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LETTER TO THE EDITOR

Surface roughness and surface-induced resistivity of gold films on mica: influence of the theoretical modelling of electron–surface scattering

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Abstract. We analyse the thickness and temperature dependence of the resistivity for several gold films on mica reported by Sambles, Elsom and Jarvis (SEJ: Sambles J R, Elsom K C and Jarvis J D 1982 *Phil. Trans. R. Soc.* A **304** 365). Data analysis proceeds according to an iteration procedure proposed recently (Munoz R C, Concha A, Mora F, Espejo R, Vidal G, Mulsow M, Arenas C, Kremer G, Moraga L, Esparza R and Haberle P 2000 *Phys. Rev.* B **61** 4514; Munoz R C, Vidal G, Kremer G, Moraga L, Arenas C and Concha A 2000 *J. Phys.: Condens. Matter* **12** 2903), that permits the calculation of the temperature-dependent bulk conductivity $\sigma_0(T)$ from the parameters δ (r.m.s. roughness amplitude) and ξ (lateral correlation length) that describe the surface roughness. To assess the influence of the theoretical modelling of the electron–surface scattering, we use the theory of Tesanovic, Jaric and Maekawa (TJM), the theory of Trivedi and Aschroft (TA) and the modified theory of Sheng, Xing and Wang (mSXW). With the parameters δ and ξ measured for a 70 nm gold film deposited on mica, under similar conditions of evaporation, all three models reproduce approximately the thickness and temperature dependence of the resistivity (between 4 K and 300 K) of the SEJ films without using any adjustable parameter. Agreement between theory and experiment improves according to the sequence TJM, TA, mSXW.

One of the fundamental problems in solid-state physics that has attracted the attention of researchers for over sixty years relates to the effect of electron–surface scattering on the transport properties of thin metallic and semiconducting films. One of the central issues is how the surface of the structure affects its electrical transport properties, when one or more of the dimensions characterizing the structure are comparable to or smaller than the mean free path of the charge carriers: what is known as 'size effects'.

The experimental work related to size effects in thin metal films has for many decades relied on the method of:

(i) preparing families of samples of the same material but of different thickness under similar conditions of evaporation;
(ii) measuring one or more of the transport properties of the different members of the family, most commonly the resistivity;

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(iii) fitting the parameters provided by theory (specularity parameter R, bulk conductivity σ_0 and in some cases the r.m.s. roughness amplitude) to the thickness and/or the temperature dependence of the data.

The possibility of measuring directly the parameters that characterize the surface roughness on a nanometric scale with a scanning tunnelling microscope (STM) [1], together with the development of theories that permit the calculation of the film conductivity σ in terms of the bulk conductivity σ_0 and of the parameters (δ, ξ) that describe the surface roughness profile—where δ represents the r.m.s. roughness amplitude of the average height–height autocorrelation function (ACF) and ξ represents its lateral correlation length—permit a reversal of this trend of data analysis based solely on parameter fitting.

There are several quantum theories that permit the calculation of the film conductivity σ in terms of the bulk conductivity σ_0, the bulk mean free path ℓ and the parameters (δ, ξ) that describe the roughness profile: the theory of Trivedi and Aschroft (TA; reference [2]), the theory of Tesanovic, Jaric and Maekawa (TJM, reference [3]) and the modified theory of Sheng, Xing and Wang (mSXW; reference [1]). Whichever theory we choose to describe electron–surface scattering, we encounter the practical difficulty that to calculate the film conductivity σ we need to know the parameters σ_0 and ℓ that characterize the bulk—where σ_0 represents the conductivity and ℓ represents the mean free path *that would be observed in a film having the same concentration of impurities as the thin film, but thick enough such that the effect of electron–surface scattering can be neglected*—and these parameters are not known *a priori*. Knowledge of the parameters δ and ξ describing the surface roughness profile permits the determination of σ_0 and ℓ through an iteration process that departs from the parameter fitting that has dominated the analysis of size-effect data for decades, an iteration process that proceeds as follows [4, 5].

