Resonant Kondo Scattering of Weakly Localized Electrons

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Measurements of the magnetic scattering time in very thin Cu films (thickness $t \simeq 5$ nm), homogeneously doped with magnetic Cr atoms (concentration $\lesssim 100$ ppm), are reported. The weak electron localization causes an anomalous magnetoresistance which is very sensitive to the spin-flip scattering of the conduction electrons. The maximum in the temperature dependence of the spin-flip scattering rate is in excellent agreement with the Suhl-Nagaoka approximation for the resonant Kondo scattering in the vicinity of the Kondo temperature $T_K \simeq 2$ K.

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During the last few years, weak electron localization (WEL) has emerged as a new and very reliable method for the study of characteristic scattering times in thin metal films.[1] Measurements of the magnetoresistance in a field perpendicular to the film plane allow a direct calculation of the inelastic, the spin-orbit, and the magnetic scattering times of the conduction electrons. While the inelastic and spin-orbit scattering mechanisms have been investigated in detail,[2] the possibility of the study of spin scattering by local magnetic moments has only been explored recently.[3-5] Although the Kondo effect was introduced more than twenty years ago,[6] the anomalous scattering in dilute magnetic alloys is still receiving considerable theoretical and experimental interest.[7] A detailed understanding of the conduction mechanism in heavy-fermion materials may, e.g., benefit from a better understanding of the Kondo problem.[8]

In this Letter, we show that WEL measurements in Cr-doped Cu films directly reveal the temperature dependence of the Kondo spin-flip scattering in the vicinity of the Kondo temperature $T_K \simeq 2$ K. The transport, magnetic, and thermodynamic properties of bulk Kondo alloys depend in a complex and indirect way upon the Kondo scattering of the conduction electrons.[9] On the other hand, our WEL measurements of dilute CuCr alloys allow a direct observation of the resonant spin-flip scattering around T_K.[10]

WEL in a thin metal film results from an enhanced backscattering probability due to the constructive interference between electron waves traveling along time-reversed paths.[1] Inelastic scattering causes a destruction of the constructive interference after a characteristic time τ_{in}. Magnetic impurities (characteristic scattering time τ_m) also destroy the WEL interference: Scattering of the electron spin by a local magnetic moment destroys the time-reversal symmetry and causes a phase shift between the interfering electron waves. The combined effect of inelastic and magnetic scattering is described by the phase-breaking time τ_ϕ defined as

$$\tau_\phi^{-1} = \tau_{in}^{-1} + 2\tau_m^{-1}. \quad (1)$$

Since superconductivity also results from an enhanced correlation between time-reversed states (Cooper pairs), the pair breaking in a superconductor by magnetic scattering is very similar to the destruction of WEL. Within the Suhl-Nagaoka approximation for the Kondo effect,[10] the lifetime of the Cooper pairs is determined by the temperature-dependent spin-flip scattering rate[11]:

$$\tau_{sf}^{-1} = \frac{c}{2\pi\hbar N(E_F)} \frac{\pi^2 S(S+1)}{\pi^2 S(S+1) + \ln^2(T/T_K)}, \quad (2)$$

where c, $N(E_F)$, and S represent, respectively, the impurity concentration, the density of states near the Fermi energy E_F (for one spin direction), and the impurity spin. Although the equality $\tau_m = \tau_{sf}$ has only been obtained[12] up to the third Born approximation (only valid for $T \gg T_K$), our WEL experiments strongly indicate that τ_m can be identified with τ_{sf} at all temperatures. WEL turns out to be a unique and powerful tool for the determination of small variations of the magnetic scattering rate. It is important to note that Eq. (2) is only valid in the single-impurity limit, where Ruderman-Kittel-Kasuya-Yosida interactions between magnetic moments can be neglected. Since a meaningful depression of the transition temperature in superconducting Kondo alloys only occurs for impurity concentrations $c \gtrsim 1$ at.%, the superconducting method is a less useful check of the validity of Eq. (2).

Strong spin-orbit scattering causes a destructive interference[13] between the backscattered electron waves (weak antilocalization). The spin-orbit scattering is important when the scattering rate

$$\tau_1^{-1} = \tau_\phi^{-1} + \tfrac{4}{3}(\tau_{s.o.}^{-1} - \tau_m^{-1}) \quad (3)$$

is much larger than the phase-breaking rate τ_ϕ^{-1}. From Eq. (3) it is obvious that magnetic scattering counteracts the influence of the spin-orbit interaction.

