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1996 J. Phys.: Condens. Matter 8 1381

(http://iopscience.iop.org/0953-8984/8/10/010)

J. Phys.: Condens. Matter **8** (1996) 1381–1387. Printed in the UK

Resonant Kondo scattering in Cu(Fe) thin film

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Received 10 October 1995

Abstract. The spin-flip rate of scattering resulting from the presence of Fe atoms in a very thin Cu film is determined as a function of the temperature between 1.6 K and 40 K by means of weak electron localization. The spin-flip scattering rate shows a maximum at 18 K which can be interpreted as resonant Kondo scattering. From a comparison of the Suhl–Nagaoka approximation with the present data a Kondo temperature $T_K \approx 15$ K is found.

1. Introduction

The phenomenon of weak electron localization has been developed into a very reliable method for the study of different scattering mechanisms in disordered thin films [**?**]. Weak electron localization is a direct consequence of quantum interference between electron waves travelling along time-reversed paths. The presence of a transverse magnetic field $\boldsymbol{H}$ modulates the phase difference between the interfering electron waves and produces an anomalous magnetoresistance (AMR), which is given by the Hikami, Larkin and Nagaoka [**?**] expression:

$$\frac{R_\square(H) - R_\square(0)}{R_\square^2(0)} = \frac{e^2}{2\pi^2\hbar}\left\{\frac{1}{2}f\left(\frac{H}{H_\phi}\right) - \frac{3}{2}f\left(\frac{H}{H_\phi^*}\right)\right\}$$
$$f(x) = \ln(x) + \psi\left(\frac{1}{2} + \frac{1}{x}\right) \tag{1}$$

where $R_\square$ is the resistance per square of the thin film, ψ is the digamma function, and H_ϕ and H_ϕ^* are temperature-dependent characteristic magnetic fields defined in the following manner:

$$H_\phi(T) = H_{in}(T) + 2H_{sf} \tag{2}$$

$$H_\phi^*(T) = H_\phi(T) + \frac{4}{3}\left(H_{so} - H_{sf}\right). \tag{3}$$

The characteristic fields H_x are equivalent to the scattering rates τ_x^{-1}, the connection between H_x and τ_x^{-1} being given by the relation

$$H_x = \frac{\hbar}{4eD}\tau_x^{-1} \tag{4}$$

where $D = \frac{1}{3}v_F^2\tau$ is the diffusion constant, v_F the Fermi velocity and τ the elastic scattering lifetime. In equations (**??**) and (**??**) the field H_{in} corresponds to the inelastic scattering

0953-8984/96/101381+07$19.50

rate τ_{in}^{-1}, H_{so} to the spin–orbit scattering rate τ_{so}^{-1}, H_{sf} to the spin-flip scattering rate τ_{sf}^{-1}, and H_{ϕ} to the phase-breaking rate τ_{ϕ}^{-1}.

The measurement of the transverse magnetoresistance allows a direct determination of the different scattering rates of the conduction electrons. Inelastic scattering, due to the electron–phonon and electron–electron interaction, causes a destruction of the interference after the characteristic time τ_{in}. The flipping of the electron spin after the interaction with a local magnetic moment of a magnetic impurity also destroys the weak-localization interference after the characteristic time τ_{sf}. The combination of inelastic and spin-flip scattering is described by the characteristic phase-breaking time τ_{ϕ}, defined as

$$\tau_{\phi}^{-1} = \tau_{in}^{-1} + 2\tau_{sf}^{-1} \tag{5}$$

which corresponds to the average time for which an electron diffuses between two scattering events.

Besides the inelastic and spin-flip scattering mechanisms, spin–orbit coupling also has a pronounced effect on the weak-localization AMR. When spin–orbit scattering is strong the AMR is positive (antilocalization), while when it is weak the AMR is purely negative. The spin–orbit interaction is important when the scattering rate

$$\tau_{\phi}^{*-1} = \tau_{\phi}^{-1} + \frac{4}{3}\left(\tau_{so}^{-1} - \tau_{sf}^{-1}\right) \tag{6}$$

is much larger than the phase-breaking rate τ_{ϕ}^{-1}. Equation (**??**) shows that spin-flip scattering counteracts the influence of spin–orbit scattering.

The interaction of conduction electrons with a localized spin $\boldsymbol{S}$ is usually described in terms of an s–d exchange potential

$$V_{ex} = J\boldsymbol{S}\cdot\boldsymbol{\sigma}$$

where $\boldsymbol{\sigma}$ is the spin of the conduction electron and J the coupling constant. The scattering from such a potential may be separated into a spin-flip and a non-spin-flip part. It is the spin-flip scattering that destroys the interference between two time-reversed electron paths and so weakens the effect of weak localization.