As a first approximation, $\ell(T)$ corresponding to each temperature, is calculated from $\ell_1(T) = \sigma(T)mv_F/(ne^2)$, where $\sigma(T) = 1/\rho(T)$ is the conductivity of the film measured at temperature T, m is the electron mass, v_F is the Fermi velocity, n the electron density and e the electron charge. This value $\ell = \ell_1$ is used to compute a first estimate of $[\sigma(T)/\sigma_0(T)]_1$, employing the roughness parameters (δ, ξ) to determine the increase in resistivity $\rho_0(T)/\rho(T) = \sigma(T)/\sigma_0(T) = q(T) < 1$ induced by electron–surface scattering, according to whichever theory we choose to describe the size effects in metal films. A corrected value for ℓ can then be calculated from $\ell_2 = \ell_1[\sigma_0(T)/\sigma(T)]_1$, and a new value of $[\sigma(T)/\sigma_0(T)]_2$ can be computed using the parameters (δ, ξ) and the theory with $\ell = \ell_2$. This process is repeated until the values of $[\sigma(T)/\sigma_0(T)]_j$ and $[\sigma(T)/\sigma_0(T)]_{j+1}$ for two successive iterations j and $j + 1$ differ by no more than 0.01%.

An interesting situation arises in metal films that satisfy the following conditions:

(a) grain-boundary scattering is negligible when compared to electron–surface scattering,
(b) electron–surface scattering taking place at the lower surface of the film (in contact with the substrate) is negligible when compared to electron–surface scattering taking place at the upper (exposed) surface of the film and
(c) the resistivity arising from electron–impurity scattering at 300 K is small compared to that arising from electron–phonon scattering at the same temperature.

In samples that satisfy these conditions, the electron scattering mechanisms that give rise to the observed film resistivity are electron–impurity scattering, electron–phonon scattering and electron–surface scattering at the upper surface of the film. The first two scattering mechanisms give rise to the bulk resistivity. For such films, if the theory used to compute the increase in resistivity induced by electron–surface scattering from the parameters (δ, ξ) that

characterize the surface roughness is correct, then the temperature-dependent bulk resistivity $\rho_0(T) = 1/\sigma_0(T)$ computed through the iteration process outlined above should agree with that expected from electron–phonon scattering plus electron–impurity scattering in the crystalline material. Consequently, the temperature dependence of $\rho_0(T)$ determined according to the iteration process sketched above should be consistent with a Bloch–Grüneisen description of the resistivity $\rho_0(T)$ in the bulk [6]:

$$\rho_0(T) = \rho_R + A\left(1 + \frac{BT}{\theta - CT}\right)\phi\left(\frac{\theta - CT}{T}\right) \qquad \text{with } \phi(x) = 4x^{-5}\int_0^x \frac{z^5 \exp(z)}{(\exp(z) - 1)^2}\,\mathrm{d}z \tag{1}$$

where ρ_R stands for the temperature-independent residual resistivity attributed to impurities (determined by the thin-film resistivity $\rho(4)$ measured at 4 K and by the ratio $\rho_0(4)/\rho(4)$ predicted by theory). But if $\rho_0(T) = 1/\sigma_0(T)$ determined through this iteration process turns out to be consistent with a Bloch–Grüneisen description, then the observed film resistivity should agree with $\rho(T) = \rho_0(T)/q(T)$, *where $\rho_0(T)$ is the bulk resistivity given by equation (1) and $q(T)^{-1}$ represents the increase of resistivity induced by electron–surface scattering predicted by theory*. This last criterion provides a powerful tool for testing different theories of size effects in metal films. For if the parameters (δ, ξ) chosen to describe the surface roughness and the theory chosen to describe the size effects are correct, then the theory ought to be capable of describing both *the thickness and the temperature dependence* of the resistivity observed in films of different thickness *without adjustable parameters*, something that constitutes quite a stringent test.