A perpendicular magnetic field B modulates the phase difference between the interfering electron waves and produces an anomalous magnetoresistance. A detailed calculation shows that this anomalous magnetoresistance

strongly depends upon the scattering times τ_{in}, $\tau_{s.o.}$, and τ_m and is given by[14]

$$[R_\square(B)-R_\square(B=0)]/R_\square^2(B=0)$$
$$=(e^2/2\pi^2\hbar)\{\tfrac{3}{2}\ln[B/B_1(T)]+\tfrac{3}{2}\psi[\tfrac{1}{2}+B_1(T)/B]-\tfrac{1}{2}\ln[B/B_\phi(T)]-\tfrac{1}{2}\psi[\tfrac{1}{2}+B_\phi(T)/B]\}. \quad (4)$$

$R_\square$ is the resistance per square of the thin metal film, and to each characteristic scattering time τ_x there corresponds a characteristic magnetic field B_x:

$$B_x=\hbar/4eD\tau_x. \quad (5)$$

D is the diffusion constant for the elastically scattered electrons.

A thin film of a dilute magnetic alloy can be obtained by evaporation of pieces of a master Kondo alloy from a resistively heated Mo source. Because of distillation effects and a strong tendency to form magnetic clusters when the master alloy is melted, additional precautions have to be taken. We have found that successive flash evaporation of very small pieces (1 mg) of the CuCr master alloy onto a liquid-nitrogen-cooled substrate (Corning glass 7059) ensures the formation of homogeneously doped, continuous Cu films. Our films had a typical resistance per square $R_\square \simeq 30$ $\Omega/\square$ and a thickness $t \simeq 5$ nm. To avoid oxidation of the Cr impurities, the CuCr films are evaporated in a background pressure $p \lesssim 10^{-5}$ Pa and immediately covered with a 100-nm-thick protective layer of SiO. A four-terminal pattern for the resistance measurements was obtained by condensation of the films through a metal mask. Very small resistance variations could be measured by means of an ac resistance bridge and a lock-in amplifier.

The actual concentration c of the Cr impurities in the thin films can be estimated from the logarithmic Kondo anomaly in the temperature dependence of $R_\square$. Since WEL and disorder-enhanced Coulomb repulsion[15] between the conduction electrons also cause a logarithmic divergence, the total normalized resistance increase for a change in temperature of one decade is given by

$$\Delta R_\square=\alpha(e^2/2\pi^2\hbar)R_\square^2\ln 10+\Delta\rho_K/t. \quad (6)$$

The constant α depends upon the ratios $\tau_{in}/\tau_{s.o.}$ and τ_{in}/τ_m.[15] From a systematic study of very pure Cu films ($c<1$ ppm) prepared under similar conditions we found that $\alpha=1.05\pm0.05$ for $20\ \Omega/\square<R_\square<40\ \Omega/\square$. The variation of the resistivity ρ_K caused by the Kondo effect for Cr-doped Cu has been measured previously for bulk CuCr alloys. In the single-impurity limit $\Delta\rho_K=0.50\pm0.05$ nΩ cm/ppm for a temperature change of one decade.[16]

In Fig. 1, we show on the left-hand side the Kondo anomaly in the temperature dependence of the resistance for a properly prepared Cr-doped Cu film. While the initial magnetic impurity concentration c_i of the master alloy was 50 ppm, the actual concentration c calculated from Eq. (6) turns out to be 40 ± 5 ppm. On the right-hand side of Fig. 1, we also show the Kondo anomaly for a film obtained by evaporation of larger pieces of a master alloy with $c_i=100$ ppm onto a substrate held at room temperature. The fine structure is probably caused by the presence of small Cr clusters, each having a characteristic magnetic ordering temperature, where the spin-flip scattering reaches a maximum. The average logarithmic slope indicates a concentration of active Kondo scatterers which is only 30 ± 3 ppm. In a magnetic field $B=2$ T, the fine structure disappears, probably because of the freezing out of the spin scattering.

In Eq. (6), we have neglected a possible modification of the Kondo scattering by the disorder, which may cause an additional $1/T$ divergence of τ_m^{-1} for $T\gg T_K$.[12] For our $R_\square$ values, this additional $1/T$ divergence is too small to be observable (correction $\lesssim 2\%$).

Figure 2 shows the temperature dependence of the low-field anomalous magnetoresistance caused by the WEL for the homogeneously doped Cu film ($c=40$ ppm) which is also shown in Fig. 1. The positive magnetoresistance indicates the presence of an important spin-orbit scattering ($B_{s.o.}=\hbar/4eD\tau_{s.o.}=0.2$ T). This value of $B_{s.o.}$ is calculated from the maximum in the magnetoresistance curve which appears at higher fields. From Fig. 2 we conclude that in sharp contrast to pure Cu films, there occurs a saturation in the temperature dependence of the magnetoresistance for $1\ \mathrm{K}<T<4\ \mathrm{K}$, i.e., in the vicinity of T_K.

