Size Effects under a Strong Magnetic Field: Hall Effect Induced by Electron-Surface Scattering on Thin Gold Films Deposited onto Mica Substrates under High Vacuum

Raúl C. Munoz,[1] Juan Pablo García,[1] Ricardo Henríquez,[1] Ana María Moncada,[2] Andrés Espinosa,[1] Marcelo Robles,[3] Germán Kremer,[2] Luis Moraga,[2] Simón Cancino,[2] José Roberto Morales,[2] Adán Ramírez,[1] Simón Oyarzún,[1] Marco Antonio Suárez,[1] David Chen,[1] Ernesto Zumelzu,[4] and Claudio Lizama[4]

[1]*Departamento de Física, Facultad de Ciencias Físicas y Matemáticas, Universidad de Chile. Blanco Encalada 2008. Casilla 487-3. Santiago 8370415, Chile*

[2]*Departamento de Física, Facultad de Ciencias, Universidad de Chile. Las Palmeras 3425. Santiago 7800024, Chile*

[3]*Departamento de Ciencias de la Construcción, Universidad Tecnológica Metropolitana. Dieciocho 390. Santiago, Chile*

[4]*Facultad de Ciencias de la Ingeniería, Universidad Austral. General Lagos 2086. Valdivia, Chile*

(Received 10 March 2006; published 26 May 2006)

We report measurements of the Hall effect performed on 4 gold films evaporated onto mica substrates where the signal arises primarily from electron-surface scattering. The measurements were performed at low temperatures T ((4 K $\leq T \leq$ 50 K) under high magnetic field strengths B (1.5 T $\leq B \leq$ 9 T), with $\mathbf{B}$ oriented perpendicular to the films.

DOI: 10.1103/PhysRevLett.96.206803 PACS numbers: 73.20.At, 73.25.+i, 73.43.Qt, 73.50.Jt

A central issue concerning thin metallic films is how the roughness of the surface that limits the structure affects its charge transport properties when one or more of the dimensions of the structure become comparable to or smaller than the mean free path ℓ of the charge carriers in the bulk. Understanding the effect of electron-surface scattering on charge transport is still fragmentary, despite over a century of research on ''size effects'' [1].

Sondheimer published the first calculation of ''size effects'' induced by a magnetic field orthogonal to a thin metallic film [2]. He used a Bolztmann transport equation (BTE) to describe the motion of electrons in the sample, and introduced the specularity of the surface as an adjustable parameter (representing the fraction of electrons that are specularly reflected upon colliding with the rough surface). Calecki published the first theory of magnetomorphic effects on metallic films arising from electron-surface scattering that contains no adjustable parameters. Calecki used a BTE but represented electron-surface scattering in terms of a perturbation Hamiltonian (describing the perturbation induced by the two rough boundaries of the film over and above the Hamiltonian describing an electron gas confined between two parallel flat surfaces) [3]. Both theories predict that, because electron-surface scattering is expected to be the dominant electron scattering mechanism in thin metallic films, the proximity of the upper and lower rough surfaces limiting the film results in a Hall tangent $\tan(\theta) = E_H/E_L$ (E_H: transverse Hall field; E_L: longitudinal field) that depends on the film thickness t separating the two rough surfaces, when $t < \ell$. On the contrary, when $t \gg \ell$, the Hall tangent becomes independent of the thickness of the specimen and depends only on the electron scattering mechanism present in the bulk. In this Letter we report measurements of the Hall effect performed on 4 gold films of different thickness deposited onto mica substrates, immersed in a magnetic field perpendicular to the plane of the films, together with measurements of the surface roughness of the samples on an atomic scale performed with a scanning tunneling microscope (STM). The Hall signal exhibits a remarkable thickness dependence that can be attributed to electron-surface scattering, and confirms qualitatively Sondheimer's predictions. The measurement was performed at low temperatures T (4 K $\leq T \leq$ 50 K) and high magnetic fields B (1.5 T $\leq B \leq$ 9.0 T).

