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Magnetic Field Depreciation of a Kondo System

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The electrical resistance of the Kondo system $Cu_{95}Au_5$-Fe, similar to Cu-Fe, has been measured in magnetic fields to 50 kG in the temperature range 0.4–100 K. The presence of the gold constituent reduces the normal magnetoresistance effects to negligible values, and field depreciation of the Kondo state can be accurately determined. The effects of magnetic field in this respect are much larger than the earlier results by Monod would indicate.

Recently several papers have discussed anomalous spin splitting of Landau levels in Kondo and in ferromagnetic alloys, observed in the de Haas–van Alphen effect.[1–3] A number of studies concerning the effects of spin-flip scattering on the de Haas–van Alphen amplitude have appeared as well.[4–7] It is not yet clear what role the spin-splitting experiments have to play as a new tool to investigate the Kondo state, apart from the immediate fact that they depend upon the real part of the itinerant-electron self-energy. Whatever this role may be, the experimental regime accessible to such measurements involves magnetic fields sufficiently great to be a large perturbation on the zero-field Kondo system, at least for the Cu-Cr and Cu-Fe systems studied to date.[1,8] It would be desirable that effects of large magnetic fields on the spin compensation be determined by independent measurements. Unfortunately, measurements of the static magnetic susceptibility or effects which depend on it have so far been difficult to interpret.[9,10]

The electrical resistance is comparatively simple and insensitive to small concentrations of precipitated solute. However, the magnetic field experiments to date have been seriously flawed by the method of separation of normal magnetoresistance effects from the direct effect of magnetic field on the Kondo state. With a number of separate scattering mechanisms, a conglomerate measurement results which cannot be unraveled by applying a magnetoresistance factor to, e.g., the temperature-independent term. Even if the electrical resistivity in large magnetic fields were a linear combination of all scattering mechanisms, which is not the case, it is not clear what magnetoresistance factor would be appropriate to the Kondo term itself to remove the normal magnetoresistance. With this uncertainty, measurements on Cu-Fe by Monod show depreciation of the low-temperature-resistivity step by approximately 2.5% at 20 kG,[11] which compares with approximately 50% for Cu-Cr at the same field.[12] The much larger effect for Cu-Cr cannot be accounted for by scaling the field according to the ratio of Kondo temperatures, a factor of approximately 8, since the depreciation for Cu-Cr at 2.5 kG is approximately 10%. It is likely that both studies underestimate the effects of magnetic field on the spin-compensated state, with a larger error for Cu-Fe where the field is in effect much smaller with respect to the spin impurity but the same magnitude for the normal magnetoresistance.

The electrical-resistance measurements do not test the real part of the electron self-energy, but are related to the imaginary part. However, the spin compensation is negligible when the Kondo resistivity is negligible, they both increase monotonically with decreasing H and T, and both are expected to saturate near $H=0$, $T=0$. The relative strengths of field and temperature in depreciating the Kondo state, as evidenced in either real or imaginary parts of the electron self-energy, can be gauged according to

$$Sg\mu_B H_K = kT_K , \tag{1}$$

subject to confirmation by experiment. Equation (1) is obtained from the convergence condition for Abrikosov's complete summation of the "most divergent" graphs for the electron self-energy.[13] Relation (1) without the factor S has often been suggested on the basis of low-order perturbation theory,[14] since, as one can easily verify, the asymptotic form of the series expansion is not achieved until order $4S$. (This is most easily seen by drawing graphs of the Goldstone expansion of the ground-state energy.) For $S=\frac{1}{2}$, Kurata has shown that the imaginary part of the *impurity-spin* self-energy is identically zero for $H \geq H_K$ at $T=0$,[15] subject to the same limitations as the Nagaoka theory at zero field.[16] Unfortunately, no simple generalization of the Kurata theory to arbitrary S is possible, due to the well-known difficulties in representing a general spin by a fermion field.

