Weak-localization magnetoresistance in quench-condensed lithium films

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We have studied the magnetoresistance due to weak localization in quench-condensed lithium films using fields both perpendicular and parallel to the plane of the sample. Very few studies have examined the magnetoresistance in metal films in both field orientations, and some puzzling anomalies have been reported. It has been proposed that some of these anomalies are connected with spin effects. For the samples we report on here, no spin effects were evident, and no anomalies were observed. In order to characterize our samples accurately, we have developed a method for determining the thickness of a thin film from the weak-localization magnetoresistance and the sheet resistance, valid for all mean free paths. Our method allows us to measure the film thickness with an error of 8% or less without any knowledge of the mean free path or surface specularity. Once we know the thickness, we may establish a range of possible values for the impurity mean free path. Applying this analysis to our films, we find that electron transport in these samples is quasiballistic (the impurity mean free path is greater than the film thickness), despite the fact that they were quench condensed. [S0163-1829(98)10131-5]

I. INTRODUCTION

Weak localization of conduction electrons is a quantum-mechanical interference effect that gives rise, at low temperatures, to corrections to the semiclassical Drude conductivity. It is due to interference between pairs of semiclassical paths related by time reversal.[1] Applying a magnetic field destroys the time-reversal symmetry and suppresses this contribution to the conductivity. The resulting magnetoresistance reveals a wealth of information about the basic properties of the sample under study. In thin films the weak-localization magnetoresistance is strongly anisotropic: the semiclassical paths are nearly confined to a plane, and the paths couple much more strongly to a field applied perpendicular to the sample plane than to a parallel field. Measurement of magnetoresistance in both parallel and perpendicular fields provides more information about a sample than does a measurement in only one field orientation.

Although there have been several studies of weak-localization magnetoresistance in both parallel and perpendicular fields in semiconductor heterostructures,[2] and in anisotropic materials,[3] few experiments have been performed in metal films, and some of these have observed surprising anomalies. Giordano and Pennington[4] observed anomalies in the high-field magnetoresistance of gold films that obey weak localization well at lower fields, and they saw behavior in silver films and iron-doped gold films that does not agree with weak localization even in weak fields. Both Refs. 5 and 6 reported anomalies in the conduction electron g factor, as measured by the weak-localization magnetoresistance. At least some of these anomalies were conjectured to be related to spin effects, but so far no comprehensive theory has emerged to accurately account for them. We chose to work with lithium, which has very low intrinsic spin-orbit and magnetic scattering rates,[7] so that we may observe the weak-localization magnetoresistance in the absence of spin effects, and so that we may add spin effects in a controlled manner by doping our films with various impurities. In this paper, we report only on our observation of undoped films.

To characterize our samples accurately, and as an example of the additional information that can be obtained by using both perpendicular and parallel fields, we have measured the thicknesses of our films by combining the weak-localization data and the sheet resistance. This measurement is simple when the mean free path is very short compared to the film thickness. When the mean free path is comparable to or greater than the film thickness, however, extracting a measurement of the film thickness from the weak-localization magnetoresistance is more subtle. Our method, which is based on the theory of Beenakker and van Houten,[8] makes no assumption about either the surface specularity or the magnitude of the impurity mean free path, and is thus applicable to diffusive, quasiballistic, and ballistic films. We developed this procedure to characterize our quench-condensed lithium films because no other thickness measurement was available.

We also show how to measure the impurity mean free path using the semiclassical model of Fuchs and Sondheimer,[9] together with the thickness obtained from the weak-localization data. The impurity mean free path is the mean free path between collisions with bulk impurities and does not count scattering from the surfaces of the film. Our extracted value is strongly sensitive to the surface specularity, but when this is not known we can still find upper and lower bounds on the impurity mean free path. From this analysis we find that our films are quasiballistic (the impurity mean free path is greater than the film thickness) despite the fact that they were prepared by quench condensation.

