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LETTER TO THE EDITOR

Surface-induced resistivity of $CoSi_2$ films and violations of Mathiessen's rule

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Abstract

We analysed the residual resistivity data for more than 40 films of $COSi_2$ reported by different groups using the available quantum theories of size effects in metal films. We found that the predictions of the model of Trivedi and Ashcroft (1988 *Phys. Rev.* B **38** 12298) of Tesanovic *et al* (1986 *Phys. Rev. Lett.* **57** 2760), and of the mSXW theory (Munoz *et al* 1999 *J. Phys. Condens. Matter* **11** L299) agree roughly with the data and with each other over the entire range of thickness 1 nm $\leqslant t \leqslant$ 110 nm, although the rms roughness amplitude needed to best describe the residual resistivity data is somewhat different for each model. All three models predict surprisingly similar values for the film resistivity ρ_F and for the surface resistivity ρ_S arising from electron–surface scattering. All three models indicate that Mathiessen's rule is violated in thin $CoSi_2$ films, that is, $\rho_F \neq \rho_S + \rho_B$, where ρ_B is the bulk resistivity. For 110 nm $< t <$ 10 nm, the resistivity of the film exceeds by some 25–55% the value dictated by Mathiessen's rule. And conversely, the apparent surface induced resistivity $\rho'_S = \rho_F - \rho_B$ estimated assuming the validity of Mathiessen's rule, exceeds by nearly one order of magnitude the true surface-induced resistivity ρ_S, except in the case of ultrathin films $t <$ 3 nm.

One of the fundamental problems in solid state physics that has attracted the attention of researchers for over 60 years, relates to the effect of electron–surface scattering on the transport properties of thin metallic and semiconducting films. A central issue is how the surface of the structure affects its electrical transport properties, when one or more of the dimensions

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characterizing the structure are comparable to or smaller than the mean free path of the charge carriers, i.e., what is known as 'size effects'.

The theoretical work concerning size effects focused for many decades on the Fuchs–Sondheimer (FS) theory, in which the electron motion is modelled by a Boltzman transport equation (BTE) and the effect of the rough surface is incorporated into the boundary conditions that must be satisfied by the electron distribution function obeying a BTE via a specularity parameter R, that represents the fraction of electrons $0 \leqslant R \leqslant 1$ that are specularly reflected upon colliding with the rough surface [1]. It is well known that this approach is inadequate for very thin, high purity samples where the film thickness t is much smaller than the bulk mean free path ℓ. On the one hand, for ultrapure thin films, the conductivity of the film is expected to exhibit a stepwise increase with increasing film thickness each time the thickness increases by half a Fermi wavelength, as a consequence of the quantization of the electronic energy levels induced by confinement of the electron gas between two parallel potential barriers. This is known as quantum size effects (QSE). Observations consistent with these expectations have been reported in Pt films evaporated onto glass slides [2]. The modelling of electron motion by a BTE with FS boundary conditions does not account for QSE. Moreover, the resistivity of ultrathin $CoSi_2$ films observed at 4 K has been reported to increase sharply as the thickness of the film decreases below 10 nm, *in a way that cannot be accounted for by the classical model no matter what reflectivity R is used in the FS formalism* [3]. On the other hand, in the limit $\ell \to \infty$ the FS conductivity diverges as $\ln(\ell/t)$, implying that when the conductivity of the film is limited only by electron–surface scattering, there is no dissipation, an unphysical result that arises from the omission of quantum effects in the classical theory.

To overcome these shortcomings, a number of quantum transport theories have been published that permit the calculation of the increase in resistivity arising from electron–surface scattering, in terms of the parameters that characterize the roughness of the surface [4–7]. One of the fundamental issues concerning size effects relates to the resistivity ρ_S induced by electron–surface scattering in the absence of electron scattering in the bulk, and to the bulk resistivity ρ_B due to electron–impurity scattering and electron–phonon scattering in the absence of electron–surface scattering. The question is whether the resistivity ρ_F measured on a film where both electron-scattering mechanisms are present (bulk scattering and surface scattering) obeys Mathiessen's rule, e.g. whether ρ_F satisfies $\rho_F = \rho_S + \rho_B$. Mathiessen's law is a powerful rule that applies to many electron-scattering mechanisms in crystalline solids.