During the last decade, several theories of size effects (in the absence of a magnetic field) were published; to mention a few, see Refs. [4], [5] and references therein. Few theories of magnetomorphic effects on metallic samples have been published after Calecki's [6]. Experimental investigation of magnetomorphic effects on nonmagnetic metallic specimens published after Calecki's work is also scarce [7,8]. Calecki's seems to be the only formalism available that predicts a Hall effect in thin metallic films arising from electron-surface scattering, in terms of parameters that characterize the roughness of the surface that can be measured in an independent experiment with a STM. The relevant scale of length over which corrugations are expected to scatter electrons is set by the Fermi wavelength, which for Au is 0.52 nm. Except for work recently published [9], transport measurements as well as surface roughness measurements performed on the same samples on the relevant scale of length are not available. Such measurements are interesting, as a way of contrasting theory and experiment regarding size effects *without using adjustable parameters*.

Details of the sample preparation have been published [9]. Summarizing, we prepared films of different thickness, starting from gold 99.9999% pure evaporated at 3 nm/min from a tungsten basket filament onto freshly cleaved mica substrates in a high vacuum evaporation chamber (vacuum of 1.0×10^{-5} Pa). The mica was preheated to 270 °C; the

TABLE I. Structural and electrical characterization of gold films evaporated onto mica substrates.

Thickness	Lateral dimension of the grains	rms roughness amplitude	Lateral correlation length	Resistivity at 4 K	Resistivity at 295 K
t [nm]	[nm]	δ [nm]	ξ [nm]	$\rho(4)$ [nΩ m]	$\rho(295)$ [nΩ m]
69	167 ± 19	0.17	10.9	7.01	29.3
93	240 ± 24	0.17	10.1	4.72	26.4
150	255 ± 28	0.16	12.2	3.27	24.8
185	290 ± 41	0.29	7.65	2.14	23.6

films were annealed for 1 h at 270 °C after evaporation. The films exhibit a room-temperature resistivity $\rho(295)$ a few percent in excess of the resistivity of 22.5 nΩ m expected from electron-phonon scattering at 295 K [10]. For completeness, we summarize in Table I results of the structural and electrical characterization of the films. The thickness of the samples was measured (to an accuracy of 5%) recording the Rutherford backscattering (RBS) spectra of 2 MeV alpha particles from a van de Graaff accelerator. The average lateral dimension of the grains was measured with a transmission electron microscope (TEM). The parameters (δ, ξ) corresponding to a Gaussian representation of the roughness profile $f(x, y) = \delta^2 \exp[-(x^2 + y^2)/\xi^2]$ [where (x, y) represent the in-plane coordinates, δ represents the rms roughness amplitude, and ξ the lateral correlation length] of the surface of each sample were measured with a STM exhibiting atomic resolution. Transport measurements were performed inserting the samples into a copper block in a superconducting magnet, whose temperature was maintained within ± 0.1 K. Note that cooling to 4 K decreases the resistivity of the films by 1 order of magnitude, leading to a $\rho(4)$ that differs by at least a factor of 3 between the thinnest and thickest film, in spite of the fact that the corresponding $\rho(295)$ do not differ by more than 30%. Since at 4 K the phonons are frozen out and the average lateral dimension of the grains is larger than the film thickness in all samples, at 4 K the mean free path is primarily determined by electron-surface scattering [9].

The resistivity, magnetoresistance and Hall voltage was measured injecting a current of 1.3 mA and 210 Hz across terminals A–B (Fig. 1). The transverse voltage was measured with a SR-830 lock-in amplifier (Stanford Research) from the signal arising between the variable arm of a 1 KΩ, 10 turn precision potentiometer (connected across terminals C–D), and terminal F (Fig. 1). The variable arm of the potentiometer was used to null the transverse voltage signal in the absence of a magnetic filed, and was used to null again the signal with the magnetic field on. The Hall voltage was determined from the change in the position of the wiper arm of the potentiometer required to cancel out the deflection induced by the magnetic field. At 4 K and 9 Tesla, the Hall voltage indicates that the product $\omega\tau$ (where ω is the cyclotron frequency, τ is the average time between collisions) ranges between 0.14 and 0.45.

