Longitudinal magnetoresistance of thin gold films deposited on mica arising from electron-surface scattering

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We report the measurement of the longitudinal magnetoresistance, performed on 4 thin gold films of different thickness evaporated onto preheated mica, where the signal can be univocally attributed to electron-surface scattering. The magnetoresistance exhibits a marked thickness dependence: at 4 K and 9 Tesla it is about 2.6% for the thinner (72 nm) film, and about 13.5% for the thicker (266 nm) film. The observed magnetoresistance is at variance with the predictions of the theory of longitudinal magnetoresistance of Way and Kao [Phys. Rev. B **5**, 2039 (1972)].

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I. INTRODUCTION

The present work is a continuation of research concerning magnetomorphic effects attributable to electron-rough surface scattering already published.[1–4] The first paper[1] contains in Sec. 3.6 a preliminary report on the transverse magnetoresistance observed at 4 K in a family of thin gold films deposited onto mica substrates (with the magnetic field **B** oriented perpendicular to the films), that was presented as part of an invited talk at a PASI meeting held in Merida, Venezuela, in March 2004 (the experiments were started in mid 2003). The second paper[2] reports the results obtained upon completion of that experiment for $0 \leq \mathbf{B} \leq 9$ Tesla, $4\ \mathrm{K} \leq T \leq 50\ \mathrm{K}$.

In the course of the first exploratory experiments performed in 2003, we found a sizable voltage drop induced by the presence of the magnetic field **B**, when the textbooks in Solid State Physics predicted a null magnetoresistance for an electron gas described by a spherical Fermi surface.[5] We realized that the signal observed was either due to departures of the Fermi surface of gold from a perfect sphere, or else to "size effects" reflecting the effect of electron surface scattering under the presence of a strong magnetic field. The thickness dependence of the magnetoresistance signal suggested that we were probably measuring the effect of electron-surface scattering, hence we decided to repeat the experiments several times before publishing.

There are two theories available that predict the transverse magnetoresistance of metallic films. The theory of Sondheimer[6] and the theory of Calecki.[7] Both theories are based upon a description of the electronic motion provided by a Boltzman transport equation (BTE). The main conclusions that can be drawn from Refs. 1 and 2 are that the theories available describe rather accurately the temperature and thickness dependence of the resistivity observed in the absence of a magnetic field at temperatures T such that $4\ \mathrm{K} \leq T \leq 50$ K, but fail to describe the transverse magnetoresistance measured on the same sample.

In the case of Sondheimer´s, the theory contains a set of adjustable parameters. Once these parameters have been adjusted (to describe the temperature and thickness dependence of the resistivity observed in the absence of magnetic field), there are no more fitting parameters left, therefore the discrepancy between theory and experiment regarding the predicted transverse magnetoresistance constitutes a strong evidence of the inadequacy of the theory.

Calecki´s is the other theory available, and it contains *no adjustable parameters*. The effect of the rough surface is represented via a perturbation Hamiltonian (that depends on the r.m.s. surface roughness amplitude δ and on the lateral correlation length ξ), that mixes electronic quantum states belonging to different sub-bands ν and ν'. The sub-bands labeled by index ν are induced by the quantization of the electron momentum perpendicular to the surface of the film, when the electron gas is confined between two parallel planes. Sub-band mixing leads to a set of coupled equations of motion where the effect of electron-surface scattering described by Boltzman collision operator is contained in a non-diagonal "collision time" matrix $T(\nu, \nu')$.

The comparison between theory and experiment requires:

(i) Measuring the parameters (δ, ξ) that characterize the roughness of the surface on an atomic scale on each sample—a measurement we did perform; and

(ii) Either solving numerically the set of coupled equations of motion proposed by Calecki, or else using the approximate expressions provided by the author, who proposes dropping the off-diagonal elements of $T(\nu, \nu')$ under the small correlation length approximation $k\xi < 1$ (where k is the electron wave vector).

To perform the comparison between the transverse magnetoresistance predicted by Calecki and the measured values, we chose the simplest option in Ref. 2, we used the (approximate) expressions provided by the author. Under the small correlation length approximation, the predicted transverse magnetoresistance turns out to be significantly smaller than the observed values. Hence, the doubt remains regarding whether the discrepancy is due to a technical difficulty, arising from the questionable validity of the approximation $k\xi < 1$ used by Calecki to ignore the off-diagonal elements of $T(\nu, \nu')$, or else it reflects a more fundamental difficulty aris-

ing from the inadequacy of the theory. This is the content of Ref. 2.

