Electron localization and interaction effects in palladium and palladium-gold films

W. C. McGinnis* and P. M. Chaikin†
Department of Physics, University of California, Los Angeles, California 90024
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We have measured the temperature and magnetic field dependence of the resistance for thin Pd and Pd-Au films ranging from 30 to 5000 Ω/□, as well as the temperature dependence of the Hall coefficient for Pd films of about 3000 Ω/□. The magnetoresistance of the Pd-Au films is well described by an electron localization model with strong spin-orbit scattering, while the effect of electron-electron interactions is small. The Pd film magnetoresistance is explained by the same model but with much stronger magnetic impurity spin scattering than the Pd-Au samples. Finally, the Hall coefficient of the Pd films is found to be independent of temperature, which indicates that single-particle effects are dominant over many-body electron interaction effects in these films, even though both appear to contribute to the zero-field resistance.

I. INTRODUCTION

Theoretical and experimental investigations of electronic transport in two-dimensional (2D) disordered media have led to some very interesting (and in some sense unexpected) predications and observations. A variety of systems has been studied experimentally, including metal- oxide-semiconductor field-effect transistors (MOSFET's),[1,2] in which a thin layer of electrons forms the two-dimensional system, as well as various metal films, ranging from clean copper films to dirty, granular aluminum films.[3–6] The prominent feature of all these systems in the weakly localized, or metallic, regime (that is, when $k_F l \gg 1$; k_F is the Fermi wave number and l is the electron elastic mean free path) is a logarithmic increase in resistance with decreasing temperature, a behavior which is definitely nonmetallic in nature. This unusual behavior can be explained theoretically, taking into account the presence of disorder, by either of two theories: electron localization[7,8] or electron-electron many-body interactions.[9] These can both give very similar corrections to the conductivity. In order to differentiate between the two effects, then, it is necessary to look at other properties, such as magnetoresistance and Hall effect, for which the theories make different predictions. Such an untanglement of the two contributions to the zero-field conductivity has been accomplished with some success in MOSFET's (Refs. 2, 10, and 11) (in which both localization and interaction effects are clearly seen) and, to a lesser extent, in metal films.[12–14] In the following sections, the results of resistance, magnetoresistance, and Hall-effect measurements of thin Pd and Pd-Au films are presented and analyzed in terms of the localization and interaction theories.

II. EXPERIMENT

The films were formed by electron-beam evaporation of 99.99% palladium (less than 9 ppm magnetic impurities), obtained from the Materials Research Corporation, or of an alloy of palladium (42 wt. %, or 57 at. %) and gold (58 wt. %, or 43 at. %). The amount of gold used is such that the Pd d band should be almost "filled,"[15] thus suppressing its "nearly magnetic" nature. The alloy was produced in an arc furnace in an atmosphere of 99.999% argon. The films were deposited at room temperature at a pressure of 8×10^{-7} to 5×10^{-6} Torr (during the evaporation) onto Pyrex glass slides at a rate of about 0.1 Å/s (controlled by a Sloan 9000 deposition controller). The films studied ranged in thickness from 18 to 150 Å, as measured with a quartz-crystal thickness monitor. Thick silver pads (~3000 Å) for making electrical connections to the films were deposited immediately after the films were put down. This sequence of depositions was done in a single pump-down cycle. The resultant geometry is shown as part of Fig. 1.

The transport measurements were performed with the sample slide mounted on a copper block inside a double vacuum-can rig which was immersed in liquid helium. A small amount of ^{4}He or ^{3}He exchange gas was put in the inner can to provide good thermal contact to the sample.

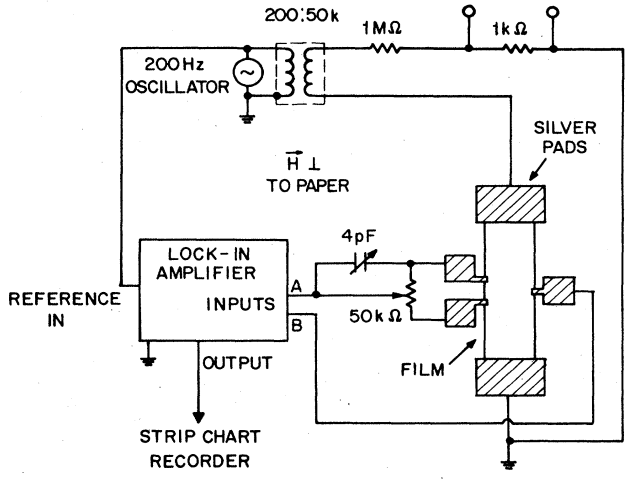

FIG. 1. Sample geometry and circuit used for measuring Hall voltage. The film is 0.96 cm long and 0.32 cm wide.

Temperatures below 1 K were achieved by condensing ^{3}He into the inner vacuum can, and then pumping on the liquid. The sample temperature was measured using either a calibrated germanium resistor, a silicon diode, or a capacitance thermometer calibrated after cool-down to 4.2 K with one of the other thermometers.

Four-probe resistance measurements of the films as a function of temperature or magnetic field were performed using an ac Wheatstone-type bridge, capable of measuring fractional changes in resistance to an accuracy of one part in 2×10^4, at a frequency of 200 Hz.

The Hall voltage was measured using a five-probe ac technique (see, for example, Ref. 16), illustrated in Fig. 1. The 50-kΩ potentiometer is adjusted so that there is zero in-phase voltage detected in zero field. The 4-pF compensation capacitor is adjusted to balance out any out-of-phase signal. The Hall voltage was measured as a function of temperature in a constant magnetic field of 1 T. Several field reversals were done at a given temperature while monitoring the lock-in–amplifier output on a strip-chart recorder. This procedure eliminates any contribution to the measured Hall voltage due to magnetoresistance. Measurements performed on a 620-Ω/□ Pd film as a function of magnetic field showed that the difference in Hall voltage between the two field orientations ($+90°$ and $-90°$ with respect to the film plane) varies linearly with the field.

