

Bulk and surface spin-orbit relaxation of the conduction electrons separated by weak-localization experiments

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Weak-localization magnetoresistance measurements allow a determination of the spin-orbit relaxation time in thin metal films. We have performed a series of experiments on films of magnesium metal evaporated on several different substrates with a range of film thicknesses, and for the first time separated the spin-orbit relaxation rate in the bulk of magnesium from that at the surface. Whereas the spin disorientation per bulk (elastic) scattering is intrinsic to the element (magnesium) under consideration, the surfaces on which the metal film is evaporated can give rise to a spin-orbit interaction that differs by more than an order of magnitude for different substrates.

The spin-relaxation time of the conduction electrons in metals without magnetic impurities has a long and interesting history.¹ The dominating contribution to spin relaxation is thought to be due to spin-orbit interaction in the various scattering processes. All experiments so far have been carried out on small samples (either thin films or small particles). Until very recently the measurements have therefore, with one possible exception,² revealed only contributions from surfaces.¹ The first clear-cut experiments,³ where bulk spin-orbit relaxation played a role were weak-localization magnetoresistance measurements on magnesium films, which are also the topic of this Rapid Communication.

The theory of spin relaxation due to the spin-orbit interaction is rather incomplete. A phenomenological parameter ϵ is normally introduced.^{4,5} $1/\epsilon$ is the average number of scattering events that the conduction electron must undergo to have its spin direction randomized. So far there has been no attempt to calculate this phenomenological number from a microscopic point of view. Abrikosov and Gorkov⁶ many years ago suggested that scattering off impurities with atomic number Z gives rise to a randomization ϵ of the spin orientation, where ϵ is proportional to the fourth power of Z , in close analogy with the well-known spin-orbit interaction of the electron orbitals in atoms. They did not distinguish between the situation where the impurity was at the surface or in the bulk of the metal. The surprising result of Ref. 1 was that there exists a trend in the experimentally determined values of ϵ , which they collected from the literature. They found that ϵ was apparently proportional to the fourth power of the atomic number of the metal itself, not just the accidental impurities at the surface. The authors took this agreement as a support of the theory of Abrikosov and Gorkov.⁶

In this paper we shall demonstrate that this conclusion is, in fact, dubious, and that for surface scattering the properties of the dielectric substrate play an important role for the value of the average number $1/\epsilon_s$ of surface scattering events that is needed to randomize the spin orientation of a conduction electron. The other conclusion we reach from our experiments is that the average number $1/\epsilon_B$ of scattering events in the bulk that is needed to randomize the spin orientation is inherent to the metal (here magnesium) under consideration. In fact, the value of ϵ_B for bulk magnesium, which we measure, comes close to the tentative suggestion of Ref. 4, $\epsilon = (\alpha Z)^4$, where α is the fine-

structure constant. This value of ϵ is, however, as already mentioned, calculated for an experimental situation, where Z is the atomic number of the bulk element under consideration.

Localization of the conduction electrons in metals leads to an increase in the electrical resistivity as the temperature is lowered. In metal films, where the film thickness d is much smaller than the inelastic diffusion length $\Lambda_i = (D\tau_i)^{1/2}$, where D is the diffusion constant and τ_i the inelastic relaxation time, the theory becomes particularly simple, and the localization (or quantum) correction to the sheet conductance is $\Delta\sigma = e^2/\pi h \ln(\tau_i/\tau_0)$, where τ_0 is the elastic (momentum) scattering time. τ_i decreases with increasing temperature and this is the reason for the observed temperature dependence mentioned. It was a breakthrough in the quantitative understanding of weak localization, when in 1981 Hikami, Larkin, and Nagaoka⁷ derived a precise formula for the two-dimensional weak-localization magnetoconductance $\sigma(B)$, based on tabulated digamma functions $\psi(\frac{1}{2} + x)$:

$$\sigma(B) - \sigma_0 = \frac{e^2}{\pi h} \left[\psi\left(\frac{1}{2} + \frac{B_0}{B}\right) - \frac{3}{2} \psi\left(\frac{1}{2} + \frac{4/3 B_{SO} + B_i}{B}\right) + \frac{1}{2} \psi\left(\frac{1}{2} + \frac{B_i}{B}\right) - \frac{1}{2} \ln \frac{B_0^2 B_i}{(4/3 B_{SO} + B_i)^3} \right]. \quad (1)$$