The same effect is important for superconductivity, where spin-flip scattering destroys the coherence between two electrons forming a Cooper pair [**?**]. According to the Suhl–Nagaoka approximation for the Kondo spin-flip scattering, the pair-breaking parameter is determined by the spin-flip scattering rate [**?**]

$$\tau_{sf}^{-1} = \frac{c}{2\pi\hbar N(\epsilon_F)}\left\{\frac{\pi^2 S(S+1)}{\ln^2(T/T_K) + \pi^2 S(S+1)}\right\} \tag{7}$$

where c, $N(\epsilon_F)$, S and T_K are the impurity concentration, the density of states at the Fermi level for one spin direction, the impurity spin and the Kondo temperature respectively. The spin-flip scattering rate τ_{sf}^{-1} has a maximum at T_K. Because of the similarity between the dephasing in weak localization and in superconductivity, it is expected that the spin-flip scattering rate entering equation (**??**) for τ_{ϕ}^{-1} will be given by equation (**??**) for a disordered metallic sample containing magnetic impurities.

Kondo maxima in the behaviour of the spin-flip scattering rate as a function of temperature have been observed experimentally in systems such as Fe on the surface and in the bulk of Au [**?**], in Cr-doped Cu films [**?**], and Co on the surface of Cu [**?**] and in Mg/Fe and Cu/Co sandwiches [**?**]. It was also proved theoretically that the presence of spin–orbit scattering does not suppress the Kondo effect, nor does it cause any observable changes in the Kondo temperature or the Kondo screening of magnetic impurities [**?**].

The purpose of the present investigation is to study the Kondo maximum of the magnetic scattering in the Cu(Fe) system.

2. Experimental procedure

The thin film of the dilute Kondo alloy was prepared by electron-gun evaporation of pieces of a master Cu(Fe) alloy (nominal concentration 60 ppm) onto a silicon nitrite substrate, held at room temperature. The pure Cu film was made from material of 99.999% purity. The pressure during the evaporation was better than 10^{-8} mbar. A quartz oscillator was used for monitoring the evaporation rate, which was calibrated using a DEKTAK II profilometer. Both films were deposited at a rate of 10 Å s^{-1} and their final thickness was $d = 100$ Å. The relative error in the determination of the thickness was 5%.

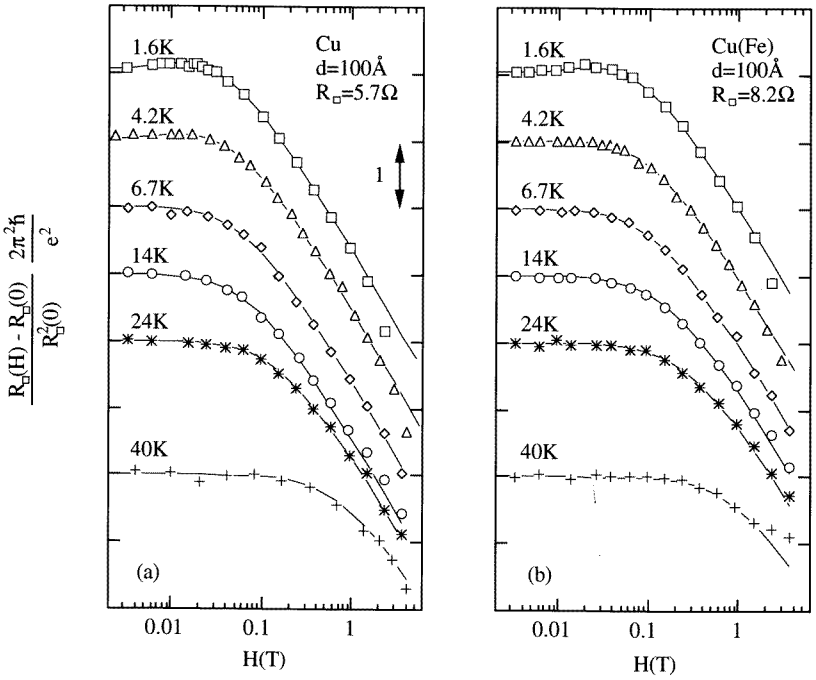

Figure 1. Normalized magnetoresistance at various temperatures for (a) a pure Cu film and (b) an Fe-doped Cu film. The solid curves represent the magnetoresistance calculated using equation (1).