III. EXPERIMENTAL RESULTS AND ANALYSIS

A. Temperature dependence of the conductivity

According to both the localization and interaction theories, the change in the 2D conductivity σ of a two-dimensional disordered system (in the weakly localized regime) in going from a temperature T to T_0 is given by[8,9]

$$\Delta\sigma=\zeta\alpha_T\ln(T/T_0)\ , \tag{1}$$

where $\zeta=e^2/2\pi^2\hbar\simeq1.23\times10^{-5}\ \Omega^{-1}$, $\Delta\sigma=\sigma(T)-\sigma(T_0)$, and α_T is the logarithmic slope (normalized by ζ). For the moment, we are neglecting the effects of spin-orbit and magnetic impurity spin scattering. In terms of the resistance R of a square section of film, Eq. (1) can be written (for $\Delta R\ll R$; that is, for $R\simeq R_0$) as

$$\frac{\Delta R(T)}{R_0^2}=\frac{R(T)-R(T_0)}{R^2(T_0)}=-\zeta\alpha_T\ln\left[\frac{T}{T_0}\right]. \tag{2}$$

The factor α_T predicted by the two theories is given in the first two columns of Table I. The value of p depends on the particulars of the dominant electron inelastic scattering mechanism (the inelastic scattering rate ω_ϵ generally depends on temperature as[17] $\omega_\epsilon\sim T^p$), while F, the average scattering matrix element between two states on the Fermi surface, is determined by the degree of screening by other electrons.[9] Estimates of the most likely values of ω_ϵ are found in Appendix A. Values of F based on the ratio k_F/κ are ~0.5 for Pd and ~0.6 for Pd-Au (see Appendix A; κ is the inverse screening length). If it is assumed that localization and interaction effects are independent (not necessarily a good assumption), then the two can be combined as shown in the third column of Table I.

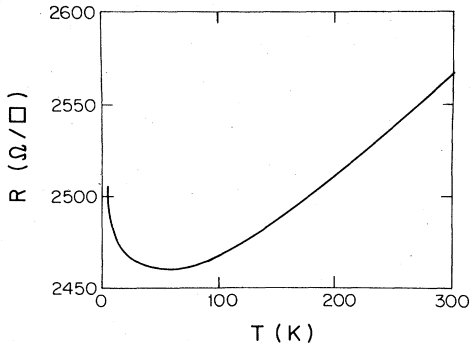

FIG. 2. Resistance as a function of temperature for an $\sim$2500-Ω/□ Pd film from room temperature down to 4.2 K.

Figure 2 shows the resistance as a function of T for a $\sim$2500-Ω/□ Pd film from room temperature down to 4.2 K. The good metallic, three-dimensional behavior ($\Delta R\sim T$) is evident at high temperatures, and the nonmetallic, two-dimensional behavior ($\Delta R\sim-\ln T$) is seen at low temperatures.

The temperature dependence of R in terms of the localization and interaction theories is then characterized by the logarithmic slope α_T. For a given material (that is, for a given amount of electron-electron screening), α_T should be a constant independent of the film resistance. If a film has a resistance R_0 at a temperature T_0, then a plot of $\zeta^{-1}\Delta R(T)/R_0$ versus $\ln T$ should be a straight line of slope $\alpha_T R_0$. A subsequent plot of these slopes versus R_0 for a set of films of various resistances should then yield a straight line of slope α_T. This procedure has been followed for Pd films ranging from 30 to 5000 Ω/□, as well as for several Pd-Au films. The resultant plots are shown in Fig. 3. The values of α_T obtained by a least-squares fit to a line are

$$\alpha_T=\begin{cases}0.97\ \text{(Pd)}\ ,\\ 0.70\ \text{(Pd-Au)}\ .\end{cases}$$

The results should be compared with Table I, keeping

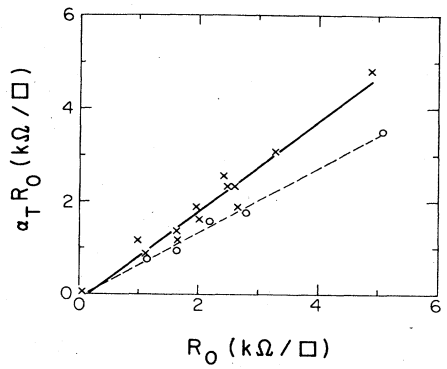

FIG. 3. Least-squares fit to a line of the logarithmic slope $\alpha_T R_0$ (from resistance versus $\ln T$ plots) as a function of resistance per square R_0 at $T=3$ K for several Pd (×) and Pd-Au films (○).

TABLE I. Predictions for the logarithmic slope α_T.

	Localization[a]	Interaction[b]	Localization plus interaction
Weak spin-orbit scattering	p	$1-\frac{3}{4}F$	$p+1-\frac{3}{4}F$
Strong spin-orbit scattering	$-\frac{p}{2}$	1	$-\frac{p}{2}+1$

[a]See Refs. 18 and 19. It is assumed here that there is one dominating inelastic scattering mechanism.
[b]See Refs. 20–23. The weak spin-orbit case is for $F/2 \ll 1$. The strong spin-orbit case does not include the contribution from the particle-particle channel (see Ref. 24).

in mind the predicted values of p (1, 2, 3, or 4) (Refs. 17 and 25–28) and F (between 0 and 1) (Ref. 9). Of course, it is expected that spin-orbit scattering is important in these materials since the scattering rate is given by[29]

$$\omega_{\text{s.o.}}=(\alpha Z)^4\omega' , \tag{3}$$

where α is the fine-structure constant, Z is the atomic number ($Z=46$ for Pd and $Z=79$ for Au), and ω' is a scattering rate related to either elastic scattering[29] or surface collisions.[30] In addition, scattering by magnetic impurity spins should also be included, particularly in Pd.[31] Within the constraints of the values of p and F, however, it is not possible to describe the temperature dependence of the resistivity as being either in the strong or weak spin-orbit limit when only a single inelastic scattering mechanism is considered, or when impurity spin scattering is ignored.

In general, more than one inelastic scattering mechanism may contribute to ω_ϵ, or magnetic impurity spin scattering may be present, in which case the localization portion of the resistance change is given by[19]

$$\frac{\Delta R(T)}{R_0^2}=\zeta\left[-\frac{3}{2}\ln\left[\frac{\omega_1(T)}{\omega_1(T_0)}\right]+\frac{1}{2}\ln\left[\frac{\omega_2(T)}{\omega_2(T_0)}\right]\right] , \tag{4}$$

where

$$\omega_1=\tfrac{4}{3}\omega_{\text{s.o.}}+\tfrac{2}{3}\omega_s+\omega_\epsilon , \tag{5}$$

$$\omega_2=2\omega_s+\omega_\epsilon ,$$

and it is assumed that ω_0, the elastic scattering rate, is large compared to $\omega_{\text{s.o.}}$ (the spin-orbit rate) and ω_s (the electron-spin—magnetic-impurity-spin rate). In other words, the logarithmic slope α_T will, in general, be a function of temperature. This effect is in fact seen in these films, with α_T slowly increasing as the temperature is lowered from ~ 5 to ~ 0.5 K. The values of α_T plotted in Fig. 3 are generally taken from the low-temperature portion of the data.

Any analysis of the individual contributions of electron localization and electron-electron interactions to the conductivity of these two-dimensional systems, based on the temperature dependence alone, is difficult. Other properties, in particular the magnetoresistance and Hall effect, are more amenable to such analysis since the predictions of the two theories are so different.

