Low-Temperature Behavior of Kondo Impurities and Check of the Fermi-Liquid Model

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The magnetic scattering rate of Fe in Mg is measured as a function of temperature by means of weak localization. It shows a strong temperature dependence which is attributed to the Kondo effect. At low temperatures Nozières predicted a Fermi-liquid behavior for the electron system plus the Kondo impurity with no magnetic scattering. Weak localization allows a check of the applicability of the Fermi-liquid theory.

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The magnetic properties of dilute and concentrated Kondo systems are attracting considerable interest. In connection with the heavy-fermion context there is a large effort to understand concentrated Kondo systems. Intensive theoretical work (see, for example, Abrahams[1] and Jones and Varma[2]) has been focused on the problem of two Kondo impurities and the results are rather puzzling. It is therefore of great interest to investigate systems with a low concentration of Kondo impurities. Hopefully they will provide a testing ground for theoretical results in this field and might eventually help to understand heavy-fermion systems.

It is a nice coincidence that weak localization provides us with a new and powerful method to measure magnetic scattering times of magnetic impurities (see the review articles by Al'tshuler *et al.*,[3] Fukuyama,[4] Bergmann,[5] and Lee and Ramakrishnan[6]). Magnetoresistance measurements of disordered thin films correspond to a time-of-flight experiment with conduction electrons which yields their inelastic lifetimes and their spin-orbit and magnetic scattering times. The effect of magnetic impurities on weak localization is quite similar to that on superconductivity. (It yields the scattering of the electronic pair amplitude by magnetic impurities and corresponds to the magnetic pair breaking in superconductivity.)

In this paper I present direct measurements of the temperature dependence of the magnetic scattering rate in a Kondo-type system. (The dependence of the superconducting transition temperature on the impurity concentration yields, in some systems, an indirect measurement of this magnetic scattering rate.[7]) I investigate Fe impurities in quench-condensed Mg. Quench-condensed Mg-Fe alloys have been investigated by Blanc and Belzons.[8] They observed a Kondo-type behavior of the resistance and estimated the Kondo temperature to be about 45 K. Recently the author[9] observed a strong screening of the magnetic scattering rate of Fe impurities in Mg. Furthermore I found an unexpected large spin-orbit scattering due to the Fe impurities which is very essential in the evaluation of the magnetic scattering rate.

In the present experiment I quench-condense a Mg/Fe/Mg/Au sandwich which consists of 30 atomic layers (atola) of Mg, 0.01 atola of Fe, and 5 atola of Mg. This corresponds roughly to an Fe concentration of 300 ppm Fe. In three further evaporation steps I condense 0.05, 0.1, and 0.5 atola of Au on top of the Mg. The Au increases the spin-orbit scattering and the different coverages with Au allow four (independent) checks of the consistency of the evaluation as we see below.

For each sandwich I measure the magnetoresistance at five temperatures (4.5, 6.5, 9.5, 14, and 20 K) in a magnetic field perpendicular to the film. By a careful annealing procedure (up to 55 K) I achieve a resistivity which is constant to within 1% for the whole series of sandwiches.

In Fig. 1(a) the magnetoresistance of the sandwich Mg/Fe/Mg is shown. The left ordinate gives the change of the resistance in ohms. Since the theory yields the change of the conductance in units of $L_{00}=e^2/2\pi^2\hbar$, I have added on the right-hand side a conductance scale in units of L_{00}. The points represent the experimental results. The curves will be discussed below. The magnetoresistance curves after the last Au evaporation are plotted in Fig. 1(b).

The shape of the magnetoresistance curves is determined by three characteristic electronic times, the inelastic lifetime τ_i, the spin-orbit coupling time $\tau_{\text{s.o.}}$, and the magnetic scattering time τ_s of the conduction electrons. The evaluation of the experiment yields two characteristic fields H_S and H_T. These fields are linear combinations of the inelastic, magnetic, and spin-orbit scattering rates. We express these rates by characteristic fields H_i, H_s, and $H_{\text{s.o.}}$, where the field H_i corresponds to the inelastic lifetime τ_i, $H_{\text{s.o.}}$ to the spin-orbit coupling time $\tau_{\text{s.o.}}$, and H_s to the magnetic scattering time τ_s. The characteristic magnetic fields H_n are equivalent to the inverse characteristic times τ_n, i.e., the scattering rates. The relation between the time τ_n and the field H_n is given by

