Resistivity, transverse magnetoresistance, and Hall voltage induced by electron-surface scattering on thin gold films deposited on mica substrates under high vacuum

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We contrast the numerical solution of the transport theory published by Calecki [D. Calecki, Phys. Rev. B **42**, 6906 (1990)], with transport data published recently [R. C. Munoz *et al.*, J. Phys.: Condens. Matter **18**, 3401 (2006); Phys. Rev. Lett. **96**, 206803 (2006)]. We use the resistivity, transverse magnetoresistance, and Hall voltage data of thin gold films deposited on mica substrates measured under high magnetic fields **B** ($1.5\ \mathrm{T} \leqslant \mathbf{B} \leqslant 9\ \mathrm{T}$) at low temperatures T ($4\ \mathrm{K} \leqslant T \leqslant 50\ \mathrm{K}$), as well as the surface roughness measured on each sample with a scanning tunneling microscope. The surprising result is that theory does provide an accurate description of the temperature dependence of the resistivity, a less accurate description of the Hall voltage observed at 4 K, but predicts a magnetoresistance at 4 K that turns out to be *several orders of magnitude smaller than observed.*

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The problem of "size effects" refers to the question of how does the roughness of the surface that limits a metallic structure affect its electrical transport properties, when one or more of the dimensions of the structure are comparable to or smaller than the mean free path ℓ of the charge carriers in the bulk. Despite over a century of research on size effects,[1] the effect of electron-surface scattering on charge transport is a fundamental problem in solid state physics that still remains open.

We recently published the first simultaneous measurements of resistivity, transverse magnetoresistance, and Hall voltage arising primarily from electron-surface scattering on a family of gold films of different thickness evaporated onto preheated mica substrates under high vacuum.[2] There are two theories available to explain magnetomorphic effects arising from electron-surface scattering on metallic films immersed in a magnetic field **B** perpendicular to the surface of the film. The first was published by Sondheimer.[3] The other theory was published by Calecki.[4]

Calecki used a Boltzmann transport equation (BTE) to describe electron transport, and estimated the effect of electron-surface scattering by calculating the perturbation induced by the presence of the two rough surfaces limiting the film, over and above the Hamiltonian describing an electron gas confined within two parallel flat surfaces. The author introduced an electron distribution function $f_\nu(\mathbf{k}) = f_0(\varepsilon_{\nu\mathbf{k}}) + \phi_\nu(\mathbf{k})$ [Eq. (11) in Ref. 4] for electrons occupying each subband 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 [where z is the direction perpendicular to the film, t is the thickness of the film, $\phi_\nu(\mathbf{k})$ represents a linear function in the electric field **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 electron-rough surface scattering induces subband mixing. Consequently the Boltzmann collision operator describing electron-surface scattering *cannot be characterized by a relaxation time* τ (unless there is only one occupied subband). To circumvent such difficulty the author introduced the "collision time" matrix $T(\varepsilon)_{\nu\nu'}$ defined by Eq. (22) from Ref. 4.

Presently Calecki's formalism is the only theory describing magnetomorphic effects arising from electron-surface scattering in metallic films immersed in a magnetic field orthogonal to the film, that contains *no adjustable parameters*. The transport coefficients predicted by theory are univocally determined by the rms roughness amplitude δ and the lateral correlation length ξ characterizing the rough surface. However, a comparison between theory and experiment that takes advantage of the fact that theory contains no adjustable parameters has never been published, because of the lack of experimental work reporting measurements of both the magnetic transport coefficients as well as the surface roughness measured in the appropriate scale of length. Recent publication of surface roughness data measured in the scale of length set by the Fermi wave length (which for Au is 0.52 nm), as well as resistivity, transverse magnetoresistance, and Hall voltage data measured on each member of a family of thin gold films,[2] permits a reversal of this trend. It makes possible *the first cross check between theory and experiment involving several transport coefficients*, a comparison that was already performed in the case of the other formalism available, the theory of Sondheimer.[3] In this paper we report the first comparison between Calecki's theory and experimental data involving resistivity, transverse magnetoresistance, and Hall voltage measured on a family of gold films of different thickness.

