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CuCo: A new surface Kondo system

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The magnetic scattering rates of dilute Co atoms both on the surface and in the bulk of Cu have been investigated by the method of weak localization. For Co on the surface of Cu the temperature dependence of the magnetic scattering rate shows a clear maximum at 23 K. This demonstrates that Co on the surface of Cu is a Kondo system with a Kondo temperature $T_K = 23$ K. However, covering the Co with about three atomic layers of Cu removes the magnetic scattering.

It has been more than twenty years since the Kondo effect was discovered in dilute magnetic alloy systems.[1] The temperature dependence of the magnetic scattering rate was predicted to have a maximum at the Kondo temperature T_K (see, for example, Ref. 2). Measurements in superconducting alloys showed a decrease of this rate with an increase of temperature for $T > T_K$.[3] Only recently, Peters, Bergmann, and Mueller[4] and Haesendonck, Vranken, and Bruynseraede[5] independently applied the method of weak localization to the Kondo systems AuFe and CuCr and found for the first time the maxima of the magnetic scattering rates at $T_K = 1$ K and $T_K = 2$ K, respectively.

Weak localization is a new and powerful method to study characteristic scattering times in thin disordered metal films (see, for example, the review articles in Refs. 6–8). Magnetoresistance measurements correspond to a time-of-flight experiment which yields the inelastic, spin-orbit, and magnetic scattering rates. The fact that the coherence of the conduction electrons is destroyed by magnetic scattering roughly after the magnetic scattering time results in an increase of the width of the magnetoresistance curves and thus provides a probe of Kondo impurities.

Dilute Co atoms in the bulk of Cu show no magnetic moment. If we want to describe this as a Kondo system, its Kondo temperature is about 500 K.[9] On the other hand, we are going to show that Co atoms on the surface of a Cu film represent a very interesting new Kondo system. In this paper, we will report our results for both cases.

Our experimental apparatus has been described elsewhere.[10,11] When weak localization is employed it is favorable to use thin disordered films. The films are quench condensed onto a quartz substrate which is kept at helium temperature in a vacuum better than 10^{-11} torr. The thicknesses of the films are registered by the frequency changes of a quartz oscillator.

We divide our experiments into two groups. In the first group, we investigate how Co atoms on the surface and in the bulk of Cu affect the spin-orbit scattering rate and the magnetic scattering rate at 4.5 K. In the second group, we investigate the temperature dependence of the magnetic scattering rate for Co on the surface of Cu.

As an example of the first group of experiments, we evaporate in the beginning 11.0 nm of Cu which is then annealed to 40 K and shows square resistance $R_0 = 112.4$ Ω. Next we evaporate 0.005 atomic layers (atola) of Co atoms on top of it. The small amount of Co atoms cannot be calibrated directly by the quartz oscillator. So we first heat the Co wire to higher rates up to 1 atola/min. The logarithm of the evaporation rate is then plotted versus the heating power, and a straight line is obtained which is extrapolated to the desired small rate. With this rate, we evaporate Co for 6 s. The error of the thickness is about 20%. The sandwich of Cu/Co is covered with 0.64 nm (2.8 atola) of Cu again, yielding a resistance per square $R_0 = 97.7$ Ω. Finally, the sandwich of Cu/Co/Cu is covered with 0.33 atola of Au. The relevant parameters are collected in Table I.

By applying a magnetic field perpendicular to the film, magnetoresistance measurements are performed after each step of evaporation and the results are shown in Fig. 1. The points are experimental results. The theoretical curves will be discussed below. The left ordinate gives the change of the resistance in Ω. A conductance scale in units of $L_{00} = e^2/2\pi^2\hbar$ is also given on the right-hand side. The horizontal scales for magnetic fields are shown immediately to the right of the curves. The upper curve represents the results for the pure Cu film. The second curve gives the results for the Cu/Co film and shows a broadening due to the Co atoms. This means that the Co atoms are magnetic on the surface of Cu. The third curve corresponds to Cu/Co/Cu and its scale is the same as that for the pure Cu. This shows that coverage of the Co with 2.8 atola Cu removes the magnetic broadening almost completely. The lower curve represents the Cu/Co/Cu/Au

TABLE I. The thickness, square resistance, τ_i, $\tau_{s.o.}$, and τ_s at 4.5 K for the sandwich Cu/Co/Cu/Au. (The product $H_n \tau_n$ varies for the sandwich between 0.59 and 0.53 ps T.)