This saturation becomes obvious when we plot the temperature dependence of the characteristic field $B_\phi=\hbar/4eD\tau_\phi$ calculated using Eq. (4) (see curves in Fig.

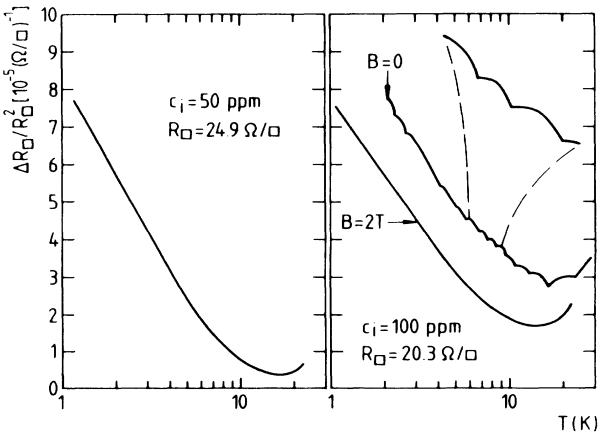

FIG. 1. Temperature dependence of the normalized resistance variation for a homogeneously doped CuCr film (initial concentration $c_i=50$ ppm) and a CuCr film showing cluster effects (initial concentration $c_i=100$ ppm).

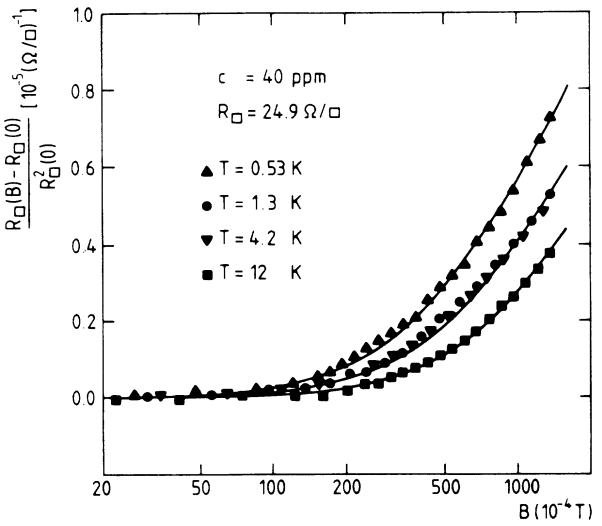

FIG. 2. Low-field normalized magnetoresistance at different temperatures for a CuCr film. The curves represent the calculated magnetoresistance [Eq. (4)].

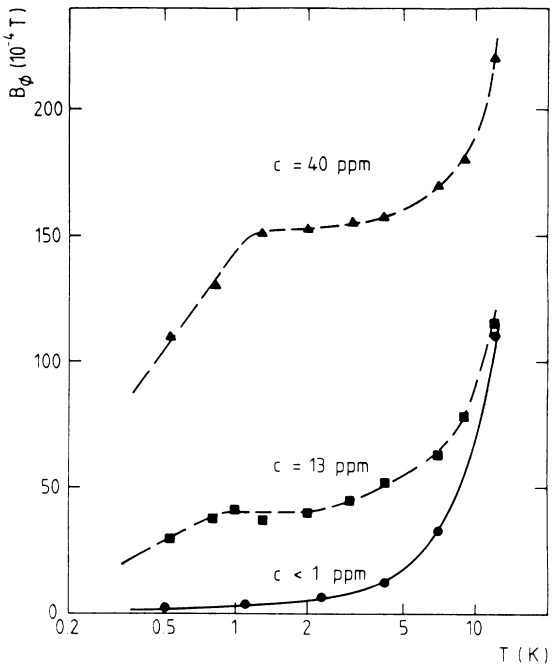

FIG. 3. Temperature dependence of the characteristic field $B_\phi \propto \tau_\phi^{-1}$ for a pure Cu film ($c < 1$ ppm) and two Cr-doped Cu films. The full curve for the pure Cu film is given by Eq. (7). The dashed curves for the doped samples are only guides to the eye.

2). The result is shown in Fig. 3. There we also show the temperature dependence of B_ϕ for a Cr-doped film with $c = 13 \pm 2$ ppm and $R_\square = 33.1$ Ω/□ and for a pure Cu film (prepared by evaporation of 99.9999% pure Cu) with $c < 1$ ppm and $R_\square = 31.1$ Ω/□. The full curve that nicely fits the data for the pure Cu film was calculated by use of a power-law expansion