A four-terminal DC method was used for measuring the film resistance. The current was kept as low as possible to avoid effects related to electron heating. This was proved by measuring the logarithmic temperature dependence of the film resistance, which is caused by the Coulomb interaction [**?**]. The low-temperature measurements, in the range from 1.6 K to 40 K, were carried out in a conventional stainless-steel cryostat. Magnetic fields up to 6 T were supplied by a superconducting magnet.

3. Results and discussion

Figures 1(a) and 1(b) show the transverse magnetoresistance of a pure Cu thin film and a film of Cu(Fe) alloy respectively, normalized in units of $e^2/2\pi^2\hbar$, at various temperatures. The full curves represent the magnetoresistance calculated using the Hikami, Larkin and Nagaoka theory (equation (**??**)).

The magnetoresistance measurements for the pure Cu film show a weak positive component at temperatures 1.6 K and 4.2 K and fields up to 0.02 T, which indicates the presence of spin–orbit scattering. The corresponding characteristic field H_{so}, as well as the inelastic field H_{in}, were estimated by fitting the theoretical expression (**??**) to the experimental data with two adjustable parameters: H_ϕ and H_ϕ^* . Only data up to magnetic fields of 1 T were used in the fitting procedure, in order to exclude possible contributions to the magnetoresistance at higher fields from the enhanced electron–electron interaction [**?**, **?**]. The value of the spin–orbit field was $H_{so} = 3 \times 10^{-3}$ T. The spin-flip scattering field H_{sf} was assumed to be zero in the pure Cu film, as the concentration of magnetic impurities was very low. At higher temperatures the magnetoresistance is negative throughout the magnetic field range, so, in order to extract the values of the characteristic fields H_ϕ and H_ϕ^* from the data, one has to assume that the spin–orbit field does not vary much as the temperature increases. This assumption is supported by previous measurements on Cu films [**?**]. Following this procedure the values of H_{so} and H_{in} could be estimated for all temperatures.

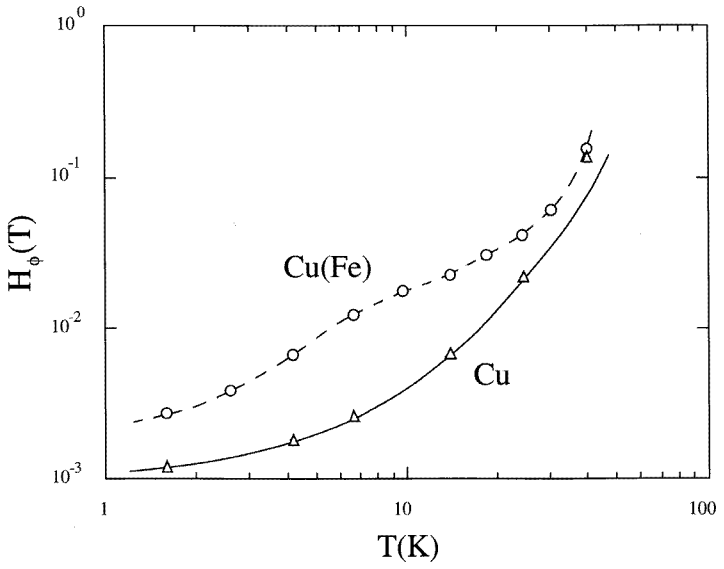

Figure 2. The temperature dependence of the dephasing field $H_\phi(T)$ for a pure Cu film and an Fe-doped Cu film. The solid curve represents equation (8) for the field $H_{in}(T)$ of the pure Cu film, while the dashed curve is only a guide to the eye.

In the Cu film with additional Fe impurities, the positive magnetoresistance was observed at temperatures lower than 4.2 K. This implies that the dephasing field H_ϕ is higher in this film, mainly due to the increased spin-flip scattering from the localized magnetic moments of the Fe ions. The characteristic fields H_ϕ and H_ϕ^* could be obtained throughout the temperature range by the same procedure as described above. The value of the difference $H_{so} - H_{sf}$, deduced from H_ϕ^*, was found to decrease by about 20% as the temperature increased. This supports the assumption that spin-flip scattering is increased by the presence of the Fe.