Film	d (nm)	R_0 (Ω)	τ_i (ps)	$\tau_{s.o.}$ (ps)	τ_s (ps)
Cu	11.0	112.4	42	9.8	· · ·
CuCo	11.0	111.7	42	3.6	18
CuCoCu	11.6	97.7	42	5.8	1080
CuCoCuAu	11.7	95.6	42	1.7	1060

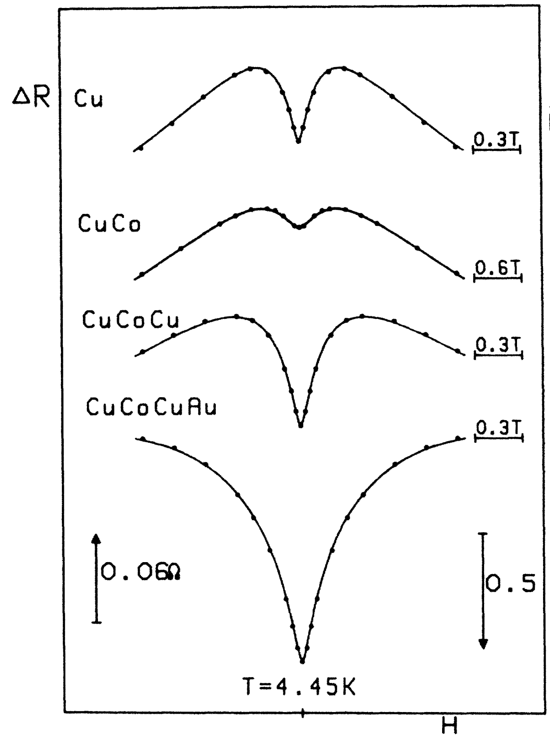

FIG. 1. The magnetoresistance of the Cu/Co/Cu/Au sandwich. The upper curve represents the pure Cu film with a thickness of 11.0 nm. The second corresponds to a coverage of the Cu with 0.005 atola of Co. The third curve is obtained after a coverage of the Co with 0.64 nm of Cu. The lower curve gives the data after a further coverage with 0.33 atola Au. The points are from the experiment and the curves are calculated with the theory by Hikami, Larkin, and Nagoka which yields $1/\tau_i$, $1/\tau_{s.o.}$, and $1/\tau_s$. The left ordinate gives the change of the resistance in Ω. A conductance scale in units of $L_{00} = e^2/2\pi^2\hbar$ is also given on the right-hand side. The horizontal scales for magnetic fields are shown immediately right to the curves.

sandwich. The coverage with Au atoms introduces a strong spin-orbit scattering and therefore a positive magnetoresistance in the whole field range. (This increase of the spin-orbit scattering yields an additional check of the consistency of the evaluation.)

The second group of experiments studies the behavior of Co atoms on the surface of Cu. According to our experience from previous experiments the evaluation of the magnetoresistance curves is optimal when they show a transition from positive to negative magnetoresistance with increasing field. To achieve this goal the spin-orbit scattering time must be shorter than the dephasing times of the electrons. Since we want to measure the magnetoresistance up to 35 K where the inelastic lifetime becomes relatively short, we need an even shorter spin-orbit scattering time. We achieve the strong spin-orbit scattering by preevaporating a sandwich of Au/Cu/Au, each with about 1 atola, before the host film Cu is made. We denote this "alloy" in the following by (Au*). The technique of using Au to enhance spin-orbit scattering is based upon our experience that this does not have any influence on the exact values of the magnetic scattering rate $1/\tau_s$. Then we evaporate on top of the alloy (Au*) 6.6 nm Cu. The annealing of the film of (Au*)/Cu to 50 K yields a resistance per square of about 102.4 Ω. Then magnetoresistance measurements are performed at eight temperatures in the temperature range from 4.5 to 35 K. Afterwards 0.01 atola of Co is evaporated on top of the (Au*)/Cu. Magnetoresistance measurements are done for 14 temperatures between 4.5 and 35 K accordingly. Figure 2 shows selected magnetoresistance curves for the (Au*)/Cu/Co sandwich.

