Surface-Induced Resistivity of Ultrathin Metallic Films: A Limit Law

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(Received 20 June 1988)

We study the variations of the electrical conductivity σ with thickness d of ultrathin metallic films. In the limit $\xi k_F \ll 1$, where ξ is the correlation length describing the film surface roughness and k_F is the electron Fermi wave vector, we show that σ follows a universal law, $\sigma \sim d^{2.3}$, independent of any adjustable parameter. This law accounts for recent experimental data on $CoSi_2$ down to $d = 10$ Å. Moreover, the measurements of σ are well fitted when we introduce into its theoretical expression the values of the surface roughness parameters recently estimated from electron microscopy.

PACS numbers: 73.60.Aq, 72.10.Fk, 73.50.Dn

The problem of the influence of surfaces on the electrical conductivity of thin metallic films was first tackled by Thomson.[1] Later, Fuchs[2] and Sondheimer[3] improved Thomson's formulation. All these theories, which were reviewed by Ziman,[4] are developed in a classical framework. Following ideas on surface roughness encountered in the work of Ziman,[5] Prange and Nee[6] gave the first quantum treatment of the surface-limited resistivity. This theory was applied to semiconductors[7-9] for which molecular-beam epitaxy allows one to obtain quantum wells thinner than 100 Å. The same technique has been applied to prepare metallic samples of $CoSi_2$ as thin as 60 Å; Hensel *et al.*[10] measured conductivity and fitted their results with a quantum surface scattering theory put forward by Tesanovic, Jaric, and Maekawa.[11] More recently, Badoz *et al.* and others have extended the conductivity measurement in $CoSi_2$ films to thicknesses down to 10 Å.[12-15] However, this raises some new theoretical questions because for such small thicknesses, the theory of Ref. 11 gives a variation too smooth to account for the new conductivity data.

First of all, we note that the number of subbands occupied by the electron gas is much higher in a metallic film than in a semiconducting quantum well. In the later case, due to the small density of charge carriers, only one to two subbands play a role; conversely, in $CoSi_2$ samples considered hereafter, the number N of occupied subbands can be calculated by the formula $N \approx (3n/\pi)^{1/3} d$, where n is the carrier concentration. For $n = 3\times 10^{22}$ cm^{-3}, $N \sim 0.3d$ with the thickness d expressed in Å; thus in the range $10 < d < 200$ Å, N changes from 3 to 60. High values of N imply significant intersubband electron transitions; their influence on conductivity can be studied by the same techniques that Siggia and Kwok[16] used to take account of intervalley scattering in silicon. The aim of this Letter is to generalize the Prange-Nee formulation of scattering by surface roughness, to adapt the Siggia-Kwok method to the case of metals, and to explain the experimental data on $CoSi_2$ as published in Refs. 12-15. In particular, we will point out an entirely general law for the dependence of σ on thickness d.

Expression of the conductivity.— In an ideal film with thickness d, the surfaces are perfect planes, perpendicular to the z axis and defined by the equations $z = \pm \frac{1}{2} d$. In that case, the Hamiltonian of a charge carrier (in $CoSi_2$ the carriers are holes) may be written

$$H_0 = p^2/2m + V\Upsilon(z - \tfrac{1}{2}d) + V_-\Upsilon(-z - \tfrac{1}{2}d), \quad (1)$$

where $\Upsilon(z)$ is the step function and V (V_-) is the potential height outside the well for $z > \frac{1}{2}d$ ($z < -\frac{1}{2}d$). Eigenfunctions and eigenenergies of H_0 are simply

$$\langle \mathbf{r} \mid \nu\mathbf{k}\rangle = S^{-1/2} e^{i\mathbf{k}\cdot\boldsymbol{\rho}} \phi_\nu(z), \quad E_{\nu\mathbf{k}} = E_\nu + \hbar^2 k^2/2m. \quad (2)$$

S is the are of the film surfaces, ν is the subband index, and $\boldsymbol{\rho}$ and $\mathbf{k}$ are two-dimensional vectors in direct and reciprocal spaces, respectively. If one of the two surfaces is not perfect, for example the one near $z = \frac{1}{2}d$, its equation becomes $z = \frac{1}{2}d + f(\boldsymbol{\rho})$ with $f(\boldsymbol{\rho}) \ll d$. (We can take account easily of both surfaces, but in order to simplify what follows we assume only one nonideal surface to exist.) The Hamiltonian is now changed into $H_0 + U$, where

$$U = V\{\Upsilon[z - \tfrac{1}{2}d - f(\boldsymbol{\rho})] - \Upsilon(z - \tfrac{1}{2}d)\} \approx -Vf(\boldsymbol{\rho})\delta(z - \tfrac{1}{2}d). \quad (3)$$

Calculations will be made to lowest order in the surface roughness $f(\boldsymbol{\rho})$.