To overcome difficulties in relating magnetoresistance measurements to fundamental properties of a Kondo system, a number of experiments have been performed on a series of polycrystalline $Cu_{95}Au_5$-Fe alloys. In zero field the presence of the gold constituent allows accurate determination of the Kondo resistivity to higher temperatures by reducing departures from Matthiesen's rule when

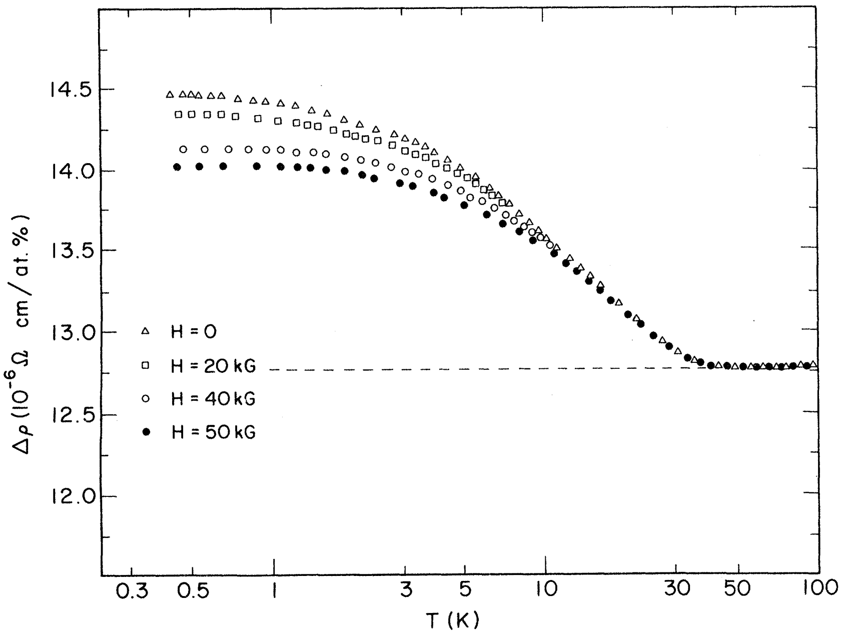

FIG. 1. Dependence of the function $\Delta\rho = [\rho(\mathrm{Cu}_{95}\mathrm{Au}_5\text{-Fe}) - \rho(\mathrm{Cu}_{95}\mathrm{Au}_5)]/c$ on temperature and magnetic field.

separating the electron-phonon resistivity. What is most important for the purpose of this communication, however, is that at fields up to 50 kG and temperatures as low as 0.4 K, measurements on a $\mathrm{Cu}_{95}\mathrm{Au}_5$ reference alloy show no magnetoresistance at the 10^{-4} level. We therefore conclude that for all scattering mechanisms in $\mathrm{Cu}_{95}\mathrm{Au}_5$-Fe alloys, with very little Fe, the normal magnetoresistance effects can safely be regarded as negligible. In Fig. 1, the effect of magnetic field on $\Delta\rho = [\rho(\mathrm{Cu}_{95}\mathrm{Au}_5\text{-Fe}) - \rho(\mathrm{Cu}_{95}\mathrm{Au}_5)]/c$ is shown. The Fe concentrations were 0.04 and 0.08 at.% with negligible concentration dependence for $\Delta\rho$. In *resistance* measurements the precision was 10^{-4} and the absolute accuracy 10^{-3}. $\Delta\rho$ was measured directly with ternary and reference alloys in excellent thermal contact, using two opposed specimen currents according to geometric factors accurate to considerably better than 0.5%. Measurements on two separate pairs for 0.08-at.% Fe yielded the same results to considerably better than this level. Temperature was measured using a single four-lead fine-carbon-grain thermometer from 0.4 to 100 K. The magnetic field was provided by a superconducting solenoid in the persistent mode. Voltages were measured to 10^{-8} V with a current-reversing potentiometer-galvanometer-amplifier technique using specimen currents of 15 mA or less. For all of the measurements we have $\Delta\rho \gtrsim 0.1\rho(\mathrm{Cu}_{95}\mathrm{Au}_5)/c$. Details of the specimen preparation will be published later, and I comment only that starting materials were the purest presently available, and checks of sample homogeneity by electron-microprobe analysis indicate excellent specimen quality.