The full curves are the theoretical fits resulting from the Hikami-Larkin-Nagaoka (HLN) theory.[12] This theory has been intensively discussed in the above cited review articles and many preceding papers. Therefore here we recall only the essential physics of weak localization. It describes the interference of two partial waves of the same electron. In real space the two partial waves propagate on a closed loop in opposite directions.[6] 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. By means of magnetoresistance measurements one determines the phase-coherence time of the conduction electrons. The actual shape of the magnetoresistance curves is determined by two independent parameters, the dephasing rates for singlet and triplet electron pairs. They are combinations of

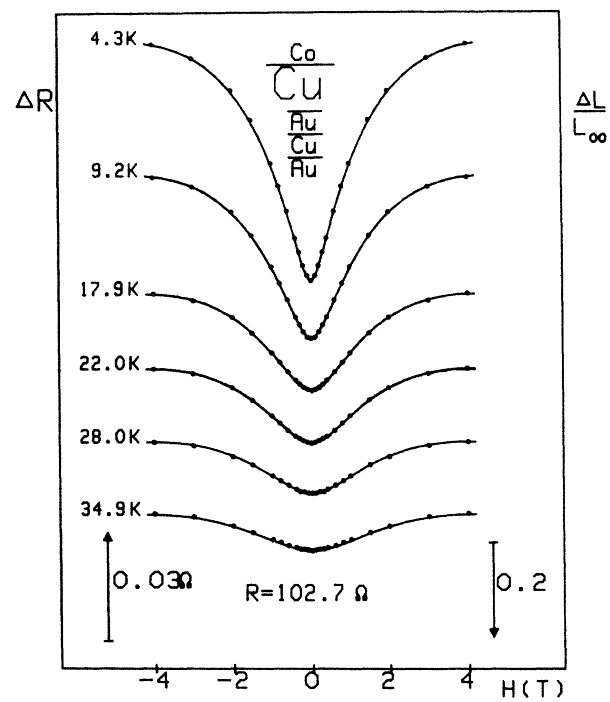

FIG. 2. Selected magnetoresistance curves for the sandwich of (Au*)/Cu/Co. The points are from the experiment and the curves are from the weak localization theory. The vertical arrows give the scales for the square resistance and the conductance change. The magnetic-field scale is given at the bottom.

three scattering rates: the inelastic, the magnetic, and the spin-orbit scattering rates $1/\tau_i$, $1/\tau_s$, and $1/\tau_{s.o.}$. The procedure for the evaluation has been described in previous papers (see, for example, Ref. 13). Therefore, here we give only a short sketch.

In the first experiment (the Cu/Co/Cu/Au sandwich), the evaluation is done as follows. (1) Pure Cu: We determine the inelastic and spin-orbit scattering rates $1/\tau_i$ and $1/\tau_{s.o.}$ for the pure Cu film. (2) Cu/Co: The Co atoms at the surface introduce a magnetic scattering rate $1/\tau_s$ and an additional spin-orbit scattering rate $\Delta(1/\tau_{s.o.})$. We can take the inelastic rate $1/\tau_i$ from the pure Cu film and determine $1/\tau_s$ and $1/\tau_{s.o.}$ for the Cu/Co sandwich. (3) Cu/Co/Cu: The Cu on top of the Co changes the magnetic and spin-orbit scattering rates due to the Co atoms. Again we take the inelastic scattering rate $1/\tau_i$ from the pure Cu and determine the $1/\tau_s$ and $1/\tau_{s.o.}$ for Cu containing bulk Co impurities. (4) Cu/Co/Cu/Au: Similarly we take $1/\tau_i$ from the pure Cu film and determine $1/\tau_s$ and $1/\tau_{s.o.}$. Coverage with Au should leave $1/\tau_i$ and $1/\tau_s$ unchanged and permits a check of the consistency of the evaluation. The evaluated results are listed in Table I and prove this consistency.

