Quantum size effect in thin metal films

G. Govindaraj and V. Devanathan
Department of Nuclear Physics, University of Madras, Guindy Campus, Madras 600 025, Tamil Nadu, India
(Received 8 January 1986; revised manuscript received 27 May 1986)

The quantum size effect for electrical conductivity in thin metal films is studied using the Kubo formalism. An expression for the electrical conductivity is obtained for the scattering of electrons by randomly distributed impurity centers represented by a screened Coulomb potential. It is shown that the electrical conductivity has an oscillatory dependence on the thickness. The period of oscillation calculated for thin aluminum and copper films is in good agreement with the results Sandomirskii obtained for semimetals.

The quantum size effect (QSE) is the oscillatory dependence of the thermodynamic and kinetic characteristics of a thin film on thickness, and it occurs when the de Broglie wavelength becomes comparable to the film thickness. The QSE arises from the quantization of the electron quasimomentum. Hitherto a number of theoretical attempts have been made to study the QSE in thin films.[1-16] The QSE and its practical applications have been discussed in a review article by Elinson, Volkov, Lutskii, and Pinsker.[1] These authors have indicated that the QSE is manifested in the microscopic characteristics of the carrier gas in films, such as the thermodynamical coefficients, the kinetic coefficients, optical properties, and their dependence on the film thickness. In the case of the degenerate gas, these quantities are oscillatory, while in the case of nondegenerate gas, a monotonically increasing or decreasing behavior with respect to changes in film thickness may be observed. Larson[2] has reviewed the classical and the quantum size effects and their relationship to the transport coefficient in metals.

Even though the general condition for the observation of the QSE is known for thin films, there is no explicit calculation for the same in the case of metallic thin films. In this paper we study the QSE in thin metal films using the Kubo formalism[17] by considering scattering of electrons by randomly distributed static impurity centers with a screened Coulomb potential. An expression for electrical conductivity is obtained, and it is found that in quantum limit the conductivity becomes an oscillatory function of film thickness. The periods of oscillation for aluminum and copper thin films are calculated and are found to be one half of the de Broglie wavelength. These features are in agreement with the results of Sandomirskii[3] who has studied the QSE in semimetal films, solving the Boltzmann transport equation with the δ potential for randomly distributed static impurities.

Let us consider a metallic thin film of infinite length L along the x,y directions and a thickness d in the z direction ($-\frac{1}{2}d \le z \le \frac{1}{2}d$). In accordance with the assumed model, the single-particle wave functions and the energy spectrum are

$$\psi^{(r)}_{\mathbf{K},m} = \begin{cases} \left(\frac{2}{L^2 d}\right)^{1/2} \sin\left(\frac{m\pi z}{d}\right) \exp(i\mathbf{K}\cdot\boldsymbol{\rho}), & \text{if } m \text{ is even} \\ \left(\frac{2}{L^2 d}\right)^{1/2} \cos\left(\frac{m\pi z}{d}\right) \exp(i\mathbf{K}\cdot\boldsymbol{\rho}), & \text{if } m \text{ is odd} \end{cases} , \tag{1}$$

and

$$E_{\mathbf{K},m} = \tfrac{1}{2}[K^2 + (m\pi/d)^2] , \tag{2}$$

where m is any positive integer, $\boldsymbol{\rho} = x\hat{\mathbf{i}} + y\hat{\mathbf{j}}$, and $\mathbf{K}$ is a two-dimensional vector

$$K_x = \pm\frac{2\pi n_1}{L}, \quad K_y = \pm\frac{2\pi n_2}{L} .$$

We use atomic units ($e=1$, $\hbar=1$, $m_e=1$) throughout the calculations. The equilibrium electron concentration at $\mathbf{r}$ in the $\mathbf{K},m$ state at temperature T is given by

$$F^0_{\mathbf{K},m}(\mathbf{r}) = \psi_{\mathbf{K},m}(\mathbf{r})\psi_{\mathbf{K},m}(\mathbf{r}) f^0_{\mathbf{K},m} , \tag{3}$$

where

$$f^0_{\mathbf{K},m} = \{\exp[(E-\mu)/kT]+1\}^{-1}, \quad E = E_{\mathbf{K},m} , \tag{4}$$

and μ is the chemical potential denoting the Fermi energy E_F of thin film at absolute zero.

The density of states $D(E)$ for thin film is given by

$$D(E) = D_0(E)(E/E_n)^{1/2} ,$$

where $D_0(E) = L^2/\pi$, $E_n = \frac{1}{2}(n\pi/d)^2$, and $(E/E_n)^{1/2}$ is the integer part of $(E/E_n)^{1/2}$. The density of states $D(E)$ is discontinuous for energies corresponding to the bottom of each subband characterized by quantum number n.[9]

Let us suppose that the electrons in the film are free except for their interaction with N randomly distributed static impurities represented by screened Coulomb potential $V_s(\mathbf{r})$. Such a system in the presence of an external field

E_0 can be described by a one-electron Hamiltonian, which we write as follows:

$$H = H_1 + H_{ef} , \tag{5}$$

$$H_1 = H_0 + V_s , \tag{6a}$$

$$H_0 = P^2/2 , \tag{6b}$$

$$v_s(\mathbf{r}) = \sum_{j=1}^{N} V(\mathbf{r}-\mathbf{R}_j),\ v_s(\mathbf{r})\,|_{R_j=0} = q_0 \frac{e^{-k_0 r}}{r} , \tag{7}$$

$$H_{ef} = -\mathbf{E}^0 \cdot \mathbf{r} , \tag{8}$$

where H_0 is the kinetic energy of the electron, v_s is the potential energy due to N impurities at positions $\mathbf{R}_j$, q_0 is the charge of the impurity atom, k_0 is the screening length, and $\mathbf{E}^0$ is the applied external electric field.