From the point of view of theory, arguments have appeared in the literature that indicate that Mathiessen's rule is violated [5, p L830]. About a year later, the additivity of the scattering rates due to bulk scattering and to electron–surface scattering (equations (4.4) and (4.10) in [4]) was used to calculated $\sigma_S = (\rho_S)^{-1}$ and $\sigma_F = (\rho_F)^{-1}$. From the experimental point of view, the apparent film resistivity due to surface scattering $\rho_S' = \rho_F - \rho_B$ has been used as a measure of ρ_S [7–9], thereby tacitly assuming the validity of Mathiessen's rule.

In the process of extending the mSXW formalism [6] to compute the resistivity arising from electron–surface scattering in a metal film bounded by a rough fractal surface, we found evidence suggesting that Mathiessen's rule might be severely violated in thin metal films [10]. However, the evidence is indirect, for it arises out of calculations of ρ_F and ρ_S based upon the extension of the mSXW formalism to a rough fractal surface, instead of arising from an analysis of thin film resistivity data. Metallic and semiconducting thin films are today the object of intense, widespread research. Elucidating how severely electron–surface scattering and electron scattering in the bulk violate Mathiessen's rule in metal films, appears to be a fundamental question whose (unknown) answer may be of interest to a wide audience. In this letter we attempt to clarify how big the (expected) violations of Mathiessen's rule are that occur in thin metallic films, through the analysis of residual resistivity data published by

different groups on more than 40 $CoSi_2$ films, using the different quantum transport theories available.

A word of caution seems appropriate. Metal films are often made out of grains that coalesced having different crystalline orientations. Consequently, the (macroscopic) resistivity observed in a metal film arises out of different (microscopic) electron-scattering mechanisms, such as (a) electron–impurity scattering, (b) electron–phonon scattering, (c) electron scattering by the rough surface at the upper (exposed) and (d) lower surface of the film (surface in contact with the substrate), (e) electron–grain boundary scattering, (f) electron scattering by pinholes, dislocations and other defects. Existing quantum theories incorporate only contributions to the resistivity arising form electron-scattering mechanisms (a)–(c). Consequently, before comparing theoretical predictions with experimental data, the data should be carefully screened, in order to rule out or to minimize the effect of electron scattering mechanisms (d)–(f) *which are not included in any of the quantum theories available*.

In order to minimize contributions to the resistivity arising from electron–grain boundary scattering, the samples should be selected such that the lateral dimension L characterizing the grains that make up the samples, is at least one order of magnitude larger than the thickness t of the films [11]. To minimize contributions to the resistivity arising because of electron scattering at the rough surface of the substrate, the substrate itself should be a freshly cleaved crystal, such that the roughness is reduced to cleavage steps, that occur rather infrequently over the scale of distance of tens to hundreds of nanometres set by the electron mean free path [6]. To minimize contributions to the resistivity arising from defects, the metal film should be deposited on a crystalline substrate that closely matches the lattice constant of the film, and should be annealed. Fortunately, technical developments have made possible the fabrication of epitaxial films of $CoSi_2$ onto Si(111) with thickness ranging from 1 to some 110 nm. As a consequence of a small lattice mismatch (1.2%) between $CoSi_2$ and Si [3, 9, 12], and because of annealing at temperatures in the range 500 °C or higher [3, 8, 9, 12], $CoSi_2$ films exhibit grains with lateral dimensions L in the range of several hundred nanometres, as measured by transmission electron microscopy (TEM) [3, 12, 13]. Stochiometry and defect concentration are often monitored by TEM [3, 8, 9, 12, 13] and by Rutherford back scattering (RBS) [8, 12, 13]. In a study of crystallinity of a 170 nm thick $CoSi_2$ film, the ratio of $\langle 111 \rangle$ channelling to random yield measured using RBS was found to be 2% '... not only the best value reported for a silicide film, but is among the best reported for any crystalline material' [12, p 684]. Consequently the system is well suited as a testing ground of different theories of size effects in metal films, for $CoSi_2$ is known to be a metal.