B. Magnetic field dependence of the conductivity

The signature of localization and interaction effects in the magnetoresistance of two-dimensional disordered systems appears in the high-field regime (beyond the characteristic field at which the behavior is no longer quadratic), where we expect

$$\frac{\Delta R(H)}{R_0^2}=\frac{R(H)-R(0)}{R^2(0)}=-\zeta\alpha_H\ln(H)+\text{const} . \tag{6}$$

The theoretically predicted values for the logarithmic coefficient are given in Table II for both the perpendicular and parallel field, where g is the electron-electron interaction constant (related to the screening parameter F; see Appendix A) and the spin-orbit scattering is considered strong when $\omega_{\text{s.o.}} \gg \omega_\epsilon, \omega_s$. The interaction constant g is estimated to be 0.07 for Pd and Pd-Au (see Appendix A).

The logarithmic dependence of the magnetoresistance on H at high fields is seen in Fig. 4 for Pd and Pd-Au films with resistances of 34 and 77 $\Omega/\square$, respectively. The sample numbers listed in this and subsequent figures refer to Table V. In Figs. 4–8, a "perpendicular" magnetic field is one which is perpendicular to the film plane, while a "parallel" field is parallel to the film plane and perpendicular to the current direction. The coefficients α_H (for $H>0.8$ T) for the Pd-Au film of Fig. 4 and for Pd sample 1 are

$$\alpha_{H_\perp}=\begin{cases}-0.69 \text{ (Pd)} ,\\ -0.57 \text{ (Pd-Au)} ,\end{cases}$$

$$\alpha_{H_\parallel}=\begin{cases}-0.86 \text{ (Pd)} ,\\ -0.91 \text{ (Pd-Au)} .\end{cases}$$

(The behavior of the Pd film of Fig. 4 will be discussed later.) From Table II, and the above value of g, it is seen that the experimental values of α_H are far too big if the results are to be explained in terms of the interaction

TABLE II. Predictions for the logarithmic slope α_H. (Loc. denotes localization, Int. denotes interaction.)

	Perpendicular field		Parallel field	
	Loc.[a]	Int.[b]	Loc.[c]	Int.[d]
Weak spin-orbit scattering	1	$-g$	2	$-2g$
Strong spin-orbit scattering	$-\frac{1}{2}$	$-\frac{g}{4}$	-1	$-\frac{g}{2}$

[a]See Ref. 19.
[b]See Ref. 32.
[c]See Refs. 33 and 34.
[d]See Ref. 35.

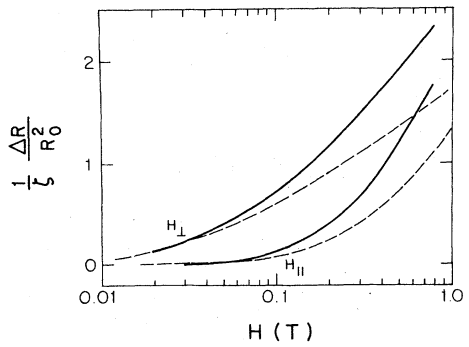

FIG. 4. Magnetoresistance of low-resistance-per-square Pd (solid lines, $R_0=34\ \Omega/\square$ and $T=0.55$ K) and Pd-Au (dashed lines, sample 5 with $R_0=77\ \Omega/\square$ and $T=0.68$ K) films in perpendicular and parallel magnetic fields. R_0 is the resistance per square in zero field, and $\Delta R=R(H)-R_0$.

theory alone. The results agree fairly well, however, with the predictions of localization theory for the expected case of strong spin-orbit scattering. (The parallel-field values are somewhat small because the high-field regime has not fully been reached.) The effect of spin-orbit coupling on the magnetoresistance of two-dimensional metal films has been clearly demonstrated experimentally by Bergmann.[6]

One obvious difference between the Pd and Pd-Au films is the magnitude of the perpendicular-field magnetoresistance (for the same resistance per square). At the same time, the parallel-field magnetoresistance is approximately the same for the two metals. Figure 5 demonstrates this point for films with resistances of approximately 70 and 1130 $\Omega/\square$. The perpendicular-field result can be qualitatively explained by localization theory in terms of the difference in the scattering rates ω_2 of Pd and Pd-Au. The relevant localization expressions for the magnetoresistance for both field orientations are[19,33,34]

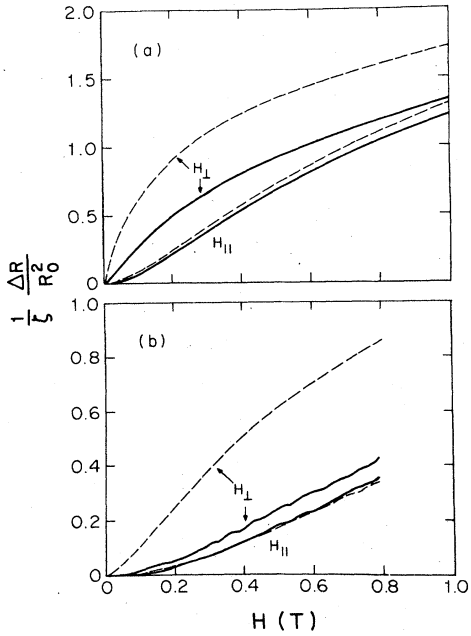

FIG. 5. Magnetoresistance of Pd (solid lines, samples 1 and 2) and Pd-Au (dashed lines, samples 5 and 6) films with (a) $R_0\simeq 70\ \Omega/\square$ and (b) 1130 $\Omega/\square$, respectively, in perpendicular and parallel magnetic fields at $T=0.68$ K. R_0 is the resistance per square in zero field, and $\Delta R=R(H)-R_0$.

$$\frac{1}{\zeta}\frac{\Delta R(H_\perp)}{R_0^2}=-\frac{3}{2}\left[\psi\left[\frac{1}{2}+\frac{\omega_1}{\omega_a}\right]-\ln\left[\frac{\omega_1}{\omega_a}\right]\right]+\frac{1}{2}\left[\psi\left[\frac{1}{2}+\frac{\omega_2}{\omega_a}\right]-\ln\left[\frac{\omega_2}{\omega_a}\right]\right], \quad (7a)$$

$$\frac{1}{\zeta}\frac{\Delta R(H_\parallel)}{R_0^2}=-\frac{3}{2}\ln\left[1+\frac{\omega_H}{\omega_1}\right]+\frac{1}{2}\ln\left[1+\frac{\omega_H}{\omega_2}\right], \quad (7b)$$

where

$$\omega_a=\frac{4eDH}{\hbar c}, \quad \omega_H=\frac{D}{48}\left[\frac{4eHd}{\hbar c}\right]^2, \quad (8)$$

and it is assumed that ω_0, the elastic scattering rate, is large compared to the other scattering rates (D is the diffusion constant and d is the film thickness). Equations (7) do not include Zeeman-splitting effects (see Appendix B for the full expressions), since they are expected to be small for the case of strong spin-orbit scattering[36] [see Eq. (B3)].

