Electron scattering times from weak localization studies of Au-Pd films

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(Received 14 February 1986)

Measurements of weak localization effects have been used to determine various electron scattering times in Au-Pd films with different thicknesses d and resistivities ρ, which have been prepared by sputtering and thermal evaporation. In all cases the inelastic scattering appears to be due to two-dimensional electron-electron scattering in the presence of disorder. For the sputtered films the magnitude of the inelastic scattering time τ_i is in reasonable agreement with the theory. However, a similar analysis of the results for the evaporated films seems to indicate that in this case the magnitude of τ_i is not consistent with the theory. The spin-spin scattering time τ_s was found to vary as d and ρ were changed. The observed variation of τ_s does not appear to be consistent with current theories.

I. INTRODUCTION

It is now fairly well established that the electrical properties of disordered metals are dominated by two effects, localization and electron-electron interactions.[1-6] A good deal of work in this area, especially in effectively two-dimensional systems,[4] i.e., thin films, has shown that the magnetoresistance in small magnetic fields is controlled by what has come to be known as weak localization. It has also been shown that measurements of weak localization can be used to determine various electron scattering times. These scattering times are in many cases difficult or impossible to measure with other techniques, and thus weak localization has the potential to become a powerful tool in the study of scattering processes in metals. We have recently reported results for the inelastic scattering time, τ_i, and the spin-spin (also called magnetic impurity) scattering time, τ_s, for sputtered Au-Pd films of various thicknesses.[7] Some features of those results were unexpected, and in this paper we describe further experiments which were designed to study in more detail the variation of τ_i and τ_s as the thickness, the resistivity, and the method of preparation of the Au-Pd were varied.

II. RESULTS

The essential features of our experiments and the analysis were the same as in our earlier work,[7] and will therefore not be discussed in detail here. The samples were thin films with thicknesses in the range 100—250 Å, which were prepared by either dc sputtering $Au_{40}Pd_{60}$, or thermal coevaporation of $Au_{60}Pd_{40}$. Both of these will be referred to as Au-Pd in the following. The as-prepared films had resistivities of 500 and 50 $\mu\Omega$ cm, respectively, at room temperature, and 480 and 46 $\mu\Omega$ cm at low temperatures. In addition, the resistivities of some of the sputtered samples were varied by annealing, which was accomplished by heating to typically 150 °C in an Ar atmosphere. In most cases glass substrates were used, although for a few samples quartz was employed. In the discussions below, the substrates will be assumed to be glass unless specifically stated otherwise. The magnetoresistance was measured for fields less than 10 kG, and temperatures in the range 1.5—15 K using standard techniques.[7]

The analysis was conducted in the same manner as in our previous work.[7] The results for the magnetoresistance were first fit to the full theoretical expressions,[8,9] in which the spin-orbit scattering time τ_{so}, and the phase breaking time τ_ϕ (which is a combination of τ_i and τ_s; see below), were both allowed to vary. The fits indicated that, as expected, the spin-orbit scattering in Au-Pd is very strong, and we were only able to set an upper limit of approximately 4×10^{-13} sec on τ_{so}. In the strong spin orbit scattering limit, which is appropriate in this case, the prediction for the magnetoresistance as a function of field is independent of the actual value of τ_{so}, and is given by[8,9]

$$\frac{\Delta R_\square}{R_\square^2}=\frac{e^2}{4\pi^2\hbar}\left[\psi\left[\frac{1}{2}+\frac{H_\phi}{H}\right]-\ln\left[\frac{H_\phi}{H}\right]\right]. \quad (1)$$

Here $R_\square$ is the sheet resistance, ψ is the digamma function, and the "phase-breaking" field H_ϕ is defined by

$$H_\phi=H_i+2H_s\ , \quad (2)$$

with $H_i=\hbar/4eD\tau_i$, etc., for τ_s. Note again that in obtaining (1) we have assumed that τ_{so} is much less than τ_i and τ_s, and also that the spin scattering is isotropic. The results given in this paper were all obtained from fits to (1), but fits to the full theory [including τ_{so} (Refs. 8 and 9)] gave essentially identical results.