II. THEORY

The weak-localization correction to the sheet conductance of a film in a perpendicular magnetic field is given by[1]

$$\delta g(B_z)=\frac{e^2}{h}\frac{1}{\pi}\left\{\psi\left(\frac{1}{2}+\frac{\hbar c}{4eD\tau_\phi B_z}\right)-\psi\left(\frac{1}{2}+\frac{\hbar c}{4eD\tau B_z}\right)\right\}, \quad (1)$$

where ψ is the digamma function, D is the diffusion constant, τ is the elastic-scattering time, and τ_ϕ is the inelastic dephasing time. For fields less than about 1 T, the dependence on τ is negligible, and the magnetoconductance only depends on one undetermined parameter, the product $D\tau_\phi$, which is conventionally defined as the square of the dephasing length,

$$L_\phi^2 = D\tau_\phi .$$

In a parallel field, we can write the magnetoconductance in the weak-field limit as[1,10]

$$\delta g(B_x) = \frac{e^2}{\pi h}\left\{ \ln\left[1+\frac{1}{3}\left(\frac{e}{\hbar c}\right)^2 (a_{\rm eff}L_\phi)^2 B_x^2\right] - \ln\left[\frac{\tau_\phi}{\tau}\right]\right\}. \tag{2}$$

The parameter $a_{\rm eff}$ is an effective film thickness. It is a measure of the coupling between the semiclassical paths of the conducting electrons and the parallel magnetic field. (We will come back to this coupling term later.) Again, the term involving τ has no field dependence, and the only parameter that affects the magnetoconductance is the product $a_{\rm eff}L_\phi$. Thus, by combining the parallel and perpendicular weak-localization data, we can determine both the dephasing length L_ϕ and the effective thickness $a_{\rm eff}$.

In mixed fields, the field component parallel to the plane of the sample has the effect of shortening the dephasing length,[11]

$$\delta g(B_x,B_z) = \frac{e^2}{\pi h}\left\{ \psi\left[\frac{1}{2} + \frac{\hbar c}{4eL_\phi^2(B_x)B_z}\right] - \psi\left[\frac{1}{2} + \frac{\hbar c}{4eD\tau B_z}\right]\right\}, \tag{3a}$$

where

$$\frac{1}{L_\phi^2(B_x)} = \frac{1}{L_\phi^2} + \frac{1}{3}\left(\frac{e}{\hbar c}\right)^2 a_{\rm eff}^2 B_x^2. \tag{3b}$$

For very thin films, those with thicknesses comparable to or even less than the conduction electrons' mean free path, surface effects can play an important role in electron transport. When the film thickness is much greater than the mean free path, electron transport is said to be diffusive, while in the opposite limit it is said to be ballistic. The case where the thickness is between these two extremes is referred to as the quasiballistic regime. Expressions (1)–(3) are valid regardless of whether transport is ballistic, quasiballistic, or diffusive. In the diffusive regime, $a_{\rm eff}$ is equal to the film thickness. The relationship between $a_{\rm eff}$ and the sample thickness in the ballistic or quasiballistic regime is more subtle and will be discussed in detail below.

III. EXPERIMENTAL PROCEDURE

Figure 1 shows a schematic of the quench condensation system. It fits into a conventional, top-loading ^{4}He cryostat. A copper, water-cooled electromagnet fits around the outside of the cryostat, and provides fields perpendicular to the plane of the sample. Parallel fields were provided by a superconducting Nb-Ti Helmholtz coil inside the cryostat. The coil forms for the Helmholtz pair and the sample stage were machined out of a single block of aluminum to provide an accurate and consistent alignment of the field with the plane of the sample. This alignment is critical for a parallel field measurement, because even a slight misalignment introduces a field component perpendicular to the plane of the film, producing a large additional, unwanted magnetoconductance signal. The alignment was checked for each sample by holding the nominally parallel field fixed, and varying the perpendicular field to find the magnetoconductance minimum (i.e., the point where the applied perpendicular field cancels the perpendicular component introduced by the Helmholtz pair). In all cases, the parallel field was found to be aligned with the plane of the sample to an accuracy of 0.6° or better. The perpendicular component produced by this misalignment introduces a negligible additional magnetoconductance. Alignment of the perpendicular field is not critical.