Since different theories of electron–surface scattering predict different values for $q(T) = \rho_0(T)/\rho(T)$ for the *same* set of parameters (δ, ξ) characterizing the roughness of the surface, it seems interesting to find out whether *any* of the available theories describing size effects in metal films is capable of reproducing, at least approximately, the temperature and the thickness dependence of the data, for samples that satisfy conditions (a)–(c). Concerning condition (a), we might expect grain-boundary scattering to be negligible compared to electron–surface scattering, when the lateral dimension D that characterizes the grains that make up the sample is about one order of magnitude larger than the film thickness t, for then the electrons are expected to undergo an average of several collisions with the upper/lower surface of the film before colliding with the boundary of a grain. Concerning condition (b), we might expect the electron–surface scattering at the surface of the film in contact with the substrate to be negligible compared to electron–surface scattering at the upper surface of the film in films that have been grown onto an insulating cleavable crystalline substrate (mica), such that the roughness contributed by the substrate consists of cleavage steps that occur infrequently over the scale of distance of a few hundred nm probed by the electrons in their motion through the metal film.

Sambles, Elsom and Jarvis (SEJ) published measurements of the resistivity of several films of different thickness deposited by thermal evaporation of gold on mica, which led to samples in which D is in the region of several hundred nm (figures 1(c), 1(d) of reference [7]). The SEJ samples whose thickness t satisfies $t \leqslant 100$ nm also satisfy the condition that D is about an order of magnitude larger than t; therefore these samples satisfy conditions (a) and (b). The resistivities of the SEJ-35 nm, SEJ-53 nm, SEJ-80 nm and SEJ-126 nm films at 300 K are some 15% to 30% larger than $\rho_I(300) = 22.49$ nΩ m expected purely from electron–phonon scattering in crystalline gold [6]; therefore these SEJ samples also satisfy condition (c).

We reported recently surface roughness and resistivity measurements performed on a gold film 70 nm thick thermally evaporated onto a mica substrate under conditions of evaporation (temperature of the substrate: 300 °C; speed of evaporation: 6 nm min^{-1}) quite close to

those used by SEJ (temperature of the substrate: 280 °C; speed of evaporation: 5 nm min^{-1}), except for the fact that SEJ used as starting material 99.9999% pure gold—purity two orders of magnitude better than that of the gold wire (99.99% pure) that we used as starting material. Since the surface roughness is expected to depend primarily on the conditions of evaporation (on the speed of evaporation and on the temperature of the substrate), and is expected to be independent of whether the concentration of impurities in the sample is in the range of 1 part in 10^4 or 1 part in 10^6—despite the fact that a small concentration of impurities is likely to influence the observed film resistivity—we decided to analyse the SEJ data as if the SEJ samples had a surface roughness similar to that of our gold films, using the parameters that describe the average surface roughness of a 70 nm thick gold film deposited on mica recently measured on a nanometric scale [1, 5].

