Kondo Maximum of Magnetic Scattering

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The magnetic scattering of Fe atoms in Au is measured as a function of temperature in the range between 0.08 and 4 K by means of weak localization. We find a strong temperature dependence with maximum scattering at the characteristic temperature $T_K \approx 1$ K of this well-known Kondo system. This is a new and direct experimental approach to observation of the theoretically predicted Kondo maximum and is analogous to pair-breaking experiments in superconductivity.

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During the last few years weak localization has been developed into a powerful new method in solid-state physics (see, for example, Bergmann[1] and Lee and Ramakrishnan[2]). Magnetoresistance measurements on disordered thin films correspond to a time-of-flight experiment with conduction electrons, yielding their inelastic lifetime, and spin-orbit and magnetic scattering times. In particular, the opportunity of directly observing magnetic scattering rates promises new insights into the Kondo problem which is recently again receiving considerable interest.[3-5]

The scattering of conduction electrons by Kondo impurities can be separated into two contributions, the spin-nonflip and the spin-flip parts. Only the latter causes dephasing in superconductivity and in weak localization. This has been intensively studied in superconducting alloys where it is possible to obtain this rate from the T_c depression at different concentrations[6] and the dependence of H_{c2} on temperature.[7] These measurements show an increase of the spin-flip rate with decreasing temperature[8] in the temperature range above the Kondo temperature T_K. The predicted maximum of the rate at T_K and its decrease for $T < T_K$ could, however, not be verified by this method. Therefore, the theoretical prediction of a maximum in the spin-flip scattering waited almost 20 yr for an experimental proof. Using the method of weak localization we find for the first time a maximum of the spin-flip rate in the Kondo system *Au*Fe and a clear decrease of this rate below T_K.

Weak localization as a method to measure the spin-flip rate has a number of advantages over the superconducting experiments. It does not require a superconducting host, nor does it need different samples for every point in the T dependence. It can be applied to convenient values of T_K and small concentrations (to stay within the single-impurity limit) and measures the temperature dependence of the spin-flip scattering rate in a wide temperature range.

When weak localization is employed it is favorable to use disordered thin films.[1] To prepare very homogeneous and highly disordered films, they are quench condensed inside a dilution refrigerator while the substrate is maintained at 4 K. This has the very desirable feature that one can produce multilayers step by step and analyze them after each evaporation step. Therefore, it is possible to separate the additional scattering caused by a small fraction of an atomic layer of Fe atoms from the total phase-breaking scattering. It is not necessary to obtain the information for the Au background from a different film.

The *Au*Fe system is well suited to the investigation of the spin-flip scattering around its Kondo temperature T_K. First of all it is a well-known Kondo system with $T_K \approx 1$ K, which is low enough to stay below the region of quadratic temperature dependence of the inelastic electron-phonon scattering in these disordered films,[9] yet by using a dilution refrigerator we are also able to get below T_K by more than an order of magnitude. Because of the large spin-orbit scattering in Au the triplet channel (see below) does not contribute to the magnetoresistance, so that we measure the pure singlet scattering rate which is essentially the only adjustable parameter in the fit for the magnetoresistance curves.

By our operating the refrigerator without the use of exchange gas it works as a cryopump and provides an ultrahigh vacuum. The magnetoresistance measurements are performed in the temperature range between 80 mK and 4 K in fields up to 1.4 T, but in order to avoid polarization of the spins the magnetoenergy $\mu_B H$ is not allowed to exceed the thermal energy $k_B T$ in our experiments. For the resistance measurements we use an ac bridge technique with a Dekatran DT 80 ratio transformer and a PAR model 124 lock-in amplifier, operated at about 1 kHz. The signal is calibrated against an 8-Ω standard resistor. The excitation is low enough that the electron temperature does not exceed the measured pho-

non temperature. This has been proved by measurements of the logarithmic temperature dependence of the film resistance which is caused by the Coulomb interaction. Our films are 0.5 mm wide and have an aspect ratio of 52.

We have investigated three different films. Film A1 is a pure Au film with a thickness of 11.5 atomic layers (atola) on which we have evaporated about 0.0003 atola Fe to make film A2. The evaporation rate for this small amount of Fe is extrapolated from higher rates as described in Ref. 5. Film A3 is the result of 4 atola of additional Au. Before the start of the measurements, the films were annealed at 22 K. The resistivities of the films A1, A2, and A3 took the values (0.24, 0.24, and 0.19) $\times 10^{-6}$ Ω m, respectively.