Concerning the resistivity of $CoSi_2$ films grown on Si(111), there are several groups that have reported measurements of the residual resistivity of families of films of different thickness (e.g. the resistivity measured at 4 K, a temperature at which the resistivity arising from electron–phonon scattering is negligible because the phonons are frozen out). In figure 1 we display the raw data reported by Badoz *et al* [3], Duboz *et al* [8], Hensel *et al* [13] and, Henz *et al* [9]. The resistivity ratio shown in figure 2 of [3] was converted into the measured film resistivity by multiplying the resistivity ratio by the bulk resistivity $\rho_B = 23.5\ \mathrm{n\Omega\ m}$ quoted by the authors in [8]. The surface resistivity shown in figure 2 of [8] was converted into film resistivity by adding the bulk resistivity $\rho_B = 23.5\ \mathrm{n\Omega\ m}$ quoted by the authors. The ratio of residual to bulk resistivity displayed in figure 3 of [13] was converted into film resistivity by multiplying the resistivity ratio by the bulk resistivity $\rho_B = 26.0\ \mathrm{n\Omega\ m}$ quoted by the authors. The residual resistivity of the bare films displayed in figure 3 of [9] was converted into film resistivity by adding the resistivity $\rho_0 = 5.4\ \mathrm{n\Omega\ m}$ quoted by the authors.

In the calculation of the film resistivity predicted by different theories we used as transport parameters characterizing the bulk, the mean free path $\ell = 100$ nm determined by Hensel and

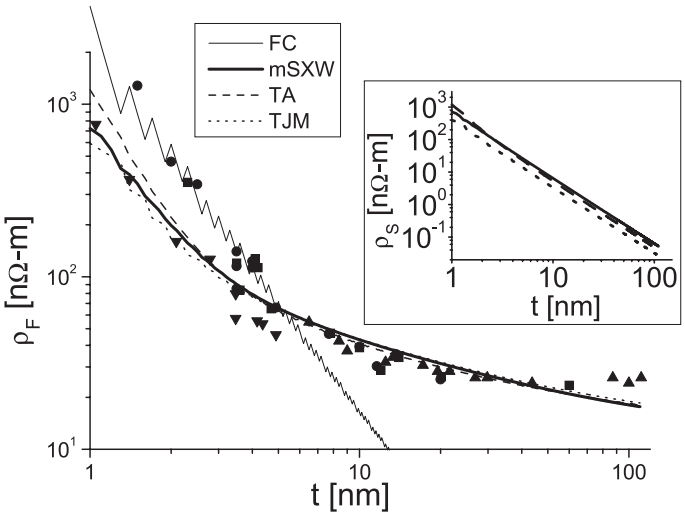

Figure 1. Film resistivity measured at 4 K plotted versus film thickness in films of $CoSi_2$. Squares: data from [3]; circles: data from [8]; triangle: data from [13]; inverted triangle: data from [9]. Dotted curve: theory of Tesanovic, Jaric and Maekawa (TJM) [5], with $\delta = 0.5$ nm. Broken curve: theory of Trivedi and Aschroft (TA) [4] with $\delta = 0.75$ nm. Thin solid curve: theory of Fishman and Calecki (FC) [7] using an exponential ACF with δ=ξ= 0.2nm. Thick solid curve: theory mSXW [6], using a Gaussian ACF with $\delta = 0.2$ nm, $\xi = 0.5$ nm. Inset: surface-induced resistivity ρ_S plotted as a function of film thickness, for each of the models TJM, TA and mSXW-Gaussian.

co-workers by measuring the magnetoresistance of a 110 nm thick film [13], and the hole density $n = 3.0 \times 10^{28}$ m^{-3} determined by Badoz and co-workers [3] by measuring the Hall constant, that turns out to be independent of film thickness. To compare theory and experiment we used a method already established, that relies on the assumption that the parameters δ, ξ (where δ stands for the rms roughness amplitude, and ξ stands for the lateral correlation length) do not depend on film thickness [2, 5, 8]. We selected for each of the theories a set of parameters (δ, ξ) that would approximately describe that data and that would lie roughly within the range of atomic dimensions, 0.20 nm $\leqslant \delta, \xi \leqslant$ 1 nm.