The dependence of the Hall tangent $\tan(\theta) = E_H/E_L$ on the magnetic field strength **B** observed at different temperatures is shown in Fig. 1. The Hall mobility $\mu_H = \frac{\partial[\tan(\vartheta)]}{\partial B}$ measured at 4 K is 0.011, 0.018, 0.032, and 0.052 $m^2/V\,s$ for the film 69, 93, 150, and 185 nm thick, respectively. It seems remarkable that the Hall mobility μ_H exhibits roughly a linear dependence with film thickness, which underlines the fact that electron-surface scattering does play a central role in determining the Hall voltage. The effect of increasing temperature is, as expected, to reduce the time elapsed between scattering events, and hence to reduce the influence of the magnetic field, thereby reducing the Hall voltage.

There are two theories available to describe the Hall voltage arising from electron-surface scattering. The first is that published by Sondheimer, who predicts $\tan(\theta) = \frac{E_H}{E_L} = \frac{\mathrm{Im}\,\varphi(s)}{\mathrm{Re}\,\varphi(s)}$, where $s = \kappa + i\beta$ is a complex variable, $\mathrm{Re}\,\varphi(s)$ and $\mathrm{Im}\,\varphi(s)$ stand for the real and the imaginary part of a

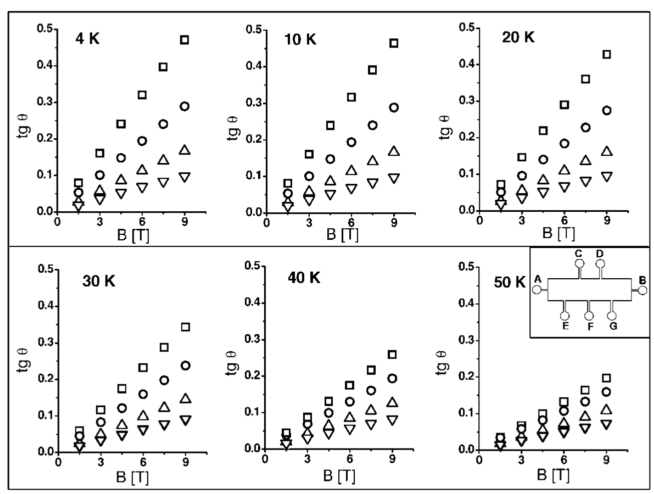

FIG. 1. Dependence of the Hall tangent $\tan(\theta) = E_H/E_L$ on the strength of the magnetic field B, at different temperatures T (4 K, 10 K, 20 K, 30 K, 40 K, 50 K) indicated in the figure, for films of different thickness. Squares: film 185 nm thick. Circles: film 150 nm thick. Triangles: film 93 nm thick. Inverted triangles: film 69 nm thick. The inset in the lowest rightmost panel indicates the shape of the sample. Dimensions of the rectangular center section of the sample are 2.5 mm × 11.0 mm; terminals E–F and F–G are 3.5 mm apart; terminals C–D are 2.0 mm apart.

complex function $\varphi(s)$. Here $\kappa(T) = t/\ell(T)$ [notice that $\ell(T)$ stands for the electron mean free path at temperature *T in the absence of electron-surface scattering*] and $\beta = t/r_0$ (r_0 is the radius of the cyclotron orbit) [2]. Setting $B = 0$ leads to $\rho(T) = \rho_0(T)/[\kappa(T)\varphi(s, B = 0)]$, where $\rho_0(T)$ is the bulk resistivity described by a Bloch-Grüneisen law [10]. However, Sondheimer considered a metal film limited by 2 rough surfaces characterized by *the same specularity.* As discussed in Ref. [9], since the lower surface is cleaved mica, which is atomically flat except for cleavage steps, the roughness of the upper gold surface is expected to dominate the resistivity, magnetoresistance, and the Hall voltage induced by electron-surface scattering. Consequently, the appropriate $\varphi(s)$ is not the function proposed by Sondheimer, but that proposed by Lucas [11],

$$\varphi(s) = \frac{1}{s} - \frac{3}{4s^2}\int_1^\infty \left(\frac{1}{t^3} - \frac{1}{t^5}\right)\frac{[1 - \exp(-st)][2 - P - Q + (P + Q - 2PQ)\exp(-st)]}{1 - PQ\exp(-2st)}dt, \qquad (1)$$

where we set $P = 1$ to characterize the reflectivity of the mica. The fitting parameters left in the theory are Q (the specularity of the upper gold surface) and $\kappa(T)$.