The results for the magnetoresistance and the quality of the fits were similar to those shown in Ref. 7. The fits yielded the phase-breaking field H_ϕ directly (and as discussed above did not depend on the spin-orbit scattering rate). From H_ϕ the phase-breaking time τ_ϕ was determined. The quantity τ_ϕ is related to the inelastic and spin-spin times by [see (2)]

$$\frac{1}{\tau_\phi}=\frac{1}{\tau_i}+\frac{2}{\tau_s}\ . \quad (3)$$

Some results for τ_ϕ are shown in Fig. 1. Here we show results for a number of samples with different values of $R_\square$, thickness, and resistivity. The "rollover" seen at low temperatures in Fig. 1 can be understood in the following way.[10] The spin-spin time is due to scattering by magnetic impurities, and hence should be temperature independent, while τ_i is expected to increase as the temperature is decreased. Thus as the temperature is lowered and τ_i increases, τ_ϕ is dominated more and more by τ_s, and therefore approaches a constant, which from (3) is equal to $\tau_s/2$. Qualitatively similar behavior for τ_ϕ has been observed in a number of other materials, including Ag,[11] Cu,[11-13] and Ti.[14]

From the results in Fig. 1 we estimated τ_i in the following way.[15] Since we expect τ_i to vary as a power of temperature,[4-6] i.e., $\sim T^{-p}$, a plot of τ_ϕ^{-1} as a function of T^p should yield a straight line, whose slope and intercept yield the magnitude of τ_i and τ_s respectively. It was found that in all cases, i.e., for sputtered, evaporated, and annealed Au-Pd, $p \approx 1$ with an uncertainty of about 20% agreed best with the results. This is the value of p which corresponds to two-dimensional electron-electron scattering,[16-18] and this mechanism has been observed in past studies of metal films,[5,13,14,19,20] including our previous work on as-prepared sputtered Au-Pd.[7] Results for the magnitude of τ_i at 1 K are shown in Fig. 2. The solid lines in Fig. 2 are drawn proportional to $R_\square^{-1}$, and to within the uncertainties, all of the results for the sputtered films, both as-prepared and annealed, fall on common curves of this form. This is in agreement with the theory, which predicts[17]

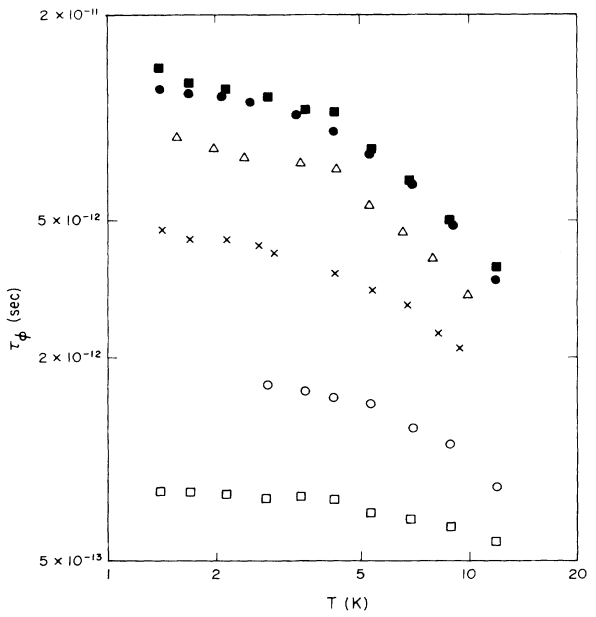

FIG. 1. Phase-breaking time, τ_ϕ, as a function of temperature for several samples: a sputtered, annealed sample with $R_\square = 157\Omega$ (■); a sputtered, annealed sample with $R_\square = 153\Omega$ (●); a sputtered, as-prepared sample with $R_\square = 327\Omega$ (△); a sputtered, as-prepared sample with $R_\square = 381\Omega$ (×); an evaporated sample with $R_\square = 27\Omega$ (○); an evaporated sample with $R_\square = 38\Omega$ (□).