$$B_\phi(T) = A_0 + A_1 T + A_3 T^3. \quad (7)$$

The constant A_0 corresponds to a small amount of magnetic scattering caused by residual magnetic impurities (Cr,Fe,Ni, . . .) or by paramagnetic surface states. The linear and cubic terms correspond respectively to the disorder enhanced electron-electron scattering[17] and to the electron-phonon scattering. The temperature dependence of the Kondo scattering in the doped Cu films can be obtained by subtraction of the $B_\phi(T)$ values for the clean Cu film from the $B_\phi(T)$ values for the doped films [see Eqs. (1) and (5)]. The small difference in the $R_\square$ values can be taken into account in a straightforward way since $A_0 \propto 1/D$, $A_1 \propto 1/DR_\square$, and $A_3 \propto 1/D$. Using this procedure, we can calculate the temperature dependence of the magnetic scattering rate $\tau_m^{-1} \propto B_m$ shown in Fig. 4 [the diffusion constant is calculated from the resistance ratio $R(300\text{ K})/R(4.2\text{ K})$]. The full curves have been calculated by our using the Suhl-Nagaoka approximation for the spin-flip scattering rate [Eq. (2)] assuming an impurity spin $S = \frac{1}{2}$. $T_K = 3$ K for the 40-ppm film and $T_K = 2$ K for the 13-ppm film. These values for T_K are in good agreement with the value $T_K = 2$ K obtained from measurements on bulk samples.[9] The absolute value of the magnetic scattering rate is for both samples 1.6 times larger than the theoretical value calcu-

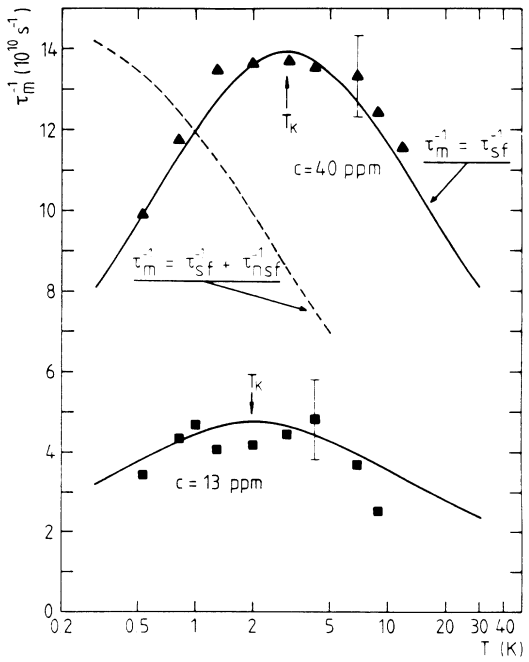

FIG. 4. Temperature dependence of the magnetic scattering rate τ_m^{-1} for the two CuCr samples which are also shown in Fig. 3. The full curves represent the Suhl-Nagaoka approximation [Eq. (2)] for the spin-flip scattering, while the dashed line also takes into account the spin-nonflip scattering.

lated from Eq. (2) if we assume the free-electron value for $N(E_F)$. If we also take into account the spin-nonflip scattering rate[10] τ_{nsf}^{-1} to calculate τ_m^{-1} ($\tau_m^{-1} = \tau_{sf}^{-1} + \tau_{nsf}^{-1}$ is proportional to the Kondo resistivity ρ_K), the agreement with the experiment is very poor as shown by the dashed line in Fig. 4 for the 40-ppm sample. We may therefore conclude that the destruction of the WEL in our Cr-doped Cu films is governed by the spin-flip part of the magnetic scattering. Moreover, the observed $\tau_{sf}^{-1}(T)$ dependence near T_K is in excellent agreement with the Suhl-Nagaoka approximation which starts from the *s-d* exchange model[10] for the magnetic scattering. The simple *s-d* model may fail[7] to describe the $\tau_{sf}^{-1}(T)$ variation in the nonmagnetic limit ($T \ll T_K$).

For small impurity concentrations our procedure to calculate $B_m \propto \tau_m^{-1}$ (see Fig. 3) becomes less accurate, so that WEL cannot provide useful information for $c < 10$ ppm. For higher concentrations ($c > 100$ ppm), the information about the Kondo scattering becomes more reliable, but magnetic interaction effects may no longer be neglected. The results of more detailed experiments in this regime, where spin-glass ordering may occur, will be published elsewhere. For the 40-ppm sample, interaction effects can still be neglected since the scattering rate is, as expected, 3 times larger than for the 13-ppm sample.

To summarize, we have developed a reliable method to prepare quasi-two-dimensional metal films which are homogeneously doped with magnetic impurities. Measurements of the magnetoresistance caused by the WEL in these films enable us to determine the temperature dependence of the Kondo spin-flip scattering rate, which is in excellent agreement with the Suhl-Nagaoka approximation. Similar WEL measurements may also be used to study the influence of Ruderman-Kittel-Kasuya-Yosida interactions between impurities in more heavily doped films.

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