These effects of electron-surface scattering were so unexpected, that we decided next to search for a *Hall effect also induced by electron surface scattering* during the second half of 2005; the results are reported in Ref. 3. The main conclusion that can be drawn from Ref. 3 is that the Hall tangent (and therefore the mean electron collision time) observed at 4 K depends linearly on film thickness. Hence the scattering mechanism giving rise to the Hall effect is, indeed, electron-surface scattering. This had been predicted by Sondheimer in 1950 but had never been reported.

To elucidate whether the discrepancy between the transverse magnetoresistance predicted by Calecki and the experimental data was a consequence of the small correlation length approximation, we proceeded to solve numerically the equations of motions contained in Calecki´s paper, without any approximation whatsoever. The result, is that the theory describes rather accurately the temperature and thickness dependence of the resistivity observed in the absence of magnetic field, it provides a fair description of the Hall voltage observed at 4 K, but the transverse magnetoresistance turns out to be orders of magnitude smaller than observed. Therefore, the discrepancy between theory and experiment regarding the predicted transverse magnetoresistance is not a consequence of the small correlation length approximation, it can be considered as a strong evidence of the inadequacy of Calecki´s theory. This is reported in Ref. 4.

These series of experiments have some interesting consequences:

(a) First, measuring routinely the Hall tangent at 4 K, can now be used as a tool to identify if electron-surface collisions are, indeed, the dominant electron-scattering mechanism at 4 K.

(b) Second, the somewhat mysterious transverse magnetoresistance in the gold samples reported in Refs. 1 and 2, which was *tentatively* interpreted as due to electron collision with the surfaces, arises from electron-surface scattering, for in these samples the Hall tangent does, indeed, depend linearly on film thickness.

(c) As a direct consequence of (b), the observed magnetoresistance ought to have different values depending on the orientation of the electric field **E** relative to the magnetic field **B**. When **B** is perpendicular to the surface of the film, we have the results informed in Ref. 2. As stated above, both theories fail to describe the temperature and thickness dependence of the resistivity as well as the temperature and thickness dependence of the transverse magnetoresistance observed on the same samples (this is the first time that both transport coefficients—together with the surface roughness measured on an atomic scale) are measured on the same samples.

Because of conclusion (c) from the preceding paragraph, when **B** is parallel to **E**, electron-surface scattering leads yet to a different (longitudinal) magnetoresistance. This is also true when **E** is orthogonal to **B**, and both fields are contained within the plane of the film, the so called MacDonald configuration. This suggested to us pursuing the measurement of the longitudinal magnetoresistance as well, performing another series of experiments. To carry out these experiments we had to prepare a new set of samples, and had to repeat the experiments several times.

In this paper, we report the first measurement of the longitudinal magnetoresistance performed on 4 gold films deposited onto mica substrates arising from electron-surface scattering, in samples where the contributions arising from the bulk are negligible. In order to verify that the contributions arising from the bulk are, indeed, negligible and that the signal can be unequivocally attributed to electron-rough surface scattering, we measured the Hall effect at 4 K on the same samples with the magnetic field orthogonal to the films. In all 4 samples the Hall mobility turns out to depend linearly on film thickness, evidence that has been considered the finger print of electron-rough surface scattering.[3] The report on these experiments and the comparison with the only theory available that we are aware of,[8] is the content of this work. We stress again, as in the case of transverse magnetoresistance, that the only theory available contains a set of parameters that can be adjusted to describe the temperature and thickness dependence of the resistivity data recorded in the absence of magnetic field. After such adjustments are made, there are no more fitting parameters left. Any discrepancy between theory and experiment regarding the predicted longitudinal magnetoresistance will become a strong evidence of the inadequacy of the theory.

II. EXPERIMENTAL

In our samples, in order to be able to observe magnetoresistance arising from "size effects," the samples must to be such that the magnetoresistance arising from electron-scattering mechanisms other than rough surfaces do not mask the effect of electron-rough surface scattering. For "size effects" to become measurable and dominant at 4 K, it is desirable that the samples satisfy the following requirements:

(a1) The films must be made up of ultrapure metal,

(a2) the samples must exhibit the lowest possible concentration of morphological defects (that could give rise to electron scattering by defects that could result in an unwanted magnetoresistance signal),

(a3) the samples should have a thickness of an order of magnitude comparable to the electron mean free path in the bulk at room temperature (in crystalline gold, $\ell_0 \approx 38$ nm at 295 K).