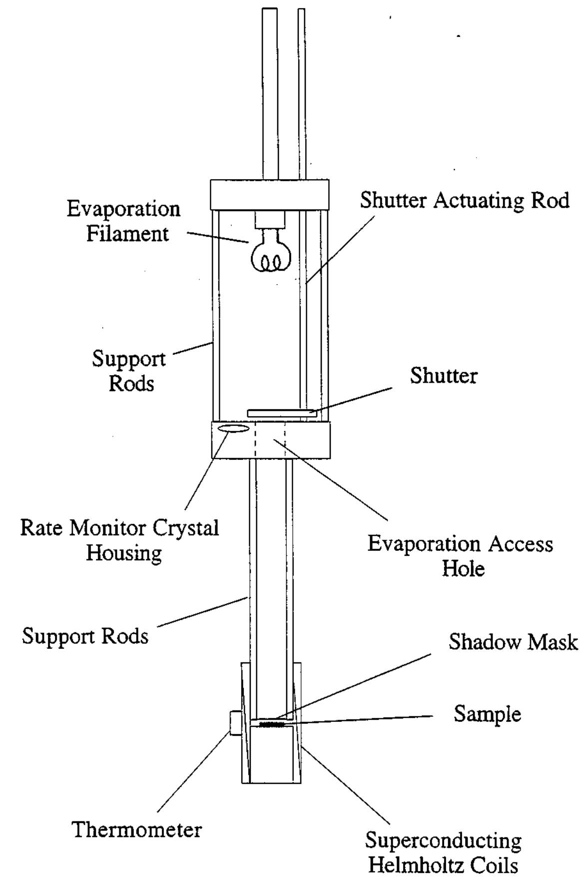

FIG. 1. Experimental apparatus. This unit goes into a conventional ^{4}He cryostat.

The films were grown under a vacuum of 10^{-8} torr or better at temperatures ranging from 45 to 49 K. After growth, the samples were annealed at about 70 K for 1 h after which an exchange gas of helium was introduced to the experimental space to facilitate cooling. A crystal rate monitor provided a rough estimate of the film thickness as the samples were grown.

Resistances were measured using a four-wire, nulled lock-in technique. The magnets were driven by a GPIB power supply controlled by a Macintosh IIcx computer. The same computer was used to collect the data. Residual noise in the 1-Hz pass band of the lock-in was averaged away by the computer. This data collection procedure was checked

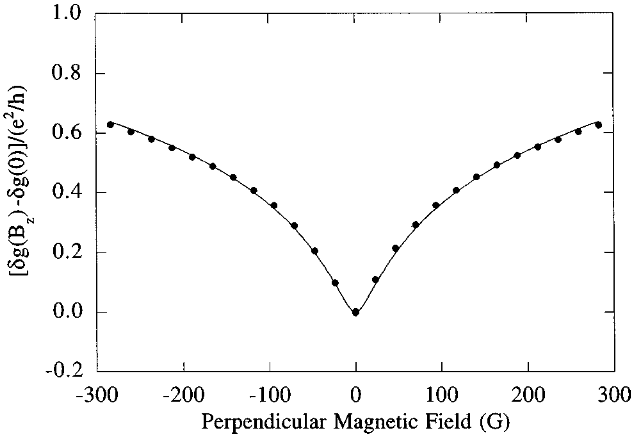

FIG. 2. Magnetoconductance of sample 1 in a perpendicular field. Dots are data. Lines are a fit to the spinless weak-localization theory given in Eq. (1).

against analog methods. (Analog methods were not used to collect the final data because they did not include signal averaging and were noisier than the digital method.) Data were collected at two temperatures, 4.0 K (the boiling point of liquid helium in Boulder) and 2.2 K.

IV. RESULTS

Figure 2 shows the magnetoconductance in a perpendicular field for sample No. 1. Figure 3 shows the magnetoconductance of sample 1 in a parallel field. Figure 4 shows the magnetoconductance of the same sample (No. 1) in a mixed field, where the parallel field was held fixed at 3 kG, and the perpendicular field was swept. In each case, dots are experimental data, and lines are theory. For the data taken in purely perpendicular fields, we have adjusted L_ϕ to produce a fit. For data taken in purely parallel fields, we varied the product $a_{\rm eff}L_\phi$ to achieve a good fit. In each case, agreement between theory and experiment is good. The fit values for each of the samples are reported in Table I. For all of the films we report here, the fits were of comparable quality. No additional parameters were adjusted to fit the mixed field data. We took the value of L_ϕ and $a_{\rm eff}$ obtained from Fig. 2, and calculated