It seems appropriate to point out that Calecki used in his work, the small correlation length approximation $k\xi < 1$ (where k stands for the electron wave vector) to deduce a diagonal form for the "collision time" matrix $T(\varepsilon)_{\nu\nu'}$. As will be shown below, the resistivity predicted under the approximation $k\xi < 1$, turns out to be about 2 orders of magnitude larger than observed. To understand why such discrepancy arises it seems appropriate to recall that, for many metals, the Fermi wave length λ_F is of the order of a few atomic diam-

eters. In such metals the approximation $k_F\xi = 2\pi\xi/\lambda_F < 1$ used by the author is severely violated, for ξ is also expected to be of the order of, or larger than, a few atomic diameters. In gold $k_F = 12.1\ \mathrm{nm}^{-1}$, and in our gold films ξ ranges from 7.6 to 12.2 nm (Table I from Ref. 2), hence $90 < k_F\xi < 145$. For such values of $k_F\xi$, replacing the Fourier transform of the height-height autocorrelation function $F(k_F\xi)$ for its value at the origin $F(0)=\pi$ (appropriate for a Gaussian autocorrelation function), leads to an overestimation of $F(k_F\xi)$ by as much as two orders of magnitude. This approximation gives rise to an underestimation of the elements of the collision time matrix $T(\varepsilon)_{\nu\nu'}$ [that are inversely proportional to $F(k_F\xi)$], hence to an overestimation of the resistivity induced by electron-surface scattering.

To avoid the "tour de force" implied by the small correlation length approximation $k\xi < 1$, and to perform a meaningful comparison between theory and experiment, we use the full nondiagonal form for the matrix $T(\varepsilon)_{\nu\nu'}$ contained in Ref. 4. The goal is to elucidate, for the first time, whether or not theory is capable of describing the experimental *data involving several transport coefficients, once the transport equations contained in the theory are solved numerically, using as input the surface roughness measured on each sample*. We abandon the small correlation length approximation $k\xi < 1$, an approximation that would render questionable any comparison between theory and experiment.

Details of the sample preparation, of the morphology and structure of the gold films, of the measurement of resistivity, magnetoresistance, Hall voltage, and the surface roughness parameters appropriate to each sample, can be found in Ref. 2. The resistivity, transverse magnetoresistance and Hall tangent predicted by theory can be calculated from $\rho = (\sigma_o)^{-1}$, $\frac{\Delta\rho}{\rho} = \frac{(\sigma_0-\sigma_2)\sigma_0}{(\sigma_0-\sigma_2)^2+\sigma_1^2} - 1$, and $\tan(\theta) = \frac{E_H}{E_L} = -\frac{\sigma_1}{\sigma_0-\sigma_2}$, where E_H stands for the transverse (Hall) field, E_L stands for the longitudinal field, σ_0, σ_1, and σ_2 are given by

$$\sigma_0 = \sum_\nu \frac{n_\nu q^2}{m} \sum_{\nu'} \langle T_{\nu\nu'}(\varepsilon)\rangle_\nu,$$

$$\sigma_1 = \omega_C \sum_\nu \frac{n_\nu q^2}{m} \sum_{\nu'} \langle [(1+\omega_C{}^2 T^2(\varepsilon))^{-1} T^2(\varepsilon)]_{\nu\nu'}\rangle_\nu,$$

$$\sigma_2 = \omega_C{}^2 \sum_\nu \frac{n_\nu q^2}{m} \sum_{\nu'} \langle [(1+\omega_C{}^2 T^2(\varepsilon))^{-1} T^3(\varepsilon)]_{\nu\nu'}\rangle_\nu,$$

which are Eqs. (32)–(34) from Ref. 4, where $\omega_C = qB/m$ stands for the cyclotron frequency, q is the electron charge, B is the magnetic field, m is the electron effective mass, n_ν is the electron density in subband ν, $T(\varepsilon)_{\nu\nu'}$ is the collision time matrix and the symbol $\langle\psi\rangle_\nu$ stands for the average of the quantity ψ over subband ν [given by Eq. (29) in Ref. 4]. In the limit of small correlation lengths (e.g., $k\xi < 1$), $T(\varepsilon)_{\nu\nu'}$ becomes diagonal, for in this case electron-surface scattering causes the distribution function $f_\nu(\mathbf{k})$ to relax towards the Fermi-Dirac distribution function $f_0(\varepsilon_{\nu\mathbf{k}})$ with a "relaxation time" τ_ν associated with subband ν. τ_ν is given by

$$T_{\nu\nu'}(\varepsilon_F) = \frac{m}{\pi^5\hbar}\frac{6t^6}{\nu_F(\nu_F+1)(2\nu_F+1)}\frac{1}{\delta^2\xi^2\nu^2}\delta_{\nu\nu'} = \tau_\nu(\varepsilon_F)\delta_{\nu\nu'} \tag{1}$$

[Eq. (64) from Ref. 4] for the case of a Gaussian autocorrelation function $f(x,y) = \frac{1}{S}\iint h(x+u,y+v)h(u,v)dudv = \delta^2\exp\left(-\frac{x^2+y^2}{\xi^2}\right)$, where $h(x,y)$ is the height of the random rough surface (measured with a scanning tunneling microscope) at a point designated by the in-plane coordinates (x,y), S is the surface of the sample, and ν_F is the number of occupied subbands.

