FIG. 7. γ' and δ', the coefficients of the two interaction terms, as a function of $A(\mu\Omega\,\mathrm{cm})$. The approximate concentration, taken as equal to $A/7.6$ at. %, is also given. The scale is logarithmic.

alloys with concentrations greater than that of 0.1-at. % Fe. However, from Fig. 3 some general features can be seen in these more concentrated alloys. At temperatures which are well below the resistance maximum the resistance can be seen to increase linearly with the temperature and roughly independently of the concentration in agreement with the predictions of Harrison and Klein[9] based on a simple $P(H)$ model with $P(0)$ proportional to $1/c$. At the very lowest temperatures the resistivities of the three most concentrated alloys show a slight curvature. The low-temperature resistivity reflects the $P(H)$ curve, and the curvature indicates a dip in the $P(H)$ curve for low fields. The $P(H)$ curve is also reflected in the low-temperature specific heat. Low-temperature specific-heat measurements on some concentrated *Au*Fe alloys by Dreyfus *et al.*[21] also indicate a similar dip in the $P(H)$ curve for low fields. This is being examined in more detail with resistivity measurements on some more concentrated *Au*Fe alloys.

VI. CONCLUSIONS

We have studied the electrical resistivity of a number of dilute *Au*Fe alloys over a wide range of concentrations. It was found in I that it was possible to fit the very dilute alloys to an expression due to Hamann[15] using a Kondo temperature of 0.24 °K and an impurity spin of 0.77. The more concentrated alloys studied here showed departures from Hamann's equation due to interactions between the impurities and it has been assumed that these modify the resistivity by a factor $[1-\gamma'/T^2+\delta'/T^4]$. The factor contains the first terms of a moment expansion in $1/T^2$. When one approaches the resistance maximum, higher-order terms have to be taken into consideration. Because these terms were neglected, the analysis can be considered valid only above the temperature of the resistance maximum and one would also expect fairly substantial errors in the coefficient of $1/T^4$. When internal fields are not present in the concentrated *Au*Fe alloys, they obey the Hamann equation in the same manner as the dilute alloys. However, the analysis described in this paper does not depend on the Hamann function, $F(T)$ being a unique fit to the dilute alloys.

From the analysis it has been found that the spin resistivity only increases linearly with the concentration of impurities up to about 0.01-at. % Fe, after which there is a systematic deviation away from linearity. This has been interpreted to be due to correlations between spin-compensating states, the temperature range of the analysis being only about an order of magnitude greater than the Kondo temperature of *Au*Fe. The resistivity measurements are supported by susceptibility measurements which indicate that there is still a large amplitude of the conduction-electron polarization at 2 °K. From the coefficients of the two interaction terms γ' and δ', it has been deduced that the internal field distribution in the *Au*Fe system approximates either a Gaussian or alternatively a cut-off Lorentzian, but where the cutoff increases linearly with the concentration. The effect of deviations from Matthiessen's rule prevented an analsis on alloys with concentrations greater than 0.1-at. % Fe. In these more concentrated alloys it was observed that the resistivity increased linearly with temperature and with a slope which is independent of the concentration, in agreement with theoretical prediction.

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Trigonal Warping of the Energy Surfaces in Tellurium

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The period of the Shubinkov–de Haas oscillations, when the magnetic field is applied in the plane perpendicular to the c axis of single-crystal heavily doped Te($p = 4.8\times10^{18}$ cm^{-3}), is observed to depend on the azimuthal angle. This angular dependence gives clear experimental evidence of trigonal warping of the constant-energy surfaces about the k_z axis in the Brillouin zone.

We present clear experimental evidence that the constant-energy surfaces in crystalline Te exhibit trigonal warping about the k_z axis of the Brillouin zone. This type of warping is expected on the basis of recent theoretical calculations[1,2] of the band structure, but has not been observed unambiguously, heretofore, experimentally. Our evidence is deduced from the angular dependence of the Shubnikov–de Haas (SdH) period as the applied magnetic field is rotated in the plane perpendicular to the crystallographic c axis.

Theoretical and experimental results[3–8] to date indicate that the constant-energy surfaces of crystalline p-Te consist of pairs of neighboring ellipsoids which are prolate in the k_z direction. In highly doped samples, these ellipsoids merge, as shown in Fig. 3(a), and the Fermi surface becomes dumbbell shaped. The center of this dumbbell is located at the H point of the Brillouin zone as in Fig. 3(c). Recent calculations[1,2] of the valence-band structure near the H point predict the existence of trigonal warping about the k_z axis. In particular, the $E(k)$ relation of the uppermost valence band was found to be of the form

$$E(k) = Ak_\perp^2 - Bk_\perp^4 - Ck_x(k_x^2 - 3k_y^2) + Dk_z^2 + (S_1^2k_z^2 + 4\Delta_1^2)^{1/2} - \Delta_2, \qquad k_\perp^2 = k_x^2 + k_y^2. \tag{1}$$

The third term in Eq. (1) describes the trigonal warping as may be seen when it is written in polar coordinates as $Ck_\perp^3\cos3\varphi$, where φ is the aximuthal angle. As is well known, the plane of the orbit of a charge carrier in a strong magnetic field is perpendicular to the direction of the applied field. Our method of studying the trigonal warping is to cause a hole to traverse an orbit in the plane par-