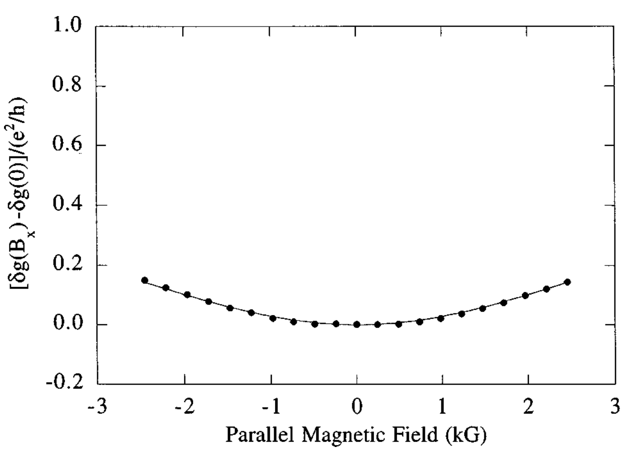

FIG. 3. Magnetoconductance of sample 1 in a parallel field. Dots are data. Lines are a fit to the spinless weak-localization theory given in Eq. (2).

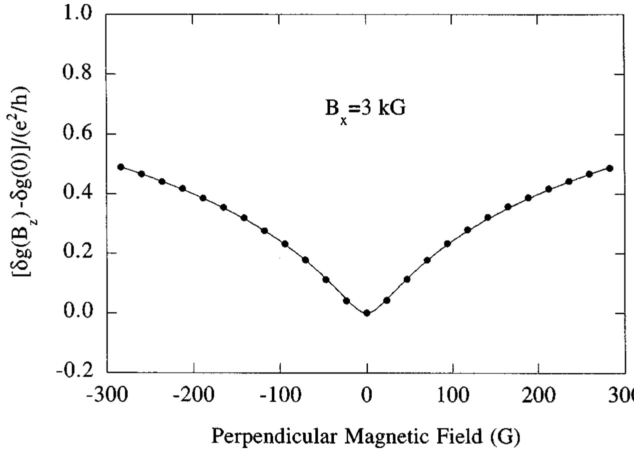

FIG. 4. Magnetoconductance of sample 1 in mixed fields. The parallel field was held fixed at 3 kG, while the perpendicular field was varied. The values of L_ϕ and $a_{\rm eff}$ determined from Figs. 2 and 3 were used to calculate $L_\phi(B_x)$, and the solid line is the magnetoconductance predicted from Eq. (3), normalized to zero for a zero perpendicular field.

the effective $L_\phi(B_x)$ from Eq. (3b), using Eq. (3a) to predict the magnetoconductance. The theory shown in Fig. 4 is that prediction, and it agrees well with the data.

V. ANALYSIS AND DISCUSSION

An excellent account of the data reported here has been obtained using the weak-localization theory without spin effects. There is no sign of spin scattering in this data. Our observations are consistent with the idea that the anomalies seen by other workers are connected with spin effects.

In order to accurately characterize our films, we have de-

TABLE I. Experimental parameters and calculations of thickness and mean free path of quench-condensed lithium films using the analysis presented. Both diffusive and specular surface scattering are considered.

					Diffusive		Specular	
Sample	T (K)	L_ϕ (μm)	$a_{\rm eff}$ (nm)	$R_\square$ (Ω)	a (nm)	l/a	a (nm)	l/a
1	4.0	0.52	6.7	3.44	13.2	5.1	12.5	1.8
	2.2	0.66		3.44				
2	4.0	0.62	10.5	2.07	19.1	3.0	18.1	1.5
	2.2	0.84	11.0	2.07	19.6	2.7	18.6	1.4
3	4.0	0.44	7.7	3.10	14.7	3.9	13.8	1.7
	2.2	0.50		3.10				
4	4.0	0.62	8.2	3.36	15.0	3.0	14.1	1.5
	2.2	1.00	8.6	3.36	15.4	2.7	14.5	1.4
5	4.0	0.75	7.9	2.14	16.1	6.1	15.2	2.0
	2.2	1.30	7.8	2.14	16.0	6.3	15.0	2.0
6	4.0	0.64	7.0	2.62	14.4	6.5	13.5	2.0
	2.2	0.97	6.8	2.62	14.1	7.2	13.3	2.1
7	4.0	0.53	7.7	2.87	14.9	4.3	14.0	1.7
	2.2	0.71	7.7	2.87	15.0	4.2	14.0	1.7
8	4.0	0.77	10.6	1.44	20.7	4.7	19.5	1.8
	2.2	1.22	10.4	1.44	20.5	4.9	19.3	1.8

veloped a procedure for measuring the thickness based on theoretical results of Beenakker and van Houten.[8] We also show how a range of possible values for the impurity mean free path may be determined, once the thickness is known. (The impurity mean free path is the mean free path between collisions with impurities. It does not count collisions with the surface.)