To compare theory and experiment, we must compute the coefficients σ_0, σ_1, and σ_2 when $T(\varepsilon)_{\nu\nu'}$ is not diagonal. To do so it becomes necessary to calculate numerically the matrix $C(\varepsilon)_{\nu\nu'}$ [defined by Eq. (19) of Ref. 4] that describes electron-surface scattering taking place at the exposed gold surface. The gold-mica interface is atomically flat except for cleavage steps that are separated by distances which are long compared to ℓ, hence the gold-mica interface is considered a specular surface.[2] The matrix elements $C(\varepsilon)_{\nu\nu'}$ are given by

$$C(\varepsilon_F)_{\nu\nu'} = \frac{S\pi^2}{4\hbar t^6}\delta^2\xi^2\left[\delta_{\nu\nu'}\nu^2 k^2{}_\nu \sum_{\mu=1}^{\nu_F}\mu^2 I_{\nu\mu} - k_\nu k_{\nu'}\nu^2\nu'^2 J_{\nu\nu'}\right] \tag{2}$$

that corresponds to Eq. (58) in Ref. 4, with $k_\nu = \sqrt{(k_F{}^2 - (\frac{\pi\nu}{t})^2)}$ [Eq. (59) in Ref. 4], where k_F is the Fermi wave vector. The symbols $I_{\mu\nu}$ and $J_{\mu\nu}$ stand for the integrals of the Fourier transform $F(k_x,k_y)$ of the height-height autocorrelation function $f(x,y)$

$$I_{\mu\nu} = \int_0^{2\pi} F(\xi\sqrt{k^2{}_\nu + k^2{}_\mu - 2k_\nu k_\mu\cos\alpha})d\alpha = 2\pi^2\exp\left[-\frac{1}{4}\xi^2(k^2{}_\nu + k^2{}_\mu)\right] I_0\left(\frac{1}{2}\xi^2 k_\nu k_\mu\right), \tag{3}$$

$$J_{\mu\nu} = \int_0^{2\pi} F(\xi\sqrt{k^2{}_\nu + k^2{}_\mu - 2k_\nu k_\mu\cos\alpha})\cos\alpha d\alpha = 2\pi^2\exp\left[-\frac{1}{4}\xi^2(k^2{}_\nu + k^2{}_\mu)\right] I_1\left(\frac{1}{2}\xi^2 k_\nu k_\mu\right), \tag{4}$$

where I_0 and I_1 are the modified Bessel function of order 0 and 1, respectively[5] [Eq. (2) has been used by Calecki to establish a limiting law governing the dependence of the conductivity of a thin metallic film on film thickness, see Eq. (15) in Ref. 6]. Rather than inverting the matrix C and the matrix $[1+\omega_C{}^2T^2(\varepsilon)]$, we used instead the approach of numerically solving the linear system of equations $CX=Y$ and $[1+\omega_C{}^2T^2(\varepsilon)]X=Y$. This method is significantly faster and reduces considerably the numerical error.

In order to compare theory and experiment, we must include in the computation of the matrix elements $C(\varepsilon)_{\nu\nu'}$, the effect of other electron scattering mechanisms acting in the bulk. As discussed in Refs. 2 and 4, electron scattering in the bulk does not induce subband mixing. Consequently, the corresponding matrix T is diagonal, for it reduces to $T = \tau_{\mathrm{BULK}}$,