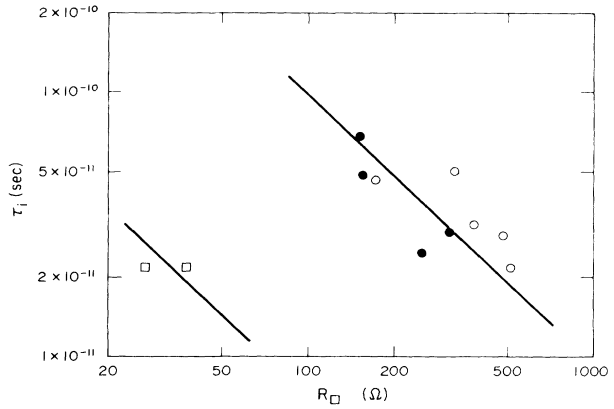

FIG. 2. Magnitude of τ_i at 1 K for sputtered, as-prepared samples (○), for sputtered, annealed samples (●), and evaporated samples (□). The solid lines are guides to the eye which are drawn proportional to $R_\square^{-1}$, which is the theoretically predicted dependence of τ_i on $R_\square$.

$$\frac{\hbar}{\tau_i} = \frac{e^2 R_\square}{2\pi\hbar} k_B T \ln\left[\frac{k_B T \tau_i}{\hbar}\right] . \tag{4}$$

The absolute magnitude of τ_i predicted by (4) is about a factor of 4 larger than the upper solid line in Fig. 2. It is thus in reasonable agreement with the experimental values for the sputtered films, especially considering that this comparison involves *no* free parameters. We also note that this level of agreement is comparable to that found in previous work.[4] However, the results for the evaporated films fall about a factor of 50 below the prediction (4), which seems much too large to be considered acceptable. The reason for this discrepancy is not clear, since according to the theory τ_i should depend *only* on the value of $R_\square$, and not on the composition of the sample or its method of preparation. Hence, the fact that the two types of samples were prepared in different manners should not, at least according to the theory, affect this comparison.

It is interesting to note that the magnitude of the inelastic "field," $H_i = \hbar/(4eD\tau_i)$ [see (2)] varies smoothly with $R_\square$. This is illustrated in Fig. 3 where it is seen that the results for *all* of the samples, sputtered and evaporated, fall approximately on a common curve, in contrast to Fig. 2. This suggests that the behavior is in fact a smooth, continuous function of $R_\square$ for all samples. It is only when the results are expressed in terms of scattering times, by essentially just normalizing the results in Fig. 3 by the diffusion constant D to obtain the scattering time, Fig. 2, that the difference between the two types of samples becomes evident. This would seem to suggest that the theory has somehow omitted a subtle dependence of τ_i on the diffusion constant, or that perhaps the factor of $R_\square$ which enters the theory (4) is for some reason different from the value measured experimentally. Another possibility is that our estimate of the diffusion constant is incorrect. However, it is hard to see why this estimate, which is based essentially on free-electron theory ideas,[21] should be accurate for the sputtered films, but fail so bad-

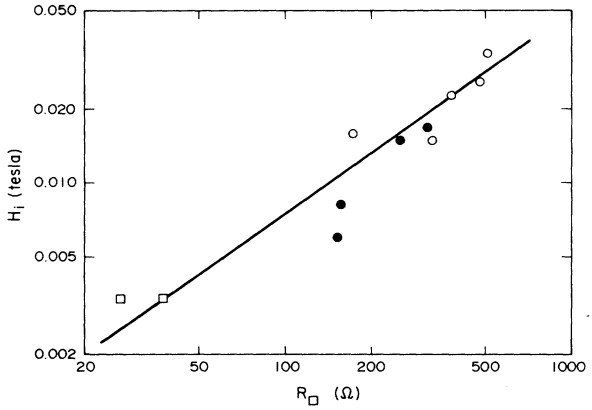

FIG. 3. Inelastic field as a function of $R_\square$, for sputtered, as-prepared samples (○), for sputtered, annealed samples (●), and evaporated samples (□). The solid line is a guide to the eye.