The only adjustable parameters here are the characteristic fields B_0 , B_i , and B_{SO} , which are related to the characteristic times τ_0 , τ_i , and τ_{SO} by the relations

$$B_0\tau_0 = B_i\tau_i = B_{SO}\tau_{SO} = \hbar/4eD = \hbar eN(E_F)R_{\square}d/8. \quad (2)$$

D is the three-dimensional diffusion constant, $D = v_F^2\tau_0/3$, R_{\square} is the film resistance per square, and d is the film thickness. τ_{SO} is the spin-orbit relaxation time, i.e., the average time it takes before the spin orientation is randomized. The last expression in Eq. (2) is derived under the assumption that the free-electron model can be used. In this paper we shall base our interpretation on the validity of the free-electron model.

In order to be able to extrapolate our results to the bulk we repeated the same measurements on many samples with different thicknesses, all made in the same evaporation.

Magnesium was selected as the first metal to be studied because it is not superconducting and because it has a low concentration of magnetic impurities. Magnesium was thermally evaporated onto a liquid-nitrogen-cooled dielectric substrate. While evaporating the magnesium film with a speed of about 5 nm/sec, a shield covering the substrate was slid back in about 6 sec, and then returned to its original position, thus producing a wedge-shaped film. The evaporation rate was determined by exposing a reference substrate for a much longer time and measuring its film thickness using an interference microscope. The main source of error in the thickness of the films was due to the oxidation of the magnesium films in air. The thickness of the oxide layer was determined by watching the change in resistance of the films as we let air into the vacuum chamber (after heating the films to room temperature). We estimate the uncertainty in the thickness to be 3 nm; this of course was most critical for the thinnest films. The sample was covered with photoresist and baked for $\frac{1}{2}$ h at 90°C. Then the photoresist was exposed through a mask to define 16 magnesium films of different thicknesses. To increase the resistance each magnesium film was shaped as a path with a total length of about 17 mm and a width of only 20 μm all in an area of $1 \times 1\frac{1}{2}$ mm².

In order to check if our magnesium films were continuous and represented bulk magnesium, we measured the Hall effect at 4.2 K and found a value of the Hall coefficient R_H , which was independent of the magnesium film thickness. The value, $R_H = -5.8 \times 10^{-13}$ $\Omega \text{ cm/G}$, comes close to measured bulk values. Our magnesium films had a long electron mean free path compared to other investigations of weak localization in the literature. This makes the weak-localization effect we are interested in more difficult to measure, but has the clear advantage that we are more likely to have films which actually represent bulk magnesium. The main problem of interpretation is thus shifted from the internal structure of the film to the surfaces of the film. Consequently, we have made a thorough study of the size effects in our films.

The resistance of a metal film can be separated into a sum of two contributions, one from the bulk, ρ_B , and one from the surface. The mean free paths in the bulk of our films, l_B , are of the order of or smaller than the thickness of the films. In this case, and assuming that the surface scattering is diffuse, i.e., the drift velocity of the electrons is on the average lost in a collision with the surface, the following expression for the film resistivity (the one measured) is easily derived:⁸

$$\rho = \rho_B + \frac{3}{8}\rho_B/d = \frac{m}{ne^2} \left(\frac{1}{\tau_B} + \frac{3}{8} \frac{v_F}{d} \right), \quad (3)$$

where the last term in parentheses is the scattering rate $1/\tau_s$, due to the presence of the surface. The last equality sign assumes the validity of the free-electron model, since n is the electron density and m is the free-electron mass. In order to compare our results with Eq. (3), we have plotted ρd vs d for all samples. Figure 1 shows an example for a wedge-shaped magnesium film evaporated on soda glass. 12 out of the 16 possible paths made out of this wedge were electrically coherent, and they are each represented by one measuring point in Fig. 1. These experimental points define a straight line as drawn in Fig. 1. The intercept along the ρd axis is, according to Eq. (3), just $\frac{3}{8}\rho_B l_B$. According to the