I regard the field dependence of electrical resistivity in Fig. 1 as indicative of field depreciation of the spin-compensated state in a similar sense to temperature depreciation of the resistivity. Furthermore, measurements at zero field show the $\mathrm{Cu}_{95}\mathrm{Au}_5$-Fe alloy to be representative of the Cu-Fe alloy but with T_K reduced by a factor of approximately 0.65.[17, 18] Field depreciation of the Kondo resistivity is much greater than indicated by Monod's earlier results for Cu-Fe,[11] with approximately 30% depreciation at 50 kG near $T = 0$. In Fig. 1, 50 kG is equivalent to 5 K with respect to depreciating the resistivity. (The observed field-to-temperature ratio should not change with T_K if S remains the same.) Assuming $S = \frac{3}{2}$ in Eq. (1) for Fe in either Cu[19] or $\mathrm{Cu}_{95}\mathrm{Au}_5$ leads to a ratio 5 kG/K. The 10-kG/K ratio of Fig. 1 would require $S \approx 0.75$, a value which was obtained by Loram *et al.* on the basis of their *zero-field* results compared to the Hamann theory.[17, 20] The correct value for S perhaps cannot be obtained with assurance from zero-field resistivity measurements and at low temperatures may be affected by electron-electron interactions not included in the present theoretical structure. However, this agreement does strongly suggest that temperature and field depreciation of the Kondo resistivity are accurately related by Eq. (1).

Heeger has estimated a scaling relation 50 kG/(20 K) by combining NMR and Mössbauer data,[21] which would require the unrealistic value $S \approx 3$ in Eq. (1). The uncertainty associated with Heeger's estimate is both large and difficult to estimate accurately, particularly with regard to other contributions to the NMR linewidth.

With regard to the earlier discussion of the normal magnetoresistance, for Cu-Cr the spin-compensated state is probably considerably more depreciated at 25 kG than the magnetoresistance results of Daybell and Steyert would suggest,[12] although the error may be considerably smaller than for Cu-Fe. Their results for Cu-Cr show 25 kG as at least approximately equivalent to 2.5 K with respect to depreciation of the Kondo-resistivity step, the same 10-kG/K ratio as I have obtained for Cu-Fe or $Cu_{95}Au_5$-Fe, with depreciation of comparable magnitude. Although values for S at low temperatures are a major uncertainty, I conclude that the general relation of Eq. (1) is at least approximately correct. These measurements remove the ambiguity which previously existed for Cu-Fe,[11, 21] regarded as a model Kondo system.

For Cu-Cr[1] and Cu-Fe[8] the de Haas–van Alphen field regime $H \gtrsim 30$ kG corresponds to considerable depreciation of the Kondo resistivity. The form of the perturbation expansions for resistivity vs $\langle S_z \rangle$ at temperatures greater than T_K and fields above H_K[14] suggests that in these regimes spin compensation is even smaller than the resistivity would suggest. For the Cu-Cr system with $H_K \approx 20$ kG, spin compensation should be small for fields greater than 30 kG, with a large almost "bare" spin more characteristic of a non-Kondo regime in the spin-splitting experiments.[1, 2]

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[1] P. T. Coleridge and I. M. Templeton, Phys. Rev. Letters 24, 108 (1970).

[2] S. Hornfeldt, J. B. Ketterson, and L. R. Windmiller, Phys. Rev. Letters 23, 1291 (1969).

[3] H. Miwa, T. Ando, and H. Shiba, Solid State Commun. 8, 2161 (1970).

[4] F. T. Hedgcock and W. B. Muir, Phys. Rev. 129, 2045 (1963).

[5] G. Holt and A. Meyers, Physik Kondensierten Materie 9, 23 (1969).

[6] H. Nagasawa, Solid State Commun. 7, 257 (1969).

[7] B. E. Faton and W. B. Muir, Phys. Rev. Letters 20, 732 (1968).

[8] P. T. Coleridge and I. M. Templeton (private communication) (results of de Haas–van Alphen experiments for Cu-Fe).

[9] A. J. Heeger, Solid State Phys. 23, 385 (1969).

[10] J. L. Tholence and R. Tournier, Phys. Rev. Letters 25, 867 (1970).

[11] P. Monod, Phys. Rev. Letters 19, 1113 (1967).

[12] M. D. Daybell and W. A. Steyert, Phys. Rev. Letters 20, 195 (1968).

[13] A. A. Abrikosov, Physics 2, 61 (1965).

[14] Reference 9, pp. 355 and 356.

[15] Y. Kurata, Progr. Theoret. Phys. (Kyoto) 43, 621 (1970).

[16] Y. Nagaoka, Phys. Rev. 138, A1112 (1965); Progr. Theoret. Phys. (Kyoto) 37, 13 (1966).

[17] J. W. Loram, T. E. Whall, and P. J. Ford, Phys. Rev. B 2, 857 (1970).

[18] Reference 9, p. 379.

[19] C. M. Hurd, Phys. Rev. Letters 18, 1127 (1967).

[20] D. R. Hamann, Phys. Rev. 158, 570 (1967).

[21] Reference 9, p. 401.