$$H_n\tau_n=\hbar e\rho N/4, \qquad (1)$$

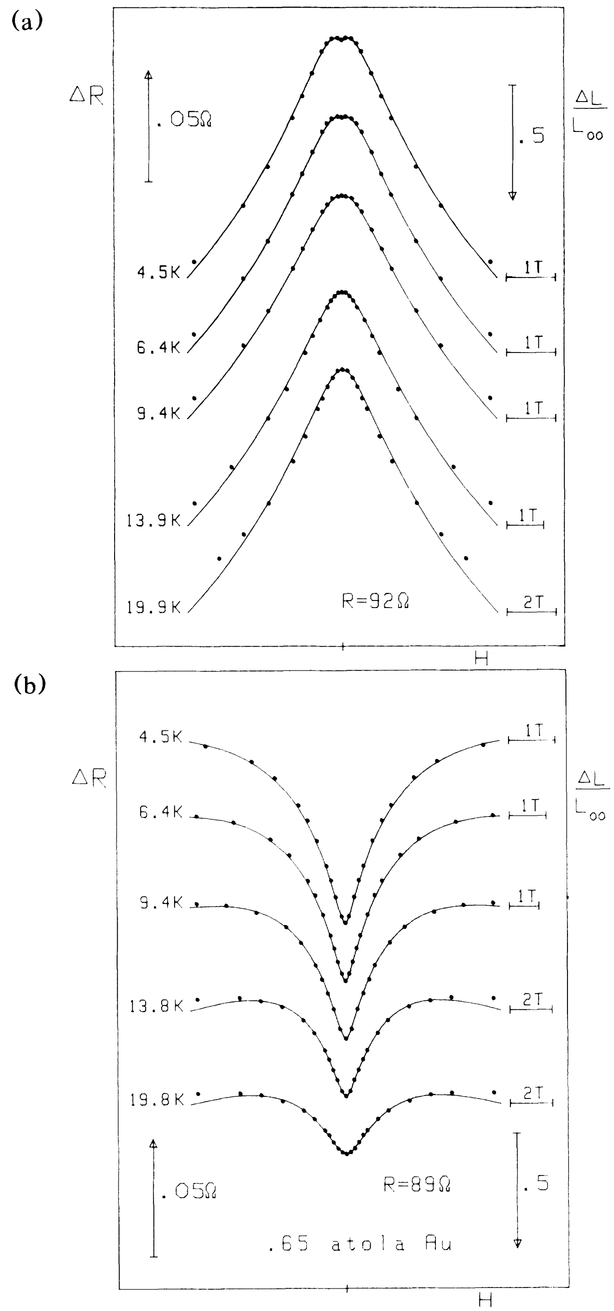

FIG. 1. (a) The magnetoresistance of a Mg/Fe/Mg sandwich with 0.01 atola of Fe. The right arrow gives the units of the conductance. The points represent the experimental results. The curves are calculated with the theory. (b) The magnetoresistance of the same sandwich after deposition of 0.65 atola of Au. The points represent the experimental results. The curves are calculated with the same parameter set as in (a) with the exception of the value $H_{\text{s.o.}}$, the spin-orbit scattering strength, which is produced by the deposition of Au.

where ρ is the resistivity of the film and N is the density of states at the Fermi level (for both spins). For my sandwich the product has the value $H_n \tau_n = 0.42$ ps T$=0.42\times 10^{-12}$ s T.

I use the theory by Hikami, Larkin, and Nagaoka[10] to evaluate the experimental magnetoresistance data. For the discussion later I rewrite their equation for the magnetoconductance ΔL_{wl} in the following form:

$$\Delta L_{wl}/L_{00} = \tfrac{3}{2} f_2(H/H_T) - \tfrac{1}{2} f_2(H/H_S), \tag{2a}$$

where $f_2 = (x) = \ln(x) + \Psi(\frac{1}{2} + 1/x)$, Ψ is the digamma function, and H is the applied magnetic field. The "singlet field" H_S and the "triplet field" H_T are composed of the inelastic, magnetic, and spin-orbit rates, or, more precisely, their corresponding fields:

$$H_S = 2H_s + H_i,$$
$$H_T = \tfrac{4}{3} H_{\text{s.o.}} + \tfrac{2}{3} H_s + H_i. \tag{2b}$$

For the interpretation of the singlet and triplet fields H_S and H_T it is helpful to recall the physics of weak localization. It describes the interface of two partial waves of the same electron. In real space the two partial waves propagate on a closed loop in opposite directions.[3] The interference intensity is equal to the product of the two amplitudes $a_1 a_2^*$. Formally one may reverse the motion of the second partial wave and obtain the product of the amplitude of an electron and its time-reversed counterpart. It is this pair amplitude which one also obtains in the propagation of the Cooper pair in superconductivity. If one includes the spin of the electron in this description then one finds that the interference intensity is mathematically identical with the difference of the triplet and the singlet electron-pair amplitudes (the first has the relative weight of $3 = 2S+1$). The singlet amplitude decays exponentially as a function of time with characteristic decay time τ_S [which corresponds to the field H_S according to Eq. (1)] while the triplet amplitude decays with characteristic time τ_T (corresponding to H_T).