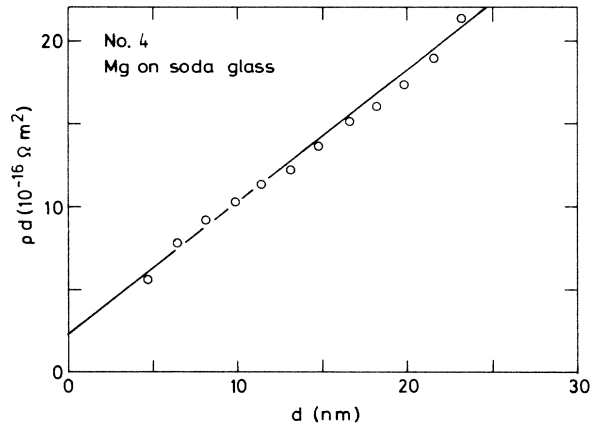


FIG. 1. The electrical resistivity times the film thickness for a series of magnesium paths plotted vs the film thickness d of each film. The straight line represents the result, Eq. (3), of size effect theory with diffuse scattering from the film surfaces. The substrate here is soda glass.

free-electron model this number is given by 2.45×10^{-16} m² Ω for magnesium. All our measurements turn out to give rough agreement with this number. However, one should bear in mind that the thickness of our films is not accurate enough to determine this number to better than within about a factor of 2, so the agreement obtained by this intercept in Fig. 1 is certainly fortuitous. However, the agreement with Eq. (3) is certainly a strong support that our magnesium films have bulk properties and that the assumptions leading to Eq. (3) are fulfilled. The slope of the line drawn in Fig. 1 determines ρ_B , and again using the free-electron model, we can determine $1/\tau_B$ and $1/\tau_s = 3v_F/8d$. In Table I we show results for four samples each with a complete set of paths. Three different substrates have been used, and for each of the four sets of paths we list the obtained values of $3\rho_B l_B/8$, ρ_B , and $1/\tau_B$.

For each of the four samples in Table I we made magnetoresistance measurements on all the 9–12 paths to determine the value of B_i and B_{SO} from Eq. (1) by a simple method of analysis.⁹ Subsequently, we determined $1/\tau_i$ and $1/\tau_{SO}$ from relation (2).

All these measurements were repeated at four different temperatures, and whereas $1/\tau_i$ was found to be proportional to the temperature, $1/\tau_{SO}$ was found to be insensitive to changes in the temperature. In Fig. 2 we have plotted the spin-orbit relaxation rate determined from our magnetoresistance measurements as a function of the inverse film thickness for three sets of measurements on different types of substrates. One sees that the spin-orbit relaxation rate is very different for the different substrates.

Similarly to the separation of the transport (elastic) relaxation rate into a bulk and a surface part, we now separate the spin-orbit relaxation rate into a bulk and a surface part, which we in turn assume are each proportional to the elastic bulk and surface part of the elastic relaxation rate as determined above. We thus write

$$\frac{1}{\tau_{SO}} = \frac{\epsilon_B}{\tau_B} + \frac{\epsilon_s}{\tau_s} = \frac{\epsilon_B}{\tau_B} + \frac{3v_F\epsilon_s}{8d}, \quad (4)$$

where ϵ_B and ϵ_s are the randomization of the spin orientation in each elastic bulk and surface scattering due to the

TABLE I. Measurements of $3\rho_B l_B/8$, ρ_B , $1/\tau_B$, ϵ_B , ϵ_s for four different substrates. The theoretical value of $3\rho_B l_B/8$ is $2.45 \times 10^{-16} \Omega \text{ m}^2$. v_F is $1.58 \times 10^6 \text{ ms}^{-1}$.