For the expected case of large spin-orbit scattering, the terms in Eqs. (7) which depend on ω_2 will dominate the terms which depend on ω_1. That is, the perpendicular-field magnetoresistance will be determined by the ratio ω_2/ω_a. For example, in the high-field limit $\omega_1\gg\omega_a\gg\omega_2$, Eq. (7a) becomes

$$\frac{1}{\zeta}\frac{\Delta R(H_\perp)}{R_0^2}=\frac{1}{2}\ln\left[\frac{\omega_a}{\omega_2}\right]. \quad (9)$$

Since the value of ω_a is approximately the same for Pd and Pd-Au, the smaller observed perpendicular magnetoresistance of Pd indicates a larger ω_2 than for the Pd-Au. This would be realized if, for example, the magnetic spin scattering were stronger in the Pd [a situation that would not be too surprising since small amounts of magnetic impurities form "giant moments" in Pd (Ref. 37)]. The parallel-field magnetoresistance is not as easily explained, however, since it similarly should depend on the ratio ω_2/ω_H. In the high-field limit $\omega_1\gg\omega_H\gg\omega_2$, Eq. (7b) becomes

$$\frac{1}{\zeta}\frac{\Delta R(H_\parallel)}{R_0^2}=\frac{1}{2}\ln\left[\frac{\omega_H}{\omega_2}\right]. \quad (10)$$

Thus, the equal values of parallel-field magnetoresistance for the two metals point to equal values of ω_2/ω_H (with ω_H approximately the same for Pd and Pd-Au), which is inconsistent with the conclusion reached for the perpendicular field.

The dependence of the magnetoresistance on film resistance [or the elastic scattering rate ω_0, which enters Eqs. (7) through the diffusion constant D] is illustrated in

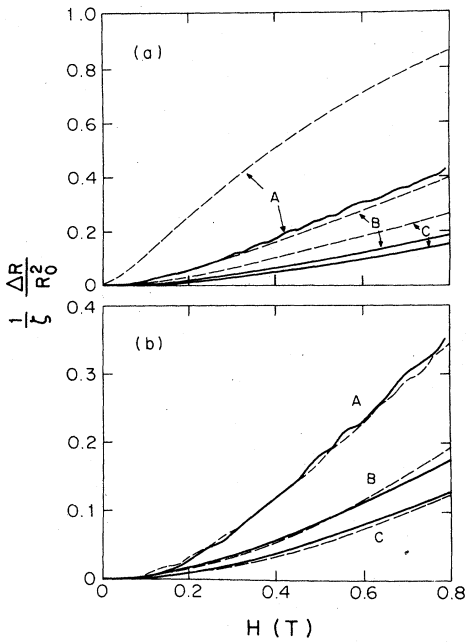

FIG. 6. Magnetoresistance of Pd (solid lines, samples 2—4) and Pd-Au (dashed lines, samples 6—8) films in perpendicular and parallel magnetic fields at $T \simeq 0.6$ K. The zero-field resistances per square R_0 are A, 1130; B, 2720; and C, 5300 Ω/□, and $\Delta R = R(H) - R_0$.

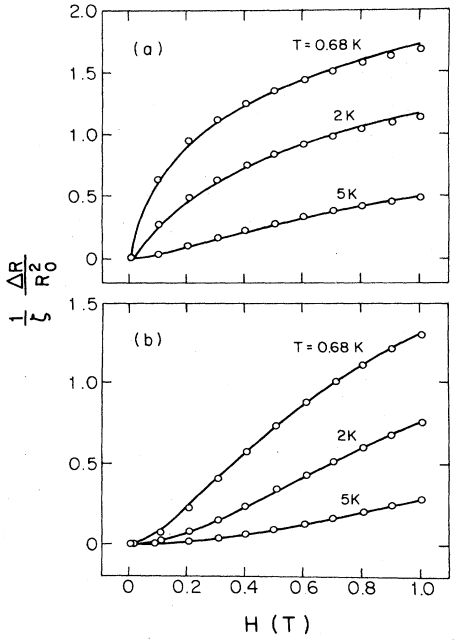

FIG. 7. Magnetoresistance of a Pd-Au film (solid line, sample 5 with a zero-field resistance R_0 of 77 Ω/□) at various temperatures in (a) perpendicular and (b) parallel magnetic fields, with $\Delta R = R(H) - R_0$. A least-squares fit to the localization expressions (see discussion in text) yields the points shown.

Fig. 6. The general trend is that the lower the resistance, the lower the characteristic field H_c at which the behavior changes from quadratic to logarithmic. This is to be expected, in terms of localization theory, since both ω_a and ω_H are proportional to ω_0^{-1} (with $R \sim \omega_0$), and H_c depends on the ratio ω_2/ω_a or ω_2/ω_H [as illustrated by Eqs. (9) and (10)]. Note that the difference between the perpendicular and parallel magnetoresistance, as well as the difference between the perpendicular-field results for Pd and Pd-Au, therefore becomes much less pronounced in films with high resistance per square.[38]

The magnetoresistance of the two low-resistance-per-square Pd and Pd-Au films at various temperatures is shown in Figs. 7 and 8. From localization theory, the temperature dependence can be attributed mainly to the T dependence of ω_ϵ, the inelastic scattering rate ($\omega_\epsilon \sim T^p$). Least-square fits of the data to the localization predictions [Eqs. (7)] are shown as points in the figure. Because fits to the full expressions of Eqs. (7) are rather insensitive to the spin-orbit scattering rate (as long as it is large compared to ω_a and ω_H), only the terms involving ω_2 were used in the final fit. The perpendicular-field data was fit by varying ω_2 using an estimated value of ω_0 (based on the film resistivity; see Appendix A). The resulting value of ω_2 was then used to fit the parallel-field data by varying the thickness d. It should be noted that the value of ω_2 determined by these fits depends on the value of ω_0 used. The reason is that the magnetoresistance expressions are functions of D/ω_2 [that is, the fit is really to the characteristic length $L_2 = (D/\omega_2)^{1/2}$]. The value of L_2 thus determined is large compared to the film thickness (see Table VI), which is consistent with the assumed two-