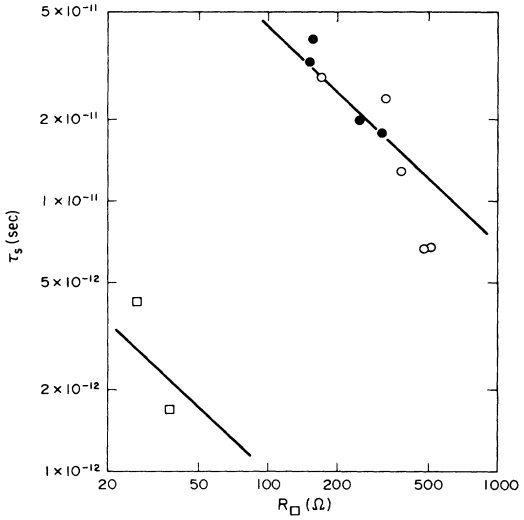

FIG. 4. Magnitude of τ_s for sputtered, as-prepared samples (○), for sputtered, annealed samples (●), and evaporated samples (□). The solid lines are guides to the eye which are drawn proportional to $R_\square^{-1}$.

ly for the evaporated films. Since the evaporated films have a lower resistivity by as much as a factor of 10, we would expect that the diffusion constants should also differ by approximately this factor, independent of the manner used to estimate the diffusion constant. In addition, measurements of interaction effects in zero magnetic field[22] can also be used to provide an independent estimate of D for sputtered and evaporated Au-Pd. Those experiments showed that sputtered and evaporated samples with the same resistivity exhibit interaction effects of the same magnitude, which implies that such samples have the *same* diffusion constants. This is strong support for our method of estimating D for these films. It is thus very hard to see how the discrepancy with the theory in Fig. 2 could be due to any mis-estimate of the diffusion constant.

Results for the spin-spin scattering time τ_s are given in Fig. 4. The value of τ_s is of order 10^{-11} sec, in good accord with the spin-spin scattering times observed in other systems.[11–14] However, we see that for the sputtered samples τ_s varies approximately as $R_\square^{-1}$. This is quite surprising since τ_s should be a "bulk" property, and therefore not depend on the film thickness. For the as-prepared samples, $R_\square \sim d^{-1}$ where d is the film thickness. One could argue that if the spin-spin scattering occurred predominantly at the surface of the film, then τ_s should also vary as $R_\square^{-1}$, since surface scattering would become more important (and more probable) as d is decreased. This explanation can be tested in the following way. We see from Fig. 4 that the annealed sputtered films also fall on the same common curve as the as-prepared sputtered samples. Since annealing changes $R_\square$ but not d, these results are *not* consistent with an explanation involving surface scattering. While nearly all of our samples were on glass substrates, we also prepared several on quartz, with the idea that perhaps the spin-spin scattering was due to magnetic impurities in the glass. However, the samples on glass and quartz substrates displayed quite similar behavior. Since the quartz substrates contained far fewer (magnetic) impurities, this suggests that scattering from the substrate did not contribute significantly to the spin-spin scattering. Returning to Fig. 4, we see that the results for the evaporated films are well below those for the sputtered samples, but also show the same trend; namely, that τ_s becomes smaller as $R_\square$ is increased. The change observed in going from the sputtered to the evaporated films could easily be due to a difference in the impurity content of the starting materials, which as noted above, was different in the two cases.

Our results seem to suggest that spin-spin scattering may be more complex than has been assumed to date. The theoretical models which have been considered have all assumed that the impurity spins fluctuate independently with time. This is certainly an oversimplification, since, for example, the interactions between impurities seem certain to make the magnitude of these fluctuations temperature dependent. While this fact has been pointed out by previous workers,[23] it does not appear to have been addressed theoretically. It is also possible that there is some other, hitherto unidentified, source of electron scattering which is both temperature independent and also dependent on the value of $R_\square$. Yet another possibility is that the strength of the impurity scattering which is responsible for τ_s is very sensitive to the metallurgical properties of the films, which are in turn a function of both thickness and annealing, etc. In any case, the consideration of more realistic models of spin-spin scattering would clearly be of interest.