I evaluate the experimental magnetoresistance curves with use of Eq. (2a) and the definition of the characteristic fields. From the form of Eq. (2a) it is obvious that it is a two-parameter equation. Therefore the evaluation of the magnetoresistance curves can only yield two of the three fields H_i, $H_{\text{s.o.}}$, and H_s. We know, however, the inelastic field H_i from the first measurement of the pure Mg film as a function of temperature. Therefore the magnetoresistance of the Mg/Fe/Mg sandwich yields the other two fields H_s and $H_{\text{s.o.}}$ at the various temperatures. The Au coverage changes neither H_i nor H_s and therefore the Mg/Fe/Mg sandwiches with different Au coverages differ only in the spin-orbit scattering field $H_{\text{s.o.}}$. The evaluation yields, for the total Au coverages of 0, 0.05, 0.15, and 0.65 atola Au, the following values for $H_{\text{s.o.}}$ at 4.5 K: 0.09, 0.22, 0.43, and 1.05 T. The $H_{\text{s.o.}}$ values show only a weak temperature dependence which will be discussed below in connection with the Fermi-liquid model.

Figures 1(a) and 1(b) show that one obtains good agreement between the experiment and the Hikami-Larkin-Nagaoka theory[10] for the Mg/Fe/Mg/Au sandwiches with zero and the largest Au coverage. For all four Au coverages (including 0 atola Au) and all temperatures the magnetoresistance curves show the same agreement between the experiment and theory as in Fig. 1(a) and 1(b) with the use of the same set of fields for $H_i(T)$ and $H_s(T)$ and the above values for $H_{\text{s.o.}}$. Therefore we conclude that the evaluation is very consistent and H_s indeed describes the incoherent rate due to the Fe impurities in Mg.

The extracted values for the magnetic scattering field H_s as a function of temperature are plotted in the upper part of Fig. 2. H_s shows a strong temperature dependence and changes in the whole temperature range roughly by a factor of 2. This is the first time that such a strong temperature dependence of the magnetic scattering rate is observed. It demonstrates the anomalous behavior of Kondo-type impurities very clearly.

The temperature dependence of the scattering rate $1/\tau_s$, i.e., H_s, for a more dilute sandwich of Mg/Fe/Mg/Au (with 0.005 atola Fe) is plotted in the lower part of Fig. 2. Although the two sandwiches differ only by a factor of 2 in Fe concentration the corresponding fields H_s at 4.5 K differ by a factor of 4. This quadratic dependence of H_s on the Fe coverage was recently described by the author[9] and attributed to an overlap of the magnetically screening electron clouds.

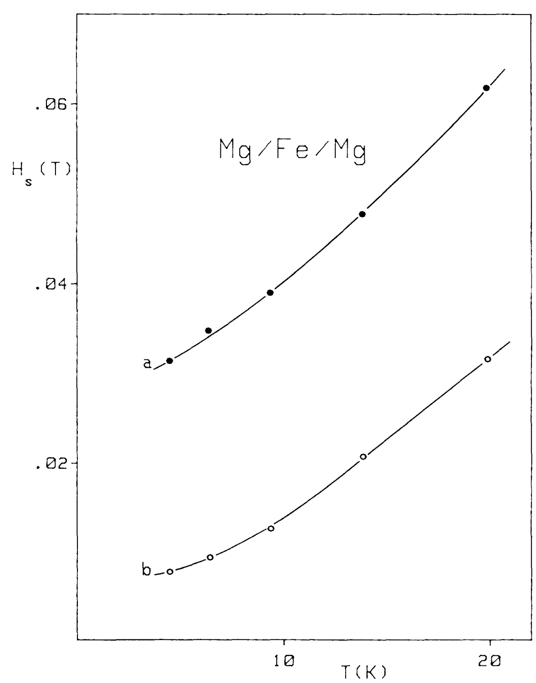

FIG. 2. The magnetic scattering strength (in units of a magnetic field) as a function of temperature. Curve *a* corresponds to 0.01 atola Fe and curve *b* to 0.005 atola Fe. The factors relating H_s and $1/\tau_s$ are 0.42 and 0.44 ps T, respectively.

Up to this point, we have discussed the properties of the magnetic impurities within the context of magnetic (and spin-orbit) scattering. I shall call this model in the following discussion the "magnetic model." This model is used in the Hikami-Larkin-Nagaoka theory[10] of weak localization. One of the fascinating properties of a Kondo system is its low-temperature behavior. On the basis of Wilson's[11] renormalization approach, Nozières[12] pointed out that a Kondo system shows Fermi-liquid behavior at low temperature. This means that the impurity loses its magnetic character. It then yields a change in the density of states and an interaction between the electrons which introduces an additional inelastic scattering. (This is restricted to low energies. Phenomena which include virtual high-energy excitations such as superconductivity may still feel the magnetic impurity.)