Through some exploratory work reported elsewhere,[2] we found the conditions of evaporation that minimize the concentration of defects (that minimize deviations of the morphology and structure of the films with respect to a thin slice of a single crystal): A minimum $\rho(295)$ is obtained when the substrate and annealing temperature are between 180 °C and 270 °C. For completeness, we briefly summarize below details of the experimental method that have been published.[2,3]

We prepared samples of gold films evaporated onto preheated mica substrates under high vacuum (HV), the thickness of the films ranges from a few tens of nm to some 260 nm. For these thickness, the contribution to the resistivity of our samples arising from electron-rough surface scattering is significant at 295 K; at 4 K it becomes dominant. We started

TABLE I. Morphological and electrical characterization of gold films. t: thickness, determined using four different methods: (i) Quartz crystal microbalance mounted on the HV evaporation station, (ii) Rutherford back scattering (RBS) spectra of 2 MeV alpha particles from a KN 3750 Van de Graaff accelerator built by HVE; (iii) profilometry, using an Alpha 500 Tencor profilometer; (iv) Tolansky optical interferometry, performed on glass slides placed close to the mica substrates on each run. D: average grain diameter (computed as the average diameter of a circle enclosing the same area, over 100 grains recorded on STM images of each sample). $\mu_H(4)$: Hall mobility measured at 4 K, determined as $\mu_H=\frac{\partial[\tan(\vartheta)]}{\partial \mathbf{B}}$ where $\tan(\theta)$ is the Hall tangent, following the method described in Ref. 3. $\rho(4)$: resistivity at 4 K. $\rho(295)$: resistivity at 295 K. $\ell(4)$: average distance traveled by the electron at 4 K between scattering events, according to Drude´s model.

Film thickness t (nm)	Grain diameter D (nm)	Hall mobility at 4 K $\mu_H(4)(T^{-1})$	Resistivity at 4 K $\rho(4)(n\Omega m)$	Resistivity at 295 K $\rho(295)(n\Omega m)$	Electron mean free path at 4 K $\ell(4)(nm)$
72	120	0.024	3.96	25.7	212
110	141	0.032	3.42	26.4	245
173	133	0.052	2.37	24.7	354
266	158	0.070	1.65	24.8	508

from gold pellets 99.9999% pure evaporated at a rate of 3 nm/min from a tungsten basket filament onto freshly cleaved mica substrates in a HV evaporation chamber (vacuum of 5.0×10^{-5} Pa). Masks were prepared such that an evaporation run yielded 4 samples for each thickness. The mica was preheated to 180 °C, the films were annealed for 1 h at 270 °C after evaporation. Grain size on each sample was measured with an OMICRON scanning tunneling microscope displaying atomic resolution running in air.

Details of the morphological and the electrical characterization of the samples are shown in Table I. The films exhibit a room-temperature resistivity $\rho(295)$ that is a few percent in excess of the resistivity of 22.5 $n\Omega$ m expected in crystalline gold at 295 K from electron-phonon scattering. Updating Drude´s model, the mean electronic collision time at temperature T is $\tau(T)=m^*/[nq^2\rho(T)]$, and the corresponding distance is $\ell(T)=v_F\tau(T)$ (where ρ is resistivity, m^* is the electron´s effective mass, q is its charge, n is the electron density, v_F is the Fermi velocity). Cooling the sample to 4 K freezes out phonons, so $\ell(4)$ (the column on the right of Table I) represents the scale of distance characterizing the structural defects that give rise to electron scattering at 4 K.

Transport measurements were performed using the 4 point method, sample geometry is depicted in the inset of Fig. 1, Refs. 2 and 3. The samples were fed a current of 1.3 mA and 210 Hz, voltage signals were red using computer controlled 830´s LIA built by Stanford research. The samples were placed in a copper block inserted in a superconducting magnet built by JANIS, the sample temperature was maintained within ±0.1 K. Two transport measurements were performed on each sample:

(i) The sample was placed with the surface of the film perpendicular to the magnetic field **B**, and the Hall effect was measured at 4 K, with the magnetic field **B** in the range $1.5\ \mathrm{T}\leq\mathbf{B}\leq9$ T;

(ii) The sample was then placed with the electric field parallel to the magnetic field, and the longitudinal magnetoresistance was measured at temperatures T in the range $4\ \mathrm{K}\leq T\leq50$ K, and magnetic field B in the range 1.5 T $\leq\mathbf{B}\leq9$ T.