To test Sondheimer's theory, we ought to determine $\kappa(T)$, and therefore we should evaluate $\ell(T)$ at each temperature T. $\ell(T)$ is determined by *the mean time between collisions* $\tau(T)$*in the bulk*, that varies with temperature according to $1/\tau = (1/\tau)_{\text{IMP}} + (1/\tau)_{\text{PHON}}$, where the first (temperature-independent) term accounts for electron scattering by impurities, and the second (temperature-dependent) term accounts for electron-phonon scattering [10]. As explained in Ref. [9], the best description of the temperature dependence of the resistivity data is obtained for $Q = 0$, hence we used this value in Eq. (1) to perform the data analysis of the Hall tangent. For each sample we adjusted $(1/\tau)_{\text{IMP}}$ to describe either $\rho(4)$ or $\tan(\theta)(4)$, using the fact that at 4 K the phonons are frozen out, hence $(1/\tau)_{\text{PHON}}$ can be neglected. To determine $1/\tau$ at $T > 4$ K, we added to $(1/\tau)_{\text{IMP}}$ the corresponding $(1/\tau)_{\text{PHON}}$ term computed from the Bloch-Grüneisen intrinsic resistivity listed in p. 1209 of Ref. [10]. The result of the analysis is interesting: (i) if $(1/\tau)_{\text{IMP}}$ is determined by adjusting it to describe $\rho(4)$, then the Sondheimer-Lucas theory provides a fair description of the temperature dependence of the resistivity of each sample, as displayed in Fig. 2(a). However, using the same parameter $(1/\tau)_{\text{IMP}}$, the theory predicts at 4 K a Hall tangent that coincides with the experimental data for the 150 and 185 nm film, but overestimates the Hall tangent for the 69 and 93 nm films, as illustrated in Fig. 3(a). For completeness, we mention that the magnetoresistance predicted turns out to be 1 order of magnitude smaller than observed, as shown in Fig. 3 of Ref. [9]. (ii) On the contrary, if $(1/\tau)_{\text{IMP}}$ is determined by adjusting it to describe $\tan(\theta)(4)$, then theory does describe appropriately the Hall tangent for all thickness as shown in Fig. 3(b), but the predicted temperature dependence of the resistivity agrees with the data only for the two thickest samples, as depicted in Fig. 2(b).

The second theory available is that published by Calecki [3]. The theory considers electrons occupying subbands with an energy $\varepsilon_{\nu\mathbf{k}} = \hbar^2(\mathbf{k}^2 + k_\nu^2)/2m$, where $\mathbf{k} = (k_x, k_y)$ represents the in-plane momentum, $k_\nu = \nu\pi/t$ represents the quantized momentum along z, the direction perpendicular to the surface of the film. The theory introduces an electron distribution function $f_\nu(\mathbf{k}) = f_0(\varepsilon_{\nu\mathbf{k}}) + \phi_\nu(\mathbf{k})$ [Eq. (11) in Ref. [3]] to describe the population of each subband, where $\phi_\nu(\mathbf{k})$ represents a linear function in $\mathbf{E}$, and $f_0(\varepsilon_{\nu\mathbf{k}})$ represents the equilibrium Fermi-Dirac distribution function. In this work, Calecki set up a BTE for $f_\nu(\mathbf{k})$ and proved that, in the presence of a magnetic field, electron-rough surface scattering leads to subband mixing; therefore, the Boltzmann collision operator cannot be characterized by a relaxation time τ, unless ν_F (the number of occupied subbands) is one. To circumvent this difficulty, the author introduced the matrix $T(\varepsilon)_{\nu\nu'}$ with dimensions of time, defined by Eq. (22) from Ref. [3]. The Hall tangent predicted by Calecki, is given by $\tan(\theta) = \frac{E_H}{E_L} = -\frac{\sigma_1}{\sigma_0 - \sigma_2}$, where σ_0, σ_1, and σ_2 are given by Eqs. (32)–(34) from Ref. [3]. As discussed in Ref. [9], in the limit of small correlation lengths (e.g., $k\xi < 1$, where k is the electron wave vector), $T(\varepsilon)_{\nu\nu'}$ becomes diagonal, and the resistivity $\rho_0 = (\sigma_0)^{-1}$, predicted at 4 K arising solely from electron-surface scattering in this limit, turns out to be about 2 orders of magnitude larger than observed. Such discrepancy could arise from an overestimation of the effect of electron-surface scattering, or could be instead a consequence of the approximation $k\xi < 1$ used to derive the diagonal form for the matrix $T_{\nu\nu'}(\varepsilon_F)$, an approximation not valid in our samples.

