Measurement of the Magnetic Scattering Time by Weak Localization

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The influence of magnetic impurities on weak localization is investigated. Thin Mg films are covered with $\frac{1}{1000}$ monolayer of Fe. A temperature-independent magnetic scattering time $\tau_s = 4.7\times 10^{-12}$ sec is obtained.

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At low temperature the conduction electrons of a metal possess a long inelastic lifetime (in a fixed energy state). In disordered thin films this yields quantum interferences in a spatially extended region, introduced by Abrahams *et al.*[1] as "weak localization." In this state the resistance shows a logarithmic anomaly with temperature. Since the calculation of its sensitivity to a magnetic field by Altshuler *et al.*[2] and Hikami, Larkin, and Nagaoka,[3] this state has become a unique probe to measure characteristic times in metals. I recently measured[4] the inelastic lifetime of the conduction electrons in Mg and the spin-orbit coupling for Mg and Mg covered with submonolayers of Au. This paper reports the measurement of the magnetic scattering time of the conduction electrons due to magnetic impurities.[5]

The dynamic behavior of magnetic ions has attracted continuous interest in solid-state physics. (i) Magnetic impurities cause an additional resistivity. (ii) They decrease or depress the superconducting transition temperature. (iii) They cause an additional Hall effect, the anomalous Hall effect. In addition, the question of whether a three-dimensional impurity is magnetic, a Kondo impurity, a local spin fluctuation, or nonmagnetic is a basic question and I believe that weak localization can contribute many basic answers to these problems. Hikami, Larkin, and Nagaoka[3] calculated the magnetoconductance of thin films in the presence of magnetic impurities (and spin-orbit coupling):

$$\frac{dL}{L_{00}} = -\left\{\psi\left(\frac{1}{2}+\frac{H_1}{H}\right)-\psi\left(\frac{1}{2}+\frac{H_2}{H}\right)+\left[\psi\left(\frac{1}{2}+\frac{H_3}{H}\right)-\psi\left(\frac{1}{2}+\frac{H_4}{H}\right)\right]/2\right\}, \quad (1)$$

$$L_{00} = \frac{e^2}{2\pi^2\hbar},$$

where H is the applied field and H_n are characteristic fields which stand for the characteristic times of the system. The connection between H_n and the corresponding relaxation time τ_n is given by $H_n\tau_n = \hbar/4eD$, where D is the diffusion constant.[6] Since the experiment yields directly the characteristic fields I prefer the use of the fields H_n in Eq. (1). The H_n are defined in the following manner:

$$\begin{aligned} H_1 &= H_0 + H_{so} + H_s, \\ H_2 &= \tfrac{4}{3}H_{so} + \tfrac{2}{3}H_s + H_i, \\ H_3 &= 2H_s + H_i, \\ H_4 &= \tfrac{2}{3}H_s + \tfrac{4}{3}H_{so} + H_i. \end{aligned} \quad (1a)$$

Here the indices stand for the following scattering processes: 0, potential scattering; i, inelastic scattering; s, magnetic scattering; and so, spin-orbit scattering. (The distinction between the x and the z components of the scattering times is neglected.) Recently Maekawa and Fukuyama[7] confirmed the calculation by Hikami, Larkin, and Nagaoka and corrected a clerical error in the final relation for H_2. For small (or very strong) spin-orbit coupling the magnetoconductance curves have roughly the shape of a bell. Its sign is positive for small spin-orbit coupling and negative for strong spin-orbit coupling. Its width is proportional to $1/\tau_i$ and of the order of $H_i = (\hbar/4eD)/\tau_i$. In the presence of magnetic scattering the width of the magnetoconductance curves increases; for $H_s > H_i$, of the order of H_s.