The magnetic field dependence of the Hall tangent measured at 4 K yields the Hall mobility $\mu_H=\frac{\partial[\tan(\vartheta)]}{\partial \mathbf{B}}$ listed in Table I. The Hall voltage measured at 4 K and 9 Tesla indicates that the product $\omega_C\tau$ (where $\omega_C=q\mathbf{B}/m^*$ is the cyclotron frequency, τ is the average time between collisions), ranges between 0.22 and 0.63. As shown in Table I, cooling to 4 K decreases the resistivity of the films by about one order of magnitude, leading to a $\rho(4)$ that differs by about a factor of 2.5 between the thinnest and thickest film, *in spite of the fact that the corresponding* $\rho(295)$ differ by less than 10%. The linear dependence of the Hall mobility on thickness observed at 4 K, indicates that the dominant electron scattering mechanism limiting the product $\omega_C\tau$ is electron collisions with the surface.

III. RESULTS

The dependence of the longitudinal magnetoresistance on magnetic field **B** at different temperatures, is displayed in Fig. 1. The temperature dependence of the resistivity observed in the absence of magnetic field, is displayed in Fig. 2.

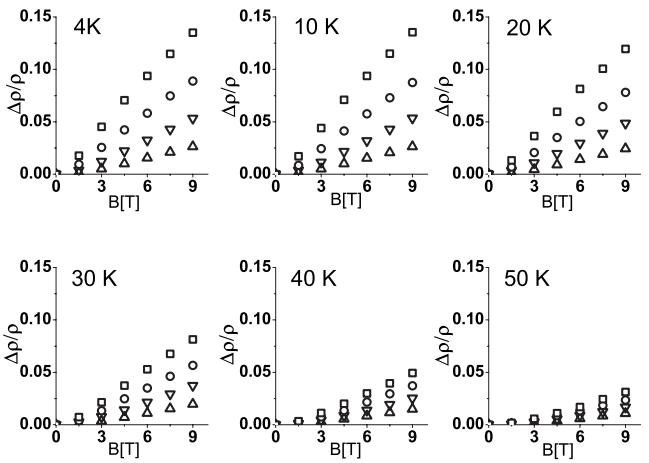

FIG. 1. Dependence of the longitudinal magnetoresistance on the magnetic field B, at different temperatures T (4, 10, 20, 30, 40, and 50 K), indicated in the figure. Squares: film 266 nm. Circles: film 173 nm. Inverted triangles: film 110 nm. Triangles: film 72 nm.

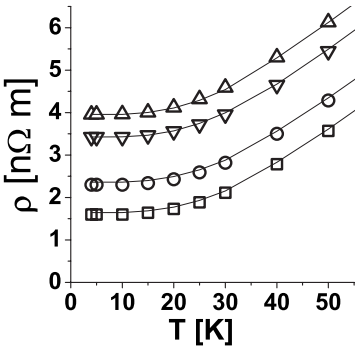

FIG. 2. Temperature dependence of the resistivity for films of different thickness, symbols as in Fig. 1. Solid line: prediction of Sondheimer-Lucas theory, with $P=1$ and $Q=0$. The term $(1/\tau)_{IMP}$ was adjusted for each sample; the corresponding value is $\tau_{IMP}=0.41\times10^{-12}$ [sec] for the 72 nm sample, $\tau_{IMP}=0.36\times10^{-12}$ [sec] for the 110 nm sample, $\tau_{IMP}=0.48\times10^{-12}$ [sec] for the 173 nm sample, $\tau_{IMP}=0.69\times10^{-12}$ [sec] for the 266 nm sample.

IV. DISCUSSION

Some early studies of magnetoresistance in metallic films were published before 1995, and attempts were made to compare experimental data with existing theories during this period.[9,10] The experimental method used in these early studies relies on preparing ultra-pure metallic films/wires whose thickness/diameter t ranges between a few microns and a few hundred microns. In such samples, at room temperature $t \gg \ell_0$ (where ℓ_0 stands for the electron mean free path in the crystalline bulk arising from electron-phonon scattering), hence, at 295 K size effects arising from electron-surface scattering has very little influence on charge transport. In order for size effects to fall within a measurable range, the samples must be cooled, hoping to achieve a resistivity ratio $R(295)/R(4)$ that is large enough for ℓ_0 to become comparable to or larger than t at 4 K (due to the freezing out of the phonon population). However, in this early work, comparison between theory and magnetoresistance data face two severe, interesting difficulties:

(a1) All available theories of magnetoresistance in thin metallic films are based upon the free electron model (where the bulk metal is characterized by a spherical Fermi surface) for which the magnetoresistance is zero.[5] But, as stated in Ref. 10 page 436, "In practice it turns out that one has to work with extremely pure metals at low temperatures in order to achieve a sufficiently long electron mean free path. Under these conditions all metals show a considerable magnetoresistance." Consequently, to extract the magnetoresistance arising from electron-surface scattering from the data recorded at 4 K (that certainly includes both the magnetoresistance from the bulk plus, presumably, the contribution arising from electron-rough surface scattering), the contribution arising from the bulk at 4 K must be substracted.