The set of experiments yields the following results. (i) Co on the surface shows a considerable magnetic scattering and therefore a magnetic moment. (ii) Co on the surface introduces a surprisingly large spin-orbit scattering. (iii) Co atoms in bulk Cu (which corresponds to the Co sandwiched between Cu films) show practically no magnetic scattering and therefore no magnetic moment. (iv) The spin-orbit scattering rate of the Co atoms in the bulk is reduced by a factor 3 compared with that at the surface. It is still surprisingly high for the light atom Co. (The dependence of the spin-orbit scattering on the magnetic character of impurities will be studied in more detail in the future.)

The evaluation for the sandwich of (Au*)/Cu/Co is performed in the same way as for the Cu/Co/Cu/Au sandwich. In order to have an accurate determination of $1/\tau_s$, we need a Co coverage of about 0.01 atola or more because weak localization does not provide an independent determination of $1/\tau_i$ and $1/\tau_s$. So $1/\tau_s$ must be strong enough to permit a separation of $1/\tau_i$ and $1/\tau_s$. The values of $1/\tau_s$ are plotted in Fig. 3. The error bars give the inaccuracy in the evaluation of the experimental magnetoresistance curves in the field range of Fig. 2. The magnetic scattering rate $1/\tau_s$ increases with increasing temperature to a maximum value of about $(3.2\ \text{ps})^{-1}$ at 23 K and then decreases.

Such a maximum of the spin-flip scattering is theoretically predicted by the Nagaoka and Suhl theory (see, e.g., Ref. 2 and references therein). Within this (approximate) theory the spin-flip scattering rate $1/\tau_s$ of conduction electrons is calculated and the following expression for $1/\tau_s$ is obtained:

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

A general problem using 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 pairs is essential. Therefore we expect that magnetic scattering destroys the pair amplitude in weak localization essentially as it does in superconductivity. The influence of Kondo impurities on superconductivity has been studied very intensively at the beginning of the seventies. The pair breaking by Kondo impurities and the resulting decrease of T_c is calculated by Müller-Hartmann and Zittartz.[14] They found that the pair-breaking parameter is proportional to $1/\tau_s$ as given by Eq. (1) and has its maximum value at T_K.

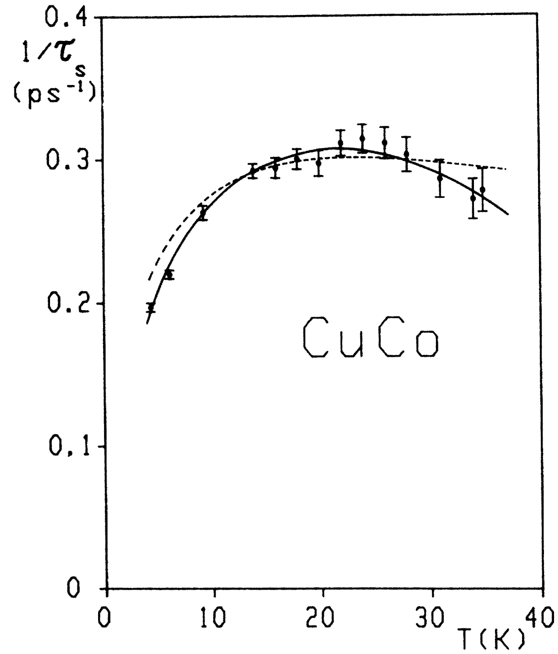

FIG. 3. The inverse magnetic scattering time as a function of temperature for 0.01 atola Co on top of (Au*)/Cu. The magnetic scattering rate shows a Kondo maximum at $T_K = 23$ K. The dashed line gives the theoretical curve according to the Nagaoka and Suhl theory [see Eq. (1)].

This theoretical curve is indicated for $S=\frac{1}{2}$ in Fig. 3 by the dashed line. It shows a maximum at the Kondo temperature T_K. The experimental behavior of $1/\tau_s$ of Co on the surface of Cu agrees qualitatively with the theoretical prediction for Kondo impurities. Therefore Co on the surface of Cu represents a new "surface Kondo" system with T_K about 23 K. We observe the same behavior in other (Au*)Cu/Co sandwiches.

In conclusion, with the method of weak localization, we have found a new surface Kondo system of CuCo which has a Kondo temperature of about 23 K. We have also found the disappearance of the magnetic scattering rate and a reduction in the spin-orbit scattering cross section by a factor of about 3 when the CuCo film is covered with Cu again.

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