Figure 1 shows some magnetoresistance curves of the Au/Fe/Au sandwich A3. Each curve has a separate field scale that is indicated at its right-hand side. The left ordinates give the change of the resistance in ohms. Since the theory yields the change of the conductance in units of $L_{00}=e^2/2\pi^2\hbar$ we have added on the right-hand side a conductance scale in these units. The points represent the experimental results; the curves are the theoretical fits resulting from the Hikami-Larkin-Nagaoka (HLN) theory. The measurements show a positive magnetoresistance in the whole field and temperature range. The small fraction of an atomic layer of iron has broadened the magnetoresistance curves. For the evaluation we used the following form of the magnetoconductance ΔL_{wl} in the HLN theory:

$$\Delta L_{wl}/L_{00}=\tfrac{3}{2}f_2(H/H_T)-\tfrac{1}{2}f_2(H/H_S),$$
$$f_2(x)=\ln(x)+\Psi(\tfrac{1}{2}+1/x). \quad (1)$$

Ψ is the digamma function and H the applied field. The H_S and H_T are defined in the following manner:

$$H_S=2H_s+H_i,$$
$$H_T=\tfrac{4}{3}H_{s.o.}+\tfrac{2}{3}H_s+H_i. \quad (2)$$

The characteristic fields H_n are equivalent to the scattering rates $1/\tau_n$ via the relation

$$H_n\tau_n=\hbar e\rho N/4. \quad (3)$$

ρ is the resistivity of the film and N the density of states at the Fermi level for both spins. The constant value of $H_n\tau_n$ for the films A1 and A2 is 0.101 T ps and for the sandwich A3 it is 0.079 T ps. In Eq. (2) the field H_i corresponds to the inelastic lifetime τ_i, $H_{s.o.}$ to the spin-orbit coupling time $\tau_{s.o.}$, and H_s to the magnetic scattering time τ_s. As pointed out by Altshuler *et al.*[10] and recently discussed by Bergmann,[11] H_S corresponds to the lifetime of the singlet-pair amplitude and H_T to the lifetime of the triplet-pair amplitude.

Because of the large spin-orbit scattering in Au the triplet term in the magnetoconductance [Eq. (11)] essentially does not contribute. All magnetoresistance curves could be well described by our taking $H_{s.o.}=1.4$ T so that we have to adjust only one parameter, H_S.

The temperature dependence of the adjusted parameter H_S is plotted in Fig. 2 on a logarithmic temperature scale. For pure Au there should be no magnetic scattering and H_S would be equal to H_i. The temperature dependence of H_S for the pure Au film A1 will be discussed elsewhere. It shows a steep decrease with decreasing temperature between 4 and 1 K, and below 1 K only a slow decrease on the logarithmic scale.

Only a few atoms of Fe (about 25 ppm) on top of the Au film change H_S considerably as is seen from the A2 data in Fig. 2. The values of H_S (and therefore the appropriate scattering rates) are increased by more than 10 mT, and below 0.5 K we now find a steep decrease of H_S with decreasing temperature. The same qualitative behavior is observed after the Fe atoms are covered with Au (film A3). The values of H_S decreased because the additional Au layers dilute the total Fe concentration. However, the scattering rate per impurity remained essentially unchanged.

To separate the influence of the Fe atoms from the Au contribution to H_S it is sufficient to take the difference of H_S between the films A2 and A1 or between A3 and A1. If we assume that the few Fe atoms only change the magnetic scattering time τ_s, this difference is twice its characteristic field H_s. Figure 3 shows this difference as

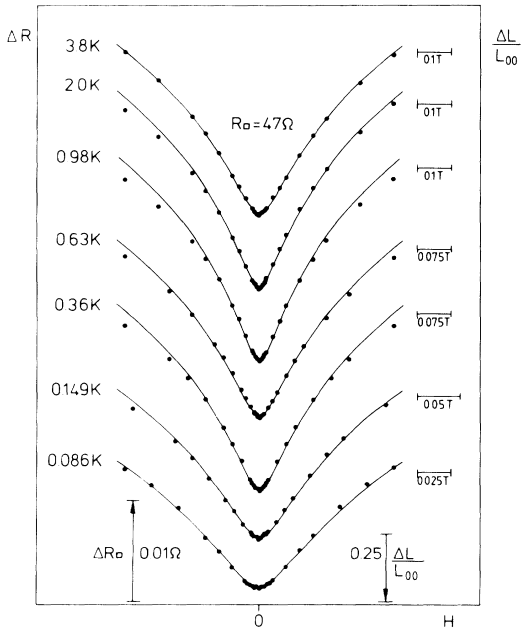

FIG. 1. Selected magnetoresistance curves of the Au/Fe/Au sandwich A3. The field units are indicated at the right-hand side of each curve; the arrows give the scales for resistance as well as conductance changes. The points represent the experimental result; curves are calculated with the HLN theory.