(a2) To perform such a substraction, it has been assumed that the magnetoresistance arising from electron-surface scattering obeys Kohler´s law. That is, the fractional change in resistivity $\Delta\rho(\mathbf{B},T)/\rho(0,T)=[\rho(\mathbf{B},T)-\rho(0,T)]/\rho(0,T)$ induced by electron-rough surface scattering caused by the presence of the magnetic field **B**, is a function f[$\mathbf{B}/\rho(0,T)$] that depends only on $\mathbf{B}/\rho(0,T)$—where $\rho(0,T)$ is the resistivity observed at temperature T in the absence of magnetic field. There is a theoretical derivation of Kohler´s rule in the bulk, that is based upon the existence of a relaxation time that accurately describes the effects of each of the electron scattering mechanisms acting in the bulk—that give rise to the observed bulk resistivity—on the electron distribution function through the Boltzmann collision operator.[5] However, the extension of Kohler´s rule to electron-surface scattering is far from obvious. Quite to the contrary, arguments have been published suggesting that the Boltzmann collision operator corresponding to electron-rough surface collisions in the presence of a magnetic field **B** cannot be represented by a relaxation time τ, for τ does not exist[7]

These early studies of galvanomagnetic effects in metallic nonmagnetic films, are complemented by experiments performed over the last two decades.[11] Regarding the theoretical description of electron-surface scattering, the pioneering work of Sondheimer[6] has been complemented by theoretical efforts published during the last 15 years, regarding electron transport in the absence of a magnetic field.[12]

We are aware of one theory that has been published to describe the longitudinal magnetoresistance arising from electron-surface scattering.[8] The theoretical prediction contains two adjustable parameters:

(i1) The ratio $t/\ell_0(T)$, and

(i2) The specularity P (the fraction $0\leq P\leq1$ of electrons that undergo a specular collision with the surfaces).

Within this theory, both surfaces of the film are characterized by the same specularity P. However, our samples are gold films deposited onto cleaved mica.

The linear dependence of the Hall mobility $\mu_H(4)$ on thickness, point to the fact that the electronic scattering mechanism controlling the resistivity at 4 K is electron-surface scattering. Moreover, electrons behave as if the collision with one of the surfaces limiting the film was a specular collision ($P=1$), for the mean distance $\ell(4)$ traveled by electrons at 4 K between scattering events is roughly equal to or larger than twice the sample thickness $2t$ (Table I). Therefore, to compare theory and experiment, we extended the formalism of Way and Kao, including two surfaces with different specularities P and Q. The conductivity of the film in the presence of $\mathbf{E}\|\mathbf{B}$ (**E**: electric field; **B**: magnetic field) was computed using a modified version of Way and Kao´s theory, where the integrals corresponding to Eq. (7) from Ref. 8, were rewritten in order to include two different specularities P and Q, and were computed using a 32 point Gaussian quadrature.

Before examining the longitudinal magnetoresistance data, we need to determine $\ell_0(T)$ from an independent experiment. To do so we first examine the resistivity data in the absence of magnetic field. Setting $\mathbf{B}=0$ in Sondheimer´s theory leads to $\rho(T)=\rho_0(T)/[\kappa(T)\varphi(s,\ \mathbf{B}=0)]$, where $\kappa(T)=t/\ell_0(T)$, $\rho_0(T)$ is the bulk resistivity described by a Bloch-Grüneisen law,[13] and $\varphi(s)$ is a function defined by Sondheimer [Eq. (19) in Ref. 6]. However, in his work, Sondheimer also considered a metal film limited by 2 rough surfaces characterized by the same specularity parameter P. Since in our samples, $\ell(4)\approx2t$, one of the surfaces behave as a

specular surface. Rather than adopting the form for $\varphi(s)$ that contains only one specularity P proposed by Sondheimer, we computed numerically $\varphi(s)$, using instead the form suggested by Lucas that contains two specularities P and Q.[3,6,14] We set $P=1$ to characterize the reflectivity of one of the surfaces limiting the film. Thus, the fitting parameters left in the theory are Q (the specularity of the other surface limiting the film) and $\kappa(T)$.