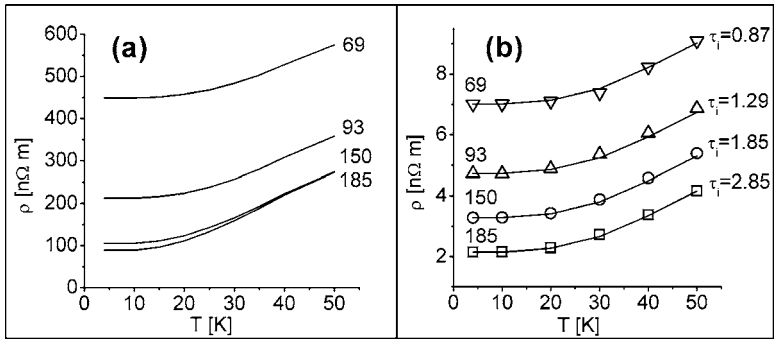

FIG. 1. (a) Solid line: Temperature dependence of the resistivity of 4 films of different thickness, predicted by Calecki's theory [with a "subband relaxation time" given by Eq. (1)], for the films having the thickness (in nm) indicated. (b) Temperature dependence of the resistivity measured on different films: Squares, film 185 nm; Circles: film 150 nm; Triangles: film 93 nm; Inverted triangles: film 69 nm. Solid lines represent the resistivity predicted on the basis of the numerical solution of the transport equations contained in Calecki's theory. The collision times τ_i (indicated in the figure in units of 10^{-13} sec) have been adjusted to describe $\rho(4)$ observed on each sample.

where τ_{BULK} represents the collision time describing electron scattering in the bulk *common to all subbands*. The matrix T is defined by $C(\varepsilon)T(\varepsilon)=F(\varepsilon)$ [Eq. (22) in Ref. 4]. Matrix $C(\varepsilon)$ depends upon the scattering rate characterizing each electron scattering mechanism, but matrix $F(\varepsilon)$ [defined by Eq. (20) from Ref. 4] is a diagonal matrix independent of the scattering mechanism. When two electron scattering mechanisms are present (for example mechanism 1—electron scattering in the bulk, and mechanism 2—electron-surface scattering), we have $C_1T_1=C_2T_2=\mathrm{F}$, hence $C_1+C_2=F(T_1^{-1}+T_2^{-1})$. Since the scattering rates due to electron-surface scattering and electron scattering in the bulk are additive, to compute the matrix C describing both mechanisms acting simultaneously, the product of $F(\varepsilon)_{\nu\nu}$ times $1/\tau_{\mathrm{BULK}}$ ought to be added to the diagonal elements $C(\varepsilon)_{\nu\nu}$ arising from electron-rough surface scattering given by Eq. (2).

Two of the electron scattering mechanisms that are relevant to the present discussion, are electron-phonon scattering and electron-impurity scattering. The inverse of the relaxation time describing these processes, can be computed according to $1/\tau_{\mathrm{BULK}}=(1/\tau)_{\mathrm{IMP}}+(1/\tau)_{\mathrm{PHON}}$, where the first (temperature independent) term accounts for electron scattering by impurities, and the second (temperature dependent) term accounts for electron-phonon scattering.[7] At 4 K the phonons are frozen out, hence electron-phonon scattering can be neglected. To determine $1/\tau_{\mathrm{BULK}}$ describing electron scattering in the bulk 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. 7. Following the procedure outlined, we adjusted $(1/\tau)_{\mathrm{IMP}}$ to describe $\rho(4)$ for each sample. The temperature dependence of the resistivity predicted by theory, computed using the numerical solution of the transport equations contained in the theory, incorporating electron-scattering in the bulk in the manner described, is displayed in Fig. 1. The magnetoresistance predicted at 4 K is displayed in Fig. 2. The Hall voltage predicted a 4 K is displayed in Fig. 3.

The resistivity arising from electron-surface scattering predicted using the diagonal form of $T(\varepsilon)_{\nu\nu'}$ [Eq. (1)] exceeds by about 2 orders of magnitude the observed resistivity, as displayed in Fig. 1(a). Nevertheless, it seems remarkable that, when the transport equations are solved numerically *without approximations* using the method described, a set of collision times τ_i can be found, that leads to an accurate description of the temperature dependence of the resistivity, as shown in Fig. 1(b). Consequently, the discrepancy between the predicted resistivity displayed in Fig. 1(a) and the observed resistivity displayed in Fig. 1(b) is a consequence of the small correlation length approximation $k\xi<1$ used to derive Eq. (1), a condition that is severely violated in our samples. Equally remarkable and surprising, is the fact that the transverse magnetoresistance predicted at 4 K computed using the parameter τ_i listed in Fig. 1(b) *turns out to be several orders of magnitude smaller than observed*, as displayed in Fig. 2. *The predicted magnetoresistance violates Kohler's rule, and is no longer monotonically increasing with increasing film thickness*, a fact that seems also remarkable. The violation of Kohler's rule is not surprising, for it can be regarded simply as a consequence of the fact that