Sample no.	Substrate	$3\rho_B l_B/8$ ($10^{-16} \Omega \text{ m}^2$)	ρ_B ($n \Omega \text{ m}$)	$1/\tau_B$ (10^{12} s^{-1})	ϵ_B (10^{-5})	ϵ_s (10^{-5})
1	Soda glass	2.4	49	119	2.1 ± 0.4	22 ± 2
2	SiO	1.4	27	65	2.6 ± 1	48 ± 4
3	Lead glass	2.0	118	285	1.9 ± 0.8	148 ± 5
4	Soda glass	2.3	80	194	2.2 ± 0.2	12 ± 1

spin-orbit interaction. This procedure for separating the spin-orbit scattering from the surface and from the bulk may need some further clarification, since it is well known that the microcrystals in evaporated films grow with the film thickness. Thus spin-orbit scattering from the grain boundaries between these microcrystals will be counted as coming from the surface, and does not enter into the extrapolat-

ed bulk spin-orbit scattering we shall now extract. The measurements of $1/\tau_{SO}$ plotted versus $1/d$ in Fig. 2 lie on a straight line for thicknesses of the magnesium films above 10 nm. For thinner films, i.e., higher values of $1/d$ than 10^8 m^{-1} , there is a tendency to saturation of $1/\tau_{SO}$, presumably due to film inhomogeneities. From the straight line we can determine ϵ_B and ϵ_s , since we have already determined $1/\tau_B$ and $1/\tau_s$. In Table I the values of ϵ_B and ϵ_s are listed for magnesium films evaporated on soda glass (two samples), on lead glass, and on previously evaporated SiO (on soda glass).

As seen from Table I the spin-orbit scattering per bulk elastic scattering, ϵ_B , is independent of the substrate and has the value 2.2×10^{-5} in our most accurate measurement so far. The spin-orbit scattering per surface scattering, ϵ_s , depends strongly on the underlying substrate, and may even vary considerably for the same type of substrate. How well cleaned the substrate is when the magnesium is evaporated is likely to be important here. It is also important to notice that if only one of the film surfaces contributes to the spin-orbit relaxation ϵ , would be twice the values given in Table I. However, the general trend that the lead glass has values of ϵ_s , a factor of 10 higher than the values for soda glass is also found in other less complete measurements we have performed. A silicon monoxide substrate gives an intermediate value of ϵ_s .

An important question now is whether our results can be related to the material parameters of magnesium. In the bulk of a metal, impurity scattering leads to the spin-orbit relaxation. The type of impurity is crucial. This has been demonstrated by Bergmann and co-workers,^{3,10} who alloyed Au atoms into the surface of magnesium films, and showed that this had a tremendous effect on the spin-orbit interaction. However, it is still an open question what spin relaxation different impurities will cause. Presumably it depends on whether the impurity is inert in the metal or not. In the case of pure magnesium films the imperfection of the crystal structure is the dominant cause of the spin-orbit scattering and the one which we observe. Presumably a large part of these scattering centers are vacancies. In a free-electron approximation it is natural to expect that a vacancy in a magnesium crystal may scatter an electron in much the same way as an electron in free space would be scattered by a magnesium atom. The last situation was the one considered by Abrikosov and Gorkov⁶ and led to the result that on average in such a scattering (isotropic case) the spin orientation is randomized by the fraction $\epsilon = (\alpha Z)^4$, which for magnesium ($Z = 12$) is 5.9×10^{-5} .

Their calculation is only an order-of-magnitude calculation, based on an extrapolation from spin-orbit effects in atoms, so our results for bulk magnesium, $\epsilon_B = 2.2 \times 10^{-5}$,