A. Thickness measurement

We have shown above how the weak-localization data can be used to measure the dephasing length L_ϕ and the effective thickness $a_{\rm eff}$. We now show how this information can be combined with the semiclassical sheet resistance to yield the true thickness a of the film. Our method requires a discussion of the weak-localization theory for the diffuse, quasiballistic, and ballistic cases.

The weak-localization magnetoresistance for a film in a parallel field was first studied by Al'tshuler and Aronov,[10] for the diffusive case, where the impurity mean free path l is much less than the film thickness a. Dugaev and Khmel'nitskii[12] later treated the ballistic limit where $l \gg a$, assuming that reflection from the film surfaces was entirely diffusive. Both papers considered only the limiting cases of very strong and very weak magnetic fields. Beenakker and van Houten[8] extended these results to include specular as well as diffuse surface scattering, and to include numerically the intermediate cases.

In this paper, we will only be concerned with the weak field limit, where the magnetic length $l_m \equiv \sqrt{\hbar c/eB}$ satisfies the condition $l_m^2 \ll al$.

In place of using Eq. (2) the parallel field magnetoconductance may be written in terms of a magnetic dephasing time

$$\delta g(B_x) = \frac{e^2}{\pi h}\left[\ln\left(1+\frac{\tau_\phi}{\tau_B}\right) - \ln\left(\frac{\tau_\phi}{\tau}\right)\right], \tag{4}$$

where

$$\tau_B = 3\left(\frac{\hbar c}{e}\right)^2 \frac{1}{D a_{\rm eff}^2 B_x^2}. \tag{5}$$

The dephasing time contains the diffusion constant D, which describes the spreading of the electrons in the film plane. This is related to the sheet resistance $R_\square$ by the Einstein relation

$$\frac{1}{R_\square a} = 2e^2 N(E_F) D, \tag{6}$$

where $N(E_F)$ is the density of levels at the Fermi surface.

The effective film thickness $a_{\rm eff}$ is equal to the actual film thickness a in the diffusive limit where $l \ll a$. When $l \gg a$, the effective thickness is given by[8]

$$a_{\rm eff} = \left(\frac{3 v_F a^3}{C_1 D}\right)^{1/2}.$$

Here v_F is the Fermi velocity of the conducting electrons, and C_1 is a dimensionless parameter that depends on the specularity of the surface scattering. (This notation follows that of Beenakker and van Houten.) For perfectly diffusive surface scattering, $C_1 = 16$, and for perfectly specular scattering, $C_1 = 12.1$.

Beenakker and van Houten evaluated the magnetic dephasing time numerically for all mean free paths,[8] and their results suggest an interpolation between the two extreme regimes,

$$\tau_B = (\tau_B)_{l \gg a} + (\tau_B)_{l \ll a}.$$

This gives an expression for the effective thickness, valid for all mean free paths, of

$$a_{\rm eff} = \frac{a}{\left(1+\dfrac{C_1 D}{3 v_F a}\right)^{1/2}}.$$

Substituting Eq. (6) for the diffusion constant yields

$$a_{\rm eff} = \frac{a}{\left(1+\dfrac{\xi C_1}{R_\square a^2}\right)^{1/2}}, \tag{7}$$

where the parameter

$$\xi = \frac{1}{6 e^2 v_F N(E_F)}$$

is independent of the specific parameters of the film. It only depends on the properties of the material from which the film is made. Solving now for the actual thickness, we find

$$a = a_{\rm eff}\left\{\frac{1}{2}\left[1+\left(1+\frac{4\xi C_1}{R_\square a_{\rm eff}^2}\right)^{1/2}\right]\right\}^{1/2}. \tag{8}$$

In practice, we use this formula to find the true thickness a from the value of $a_{\rm eff}$ determined from weak-localization measurements. If an independent and very accurate measurement of the thickness were available, this formula could be used to find C_1, and thus the specularity.