For the experimental investigation, thin Mg films with resistances of the order of 100 Ω are quench condensed at 6 K in a vacuum better than 10^{-11} Torr onto a crystalline quartz plate. The magnetoconductance of the films is measured at different temperatures. The characteristic parameters of thin Mg films such as $H_i(T)$ and H_{so} have been determined recently[4] and are confirmed in the present experiment. In Fig. 1 the magnetoconductance is shown for a Mg film with a thickness of 8.6 nm. The field is taken in units of H/H_i, where $H_i(T)$ varies as $6.25\times 10^{-4}T^2$ (T/K^2). The spin-orbit coupling field is $H_{so} = 0.0065$ T. The conductance is normalized to

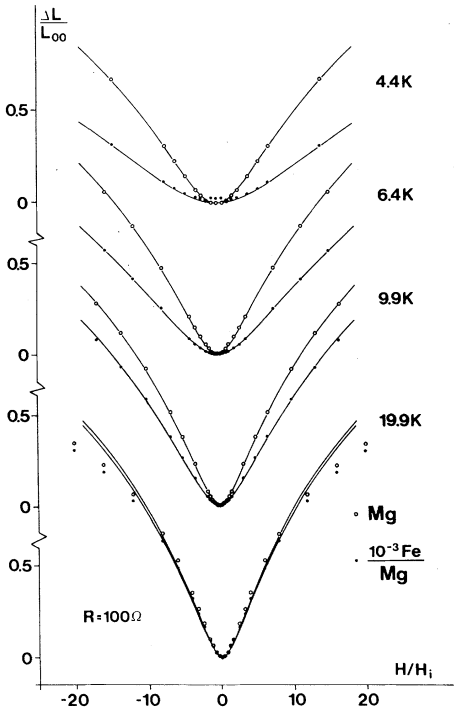

FIG. 1. The influence of $\frac{1}{1000}$ monolayer of Fe on the magnetoconductance curves of Mg. The field is normalized with respect to $H_i(T) = 6.25\times 10^{-4}T^2$ (T/K^2). The open circles show the magnetoconductance of the pure Mg film and the full circles the magnetoconductance curves after a coverage with $\frac{1}{1000}$ monolayer of Fe. The full curves give the theoretical results with $H_s = 0.075$ T.

L_{00}. The open circles give the experimental points for pure Mg at different temperatures. The solid curves are calculated with the theoretical formula of Hikami, Larkin, and Nagaoka using the above $H_i(T)$ and H_{so}. They give perfect agreement with the experimental results (with the exception of the highest temperature where thermal phonons start to interfere). This good agreement is essential for the following analysis of the Mg-Fe system. After the measurement of the magnetoconductance curves of the pure Mg film, the Mg is covered with roughly $\frac{1}{1000}$ of an atomic layer (atola) of Fe. The small coverage of Fe could not be measured directly. To achieve this coverage an Fe wire is heated so that evaporation rates between 1 and 0.1 atola/min are obtained. The logarithm of the evaporation rate as a function of the heating power of the Fe wire is extrapolated to a rate of 0.01 atola/min. With the resulting power, the Fe is evaporated for 6 sec which corresponds roughly to a coverage of $\frac{1}{1000}$ atola of Fe or an impurity concentration of 30 ppm. The small coverage of the Mg with Fe changed the magnetoconductance curves completely. In Fig. 1 the four magnetoconductance curves for the Mg-Fe system are also plotted. For each set of curves H_i is constant and the two curves are distinguished by the Fe coverage of $\frac{1}{1000}$ atola. Obviously the Fe broadens the low-temperature curves considerably whereas at high temperature its effect is small. This is due to the fact that $H_i(4.4\text{ K}) < H_s < H_i(20\text{ K})$ and the reason why such a small Fe coverage has been chosen. Since $H_i(T)$ and H_{so} are known for the Mg film one can fit H_s to the measured points. This is done for all temperatures with $H_s = 0.075$ T and the full curves give the theoretical results calculated with this value of H_s (and the values of H_i and H_{so} obtained from the pure Mg film). The small alignment of the magnetic moments in the applied field can be neglected. For example at 4.4 K the curves are evaluated in the field range of -0.2 to $+0.2$ T. This yields a relative alignment of about $\mu H/3k_BT$ which is of the order of $\frac{1}{20}$. The change in $1/\tau_s$ is essentially proportional to the square of the relative alignment and is negligible in the evaluated field range. The value of H_s corresponds to a magnetic scattering time of 4.7×10^{-12} sec. Within the framework of the theory one can also determine the temperature dependence of H_s or τ_s, respectively, as long as $H_i < H_s$. For $H_i > H_s$ the fit is too inaccurate. Such a procedure yields a temperature-independent H_s for the Mg with Fe impurities on its surface.