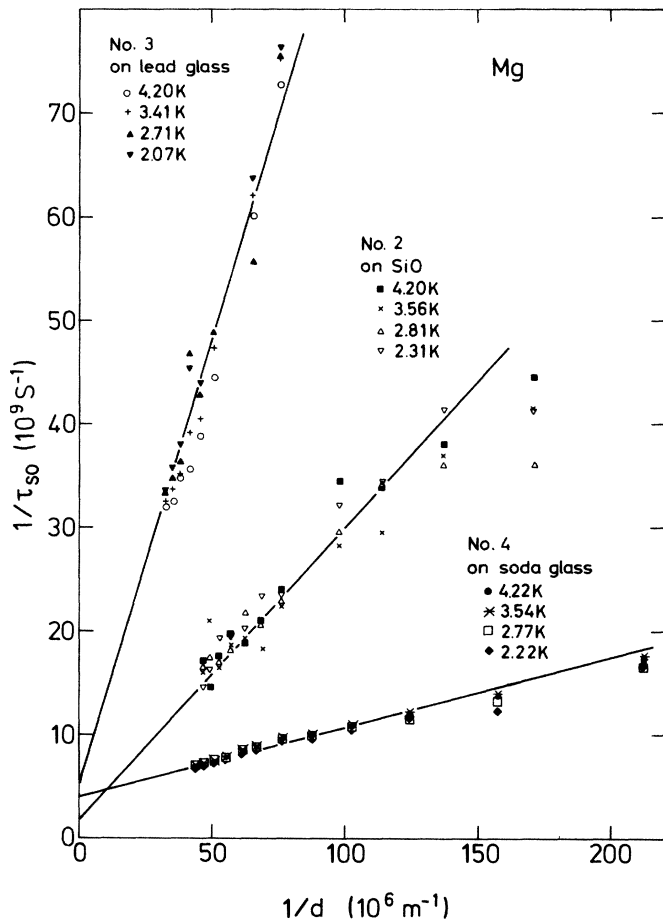


FIG. 2. The spin-orbit relaxation rate $1/\tau_{SO}$, as extracted from weak-localization magnetoresistance measurements, plotted as a function of the inverse film thickness $1/d$. The substrates used here are silicon monoxide, lead glass, and soda glass. All the films of different thicknesses have been measured at four different temperatures as indicated. The straight lines are valid theoretical fits only if the spin-orbit relaxation rate can be written as a linear combination of the bulk and surface elastic (transport) relaxation rates.

must be considered as supporting the picture we have given above. Furthermore, when it becomes important to extract the absolute value of ϵ_B we will have to make a suitable average over the Fermi surface. To justify such an enterprise a more precise theory is required. As regards the spin-orbit scattering from the film surface, there are roughly speaking two viewpoints. The electrons may be considered scattered from individual atoms, which is part of the dielectric substrates. Heavy atoms like lead will then give rise to a large spin-orbit interaction, as we observe in the case of lead glass. The other viewpoint is to let the electron interact with the dielectric material as a whole (band structure, etc.) during scattering. The spin-orbit interaction is connected with the details of the scattering process, and our observation that different dielectric substrates contribute very dif-

ferently to the spin relaxations must therefore await a more detailed theoretical investigation of scattering at interfaces.

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¹R. Meservey and P. M. Tedrow, Phys. Rev. Lett. **41**, 805 (1978).

²R. T. Schumacher and S.-K. Wang, Phys. Rev. B **10**, 2129 (1974).

³G. Bergmann, Phys. Rev. Lett. **48**, 1046 (1982); A. E. White, R. C. Dynes, and J. P. Garno, Phys. Rev. B **29**, 3694 (1984); C. van Haesendonck, M. Gijs, and Y. Bruynseraede, in *Localization Interaction and Transport Phenomena in Impure Metals*, edited by B. Kramer, G. Bergmann, and Y. Bruynseraede (Springer-Verlag, Berlin, 1985), p. 221.

⁴F. J. Dyson, Phys. Rev. **98**, 349 (1955).

⁵M. B. Walker, Phys. Rev. B **3**, 30 (1971).

⁶A. A. Abrikosov and L. P. Gorkov, Zh. Eksp. Teor. Fiz. **42**, 1088 (1962) [Sov. Phys. JETP **15**, 752 (1962)].

⁷S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).

⁸K. L. Chopra, *Thin Film Phenomena* (McGraw-Hill, New York, 1969), pp. 344–365.

⁹P. E. Lindelof, J. Nørregaard, and J. Bindslev Hansen, Z. Phys. B **59**, 423 (1985).

¹⁰G. Bergmann and C. Horriar-Esser, Phys. Rev. **31**, 1161 (1985).