Equation 8 depends only weakly on the surface specularity, and a reasonably accurate value for the film thickness can be obtained without any knowledge of the actual value of the constant C_1. It is not hard to show from Eq. (8) that the error in the thickness measurement due to uncertainty in C_1 will always be less than 8.33%. This means that we can measure the thickness of a film within 8% or so just from a weak-localization measurement and a sheet resistance measurement. No knowledge of the specularity or the impurity mean free path is required, and the method works in the diffusive, ballistic, and intermediate regimes.

B. Mean free path

We may set upper and lower bounds on the mean free path, once we have a measurement of the film thickness and sheet resistance. The conductivity of a thin film is related to the conductivity of a bulk sample with the same microscopic properties (i.e., the same material, impurity concentration, etc.) by[13]

$$\sigma_f=\sigma_b\left[1-A'(p,\kappa)\,\frac{1}{\kappa}\right],$$

where $\kappa=a/l$. The form of the function A' depends on the microscopic model used to describe conduction in a thin film. In this paper, we will use the Fuchs-Sondheimer model.[9] The parameter p describes the fraction of surface scattering events that are specular. (A perfectly specular surface would have $p=1$.) The film conductivity σ_f is defined by the observed sheet resistance and the film thickness,

$$\sigma_f=\frac{1}{R_\square a}.$$

The bulk conductivity σ_b is given by the Drude formula

$$\sigma_b=\frac{ne^2 l}{mv_F}.$$

(Recall that l is the mean free path between collisions with bulk impurities, and does not count collisions with the surface.)

We solve for κ in terms of the sheet resistance, film thickness, and parameters that depend only on the film material:

$$\frac{1}{R_\square a^2}\,\frac{mv_F}{ne^2}=\frac{1}{\kappa}\left[1-A'(p,\kappa)\,\frac{1}{\kappa}\right]. \tag{9}$$

The function A' is given in the Fuchs-Sondheimer model by

$$A'(p,\kappa)=\frac{3}{8}\,(1-p)\left[1-4(1-p)\int_0^1 \frac{x(1-x^2)}{\exp(\kappa/x)-p}\,dx\right]. \tag{10}$$

For a given sheet resistance and film thickness, we expect the minimum possible impurity mean free path to correspond with the maximum possible specularity, $p=1$, and vice versa. For $p=1$, A' vanishes, and Eq. (9) reproduces the Drude formula. For any thin film, then, a lower bound on the impurity mean free path will be set by

$$l\geqslant\frac{mv_F}{ne^2}\,\frac{1}{R_\square a}. \tag{11}$$

Solving Eq. (9) with $p=0$ gives the upper bound on the impurity mean free path. If $\kappa\gg1$ (i.e., the mean free path is short compared with the film thickness, and the surfaces do not effectively contribute to the resistance of the film), then the second order term in $1/\kappa$ in Eq. (9) may be neglected, and the Drude formula again applies. In this case, the upper and lower bounds are the same, and the mean free path is uniquely determined by Eq. (11), with the inequality replaced by an equality.

If $\kappa\ll1$, then A' is approximately

$$A'(0,\kappa)\approx\kappa-\frac{3}{4}\,\kappa^2\,\ln\!\left(\frac{1}{\kappa}\right),$$

and the upper bound on the mean free path is given by

$$l\leqslant a\,\exp\!\left(\frac{4}{3}\,\frac{mv_F}{ne^2}\,\frac{1}{R_\square a^2}\right).$$

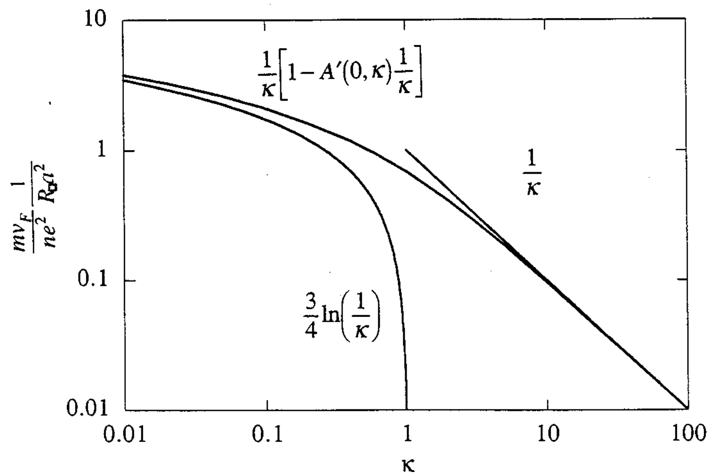

FIG. 5. Plot of the right-hand side of Eq. (6) with $p=0$, along with asymptotic expressions for very large and very small κ.