In this letter we report an analysis of the SEJ-35, SEJ-53, SEJ-80 and SEJ-126 resistivity data between 4 K and 300 K, using the parameters (i) $\delta = 0.455$ nm and $\xi = 0.480$ nm, the averages of δ and ξ over the 8×8, 10×10 and 12×12 cells of table 1 of reference [5], for a Gaussian representation of the ACF and (ii) $\delta = 0.689$ nm and $\xi = 0.233$ nm, the averages of δ and ξ over the 6×6, 8×8 and 10×10 cells of table 1 of reference [5], for an exponential representation of the ACF. The analysis proceeds according to the iteration procedure outlined above, and contains *no adjustable parameters*. To assess the influence of theoretical modelling, in the analysis of the SEJ data we used the different theories available to describe electron–surface scattering in terms of δ and ξ: TA and TJM—that use an ACF characterized by a Gaussian which for short-range correlations has been replaced by a delta function with a r.m.s. amplitude δ according to the so-called white-noise approximation—and mSXW—that allows both a Gaussian and an exponential representation of the ACF. In the case of TA theory, we computed the film resistivity as the inverse of the film conductivity (equation (4.13) in reference [2]); in the case of TJM theory, we computed the film resistivity as the inverse of the film conductivity (equation (7) in reference [3]). The film resistivity in the case of mSXW theory was computed as the inverse of the film conductivity given by equation (1) of reference [1], with the reflectivity $R(k_\parallel)$ given in reference [1] and the self-energy of the electron gas $Q(k_\parallel)$ given by equations (5) and (6) of reference [1] for the case of a Gaussian and of an exponential representation of the ACF, respectively. The results are plotted in figure 1, on the same double-logarithmic scale as was used by SEJ.

The first remarkable result—considering that *none* of the theories contain *any* adjustable parameters—is that all four models provide an approximate description of both the temperature and the thickness dependence of the data between 4 K and 300 K. The agreement between theory and experiment is about 15% or better in the TJM case, it is about 10% or better in the TA case and it is better than 7% in the mSXW case, regardless of whether we use a Gaussian or an exponential representation of the ACF. A second interesting feature is that the residual resistivities ρ_R predicted by different models for each film are comparable but are *model dependent*. What is more interesting is that the residual resistivities predicted by the same model *are different for films of different thickness*—despite the fact that the films were evaporated under similar conditions of evaporation—and decrease as the thickness of the film increases; this is at variance with the constant residual resistivity (independent of film thickness) that has been assumed for several decades in the analysis of size-effect data. This might be expected if thicker films had a smaller concentrations of impurities than thinner films, something consistent with the fact that at 4 K, the bulk mean free path ℓ determined using any of the theories grows larger as the film grows thicker.

In the case of mSXW theory, the residual resistivities predicted for a Gaussian representation of the ACF are smaller than those predicted for an exponential representation of the ACF. Consequently, the bulk resistivity $\rho_0(T)$ predicted for a Gaussian ACF turns out to