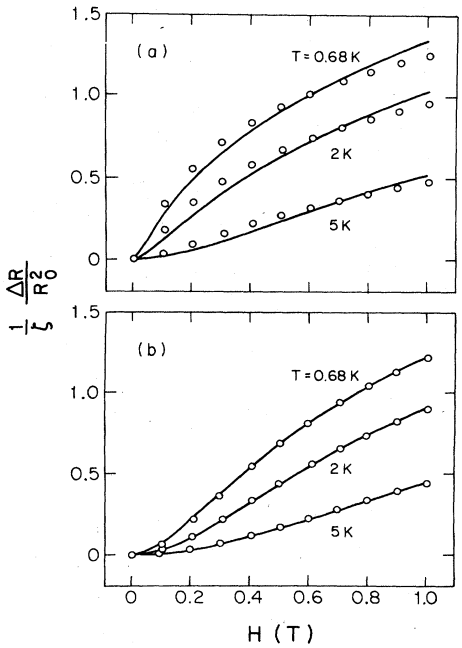

FIG. 8. Magnetoresistance of a Pd film (solid line, sample 1 with a zero-field resistance R_0 of 64 Ω/□) at various temperatures in (a) perpendicular and (b) parallel magnetic fields, with $\Delta R = R(H) - R_0$. A least-squares fit to the localization expressions (see discussion in text) yields the points shown.

dimensional nature of the films with respect to inelastic scattering. The preliminary fits, in which both ω_1 and ω_2 were varied, indicated that ω_1 (and therefore $\omega_{s.o.}$) is larger than $\sim 1\times 10^{13}$ s^{-1} for both the Pd and Pd-Au. The characteristic length[39] $L_1=(D/\omega_1)^{1/2}$ is therefore comparable to the film thickness, if not smaller, and so the films may be three-dimensional with respect to spin-orbit scattering. The 3D contribution to the magnetoresistance[40,41] is small, however, compared to that in two dimensions [Eqs. (7)].

Up until now we have assumed that ω_2 is independent of magnetic field. The spin-flip scattering of the conduction electrons by magnetic impurity spins will, however, be "frozen out" as $g_L\mu_B H/k_B T$ becomes greater than 1. Therefore, from Eq. (5), ω_2 will decrease with increasing field (particularly when ω_s is comparable to or larger than ω_ϵ). Experimentally, this effect will be seen as an increase in the magnetoresistance (at high fields) compared to that expected for the case of a field-independent ω_2. Just as the magnetoresistance increases with decreasing temperature, it also increases with increasing magnetic field (ω_2 gets smaller in both cases). In addition, for $\omega_s > \omega_\epsilon$, the decrease of ω_s with field will cause the logarithmic slope α_H to be larger in magnitude than predicted in Table II (until ω_s is again smaller than ω_ϵ). In fact, these effects are seen in Fig. 7(a) and 8(a) for $H_\perp > 0.7$ T, with the Pd film showing the largest difference between the experimental and fitted values (which indicates that the Pd film has a larger ω_s than the Pd-Au). As further proof, Pd films to which Fe was intentionally added (by making an evaporation source, using an arc furnace, with approximately 15 ppm Fe dissolved in Pd) showed enhanced values of α_H, in perpendicular field, equal to about -1.5. Even "pure" Pd films produced not long after these films (and other films with higher Fe concentrations) had large values of α_H, as seen in Fig. 4.

To avoid the problem of spin-flip scattering "freeze-out," the data were refitted using the same procedure as above, but only up to $H=0.25$ T for the 0.68- and 2-K data, and up to 0.5 T for the 5-K data. Also fitted over the full field range of 1 T were data taken at 10 K (not shown in Figs. 7 and 8). The fit at each temperature and for both the Pd-Au and Pd was much improved (with the fit virtually overlapping the data in the specified field ranges). The scattering rates ω_2 obtained from these fits are best described by the temperature dependence $\omega_2=AT^2+BT+C$, with coefficients given in Table III.

The coefficient C, which can be identified with $2\omega_s$ from Eq. (5), is much larger in the Pd film than in the Pd-Au film (as expected from the above discussion concerning the general behavior of the perpendicular-field magnetoresistance). The BT term most closely corresponds to a contribution to ω_ϵ from impurity-induced electron-electron scattering,[27,42] while the AT^2 term appears to arise from electron-phonon scattering[28] (also as modified by disorder). The nearly equal values of A for Pd and Pd-Au argues against the scattering of s electrons from spin fluctuations in the d band[43] (see also the review by de Chatel and Wohlfarth[44]). Other possible sources of inelastic electron scattering are discussed in great detail by Bergmann[45,46] and by Kaveh and Wiser.[47] Estimates of the coefficients for these samples based on the scattering mechanisms mentioned above are listed in Table III (see Table VI for details). The various parameters used to calculate the Pd-Au coefficients are based on the individual Pd and Au values (see Table IV) and the atomic fractions of each in the Pd-Au alloy (that is, $x_{\text{Pd-Au}}=0.57x_{\text{Pd}}+0.43x_{\text{Au}}$).

TABLE III. Coefficients of $\omega_2=AT^2+BT+C$.

	A (K^{-2}s^{-1})	B (K^{-1}s^{-1})	C (s^{-1})
	Experiment		
Pd-Au	5.5×10^9	7.4×10^9	4.7×10^9
Pd	4.5×10^9	1.6×10^{10}	3.1×10^{10}
	Estimated		
Pd-Au	2.8×10^8	3.6×10^9	$<3.6\times 10^9$
Pd	1.0×10^8	2.9×10^9	$<3.6\times 10^9$

The scattering rates ω_2 obtained from these fits can be used to calculate the localization contribution to the zero-field value of $\Delta R/R^2$ [given by the second term in Eq. (4) for the case of strong, temperature-independent spin-orbit scattering]. When interaction effects are included (with a contribution to α_T of 0.93 and 0.78 for these low-resistance Pd-Au and Pd films, respectively), the values of the coefficients A, B, and C obtained from the magnetoresistance data nicely account for the observed temperature dependence of the zero-field resistance (for $T<3$ K), which for these particular films gives low-temperature values of α_T of 0.49 for Pd-Au and 0.68 for Pd.

The fact that α_T is larger for Pd than for Pd-Au (see Fig. 3) can also be understood as follows. For the case of strong spin-orbit scattering, the contributions to α_T by localization and interaction effects are opposite in sign (see Table I). From the experimental coefficients listed in Table III, effective values of the exponent p at low temperatures are approximately 1 for the Pd-Au film (a value between 0 and 2) and 0 for the Pd film (since the impurity-spin scattering rate is so large). Estimates of α_T from Table I are then 0.5 for Pd-Au and 1 for Pd, which illustrates the point. Furthermore, the slow increase of α_T with decreasing temperature is understood in terms of ω_s becoming larger than ω_ϵ (see Refs. 48 and 49 for the same effect in copper and silver films).