The general expression of the conductivity of a two-dimensional degenerate gas, the charge carriers of which are

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elastically scattered by the potential U, is given (for example, in Refs. 17 or 18) in the form

$$\sigma=\frac{Sm^2e^2}{\pi^2\hbar^6 d}\sum_{\nu=1}^{N}\sum_{\nu'=1}^{N}(E_F-E_\nu)(E_F-E_{\nu'})[C^{-1}(E_F)]_{\nu\nu'}\,, \tag{4}$$

where E_F is the Fermi energy and N is the number of subbands with minimum E_ν less than E_F. $C^{-1}(E)$ is the inverse matrix of $C(E)$ defined by its elements

$$[C(E)]_{\nu\nu'}=\frac{2\pi}{\hbar}\sum_{\mathbf{k}}\sum_{\mathbf{k}'}\Big\{\delta_{\nu,\nu'}\sum_{\mu}|\langle\nu\mathbf{k}|U|\mu\mathbf{k}'\rangle|^2k^2\delta(E-E_{\nu\mathbf{k}})\delta(E-E_{\mu\mathbf{k}'}) - |\langle\nu\mathbf{k}|U|\nu'\mathbf{k}'\rangle|^2\mathbf{k}\cdot\mathbf{k}'\delta(E-E_{\nu\mathbf{k}})\delta(E-E_{\nu'\mathbf{k}'})\Big\}. \tag{5}$$

When U is given by Eq. (3), we get

$$S|\langle\nu\mathbf{k}|U|\nu'\mathbf{k}'\rangle|^2=A_\nu A_{\nu'}\int_S d^2\rho\, e^{i(\mathbf{k}'-\mathbf{k})\cdot\rho}\langle f(\rho')f(\rho'+\rho)\rangle\,. \tag{6}$$

This last expression involves parameters $A_\nu=V\phi_\nu^2(\frac{1}{2}d)$ which are easily calculated from the characteristics of the ideal film and the Fourier transform of the autocorrelation function,

$$\langle f(\rho')f(\rho'+\rho)\rangle=\frac{1}{S}\int_S d^2\rho' f(\rho)f(\rho+\rho')\,, \tag{7}$$

which characterizes the surface roughness. If Δ describes the root mean square of the height of the bumps on the surface and ξ is the roughness correlation length, the autocorrelation function for an isotropic surface can be written

$$\langle f(\rho')f(\rho'+\rho)\rangle=\Delta^2G(\rho/\xi)\,, \tag{8}$$

where the function G will be clarified hereafter. Now, defining $F(q)$ as the Fourier transform of $G(\rho)$, we get

$$S|\langle\nu\mathbf{k}|U|\nu'\mathbf{k}'\rangle|^2=A_\nu A_{\nu'}\Delta^2\xi^2F(\xi|\mathbf{k}-\mathbf{k}'|)\,. \tag{9}$$

Finally the expression for the conductivity reduces to

$$\sigma=\frac{e^2d^5}{4\pi^6\hbar\Delta^2}\sum_{\nu=1}^{N}\sum_{\nu'=1}^{N}k_\nu^2k_{\nu'}^2(D^{-1})_{\nu\nu'}\,; \tag{10}$$

k_ν is the Fermi wave vector of subband ν, $k_\nu=[(2m/\hbar^2)(E_F-E_\nu)]^{1/2}$, and D^{-1} is the inverse matrix of D defined by its elements

$$D_{\nu\nu'}=\frac{d^6\xi^2m^2}{4\pi^6\hbar^4}A_\nu\int_0^{2\pi}d\theta\left[\delta_{\nu,\nu'}k_\nu^2\sum_{\mu=1}^{N}A_\mu F(\xi k_{\nu\mu})-A_{\nu'}k_\nu k_{\nu'}\cos\theta F(\xi k_{\nu\nu'})\right], \tag{11}$$

with

$$k_{\nu\nu'}=(k_\nu^2+k_{\nu'}^2-2k_\nu k_{\nu'}\cos\theta)^{1/2}.$$

For the chosen surface roughness model, the two equations (10) and (11) give an exact expression of the conductivity (in the Born approximation). As the A_ν are proportional to m^{-1}, $D_{\nu\nu'}$ and σ are finally m independent. This point is important because it proves that it is not necessary to know the carrier mass for fitting conductivity measurements through formula (10).