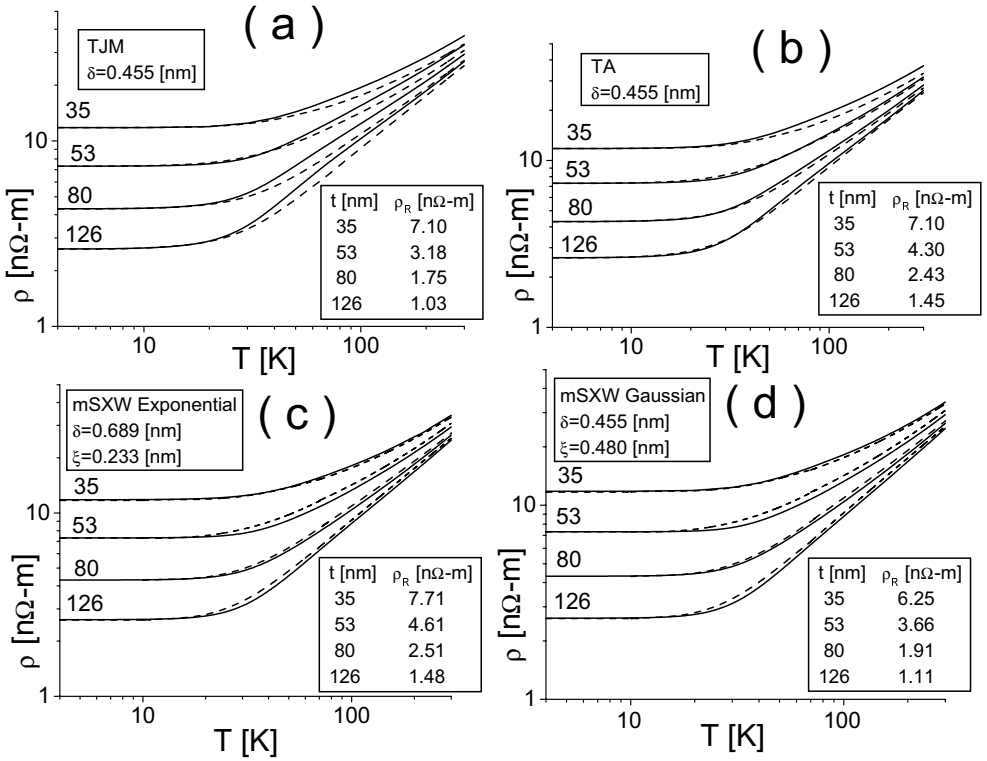

Figure 1. (a) Dashed lines: the resistivity of the 35 nm, 53 nm, 80 nm and 126 nm thick gold films on mica as reported in figure 3(a) of reference [7]. Solid lines: the film resistivity $\rho(T)$ described on the basis of a Bloch–Grüneisen model, using equation (1), ρ_R as listed and the constants $A = 12.359$ nΩ m, $B = -9.8996 \times 10^{-4}$, $C = 3.3994 \times 10^{-2}$, $\theta = 172.1$ K from reference [6] and using the ratio σ/σ_0 predicted by the theory of Tesanovic, Jaric and Maekawa (TJM; reference [3]), for an ACF described by $\delta = 0.455$ nm. (b) Dashed lines as in (a). Solid lines: the film resistivity $\rho(T)$ described on the basis of a Bloch–Grüneisen model, using equation (1), ρ_R as listed, the constants A, B, C and θ as in (a) and the ratio σ/σ_0 predicted by the theory of Trivedi and Aschroft (TA; reference [2]), for an ACF described by $\delta = 0.455$ nm. (c) Dashed lines as in (a). Solid lines: the film resistivity $\rho(T)$ described on the basis of a Bloch–Grüneisen model, using equation (1), ρ_R as listed, the constants A, B, C and θ as in (a) and the ratio σ/σ_0 predicted by mSXW theory (reference [1]), for an ACF described by $f(x, y) = \delta^2 \exp[-\sqrt{x^2 + y^2}/\xi]$ with $\delta = 0.689$ nm, $\xi = 0.233$ nm. (d) Dashed lines as in (a). Solid lines: the film resistivity $\rho(T)$ described on the basis of a Bloch–Grüneisen model, using equation (1), ρ_R as listed, the constants A, B, C and θ as in (a) and the ratio σ/σ_0 predicted by mSXW theory (reference [1]), for an ACF described by $f(x, y) = \delta^2 \exp[-(x^2 + y^2)/\xi^2]$, with $\delta = 0.455$ nm, $\xi = 0.480$ nm.

be systematically smaller than $\rho_0(T)$ predicted for an exponential representation of the ACF; this is consistent with what we found by analysing the resistivity data for our 70 nm thick gold film [5]. The increase in resistivity $q(T) = \rho_0(T)/\rho(T)$ induced by electron–surface scattering computed for a Gaussian representation of the ACF also turns out to be systematically smaller than that computed for an exponential representation of the ACF at all temperatures. The outcome is that the resistivity of the film $\rho(T) = \rho_0(T)/q(T)$ predicted for a Gaussian ACF agrees to better than 0.5% with the resistivity of the film predicted for an exponential representation of the ACF for all four films and 4 K $\leqslant T \leqslant$ 300 K. *The two representations of the ACF within mSXW theory lead essentially to the same film resistivity.*