The surface-induced resistivity ρ_S and the thin film resistivity ρ_F corresponding to TA, was computed as the inverse of equations (4.12) and (4.13) in [4], respectively. In the case of TJM, we computed ρ_F as the inverse of σ_F given by equation (7) in [5]; ρ_S was computed as the inverse of $\sigma_S = \lim_{(\ell\to\infty)} \sigma_F$. In the case of FC, ρ_S was computed as the inverse of equation (13) in [2]. FC assume that $\rho_F = \rho_S$, since bulk scattering is ignored. In the case of the mSXW model, ρ_S was computed as the inverse of the conductivity given by equation (1) in [6], with $R[u_n] = [(1 - k_F u_n Q(u_n))/(1 + k_F u_n Q(u_n))]^2$, where $u_n = \frac{n\pi}{tk_F}$, k_F stands for the Fermi wavevector, and the self-energy $Q(u_n)$ of the electron gas is given by equations (5) and (6) of [6] in the case of a Gaussian and of an exponential representation of the height–height autocorrelation function (ACF), respectively. Within the mSXW theory, the surface-limited resistivity $\rho_S = (\sigma_S)^{-1}$ is computed as the limit $\ell \to \infty$ by expanding the right-hand side of equation (1) in [6] to first order in powers of t/ℓ. This leads to

$$\frac{\sigma_S}{\sigma_B} = \frac{3t}{4\ell}\frac{1}{X_o N_C}\sum_{n=1}^{N_C}\left(\frac{1}{u_n} - u_n\right)\frac{1 + R(u_n)}{1 - R(u_n)},$$

where

$$X_C = \left(\frac{tk_F}{\pi}\right), \qquad N_C = \text{integer part of } \left(\frac{tk_F}{\pi}\right),$$

$$X_o = \frac{3}{2}\left[1 - \frac{1}{3}\left(\frac{N_c}{X_c}\right)^2\left(1+\frac{1}{N_c}\right)\left(1+\frac{1}{2N_c}\right)\right].$$

Note that in the case of thick films (films where the number N_C of occupied subbands satisfies $N_C \gg 1$), a constant reflectivity R (e.g. $R < 1$ independent of angle) leads to a finite mean free path ℓ_S limited only by electron–surface scattering, given approximately, by $\ell_S = \frac{3}{4}t\frac{1+R}{1-R}\ln(N_C)$. This quantity, although it has *no classical analogue*, can be considered as the quantum version of equation (21) in [1], where the quantum theory has removed the logarithmic divergence $\ln(\ell/t)$ of the classical FS theory.

In figure 1 we plot the film resistivity ρ_F predicted by TA for $\delta = 0.75$ nm, by TJM for $\delta = 0.5$ nm, by mSXW using a Gaussian ACF with $\delta = 0.2$ nm and $\xi = 0.5$ nm, and by FC using an exponential ACF with $\delta = \xi = 0.2$ nm. For ultrathin films $t < 10$ nm, the predictions of the mSXW model for a Gaussian ACF exceed by 20% or less the predictions corresponding to an exponential ACF characterized by the same parameters δ and ξ; for thicker films 10 nm $< t <$ 110 nm both representations of the ACF lead to a similar ρ_F to within a few per cent. Within the FC model, the mean free path for a Gaussian ACF is double the mean free path for an exponential ACF and such a relation is independent of film thickness, therefore the predicted FC film resistivity for a Gaussian ACF is one half of that corresponding to an exponential ACF with the same parameters (δ, ξ). In the inset of figure 1 we display the surface-induced resistivity ρ_S predicted by different models. In figure 2 we plot the different $\Delta\rho/\rho_F = [\rho_F - (\rho_S + \rho_B)]/\rho_F$, *a dimensionless quantity that ought to be zero for all thickness if Mathiessen's rule is obeyed*; how much $\Delta\rho/\rho_F$ departs from zero indicates the severity of the violations of the additivity rule $\rho_F = \rho_S + \rho_B$. In the inset of figure 2 we plot the ratio between ρ_S and the *apparent surface resistivity* $\rho'_S = \rho_F - \rho_B$. Should Mathiessen's rule hold, then this ratio ought to be unity independent of film thickness; the degree to which the ratio ρ_S/ρ'_S *departs from unity reflects the severity with which Mathiessen's rule is violated*.