Limiting expressions.— These results can be simplified in the special case of physical interest where the correlation length ξ is much less than k_1^{-1}, where k_1 is the largest of the Fermi wave vectors k_ν. In the limit $\xi k_1\ll 1$, we have $\xi k_{\nu\nu'}\ll 1$ for all $\nu,\nu'\le N$ and we get from Eq. (11)

$$D_{\nu\nu'}\approx\delta_{\nu,\nu'}\frac{d^6\xi^2m^2}{2\pi^5\hbar^4}F(0)k_\nu^2A_\nu\sum_{\mu=1}^{N}A_\mu\,. \tag{12}$$

Now D is a diagonal matrix whose inverse is trivial. Moreover, let us assume that the carrier wave functions are almost entirely confined inside the film; then we can take V as infinite as it is easy to verify that

$$\lim_{V\to\infty}A_\nu=\frac{\hbar^2\pi^2\nu^2}{md^3}\,.$$

Finally, in these limits, we can obtain for the conductivity

$$\sigma\approx\frac{e^2}{\hbar}\frac{d^5}{2\pi^6\Delta^2\xi^2}\frac{\pi}{F(0)}\frac{6}{N(N+1)(2N+1)}\sum_{\nu=1}^{N}\frac{k_\nu^2}{\nu^2}\,. \tag{13}$$

This expression is the key to understanding the variations of σ with d, providing that we do not forget that N and k_ν depend on the thickness d.

We remark that when $\xi k_1\ll 1$ the variations of σ with

d are not sensitive to the exact form of the autorcorrelation function that characterizes the surface roughness and are independent of the correlation length ξ. At fixed carrier density, when d increases at constant N, σ increases. If d increases more, the number of occupied subbands goes from N to $N+1$, which leads to a discontinuous decrease in σ; but this discontinuity is no larger than the dispersion of the experimental data.

Let us evaluate σ for a semiconducting film for which $N=1$; then $k_1^2=2\pi nd$, where n is the density of carriers and we recover the well-known result[9] $\sigma \sim d^6$. In the other extreme case $N \gg 1$, valid for metallic films, we would obtain

$$\sigma \sim d^2 \left[1-\frac{6}{(3n\pi^5)^{1/3}}\frac{1}{d}\right]. \tag{14}$$

This last expression, for $10 < d < 200$ Å and $n \sim 3\times10^{22}$ cm^{-3}, is equivalent to $\sigma \sim d^{2.1}$. In conclusion, the limiting expression (13) of σ shows that $\sigma \sim d^{\alpha}$ with α decreasing from 6 when $N=1$ to 2.1 when $N \gg 1$.

We are now able to compare these limiting results with the experimental law $\sigma \sim d^{2.3}$ found for $CoSi_2$ and reported in Fig. 1, in the range $10 < d < 200$ Å. The exponent $\alpha=2.3$ is well inside the two theoretical limits: $\alpha_{min}=2.1$ and $\alpha_{max}=6$. It indicates that N is obviously higher than 1 but not high enough for expression (14) to fit the data for $CoSi_2$. However, these limits were obtained with the initial assumption $\xi k_1 \ll 1$ which leads to a dependence of σ on d that is independent of the correlation length ξ. In order to check the influence of ξ on α we return to the exact Eqs. (10) and (11) and, at the same time, we will consider the magnitude of the conductivity.

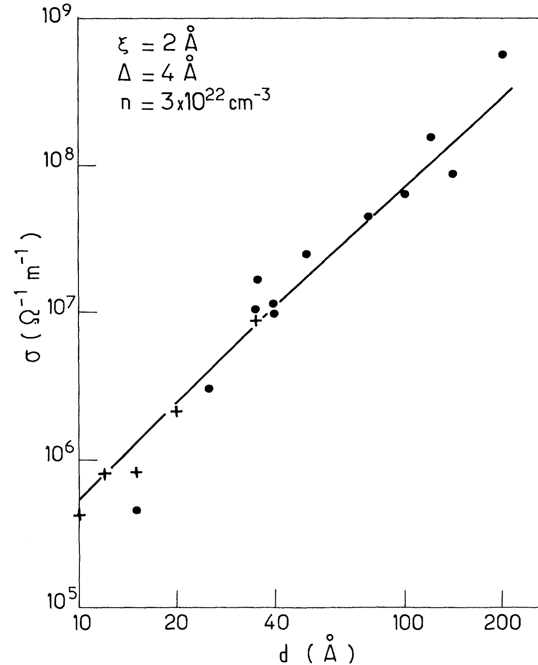

FIG. 1. Low-temperature inverse residual resistivity $\sigma(d) = [\rho(d)-\rho(\infty)]^{-1}$ as a function of the film thickness d for two sets of samples. Points indicate films obtained (as explained in Ref. 19) by solid-phase epitaxy, while crosses refer to films realized by codeposition of Co and Si (see Ref. 15). The solid line is the best fit between data and theory.