Even with the consistency between the resistance and magnetoresistance provided by these results, though, some problems remain. The apparent requirement of equal values of ω_2/ω_H for Pd and Pd-Au mandated by the parallel-field results (for the same resistance per square) has been artificially satisfied in the above fits by varying the thickness d. The necessity of this procedure has been observed in previous studies.[50] In contrast to these studies, however, the fitted value of d does not seem to depend on the measured film thickness, but rather has a value which is characteristic of the material: 242 Å for Pd-Au and 358 Å for Pd. The fitted values of d for the films listed in Table V all fall within about 10% of these values.

Finally, the fact that the parallel-field results for the two metals are the same indicates that the magnetoresis-

TABLE IV. Quantities given in the literature (bulk values).

Quantity	Pd	Au	Refs.
Atomic number Z	46	79	
Effective-mass ratio m^*/m	1.66	1.05	65,66
Fermi energy E_F (J)	1.21×10^{-18}	1.19×10^{-18}	65,66
Density of states N ($J^{-1}cm^{-3}$)	1.06×10^{41}	0.65×10^{41}	65,66
Speed of sound[a] v_s ($cm\,s^{-1}$)	2.63×10^{5}	1.20×10^{5}	68,69
Mass density ρ_M ($gm\,cm^{-3}$)	12.16	19.4	69
Debye temperature Θ_D (K)	274	165	69

[a]The value given is the transverse speed of sound. The Pd value is calculated using the longitudinal speed of sound where the ratio of longitudinal to transverse speed is (Ref. 67) $[2(1-\sigma_p)/(1-2\sigma_p)]^{1/2}$ (with a Poisson ratio $\sigma_p=0.35$).

tance arises from the same mechanism for both. This would rule out any contribution to the magnetoresistance due to Zeeman effects in an exchange-enhanced material with spin-orbit scattering.[51] In addition, calculations of the magnetoresistance due to this effect (including corrections[52] to the expressions of Ref. 51) show that, at $T=0.68$ K and $H=1$ T, $\xi^{-1}\Delta R(H=1\text{ T})/R_0^2$ is essentially zero for $\omega_{s.o.}\geq 3\times10^{12}$ s^{-1}, even for a strongly enhanced material (such as Pd). Furthermore, as seen in Fig. 6, the parallel-field magnetoresistance is strongly dependent on the film resistance per square, which would not be the case if it were due to Zeeman splitting (as calculated in Ref. 53). It should also be noted that the results shown in Figs. 4—8 cannot be explained at all in terms of interactions effects alone (for either field orientation). The magnitude of the magnetoresistance is much larger than that expected from orbital effects.[32,54] Fits of the perpendicular-field data of Figs. 7 and 8 to the magnetoresistance expression[32] describing orbital effects yield interaction constants (a direct measure of the amplitude of the effect) which are much larger than expected theoretically. In other words, the interaction contribution to the magnetoresistance, based on the estimated values of g, is much less than the localization contribution.

TABLE V. Typical experimentally measured quantities.

Sample	Thickness d (Å)	R ($\Omega/\square$) at $T=3$ K
	Pd	
1	116	64
2	22	1109
3	25	2642
4	18.5	4892
	Pd-Au	
5	145	77
6	24	1135
7	25	2787
8	23	5060

C. Hall effect

The temperature dependence of the Hall coefficient should give even more insight concerning the presence of localization and interaction effects than does the magnetoresistance. If only localization effects are important, the Hall coefficient R_H should be independent of temperature[55,56] (even for the case of strong spin-orbit scattering[57]). For the case of interaction effects being dominant, the fractional change in R_H should be twice the fractional change in resistance (due to interaction effects alone) over the same temperature range:[55,56]

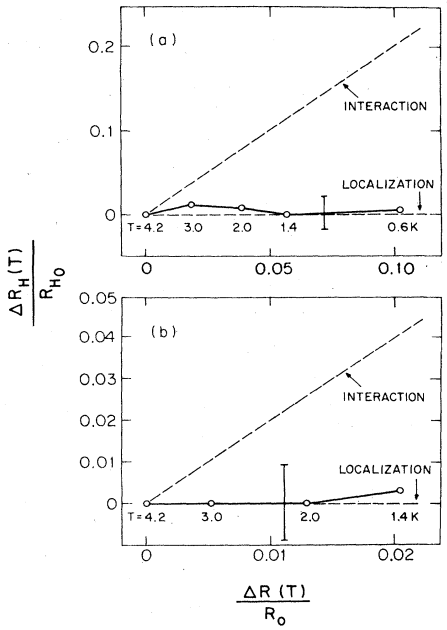

FIG. 9. Fractional change in Hall coefficient as a function of the fractional change in the resistance for Pd films with zero-field resistances per square R_0 at $T=4.2$ K of (a) 3360 $\Omega/\square$ and (b) 2506 $\Omega/\square$ (same sample as in Fig. 2). The data points are shown as circles, with the solid line serving as a guide. The experimental uncertainty in the Hall values is indicated by the error bars. The theoretical predictions are shown as dashed lines. R_{H_0} is the Hall coefficient at $T=4.2$ K, $\Delta R_H=R_H(T)-R_{H_0}$, and $\Delta R=R(T)-R_0$.

TABLE VI. Calculated quantities.

Quantity	Given by	Refs.	Sample 1	Sample 3
Fermi velocity v_F (cm s^{-1})	$(2E_F/m^*)^{1/2}$		1.27×10^8	1.27×10^8
Electron concentration n (cm^{-3})	$\frac{2}{3}NE_F$		8.55×10^{22}	8.55×10^{22}
Resistivity ρ (Ω cm)	Rd		7.4×10^{-5}	6.6×10^{-4}
D (cm^2 s^{-1})	$(Ne^2\rho)^{-1}$		5.0	0.56
l[a] (3D) (Å)	$3D/v_F$		11.7	1.3
Localization length[b] L_2 (Å)	$(D/\omega_2)^{1/2}$	17	477	160
Interaction length L_T (Å)	$(D\hbar/k_BT)^{1/2}$	9	275	92
Phonon wavelength λ_{ph} (Å)	$\hbar v_s/k_BT$		40	40
k_F (Å^{-1})	$m^*v_F/\hbar$		1.83	1.83
κ (3D) (Å^{-1})	$(4\pi e^2N)^{1/2}$		1.75	1.75
k_Fl	$3m^*D/\hbar$		22	2.4
$2k_F/\kappa$			2.1	2.1
F	$\frac{K}{\pi(1-K^2)^{1/2}}\ln\left[\frac{1+(1-K^2)^{1/2}}{1-(1-K^2)^{1/2}}\right]$, $K=(2k_f/\kappa)^{-1}$, $0<K<1$	9,71	0.47	0.47
g[c]	$\left[\frac{1}{\lambda}+\ln\left[\frac{\gamma E_F}{\pi k_BT}\right]\right]^{-1}$, $\lambda=0.24$, $\ln\gamma=0.577$	32,23	0.07	0.07