Although the raw data plotted in figure 1 exhibits some appreciable scatter, as might be expected for data reported by different groups, it seems clear that the data does not follow a simple power law dependence on film thickness t, as has been claimed by other authors on the basis of resistivity data covering a smaller range of thicknesses [2], The FC model predicts a resistivity that exhibits a sawtooth behaviour, it decreases sharply with increasing film thickness for certain thicknesses, as a consequence of the fact that the number of filled subbands that participate in conduction increases by one each time the thickness of the film increases by half a Fermi wavelength. This manifestation of QSE is also predicted by the other models TJM, TA and mSXW, although the decrease in resistivity predicted by the other models (each time the thickness increases by half a Fermi wavelength) is significantly less pronounced. The resistivity predicted by FC fails to level off with increasing thickness for 10 nm $< t <$ 110 nm.

Beyond these features, some of which have already been reported by other authors, these plots illustrate two new and remarkable results:

(i) *The values of* ρ_F *and* ρ_S *predicted by these three (apparently) very different models, roughly agree with each other and roughly describe the data*, considering the scatter of the raw data reported by different groups, over a thickness that changes by two orders of magnitude, although the rms roughness amplitude needed to best describe the resistivity data is somewhat different for each model,

(ii) *all three models indicate that Mathiessen's rule is severely violated in* $CoSi_2$ *films, and that the apparent surface resistivity* ρ'_S *in this system is about one order of magnitude*

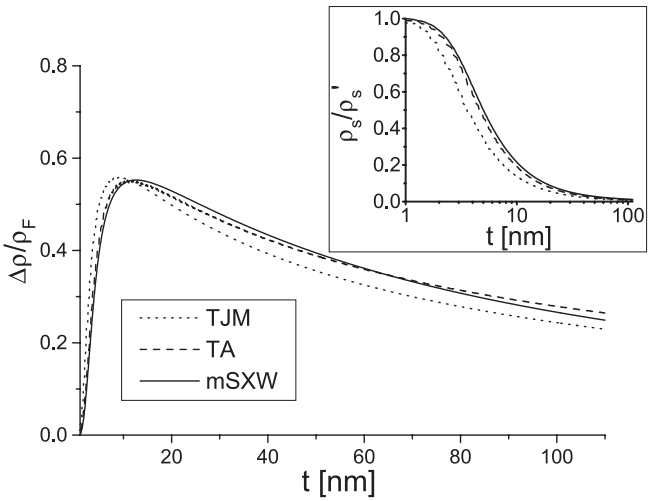

Figure 2. $\Delta\rho/\rho_F = [\rho_F - (\rho_S + \rho_B)]/\rho_F$ plotted as a function of film thickness. Dotted curve: theory of TJM, with $\delta = 0.5$ nm. Broken curve: theory of TA, with $\delta = 0.75$ nm. Solid curve: theory mSXW, using a Gaussian ACF with $\delta = 0.2$ nm, $\xi = 0.5$ nm. Inset: ratio between the surface resistivity ρ_S and the apparent surface-induced resistivity $\rho'_S = \rho_F - \rho_B$ predicted by different models, plotted versus film thickness.

larger than the surface resistivity ρ_S predicted by any of the models, except in the case of ultrathin films which are only a few nanometres thick.

As pointed out above, the fact that Mathiessen's rule is violated when the scattering mechanisms involved are electron–surface scattering and electron scattering in the bulk, has been known for over a decade. However, to our knowledge, this letter contains the first *quantitative* estimation of how severe such a violation is. The results reported here are consistent with published evidence in thin ($t < 10$ nm) $CoSi_2$ films, figure 1 of [9], that contradicts the additivity rule. Such violations become noticeable when the temperature dependent film resistivity $\rho_F(T)$ ($4\ \mathrm{K} \leqslant T \leqslant 300\ \mathrm{K}$) measured in films of different thickness, are no longer related to each other by means of a constant (temperature-independent) additive term identified as residual resistivity in the Bloch–Grunheisen model. They become apparent as the thickness of the film decreases below 10 nm and $\rho_F(4)$ becomes comparable to the resistivity ρ_{BG} expected from electron–phonon scattering at 300 K, $\rho_{BG}(300) \approx 125\ \mathrm{n\Omega\,m}$ (figure 2 of [13]).