Influence of λ *and magnitude of* σ.—In order to proceed we specify the function G occurring in the expression (8) of the autocorrelation function. We make the same choice as in Ref. 6, often used in semiconductors: $G(\rho/\xi)=\exp(-\rho^2/\xi^2)$. The matrix elements $D_{\nu\nu'}$ defined by Eq. (12) are easily expressed:

$$D_{\nu\nu'}=\tfrac{1}{2}\xi^2\nu^2k_\nu\left[\delta_{\nu,\nu'}k_\nu\sum_{\mu=1}^{N}\mu^2\exp[-\tfrac{1}{4}\xi^2(k_\nu^2+k_\mu^2)]I_0(\tfrac{1}{2}\xi^2k_\nu k_\mu)-\nu'^2k_{\nu'}\exp[-\tfrac{1}{4}\xi^2(k_\nu^2+k_{\nu'}^2)]I_1(\tfrac{1}{2}\xi^2k_\nu k_{\nu'})\right]. \tag{15}$$

$I_0(x)$ and $I_1(x)$ are modified Bessel functions.

By numerical computation we get the inverse matrix D^{-1} and introduce it into Eq. (10) for σ. Finally, we find the following: (i) At fixed ξ the ratio $s=\log(\sigma/\sigma_0)/\log(d/d_0)$ is roughly constant for the entire range $10 < d < 200$ Å. (ii) s is practically independent of ξ in the range $0 \lesssim \xi \lesssim k_1^{-1}$. We already knew that this was exact for $\xi \ll k_1^{-1}$ with s equal to 2.3; the numerical calculations show that it remains true until $\xi \approx k_1^{-1}$. (iii) For $\xi \gg k_1^{-1}$, s becomes lower than 2.3 and we cannot expect to fit the experimental data on $CoSi_2$. In conclusion, the experimental variation of σ vs d in $CoSi_2$, as presented in Fig. 1, is in good agreement with the limiting law (13).

We continue with this limiting law to check the magnitude of σ. From Eq. (13) and the data of Fig. 1, we can only determine the product $\Delta^2\xi^2F(0)$. The value of $F(0)$ depends on the choice of the function G. If we take[6] $G(\rho/\xi)=\exp(-\rho^2/\xi^2)$, we get $F(0)=\pi$. If we had chosen $G(\rho/\xi)=\exp(-\rho/\xi)$, we would have obtained $F(0)=2\pi$. Other trial functions of physical interest give $F(0)=\beta\pi$, where β is a factor of several times unity. This does not change the magnitude of σ drastically. Thus, we proceed with the Prange-Nee model and we recall that a condition for the validity of Eq. (13) is $\xi \lesssim k_1^{-1}$, where k_1^{-1} does not exceed a few angstroms. This value is very low: On this point, it is worthwhile to notice that the two kinds of samples used in the experiments, the results of which are reported in Fig. 1, differ strongly in their long-range roughness (see details in Ref. 19), without measurable influence on conductivity. The results on $CoSi_2$ reported in Ref. 14 suggest that the

surface profile is atomically rough, which is consistent with $\xi = 2$ Å. Then, from the magnitude of σ, we infer that $\Delta \approx 4$ Å; this value is almost the same as that found in Ref. 14, where the authors believe that the free surface could have "a rms roughness amplitude perhaps as large as 5 nm." Finally, in $CoSi_2$ films, for the thickness between 10 and 200 Å, expression (13) correctly predicts the variation of σ vs d; physically reasonable values of surface roughness parameters such as $\xi \approx 2$ Å and $\Delta \approx 3$ Å give a correct order of magnitude for σ.

We stress in closing that in order to study the conductivity of a metallic film as a function of its thickness d, it suffices to combine the ideas of Prange and Nee on surface roughness and a complete treatment of the system of subbands occupied by carriers. In the limit of very small correlation length, a law of variation of σ vs d is obtained for an arbitrary roughness description and without any adjustable parameters. This theory fits quite well the low-temperature conductivity measurements on $CoSi_2$ films; moreover, the surface roughness parameters recently estimated from electron microscopy allow prediction of the correct order of magnitude of the conductivity limited by surface scattering.

We wish to express our sincere thanks to P. A. Badoz, J. Lajzerowicz, J. C. Pfister, R. Rammal, R. Romestain, and E. Rosencher for stimulating discussions. Laboratoire de Spectrométrie Physique and Groupe de Physique des Solides de l'Ecole Normale Supérieure are laboratories associés au Centre National de la Recherche Scientifique.

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