III. DISCUSSION

There have been many previous studies of electron scattering times in metal films using measurements of weak localization, and the results of those studies have generally been interpreted as being in good agreement with the appropriate theories. However, there have actu-

ally been very few experiments in which a single scattering time, such as τ_i, has been carefully measured as sample properties were varied in a controlled, systematic manner. We have attempted to do just this in our experiments, and have measured both τ_i and τ_s for Au-Pd films made in different ways. The results are only in partial agreement with the theory. The inelastic scattering times observed for the sputtered films agree reasonably well with the theory (4). However, the values of τ_i for the evaporated films are substantially lower than predicted, by an amount (more than an order of magnitude) which would seem to be well outside the combined experimental and theoretical uncertainties.[24] This discrepancy is not understood, although it should be noted that our analysis has assumed that τ_s is temperature independent. As we have seen, the results for τ_s are also not understood. It is conceivable our assumption of a temperature independent τ_s may not be valid. If this is the case, then the method we have used to extract τ_i might not be appropriate, even though it seems to have yielded reasonable results in previous analyses (both by other workers, and for our sputtered samples).

The results for the spin-spin scattering times are also a puzzle. They suggest that either the spin-spin scattering process is more complicated than considered by current theories, or that perhaps there is another source of electron scattering which leads to a temperature-independent scattering time.

In conclusion, it appears that our understanding of electron scattering processes in metal films is far from complete. It is also conceivable that the "problem" lies with the theory of weak localization, since the scattering times have all been derived assuming the validity of this theory. In any case, further more detailed work in this area certainly seems to be called for.

ACKNOWLEDGMENTS

We thank D. E. Beutler for useful discussions and critical readings of the manuscript. This work was supported in part by the National Science Foundation through Grant No. DMR84-03995.

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[10]Similar behavior, i.e., a "rollover" of τ_ϕ has been observed in other studies of this kind (see, e.g., Ref. 4). It is usually ascribed to a change in the temperature dependence of τ_i (due to a change in the dominant inelastic mechanism), rather than to magnetic scattering. However, in our case this explanation seems very unlikely, since the temperature dependence of τ_i would have to be extremely weak to account for the behavior we observe. No known scattering mechanisms have values of the exponent p (where $\tau_i \sim T^{-p}$) which are small enough to be consistent with our results. Hence an explanation in terms of of magnetic scattering is much more plausible in the present case.

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[15]We note that direct least-squares fits to the data with p allowed to vary gave essentially the same results as this graphical method (see Ref. 7). Also, given possible systematic uncertainties, the best measure of the uncertainties in the derived parameters is probably not the standard error derived from the least-squares fits, but rather the sample to sample scatter in Figs. 2–4.

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[18]This result is the same as that obtained by Abrahams and coworkers. See the discussion by E. Abrahams in *Localization, Interaction, and Transport Phenomena,* Ref. 5, p. 245; E. Abrahams, P. W. Anderson, P. A. Lee, and T. V. Ramakrishnan, Phys. Rev. B **24**, 6783 (1981); H. Fukuyama and E. Abrahams, *ibid.* **27**, 5976 (1983); and H. Fukuyama, J. Phys. Soc. Jpn. **53**, 3299 (1984).

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[21]We estimated the diffusion constant from the relation $D = v_F^2 \tau_e / 3$, where τ_e is the elastic scattering time. The various parameters were estimated using free-electron theory in conjunction with the measured resistivity. We found $D = 2.2 \text{cm}^2/\text{sec}$ for the as-prepared sputtered films. The

values for the other samples scaled in the appropriate manner with the (measured) resistivity.

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[23]See, for example, *Localization, Interaction, and Transport Phenomena,* Ref. 5, and R. S. Markiewicz and C. J. Rollins, Phys. Rev. B **29**, 735 (1984); W. C. McGinnis and P. M. Chaikin, *ibid.* **32**, 6319 (1985).

[24]It is surprising that the theory agrees better with the results for the high-resistivity samples, since the theory is expected to work best in the clean (i.e. low-resistivity) limit.