$$\frac{R_H(T)-R_H(T_0)}{R_H(T_0)}=2\frac{R(T)-R(T_0)}{R(T_0)}\ ,$$

or

$$\frac{\Delta R_H}{R_{H_0}}=2\frac{\Delta R}{R_0}\ , \tag{11}$$

where all quantities are measured at a given magnetic field H. It can be shown that if both interaction and localization effects are present, such that the correction to the conductivity can be written as $\delta\sigma=\delta\sigma_L+\delta\sigma_I$ (and similarly for the Hall conductivity), then the change in the Hall coefficient is related to the resistance change by (see Ref. 58 for a similar expression in the weak-spin-orbit-scattering, high-magnetic-field limit)

$$\frac{\Delta R_H}{R_{H_0}}=\frac{2\Delta\sigma_I}{\Delta\sigma_L+\Delta\sigma_I}\frac{\Delta R}{R_0}\ , \tag{12}$$

where

$$\Delta\sigma_L=\delta\sigma_L(H,T)-\delta\sigma_L(H,T_0)$$

and

$$\Delta\sigma_I=\delta\sigma_I(H,T)-\delta\sigma_I(H,T_0)\ .$$

The coefficient relating $\Delta R_H/R_{H_0}$ to $\Delta R/R_0$ can therefore change in value as the relative importance of localization and interaction effects changes (with magnetic field, for example). Such behavior has been observed in Si MOSFET's.[59]

Figure 9 shows the fractional change in the Hall coefficient with respect to $R_H(T=4.2\ \text{K})$ for two different Pd samples for the temperature range 0.6 to 4.2 K. This is plotted against the fractional change in the resistance with respect to $R(T=4.2\ \text{K})$. Also shown is the expected change in this temperature range assuming that only localization or only interaction effects are important. The data show that there is no change in R_H in almost a decade in temperature, which suggests that single-particle localization is mainly responsible for the electronic transport behavior of these films in this temperature and magnetic field range. Such a conclusion, however, is at odds

TABLE VI. (*Continued*).

Quantity	Given by	Refs.	Sample 1	Sample 3
ω_0 (s^{-1})	v_F/l		1.1×10^{15}	9.6×10^{15}
Electron-phonon inelastic scattering rates[d] (3D) (s^{-1})	$\frac{2}{3}\left[\frac{m^*k_B}{\hbar\Theta_D}\right]^2\frac{DN}{\rho_M}\left[\frac{v_F}{v_s}\right]^3T^4$	25	1.3×10^{6}	1.5×10^{6}
	$2\pi^2\frac{a}{k_Fl}\frac{k_BT^2}{\hbar\Theta_D}$	28	2.5×10^{9}	2.3×10^{10}
	$a=\frac{nm^*}{6\rho_M}\left[\frac{k_B\Theta_D}{\hbar k_F}\frac{v_F}{v_s^2}\right]^2$			
Electron-electron inelastic scattering rate[e] (2D) (s^{-1})	$\frac{3\pi}{2}\frac{l}{\hbar d}\frac{k_BT}{(k_Fl)^2}\left\lvert\ln\left[\frac{T}{T_1}\right]\right\rvert$	27,42	1.4×10^{10}	3.8×10^{11}
	$T_1=\frac{32}{27}\frac{m^*e^4}{2\hbar^2k_B}(k_fl)^3$			
$\omega_{s.o.}$ (s^{-1})	$(\alpha Z)^4\omega_0$	29	1.4×10^{13}	1.2×10^{14}
ω_s (s^{-1})	$\frac{\text{ppm magnetic impurities}}{5\times10^{-9}\text{ s}}$	73	$<1.8\times10^{9}$	$<1.8\times10^{9}$
ω_a (s^{-1})	$\frac{4eDH}{\hbar c}$	19	7.6×10^{11}	8.5×10^{10}
ω_H (s^{-1})	$\frac{D}{48}\left[\frac{4eHd}{\hbar c}\right]^2$	34	5.1×10^{10}	2.7×10^{8}

[a] *In situ* measurements of resistance R versus film thickness d show (Ref. 70) that $R\sim1/d$, which indicates that the elastic scattering length l is limited by impurity scattering rather than being thickness limited (since, by definition, the resistance per square is $R=\rho/d\sim1/ld$), and therefore is less than d.
[b] A value of $\omega_2=2.2\times10^{11}$ s^{-1} is used, corresponding to $T=5$ K (see Table III).
[c] The bare interaction constant λ is taken to be equal to $F/2$ [compare Eqs. (14a) of Ref. 32 and (2.23) of Ref. 23].
[d] Since $\lambda_{ph}\gg l$, samples 1 and 3 are both in the dirty limit in terms of electron-phonon scattering. The thicker film has $d/\lambda_{ph}\simeq3$, and is therefore considered three-dimensional. The thinner film has $d/\lambda_{ph}\simeq0.6$, and so is marginally two-dimensional. Coupling to the substrate, however, will likely enhance the three-dimensional character of the phonons in the film.
[e] The scattering rate given here is the impurity-induced inelastic electron-electron scattering rate. Both samples are considered two dimensional ($d<L_T$) with 3D screening ($d\gg\kappa^{-1}$) and 3D diffusion ($d\gg l$). A contribution proportional to T^2 (given by Ref. 26 as $(\pi/8)[(k_BT)^2/\hbar E_F]\simeq1.5\times10^7$ s^{-1}), which follows from Landau's Fermi-liquid theory (Ref. 72), is also present but is much smaller.

with the necessity of including interaction effects to explain the temperature dependence of the zero-field resistance. Nevertheless, a temperature-independent Hall coefficient has also been observed (over a much larger temperature range) by Ovadyahu and Imry[60] in their investigation of thin indium oxide films. Others, however, have seen R_H change with temperature.[61,62]

IV. DISCUSSION

A brief comparison of the results obtained here to those presented in related papers by Markiewicz and Rollins[14] and Dumoulin *et al.*[63] is in order. The following similarities in the Markiewicz and Rollins study and the present one can be noted. Both involve Pd films whose magne-

toresistance has been fitted to localization expressions which incorporate the theoretical results of Hikami, Larkin, and Nagaoka[19] as well as those of Altshuler and Aronov.[34] The positive magnetoresistance observed is found to be due to strong spin-orbit scattering, while the contribution of interaction effects is small. There are, however, important differences between these two papers, too. Markiewicz and Rollins studied Pd which was sputtered onto Si substrates so that the films were either in the form of Pd silicides or Pd/Pd_2Si bilayers. This complication does not exist for our films since they were evaporated onto glass slides. In the first place, Pd is not known to react with SiO_2 to form silicides under normal deposition conditions,[64] and secondly, no superconducting transition (as would be expected for Pd_2Si) was observed in our films down to a temperature of 150 mK.[38] Another difference is that their thickest film (which is the bilayer film and probably the closest match to our films) has a magnetoresistance which is ~ 10 times smaller than that observed in our films (at $H=1$ T). In addition, they find that the magnetoresistance changes nonmonotonically with temperature (which they describe as a drooping effect), whereas we have found that the magnetoresistance continually increases as the temperature is lowered (down to ~ 0.6 K). Finally, the efforts of Markiewicz and Rollins were concentrated on the high-magnetic-field regime (up to 13.5 T), while in the present paper, the (0—1)-T range was studied in more detail. Also, we went to slightly lower temperatures (0.6 K compared to 1.6 K). In these respects the two investigations may be considered to be complimentary as far as the magnetoresistance measurements are concerned.