Summarizing, we have performed the *first* measurement of resistivity, transverse magnetoresistance, and Hall effect at low temperatures T (4 K $\leq T \leq$ 50 K) under high mag-

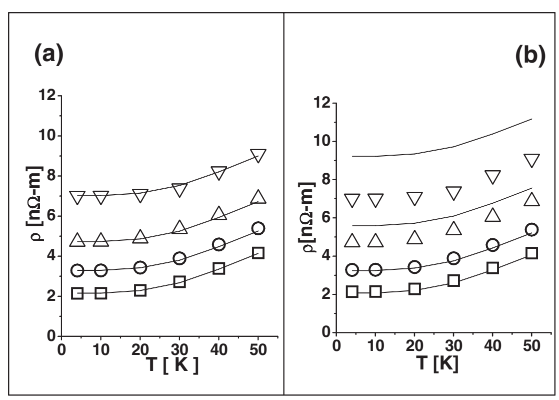

FIG. 2. Temperature dependence of the resistivity, predicted by the Sondheimer model, using: (a) $(1/\tau)_{\text{IMP}}$ adjusted to describe $\rho(4)$ for each sample. (b) $(1/\tau)_{\text{IMP}}$ adjusted to describe $\tan(\theta)(4)$ for each sample. Symbols as in Fig. 1.

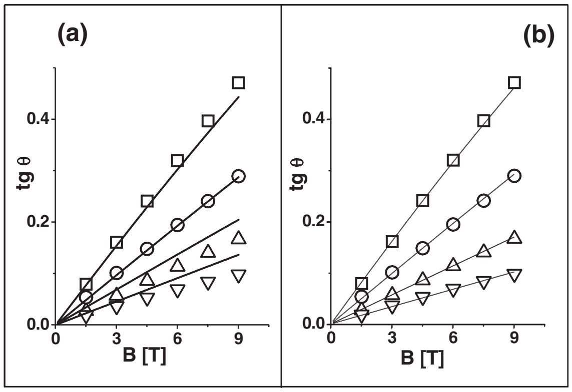

FIG. 3. (a) Magnetic field dependence of $\tan(\theta)$ observed at 4 K, symbols as in Fig. 1. Solid line represents the prediction of Sondheimer's model, adjusting $(1/\tau)_{\mathrm{IMP}}$ to describe $\rho(4)$ for each sample. The corresponding $\kappa = t/\ell$ at 4 K are 0.347, 0.295, 0.358, and 0.246 for the 69, 93, 150, and 185 nm film, respectively. (b) Magnetic field dependence of $\tan(\theta)$ observed at 4 K, symbols as in Fig. 1. Solid line represents the prediction of Sondheimer's model, adjusting $(1/\tau)_{\mathrm{IMP}}$ to describe $\tan(\theta)(4)$ for each sample.

netic fields B (1.5 T $\leq B \leq$ 9 T), on a family of four gold films evaporated onto mica substrates; in all transport experiments the signal is primarily controlled by electron-surface scattering. We also measured with a STM the surface roughness on each sample in the relevant (atomic) scale of length. Sondheimer's theory provides a fair description of the temperature dependence of the resistivity observed on all films, as well as an accurate description of the Hall voltage observed on the two thicker films at 4 K, but overestimates the Hall voltage observed on the two thinner films. However, it predicts a magnetoresistance 1 order of magnitude smaller than observed at 4 K. The theory of Calecki (in the limit of small correlation lengths $k\xi < 1$) leads to a resistivity arising solely from electron-surface scattering at 4 K which is 2 orders of magnitude larger than observed.