Figure 2 shows the temperature dependence of the adjusted characteristic field H_ϕ of the dephasing rate for the pure Cu thin film and for the Fe-doped film in a double-logarithmic

plot. For the pure Cu film there should be negligible magnetic scattering and H_ϕ is roughly equal to H_{in}. The behaviour of H_{in} versus the temperature for the pure Cu film follows a curve

$$H_{in}(T) = A + BT + CT^3 \tag{8}$$

for temperatures up to 25 K, which is represented by the solid line in figure 2. The terms of this power-law expansion correspond to different phase-breaking mechanisms. The constant term A comes from spin-flip scattering due to residual magnetic impurities or paramagnetic surface states [**?**]. The linear term is due to electron–electron collisions ($\tau_{ee}^{-1} \sim T$ in two dimensions in the presence of disorder [**?**, **?**]), while the third term represents the contribution from inelastic electron–phonon scattering [**?**, **?**]. The dephasing field H_ϕ is higher in the Fe-doped film at all temperatures, as can be seen from figure 2. Its temperature dependence is markedly different from that described by equation (**??**). Thus the spin-flip scattering rate τ_{sf}^{-1} due to the Fe impurities is significant and temperature dependent. In order to find τ_{sf}^{-1}, the values of $H_{in}(T)$ for the pure Cu film were subtracted from the $H_\phi(T)$ of the Fe-doped film. If it is assumed that the Fe impurities change only the spin-flip scattering rate, then this difference is twice the field H_{sf}. Using equation (**??**), the H_{sf}-values may be converted to τ_{sf}^{-1}, where the diffusion constant is taken as $D = \left(e^2 N(\epsilon_F) R_\square d\right)^{-1}$, $N(\epsilon_F)$ being the free-electron density of states of Cu at the Fermi energy. The small difference between the values of $R_\square$ of the two films, owing mainly to thickness measurement uncertainties, is accounted for as described by Van Haesendonck *et al* [**?**].

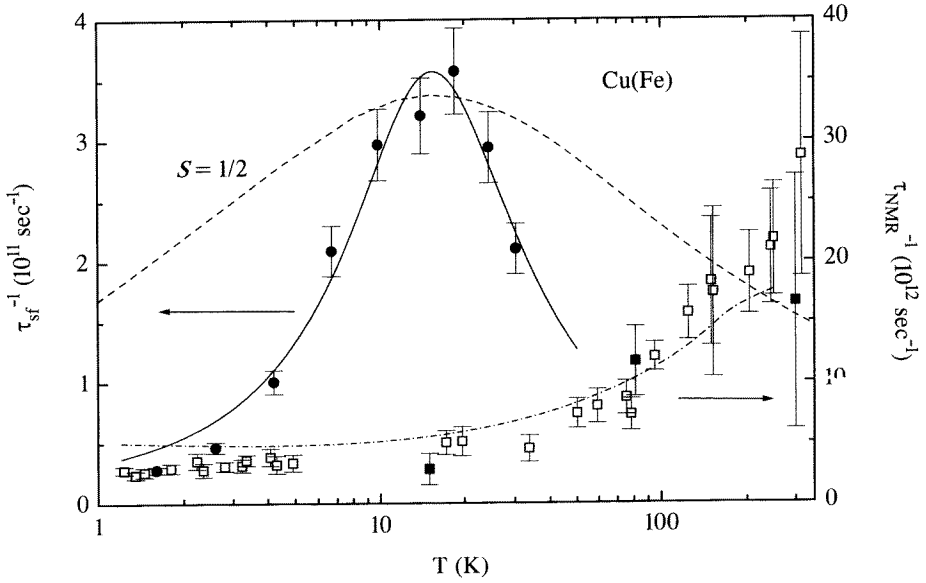

Figure 3. The temperature dependence of the spin-flip scattering rate for the Fe-doped Cu film. The spin-flip rate shows a Kondo resonance maximum at 18 K. The dashed and solid curves were calculated using the Suhl–Nagaoka approximation for $S = 1/2$ and $S = 0.072$ respectively. The adjusted T_K is 15 K in both cases. Open and full squares represent the relaxation rates of NMR and INS experiments respectively. The chain line is a fit of the theoretical result of Goetze and Schlottmann for the spin-1/2 exchange model.

Figure 3 shows the corresponding spin-flip scattering rate τ_{sf}^{-1} as a function of the

temperature in a semilogarithmic plot. The errors become higher as the temperature increases, mainly because of insufficient stabilization of the sample temperature during measurement of the magnetoresistance. τ_{sf}^{-1} first increases with increasing temperature, reaches its highest value at about 18 K and then decreases. The sharp maximum observed can be interpreted as resonant Kondo scattering at the Kondo temperature $T_K \sim 18$ K. The two curves appearing in figure 3 are calculated using the Suhl–Nagaoka theory (equation (**??**)), with an adjusted Kondo temperature of 15 K for the best fit to the experimental data. The value of S was taken as 1/2 for the dashed curve, while for the solid curve S was left as a free parameter. It turned out that S must be as low as 0.072 in order for the data of figure 3 to be fitted according to the Suhl–Nagaoka approximation.