The portion of the paper by Dumoulin *et al.* involving pure Pd (they also looked at palladium hydrides) gives results quite similar to those of the present paper as far as the resistance and magnetoresistance (perpendicular-field) measurements are concerned. Their films were produced under conditions very close to those described here (but under better vacuum, producing "cleaner" films). Dumoulin *et al.* found that the magnetoresistance, which increased monotonically as T decreased, was well described by localization theory [using the term in Eq. (7a) involving ω_2]. Their measurements were done only in a perpendicular field. Finally, they also conclude that, based on the magnetoresistance data, interaction effects must make a substantial contribution to the change of the zero-field resistance with temperature.

V. CONCLUSION

As discussed above, localization and interaction theories predict that the resistance of the thin Pd and Pd-Au films studied here should vary logarithmically in given regimes with temperature and magnetic field. The dependences are characterized by the coefficients α_T and α_H, respectively. When magnetic impurity spin scattering is strong, these quantities will no longer be constants but will depend on temperature and magnetic field. In addition, the localization theory predicts that the Hall coefficient R_H should be temperature independent, while the interaction theory predicts a fractional change in R_H which is twice as large as the fractional change in the film resistance over the same temperature range.

The values of α_T observed, 0.97 for Pd and 0.70 for Pd-Au, are most likely comprised of contributions from both localization and interaction effects. The two effects cannot be separated, however, on the basis of the temperature dependence alone. It is difficult, in fact, to conceive of a justifiable combination of inelastic scattering exponent p (based on a single scattering mechanism) and screening factor F that produce the α_T seen experimentally.

The magnetoresistance data obtained clarify the situation considerably. The logarithmic coefficients α_H for Pd-Au and Pd are in fair agreement with localization predictions for the case of strong spin-orbit scattering. The interaction theory, however, predicts values of α_H on the order of -0.03, which is not found for either material. In addition, least-square fits of the magnetoresistance to the localization theory give good agreement for the Pd-Au films, and also for the Pd films when the "freezing out" of spin-flip scattering is taken into account. The parameters obtained from these fits are consistent with the temperature dependence of the zero-field resistance when interaction effects are included. The parallel-field magnetoresistance, however, is still not fully understood. Finally, fits to the interaction expressions for the magnetoresistance give interaction constants that are much too large.

The Hall-effect measurements show that R_H is independent of temperature, which according to current theory indicates the domination of localization effects over interaction effects in these films. This observation is made over the same temperature range where the resistance varies logarithmically with T. Such a conclusion, however, is inconsistent with the apparent need to include interaction effects to explain the zero-field-resistance temperature dependence in materials which show strong spin-orbit scattering. This suggests that perhaps the localization and interaction corrections cannot simply be added together.

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APPENDIX A: ESTIMATES OF VARIOUS QUANTITIES

Tables IV—VI provide estimates of the pertinent physical parameters of the Pd and Pd-Au films studied. In Table VI estimates involving the temperature T are made for $T=5$ K, and those involving the magnetic field H are made for $H=1$ T.

APPENDIX B: LOCALIZATION EXPRESSIONS FOR THE MAGNETOCONDUCTIVITY

The following expressions for the magnetoconductivity due to localization effects incorporate orbital, Zeeman, spin-orbit, and impurity spin scattering:[19,33,34]

$$\Delta\sigma(H_\perp)=\zeta\left\{-\left[\psi\left[\frac{1}{2}+\frac{\omega_0}{\omega_a}\right]-\ln\left[\frac{\omega_0}{\omega_a}\right]\right]+\left[\psi\left[\frac{1}{2}+\frac{\omega_1}{\omega_a}\right]-\ln\left[\frac{\omega_1}{\omega_a}\right]\right]\right.$$
$$+\frac{1}{2}\left[\frac{1}{\beta}\psi\left[\frac{1}{2}+\frac{(\omega_1+\omega_2)+\beta(\omega_1-\omega_2)}{2\omega_a}\right]-\ln\left[\frac{\omega_1}{\omega_a}\right]\right]$$
$$\left.-\frac{1}{2}\left[\frac{1}{\beta}\psi\left[\frac{1}{2}+\frac{(\omega_1+\omega_2)-\beta(\omega_1-\omega_2)}{2\omega_a}\right]-\ln\left[\frac{\omega_2}{\omega_a}\right]\right]\right\}, \tag{B1}$$

$$\Delta\sigma(H_\parallel)=\zeta\left\{\ln\left[1+\frac{\omega_H}{\omega_1}\right]+\frac{1}{2}\left[\frac{1}{\beta}\ln\left[\frac{\omega_1+\omega_2+2\omega_H+\beta(\omega_1-\omega_2)}{\omega_1+\omega_2+2\omega_H-\beta(\omega_1-\omega_2)}\right]-\ln\left[\frac{\omega_1}{\omega_2}\right]\right]\right\}, \tag{B2}$$

where

$$\beta=\left[1-\left[\frac{2g_L\mu_B H}{\hbar(\omega_1-\omega_2)}\right]^2\right]^{1/2}, \tag{B3}$$

g_L is the Landé g factor, μ_B is the Bohr magneton, ω_0 is the elastic scattering rate, and the other scattering rates (assumed much smaller than ω_0) are given in Eqs. (5) and (8). These expressions are taken from Eqs. (A·9) and (A·20) of Maekawa and Fukuyama[33] (although written in a slightly different form). All types of scattering are assumed to be isotropic, so that $\omega_x=\omega_y=\omega_z=\omega/3$ for each scattering rate ω. The parallel-field expression (also see Refs. 74, 50, 71, and 14) includes the orbital effects predicted by Altshuler and Aronov[34] by making the substitution[71] $\omega_\epsilon\rightarrow\omega_\epsilon+\omega_H$ in the equations of Maekawa and Fukuyama.

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*Present address: Code 633, Naval Ocean Systems Center, San Diego, CA 92152.

†Present address: Department of Physics, University of Pennsylvania, Philadelphia, PA 19104, and Exxon Research and Engineering Company, Annandale, NJ 08801.

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