These measurements suggest that both theories available to describe size effects in the presence of a magnetic field orthogonal to the plane of a metallic film fail to provide a coherent description of the complete set of transport data. The results on resistivity, transverse magnetoresistance, and Hall voltage point to a charge transport process where electron scattering is primarily controlled by electron-surface scattering at low temperatures, and the results do agree qualitatively with Sondheimer's prediction. The agreement is satisfactory for resistivity, and less satisfactory for the Hall voltage. However, in spite of the fact that Sondheimer's formalism has been considered as the guiding theory on hundreds of papers published during several decades, the formalism fails to describe all three transport coefficients, and this can be considered the first indication of a severe shortcoming of the theory. In the case of Calecki's formalism, the failure to describe the data could arise from an overestimation of the effect of electron-surface scattering within the theory, or it could simply be a consequence of the inapplicability of the small correlation length approximation to our samples. To elucidate the origin of the discrepancies between theory and experiments, the transport equations proposed by Calecki ought to be solved numerically, computing the matrix $T(\varepsilon)_{\nu\nu'}$ for each sample. Such work is in progress.

This work was funded by FONDECYT under Contract No. 1040723.

*Corresponding author.
Present address: Departamento de Física, Facultad de Ciencias Físicas y Matemáticas, Universidad de Chile, Blanco Encalada 2008, Casilla 487-3, Santiago 8370415, Chile.
Email address: RAMUNOZ@CEC.UCHILE.CL
Telephone: 56-2-696-0148
Fax: 56-2-696-7359

[1] The first paper addressing size effects appears to be that by I. Stone, Phys. Rev. **6**, 1 (1898).
[2] E. H. Sondheimer, Phys. Rev. **80**, 401 (1950); Adv. Phys. **1**, 1 (1952).
[3] D. Calecki, Phys. Rev. B **42**, 6906 (1990).
[4] A. E. Meyerovich and S. Stepaniants, Phys. Rev. B **51**, 17 116 (1995); **58**, 13 242 (1998); **60**, 9129 (1999); J. Phys. Condens. Matter **12**, 5575 (2000); A. E. Meyerovich and I. V. Ponomarev, Phys. Rev. B **65**, 155413 (2002); **67**, 165411 (2003).
[5] G. Palasantzas, Phys. Rev. B **58**, 9685 (1998); G. Palasantzas and J. Barnas, Phys. Rev. B **56**, 7726 (1997); G. Palasantzas, Y. P. Zhao, G. C. Wang, T. M. Lu, J. Barnas, and T. J. M. De Hosson, Phys. Rev. B **61**, 11 109 (2000); G. Palasantzas and T. J. M. De Hosson, Phys. Rev. B **63**, 125404 (2001); J. A. Sánchez-Gil, V. Freilikher, I. Yurkevich, and A. A. Maradudin, Phys. Rev. Lett. **80**, 948 (1998).
[6] P. Joyez and D. Esteve, Phys. Rev. B **64**, 155402 (2001); H. Akera and T. Ando, Phys. Rev. B **41**, 11 967 (1990); **43**, 11 676 (1991).
[7] M. Jalochowski, M. Hoffman, and E. Bauer, Phys. Rev. Lett. **76**, 4227 (1996); M. Henzler, T. Lüer, and A. Burdach, Phys. Rev. B **58**, 10 046 (1998); M. Henzler, T. Lüer, and J. Heitmann, Phys. Rev. B **59**, 2383 (1999); G. M. Mikhailov, A. V. Chernykh, J. C. Maan, J. G. S. Lok, A. K. Geim, D. Esteve, and P. Joyez, Nanotechnology **11**, 379 (2000).
[8] D. Gitsu, T. Huber, L. Konopko, and A. Nikolaeva, Phys. Status Solidi A **196**, 137 (2003); O. Pfennigstorf, A. Petkova, H. L. Guenter, and M. Henzler, Phys. Rev. B **65**, 045412 (2002); J. Heremans, C. M. Thrush, Y. M. Lin, S. Cronin, Z. Zhang, M. S. Dresselhaus, and J. F. Mansfield, Phys. Rev. B **61**, 2921 (2000).
[9] R. C. Munoz *et al.*, J. Phys. Condens. Matter **18**, 3401 (2006).
[10] R. A. Matula, J. Phys. Chem. Ref. Data **8**, 1147 (1979).
[11] M. S. P. Lucas, J. Appl. Phys. **36**, 1632 (1965).