The surprisingly small value of the magnetic impurity spin ($S = 0.072$), which is used as a parameter in the Suhl–Nagaoka expression (7) in order to obtain a good agreement between experiment and theory, indicates that the Fe impurities do not behave as isolated Kondo scattering centres with a spin $S = 1/2$. This discrepancy can be attributed to the possible presence of very small Fe precipitations, which are formed during the evaporation at room temperature, where the Fe ions are probably coupled antiferromagnetically.

Recently Sacramento and Schlottmann [**?**] compared the experimental data for susceptibility, specific heat, magnetization and resistivity of dilute Cu(Fe) alloys to the exact theoretical calculation, using the Bethe *ansatz* method, for the n-channel Kondo problem. They have obtained good agreement between experimental and theoretical results for spin $S = n/2 = 2$, $T_K = 18$ K, and a g-factor of 2.

It must be mentioned here that spin relaxation rates, τ^{-1}, in the classic Kondo system Cu(Fe), are also obtained from inelastic neutron scattering (INS) [**?**] and NMR [**?**] experiments. For comparison with the present weak-localization results figure 3 includes also the relaxation rate τ_{NMR}^{-1} data taken from Alloul [**?**] (open squares: NMR data; full squares: INS data). Figure 3 shows that there is a considerable discrepancy between the present spin-flip relaxation rate τ_{sf}^{-1} data and those obtained from inelastic scattering and in particular from NMR experiments. According to figure 3 the corresponding spin relaxation rate τ_{NMR}^{-1} does not exhibit a maximum at T_K, but has a constant value for $T \ll T_K$ ($T_K \sim 30$ K [**?**]), which is larger by a factor of the order of ten than that obtained from weak-localization measurements. For $T \gg T_K$, τ_{NMR}^{-1} increases and reaches asymptotically a linear T-dependence as is expected from perturbation theory. Goetze and Schlottmann [**?**] have performed approximate calculations of the zero-field relaxation rate $\tau^{-1}(T)$ for the spin-1/2 sd model, which indicate that $\tau^{-1}(T)$ should reach the constant value $\tau^{-1} \sim k_B T_K/\hbar$ for $T \ll T_K$ and should increase for $T \gg T_K$ linearly with the temperature T, but is much larger than the Korringa value $\tau^{-1} \sim (\pi/\hbar)(JN(\epsilon_F))^2 k_B T$. J is the exchange coupling constant and $N(\epsilon_F)$ the density of states at the Fermi level. There is a progressive change from the 'strong-coupling' to 'local moment' regime of the electronic system. The chain line in figure 3 represents the fit of the theoretical results of Goetze and Schlottmann [**?**] for antiferromagnetic coupling and for $JN(\epsilon_F) = 0.200$ on the Cu(Fe) system.

The discrepancy between the weak-localization spin-flip relaxation rate τ_{sf}^{-1} and the NMR spin relaxation rate τ_{NMR}^{-1} on one hand, and the agreement between τ_{NMR}^{-1} and the theoretically calculated $\tau^{-1}(T)$ values [**?**] on the other hand, can be attributed to different mechanisms. In order to calculate the spin relaxation rate $\tau^{-1}(T)$ for a Kondo system with antiferromagnetic coupling, Goetze and Schlottmann [**?**] assume that the impurity complex has a broad, continuous excitation spectrum, which resembles that of a molecule with many levels. The existence of such a broad excitation spectrum gives the best fitting results for the

spin relaxation rate of Cu(Fe) system as shown in figure 3. A spectrum like this is excluded by assumption within the Suhl–Nagaoka approximate calculations [**?**], which for single-electron scattering lead to equation (**??**) for the spin-flip scattering rate τ_{sf}^{-1}. A general problem with the Suhl–Nagaoka approach is that the Kondo effect enters weak localization in a more complicated way than for single electrons, because one is concerned with the phase coherence of the partial waves of an electron. In weak localization one measures all events that destroy this phase coherence. One expects that magnetic scattering will destroy the pair amplitude in weak localization, essentially in the same way as it destroys the Cooper pairs in a magnetically doped superconductor.

4. Conclusion

In the present investigation the method of weak localization was used to study the magnetic scattering due to Fe impurities in Cu. It was found that the magnetic scattering has the highest value at about 18 K which is interpreted as resonant Kondo scattering. The Suhl–Nagaoka approximation agrees with the experimental data, giving a Kondo temperature of 15 K.

Acknowledgment

One of us (CP) wishes to thank the GSRT and the International Bureau of Collaboration, KFA, Jülich, for financial support.

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