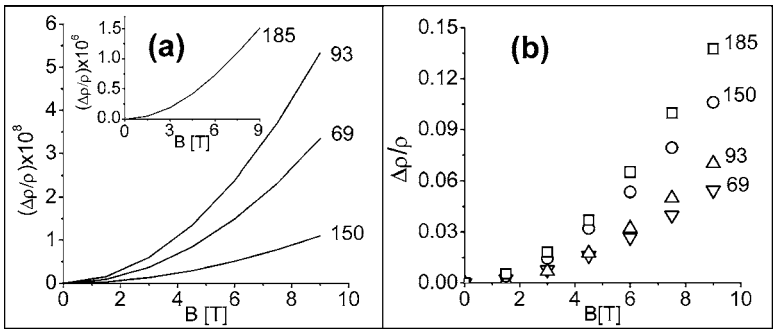

FIG. 2. (a) Transverse magnetoresistance predicted by theory at 4 K, for the 69 nm, 93 nm, and 150 nm thick, using the full nondiagonal scattering time matrix $T(\varepsilon)_{\nu\nu'}$. Inset: Transverse magnetoresistance predicted by theory at 4 K, for the film 185 nm thick. (b) Transverse magnetoresistance measured at 4 K on films of different thickness, symbols as in Fig. 1(b); data from Ref. 2.

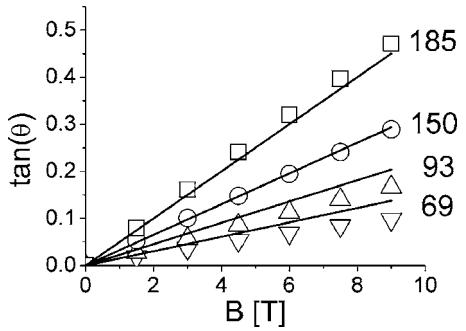

FIG. 3. Hall tangent $\tan(\theta)=E_H/E_L$ measured as a function of magnetic field B at 4 K, symbols as in Fig. 1(b); data from Ref. 2. Solid lines represent $\tan(\theta)$ predicted using the numerical solution of the transport equations contained in Calecki's theory.

within Calecki's theory, the Boltzmann collision operator cannot be represented by a relaxation time τ. Regarding the Hall tangent, displayed in Fig. 3, theory predicts at 4 K a Hall tangent that coincides roughly with the experimental data for the 150 and 185 nm film, but overestimates the Hall tangent for the 69 and 93 nm films. Such performance is strongly reminiscent of that regarding Sondheimer's theory, a model that was found to provide an accurate description of the temperature dependence of the resistivity, to provide a less accurate description of the Hall voltage observed at 4 K, but that leads to a magnetoresistance predicted at 4 K *that is one order of magnitude smaller than observed*.[2]

Another remarkable result of this comparison between theory and experiment, is the fact that the scattering times τ_i displayed in Fig. 1(b) turn out to increase roughly linearly with film thickness, and that the corresponding mobility $\mu_i = q\tau_i/m$ agrees (to within 35% or better) with the observed Hall mobility.[2] This suggests that either the concentration of impurities/defects is inversely proportional to the film thickness, or else *the mobility observed in our samples at* 4 K *is primarily determined by electron-surface scattering rather than electron-impurity scattering*. The latter interpretation would imply that electron-surface scattering *can no longer be considered a perturbation* when compared to other electron-scattering mechanisms acting in the bulk at low temperatures.

Summarizing, we have performed the first comparison between the numerical solution of the transport equations contained in Calecki's theory, and experimental data involving resistivity, transverse magnetoresistance and Hall voltage observed on a family of thin gold films evaporated onto mica substrates measured at temperatures T such that $4\ \mathrm{K} \leqslant T \leqslant 50\ \mathrm{K}$, under high magnetic fields B ($1.5\ \mathrm{T} \leqslant B \leqslant 9\ \mathrm{T}$). The surprising result is that the theory does provide an accurate description of the temperature dependence of the resistivity, it provides a less accurate description of the Hall voltage observed at 4 K, but leads to a magnetoresistance predicted at 4 K that is *several orders of magnitude smaller than observed*. These results indicate that the discrepancy between theory and experiment reported in Ref. 2, *does not arise from the small correlation length approximation* $k\xi < 1$. *It reflects instead, a shortcoming of the theory*. Such agreement in the resistivity and Hall voltage, and such sharp discrepancy in the transverse magnetoresistance, are reminiscent of the predictions based upon Sondheimer's theory.

The foregoing analysis confirms the suggestion that none of the theories currently available provide a coherent description of the complete set of transport data.[2] The failure of both models, Calecki's and Sondheimer's, may be considered the first strong, compelling evidence pointing to the need for a new, fresh theory to describe size effects arising from electron-surface scattering in metallic films in the presence of a magnetic field.

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