This Fermi-liquid theory means that there is no magnetic scattering at sufficient low temperatures but only inelastic scattering. It would be quite interesting to check whether our Kondo system at low temperature shows magnetic or only inelastic scattering. The weak-localization method is in principle capable of distinguishing between the "magnetic" and the "Fermi-liquid" model. The inelastic scattering and the magnetic scattering affect the two relevant fields H_s and H_T differently. According to Eq. (2b) the difference between H_T and H_S is

$$\tfrac{3}{4}(H_T - H_S) = H_{\text{s.o.}} - H_s. \tag{3}$$

The two models have the following properties: (i) In the case of the Fermi-liquid model, H_s is zero at sufficiently low temperature and $\frac{3}{4}(H_T - H_S)$ is equal to $H_{\text{s.o.}}$ and should be temperature independent. (ii) For the magnetic model the term $\frac{3}{4}(H_T - H_S)$ is equal to $H_{\text{s.o.}} - H_s$ and should show the same temperature dependence as H_s. In this model we have $H_s = \Delta H_S/2$ where ΔH_s is the change in the singlet field due to the magnetic impurities. If we plot $\frac{3}{4}(H_T - H_S)$ as a function of $\Delta H_S/2$ it should be a straight line either with the slope zero for the Fermi-liquid model or the slope -1 in the case of the magnetic model (provided that ΔH_s is temperature dependent).

The experiments with Mg/Fe/Mg/Au sandwiches are well suited for this check because I determined the singlet field H_S with high accuracy after the deposition of the additional Au. This yields the total H_S in the presence of the magnetic impurities and ΔH_S as well. H_S does not change with the Au coverage and therefore has the same value in the Mg/Fe/Mg sandwich. Then we can determine H_T for the Mg/Fe/Mg sandwich. Although the magnetoresistance curves show little structure, particularly at higher temperatures, the value of H_T can be determined without ambiguity. In Fig. 3 I have plotted $\frac{3}{4}(H_T - H_S)$ as a function of $\Delta H_S/2$. The experimental points lie roughly on a straight line with the slope -0.7. The uncertainty of the measurement be-

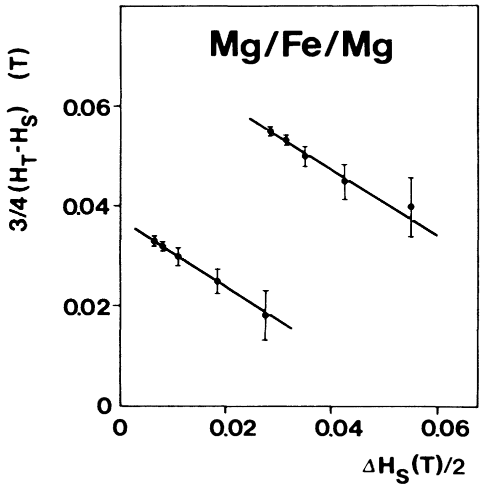

FIG. 3. $\frac{3}{4}(H_T - H_S)$ as a function of $\Delta H_S/2$. For the Fermi-liquid model of the Kondo effect the straight line should be horizontal; for the magnetic model its slope should be -1.

comes greater with increasing singlet field H_S. The data do not agree with a Fermi-liquid model in the temperature range between 4.5 and 20 K. (The Fe impurities appear to be 70% magnetic.)

We have to keep in mind that in a real experiment the spin-orbit scattering field $H_{\mathrm{s.o.}}$ is not completely temperature independent. Even in the absence of magnetic impurities the best fit between experiment and theory often yields a small temperature dependence of $H_{\mathrm{s.o.}}$ of about 15% between 4.5 and 20 K. However, this inaccuracy of the evaluation does not alter our conclusion that the experiment does not support the Fermi-liquid theory for the Fe impurities in Mg.

The deviation of the Mg/Fe/Mg data from the Fermi-liquid theory is most likely due to the interaction between the Kondo impurities. This is supported by the recent experimental result[9] that the magnetic screening of the Fe impurities in Mg depends strongly on the distance between the Fe impurities. On the other hand, it should be emphasized that this is only a first approach to check the Fermi-liquid theory for a Kondo system. At the present time the Kondo effect is not yet incorporated into the theory of weak localization. Ohkawa and coworkers[13,14] have performed calculations for a disordered thin film with Kondo impurities in second Born approximation. This theory is only applicable far above the Kondo temperature. The present experimental investigation will hopefully stimulate further theoretical work in this field.

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