This work departs sharply from the method of parameter fitting traditionally used to

analyse size-effect data. Rather than assuming that the parameters σ_0, ℓ and the surface reflectivity R characterizing the samples are the same for films of different thickness, *we calculated σ_0 and ℓ*, starting from the justifiable assumption that, because our 70 nm gold film and the SEJ films were prepared—with the exception of the purity of the starting material—under similar conditions of evaporation, then the parameters δ and ξ characterizing the surface roughness should be about the same. We used an iteration procedure published recently that permits the determination of the parameters characterizing the bulk from the knowledge of the parameters that characterize the surface roughness [4, 5]. The outcome of this analysis is that the temperature and the thickness dependence of the resistivity for these four SEJ films can be approximately accounted for in terms of a Bloch–Grüneisen model describing electron–phonon scattering and electron–impurity scattering in the bulk, corrected by the mSXW theory describing electron–surface scattering. *Grain-boundary scattering is ignored*.

It seems appropriate to mention that SEJ fitted the temperature and the thickness dependence of the data measured on six films between 2 K and 300 K, using a model containing five adjustable parameters. As a result of this fitting procedure, SEJ arrive at the conclusion that $r = \delta/\lambda_F \approx 0.05$ and $R_G \approx 0.45$ (where R_G represents the electron reflection coefficient at grain boundaries and λ_F is the Fermi wavelength (for gold, $\lambda_F = 0.52$ nm)); consequently grain-boundary scattering plays a central role in the SEJ interpretation of the data. An important element in the SEJ analysis is the use of the angle-dependent reflectivity R_S proposed by Soffer [8] (equation (5) in reference [7]):

$$R_S(\theta) = \exp\left[-\left(\frac{4\pi\delta}{\lambda_F}\cos(\theta)\right)^2\right]. \tag{2}$$

From the parameter fitting of their data, SEJ arrive at $\delta \approx 0.026$ nm, about one tenth of an atomic diameter. The value of δ measured with the STM on our 70 nm film *is about 17 times larger*. Data recorded on samples measured with the STM during the preparatory experiments described in reference [5] indicate that in a continuous film prepared by thermal evaporation of gold on mica, the roughness measured on a nanometric scale is characterized by a r.m.s. amplitude δ that is comparable to the Fermi wavelength λ_F. Therefore Soffer's model leads to essentially diffuse scattering $R_S \approx 0$ in these films, except for near grazing incidence $\theta \approx \pi/2$.

The fact that the r.m.s. surface roughness δ measured on our 70 nm film turns out to be about 17 times larger than the value inferred by SEJ from fitting the temperature and the thickness dependence of their data *using a model containing five adjustable parameters* casts doubt on the validity of both the theoretical model used in fitting the resistivity data (Soffer's reflectivity and the central role assigned to grain-boundary scattering in the SEJ films), as well as on the underlying assumptions that constitute the basis for the parameter fitting of resistivity data that has dominated the literature for decades, such as a surface reflectivity and a residual resistivity (and consequently a bulk resistivity) *which are the same for films of different thickness*. The discrepancy of nearly a factor of 20 between the measured and the inferred δ reported here casts doubt on the validity of data analysis performed by fitting parameters describing the surface roughness to a set of resistivity data, unless the fitted parameters agree with the roughness measured in an independent experiment. This discrepancy underlines the need for revisiting transport measurements on thin metallic films, and the need for cross-checking the parameters characterizing the surface roughness obtained by fitting transport data, with direct measurements of the surface roughness of the films performed on a nanometric scale with a scanning probe microscope capable of atomic resolution.

We believe this to be the first report in which the temperature dependence and the thickness dependence of the resistivity predicted by a theory, that uses as input the information contained in the surface roughness measured on a nanometric scale in an *independent experiment*, agrees

approximately with the resistivity measured for a set of thin metallic films. *The theory contains no adjustable parameters*. However, since *the roughness and the resistivity were measured on different films* prepared under similar conditions of evaporation—except for the purity of the starting material—the analysis presented might be considered as evidence supporting the mSXW theory, but certainly may not be considered a proof of its validity until the surface roughness and resistivity are measured *for the same film* for several samples of different thickness.

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