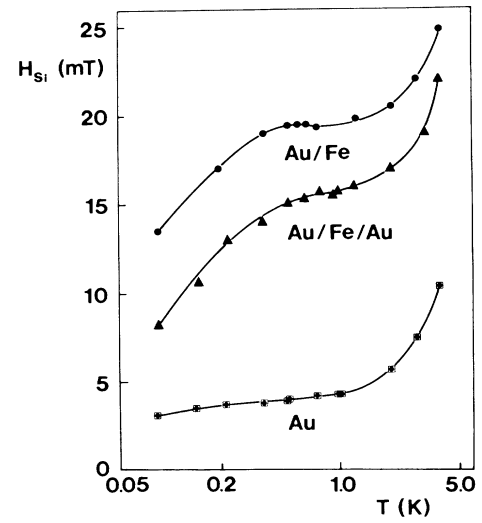

FIG. 2. The characteristic field $H_S = 2H_s + H_i$ of the dephasing rate for the singlet-pair amplitude as a function of temperature T on a logarithmic T scale. The squares indicate the values of the Au film A1, the circles the Au/Fe film A2, and the triangles the Au/Fe/Au sandwich A3.

a function of temperature; it is proportional to the pair-breaking strength of Fe in Au. For both, Fe on top of and Fe inside of Au, there is a maximum of ΔH_S at about 1 K, and below a sharp decrease. This is the first direct observation of the maximum in magnetic scattering in a Kondo system at T_K. The points above 3 K show a somewhat larger uncertainty because of the increase of the inelastic-scattering background. The maximum occurs at a temperature about the same as literature values of the Kondo temperature for the Au-Fe system, e.g., 0.88 K in the work of Daybell.[12] It is remarkable that we find within 30% the same temperature for the maxima with Fe on top of and inside of Au. Instead one might have expected that because T_K depends exponentially on the density of states and the exchange interaction it might be very sensitive to the local surroundings of the Fe atoms.

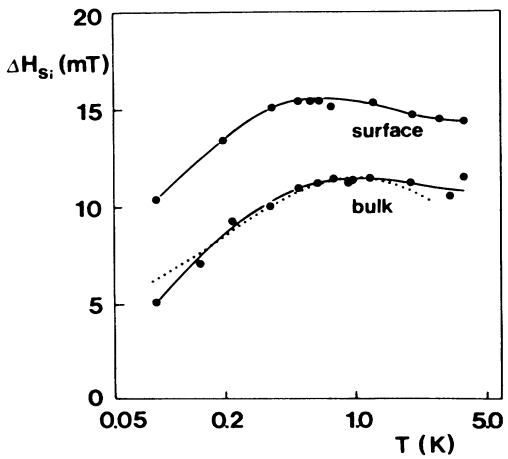

FIG. 3. The increase of H_S due to the 0.0003 atola Fe at the surface of Au (film A2) and in the bulk (film A3) as a function of temperature. ΔH_S is proportional to the singlet spin-flip scattering rate. The absolute values of the two curves differ because the additional Au layers dilute the Fe.

For Kondo systems the maximum of the spin-flip scattering is theoretically expected. Approximate calculations by Nagaoka and Suhl (see, e.g., Fischer[13] and references therein) for single-electron scattering lead to the following expression for the spin-flip scattering rate $1/\tau_s$:

$$1/\tau_s \propto [\ln^2(T_K/T) + \pi^2 S(S+1)]^{-1}. \quad (4)$$

Such behavior for $S = \frac{1}{2}$ is indicated in Fig. 3 by the dotted line. At the lowest temperatures the measured behavior is much steeper. A general problem with this approach is that the Kondo effect enters weak localization in a way more complicated than for single electrons because we are concerned with the phase coherence of two partial waves of an electron. In weak localization one measures all events that destroy this phase coherence. It is very similar to superconductivity where the coherence of two different electrons forming a Cooper pair is essential. The Maki-Thompson fluctuations, for example, are damped by the same processes that limit phase coherence in weak localization.[14] Therefore, we expect that magnetic scattering destroys the pair amplitude in weak localization essentially as it does in superconductivity. One important difference, however, is that for superconductivity there are Cooper pairs with kinetic energies up to $k_B\theta_D$ while the electrons in weak localization only have energies of about k_BT.

The influence of Kondo impurities on superconductivity was studied very intensively at the beginning of the 1970's. The pair breaking by Kondo impurities and the resulting decrease of T_c has been calculated by Müller-Hartmann and Zittartz.[15] They found that the pair-breaking parameter is proportional to $1/\tau_s$ as given by Eq. (4) and has its maximum value at T_K. The disorder which is present in our films should not be responsible for a different Kondo behavior compared to a crystalline *Au*Fe system; e.g., T_K should not be altered by more than 6% as can be estimated by use of the theoretical result of Vladar and Zimanyi.[16] Ohkawa and Fukuyama have already included the Kondo effect in weak localization up to the third order in the coupling constant J.[17] This calculation, however, is only valid for temperatures far above T_K and therefore is not applicable to our results.

We have measured the spin-flip dynamics of Fe impurities on top of and inside of Au by means of weak localization. The corresponding pair-breaking parameter shows a maximum at the Kondo temperature. We hope that our measurements will stimulate further theoretical

work on the Kondo effect and its influence on weak localization.

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