Intermediate cases, where κ is of order unity, may be treated numerically. Figure 5 shows a plot of the right-hand side of Eq. (9) with $p=0$, along with the asymptotic expressions for very large and very small κ. It is a simple matter to calculate a value for the left-hand side, and then to solve Eq. (9) graphically.

C. Experimental verification

To verify Eq. (8), we consider the results obtained by Giordano and Pennington[4] in a gold film. They reported weak-localization magnetoresistance in both parallel and perpendicular fields, and the film thickness as determined during film growth. They quoted, for a pure gold film (with no magnetic impurities), a sheet resistance of 4 Ω and a film thickness of 140 Å. We have fit their parallel field data to weak-localization theory including spin-orbit and spin-flip scattering. For parallel fields, this theory is[10,14]

$$\delta g(B_x)-\delta g(0)=\frac{e^2}{\pi h}\left\{\frac{3}{2}\ln\left[1+\frac{1}{3}\left(\frac{e}{\hbar c}\right)^2(a_{\rm eff}L_1)^2B_x^2\right]-\frac{1}{2}\ln\left[1+\frac{1}{3}\left(\frac{e}{\hbar c}\right)^2(a_{\rm eff}L_0)^2B_x^2\right]\right\}.$$

The parameters L_0 and L_1 are the singlet and triplet dephasing lengths, respectively, and are obtained from the perpendicular field magnetoconductance. We used values for L_0 and L_1 given by Giordano and Pennington, and, for our parallel field fits, we only allowed $a_{\rm eff}$ to vary. By this procedure, we found the best fit value of $a_{\rm eff}$ to be 86.5 Å. Using Eq. (8) with an effective thickness of 86.5 Å and a sheet resistance of 4 Ω gives a film thickness of 137 Å assuming specular surfaces ($C_1=12.1$), and 144 Å assuming diffusive surface scattering ($C_1=16$). Each of these values is only a few percent off from the 140 Å quoted as the actual thickness, verifying this method of measuring film thickness in this case.

D. Analysis of our films

The results of our analysis of thickness and mean free path for eight films are given in Table I. The thicknesses are consistent with very crude measurements made with the cold crystal rate monitor. We find that the impurity mean free path is greater than the film thickness, regardless of whether the surface scattering is assumed to be specular or diffusive. Thus our films are quasiballistic, with impurity mean free paths greater than the film thicknesses, despite the fact that they were prepared by quench condensation. This result is somewhat surprising, since quench condensation is often considered the method of choice for producing thin, diffusive samples. However, it has been shown before that quench condensed alkali-metal films, when prepared under the right conditions, can be quasiballistic.[15]

VI. SUMMARY AND CONCLUSIONS

We have studied the weak-localization magnetoresistance of quench condensed lithium films in both parallel and perpendicular fields. Other workers who made similar studies in metal films reported some puzzling anomalies that appear to be connected with spin effects. For the samples we report on here, no spin effects were evident, and no anomalous behavior was observed. This indicates that at least some of the anomalies reported in the literature are indeed connected with spin effects, and that we understand weak localization in the absence of spin effects.

In order to characterize our samples accurately, we have developed a procedure to measure the film thickness from the weak-localization magnetoresistance (in both parallel and perpendicular fields) and the semiclassical sheet resistance. Unlike previous analyses, this procedure is not restricted to the limiting cases of very short or very long mean free paths, and it allows us to measure the film thickness with an accuracy of 8% or better, regardless of the mean free path or surface specularity. We also show how a range of values for the impurity mean free path may be obtained once the thickness is known. When we apply this analysis to the data obtained by Giordano and Pennington,[4] we find that we can resolve one of the mysteries reported in that paper: The film thickness obtained from one of the weak localization fits did not agree with the actual film thickness. Taking into account the fact that transport was quasiballistic in that sample allows us to extract a value for the film thickness from the weak-localization fit that is in agreement with the actual film thickness.[16] Applying this analysis to our films, we find that electron transport is quasiballistic, despite the fact that the samples are quench condensed.

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