As discussed in Refs. 2 and 3, $\ell_0(\mathrm{T})$ is determined by the collision time $\tau(T)$ in the bulk, that varies with temperature according to $1/\tau=(1/\tau)_{\mathrm{IMP}}+(1/\tau)_{\mathrm{PHON}}$, where the first (temperature independent) term accounts for electron scattering by impurities/point defects, and the second (temperature dependent) term accounts for electron-phonon scattering.[2,3,13] For each sample we selected a value for the parameter Q, and adjusted $(1/\tau)_{\mathrm{IMP}}$ to describe $\rho(T)$ for 4 K$\leq T\leq$50 K, neglecting $(1/\tau)_{\mathrm{PHON}}$ at 4 K. To calculate $1/\tau$ at $T>4$ K, we added to $(1/\tau)_{\mathrm{IMP}}$ the corresponding $(1/\tau)_{\mathrm{PHON}}$ computed from the Bloch-Grüneisen intrinsic resistivity listed in page 1209 of Ref. 13. We repeated this procedure for different values of Q. The best fit to the temperature dependence of the resistivity data was obtained for $Q=0$, as shown in Fig. 2. The result is that, if $(1/\tau)_{\mathrm{IMP}}$ is adjusted to fit $\rho(T)$, then the Sondheimer-Lucas theory provides a good description of the temperature dependence of the resistivity $\rho(T)$ of each sample in the range 4 K$\leq T\leq$50 K, as shown in Fig. 2. This is consistent with our findings published in Refs. 2 and 3.

We proceed next to compute the expected longitudinal magnetoresistance arising from electron-surface scattering as a function of magnetic field **B** at 4 K, using $P=1$, $Q=0$, and the collision time $\tau(\mathrm{T})$ in the bulk needed to describe $\rho(T)$ in the absence of magnetic field, using the extended theory of Way and Kao. The theoretical predictions are displayed in Fig. 3(a); the longitudinal magnetoresistance measured at 4 K is displayed in Fig. 3(b) for comparison. It seems remarkable that although Sondheimer´s theory describes rather accurately the temperature dependence of the resistivity (at temperatures T such that 4 K$\leq T\leq$50 K) of films of different thickness in the absence of magnetic field, the theory of Way and Kao predicts at 4 K a longitudinal magnetoresistance that is an order of magnitude smaller than that observed at low fields, together with a maximum that should occur at some intermediate magnetic field that is at variance with what is experimentally observed.

In summary, in this paper, we report the measurement of the longitudinal magnetoresistance, performed on 4 thin gold films evaporated onto preheated mica, where the contribution arising from the bulk is negligible, and the signal can be univocally attributed to electron-surface scattering. Beyond the fact that there is a marked disagreement between the theoretical predictions based upon Way and Kao´s theory and experimental data on longitudinal magnetoresistance—a disagreement that is reminiscent of the discrepancies between theoretical predictions based upon Sondheimer´s theory and experimental data reported in the case of the transverse magnetoresistance as well[2]—the research reported here departs from previous work in the field performed over several decades. Based upon the fact that, on our samples, the electron collision time depends linearly on film thickness, we present in this paper an experimental method that permits direct comparison between magnetoresistance data and theoretical predictions, without invoking the use of Kohler′s law. The strong disagreement between theory and experiment reported underlines the need for a fresh theoretical description of charge transport involving electron-rough surface scattering in the presence of a magnetic field in metallic nanostructures.

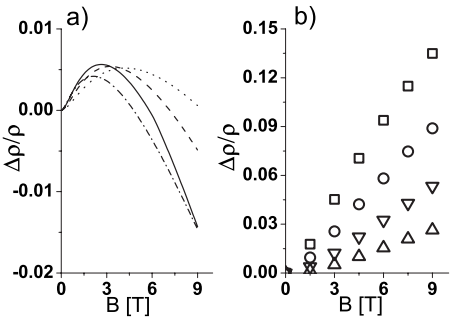

FIG. 3. (a) Magnetic field dependence of the longitudinal magnetoresistance at 4 K predicted by the modified theory of Way and Kao, with $P=1$ and $Q=0$, and the values of τ_{IMP} listed in Fig. 2. slashed-dotted line: film 72 nm thick; dotted line: film 110 nm thick; slashed line: film 173 nm thick; solid line: film 266 nm thick. (b) Longitudinal magnetoresistance measured at 4 K, symbols as in Fig. 1.

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