In a third evaporation step the Fe is covered with two atomic layers of Mg. This does not change the temperature dependence or the absolute value of H_s. The value of H_s is determined by the best fit of the theory to the experimental points. The former values of H_i and H_{so} of pure Mg are used and H_s is adjusted so that a complete coincidence between theory and experiment is achieved. H_s is plotted in Fig. 2. Then H_s is slightly changed to higher and smaller values until a clear difference between experiment and theory is recognized. This determines the error bars in Fig. 2. H_s is only reliable in the limited temperature range where $H_s > H_i$. One can, of course, increase the Fe concentration and therefore H_s. But the theory by Hikami, Larkin, and Nagaoka does not contain the influence of the magnetic energy $H\mu$ and in addition the magnetoconductance of the electron-electron interaction interferes.[8]

Further experiments have been performed with

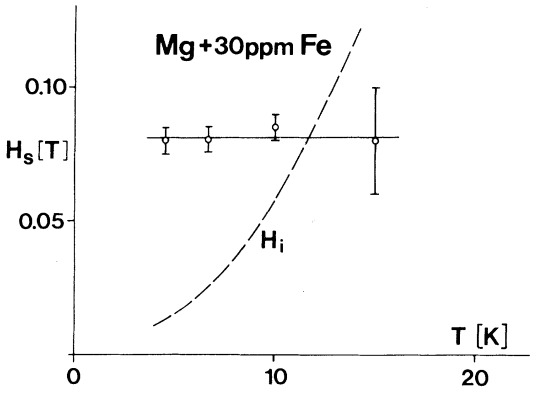

FIG. 2. Temperature dependence of the magnetic scattering field $H_s = (\hbar/4eD)/\tau_s$.

Fe on and in Cu and Au. In these systems, even smaller Fe coverages cause an equivalent broadening of the magnetoconductance curves. Since the strong spin-orbit coupling in these matrices complicates the evaluation these results will be published in detail elsewhere.

One particularly interesting application of this determination of H_s is the investigation of a Kondo system. In bulk Kondo systems the temperature dependence of $1/\tau_s$ is rather weak and only a change of about 10% to 20% per decade of temperature is expected for $S = 1$ impurities (depending on T/T_K). In two-dimensional disordered systems the Kondo effect will be altered (but the theory is not yet worked out). In the Mg-Fe system there is no noticeable temperature dependence of $1/\tau_s$, but because of the limited temperature range and the lack of theory in disordered two-dimensional Kondo systems a Kondo behavior cannot be completely excluded at the present time.

At low temperatures the magnetoconductance curves show an interesting substructure, a small maximum at zero field, as Fig. 1 demonstrates. The superposition of 2 atola of Mg enhances this maximum by a factor of 3. The origin of this structure has not yet been investigated but it is probably due to the dynamics of the Fe-Fe interaction.

The experimental determination of τ_s may be quite helpful in the search for new superconductors. It is well known that magnetic impurities depress the superconducting transition temperature by $k_B T_c = \hbar/\tau_s$. If we want to check whether a given metal like Mg or Au has a transition temperature of the order of 10^{-4} K then τ_s must be larger than 10^{-7} sec. For the Mg film investigated this means that $H_s < 5\times10^{-6}$ T. Whether the residual magnetic scattering supresses possible superconductivity of the metal under investigation could easily be detected at 0.1 K where $H_i = 6\times10^{-6}$ T. (If a Kondo effect is present the latter must be studied in detail for such an analysis.)

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[5]The first magnetoresistance measurement and study of the influence of magnetic impurities were performed on Pd [G. Bergmann, Phys. Rev. Lett. 43, 1357 (1979)]. At that time the phase sensitivity of the resistance was unknown and the measurement had been discussed in terms of a magnetic model.

[6]The diffusion constant is given by $D = 1/RdNe$, with R the resistance, d the film thickness, and N the density of states. This yields $H_n \tau_n = \hbar RdNe/4$. Assuming a homogeneous film and the free-electron model one obtains for the investigated film $H_n \tau_n = 3.75\times10^{-13}$ T s.

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