The question naturally arises: why is Mathiessien's rule violated? In the case where electron motion is described through a classical model such as that based upon BTE, the different scattering mechanisms appearing in the collision operator in BTE might be written as the sum of the different collision operators corresponding to each scattering mechanism acting alone. The resistivity arising from each electron scattering mechanism is proportional to the matrix element representing the transition rate from the initial to the final electronic state. However, writing the collision operator within the simplest approximation, the relaxation time approximation, might lead to a relaxation time τ that turns out to be momentum dependent $\tau(\boldsymbol{k})$. In such a case, the presence of two electron scattering mechanisms, each characterized by relaxation times $\tau_1(\boldsymbol{k})$ and $\tau_2(\boldsymbol{k})$, leads to a total resistivity that is proportional to the average of the inverse of the relaxation time $\langle 1/\tau\rangle = \langle 1/\tau_1\rangle + \langle 1/\tau_2\rangle$, that need not be equal to the sum $1/\langle\tau_1\rangle + 1/\langle\tau_2\rangle$, as would be required for Mathiessen's rule to hold [14].

When a quantum transport theory is employed to describe electron scattering by a rough surface, the quantum description of charge transport assigns an important role to the identity of the states occupied by the electron, as is sharply manifested in QSE. Such an important role is also reflected in the fact that, regardless of the different approximations involved, *all* quantum theories of size effects lead to a resistivity that depends explicitly upon the subband index n that identifies the quantum states participating in the conduction process. This is in contrast to bulk scattering, where the fact that the metallic sample takes the form of a thin film (and the electron momentum perpendicular to the film is quantized as a consequence of the confinement of the electron gas between two parallel potential barriers) is irrelevant, and hence the corresponding resistivity is independent of the identity of the electron states. As a result, the total resistivity is no longer the sum of the resistivities arising from each of the different channels contributing to charge transport, *neither is the resistivity given by the sum of the different resistivities arising from each electron-scattering mechanism acting alone*. As pointed out in [4], as a consequence of the fact that the electronic states are quantized and because of the importance of the identity of the electron states within the quantum description of charge transport, the additivity of the scattering rates (stemming from the statistical independence between averaging over impurities or over the phonon population in the case of bulk scattering, and averaging over surface roughness configurations in the case of surface scattering) no longer leads to the additivity of the corresponding resistivities, consequently Mathiessen's rule no longer holds.

The preceding argument implies that, in a thin metallic film, we should expect Mathiessen's rule to be violated, as a consequence of the fact that the quantization of the electronic states within the film plays a dominant role in the surface resistivity ρ_S. Then the proper question to ask seems to be the opposite: why (under what conditions) should Mathiessen's rule be valid in a metallic film, if the surface resistivity ρ_S is dominated by the quantized electron states participating in conduction? The answer is, of course, that we might expect the validity of the additivity rule to be recovered (to within a few per cent), *when counting over quantized states becomes irrelevant*, that is, *for films that are thick enough to contain hundreds of electron states*, films whose thickness is in the range of, at least, hundreds of nanometres. The tendency of $\Delta\rho/\rho_F$ to decrease with increasing thickness t, reaching a level around 20% for $t > 100$ nm displayed on figure 2, supports this conclusion.

In summary, we have analysed the residual resistivity data of more than 40 epitaxial films of $CoSi_2$ reported by different groups using different quantum theories of size effects in metal films. We found that the predictions of the model of Trivedi and Aschroft [4], of Tesanovic *et al* [5] and of the mSXW theory [6] agree roughly with the data and with each other over the entire range of thickness 1 nm $\leqslant t \leqslant$ 110 nm, although the rms roughness amplitude needed to best describe the residual resistivity data is somewhat different for each model. All three models predict surprisingly similar values for the resistivity ρ_S arising from electron–surface scattering, and they all indicate that Mathiessen's rule is severely violated in $CoSi_2$ films.

The results reported here suggest the interesting conclusion that, regardless of which theoretical model we choose to describe electron–surface scattering, *Mathiessen's rule is severely violated in thin metallic films*. They also suggest that, for the surface and bulk resistivity to be additive to within an error of the order of a few per cent, *the metallic film must be thick enough to contain hundreds of electron states, which means that the film thickness must be, at least, in the range of a few hundred nanometres*.

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