It is known that such a system can be generally described by a density operator ρ, which satisfies the Liouville equation of motion. Restricting our consideration to linear terms in the electric field we can write

$$\rho(t) = \rho_0 + \rho_1(t) , \tag{9}$$

where ρ_0 is the density operator in the absence of the electric field. If the system is homogeneous with respect to the temperature and chemical potential [$T(\mathbf{r})$ and $\mu(\mathbf{r})$ being constants], the density operator ρ_0 implies the equilibrium distribution function given by Eq. (3). Using the Kubo formalism an expression for the change in distribution function $\rho_1(t)$ is obtained and it is given by

$$\rho_1(t) = -i \int_0^{\infty} e^{-iH_1\eta} [F^0_{\mathbf{K},m}, \mathbf{E}^0 \cdot \mathbf{r}] e^{iH_1\eta} d\eta . \tag{10}$$

The mean value of the current density is given by

$$\langle J_\alpha \rangle = -\mathrm{Tr}(\rho_1 V_\alpha) . \tag{11}$$

We know that the current density $\langle J_\alpha \rangle$ is given by

$$\langle J_\alpha \rangle = \sigma_{\alpha\beta} E^0_\beta . \tag{12}$$

From Eqs. (10), (11), and (12), we have the conductivity tensor $\sigma_{\alpha\beta}$ equal to

$$\sigma_{\alpha\beta} = \int_0^{\infty} \sum_{\mathbf{K},m} \langle \mathbf{K},m \,|\, e^{iH_1\eta} V_\alpha e^{-iH_1\eta} [F^0_{\mathbf{K},m}, r_\beta] \,|\, \mathbf{K},m \rangle d\eta , \tag{13}$$

where V_α is the velocity operator. The commutator $[F^0_{\mathbf{K},m}, r_\beta]$ can be replaced by[18]

$$-i \frac{\partial F^0_{\mathbf{K},m}}{\partial P_\beta} = -i (F^0_{\mathbf{K},m})' V_\beta ,$$

where $(F^0_{\mathbf{K},m})'$ is the energy derivative. To calculate the conductivity to lowest order in the scattering potential, we can write

$$\langle \mathbf{K},m \,|\, F^0_{\mathbf{K},m}(H_1) \,|\, \mathbf{K},m \rangle = \delta_{\mathbf{K}\mathbf{K}'} \delta_{mm'} [F^0_{\mathbf{K},m}(E_{\mathbf{K},m})]' , \tag{14}$$

and Eq. (13) becomes

$$\sigma_{\alpha\beta} = \int_0^{\infty} \sum_{\mathbf{K},m} \langle \mathbf{K},m \,|\, e^{iH_1\eta} V_\alpha e^{-iH_1\eta} \,|\, \mathbf{K}_m \rangle V_\beta [F^0_{\mathbf{K},m}(E_{\mathbf{K},m})]' d\eta . \tag{15}$$

To calculate the matrix element in the integrand, we use the well known Wigner-Weisskoff damping theoretical approximation[10]

$$e^{\pm iH_1\eta} \,|\, \mathbf{K},m \rangle = e^{\pm E_{\mathbf{K},m}\eta - \Gamma_{\mathbf{K},m}\eta} \,|\, \mathbf{K},m \rangle , \tag{16}$$

where

$$\Gamma_{\mathbf{K},m} = 2\pi \sum_{\mathbf{K},m} |\langle \mathbf{K}'m' \,|\, V_s(\mathbf{r}) \,|\, \mathbf{K},m \rangle|^2 \delta(E_{\mathbf{K}',m'} - E_{\mathbf{K},m}) .$$

$V_s(\mathbf{r})$ is given by Eq. (7). The conductivity tensor $\sigma_{\alpha\beta}$ is given by

$$\sigma_{\alpha\beta} = \sum_{\mathbf{K},m} \frac{1}{2\Gamma_{\mathbf{K},m}} V_\alpha V_\beta \frac{\partial F^0_{\mathbf{K},m}}{\partial E} . \tag{17}$$

The procedure used by Abrikosov[19] with suitable modification for thin films is followed to obtain the expression for $\Gamma_{\mathbf{K},m}$. One can write, after some algebra[20]

$$\Gamma_{\mathbf{K},m} = \frac{16\pi^3 q_0^2 N_c}{L^2 d} \sum_{\mathbf{K}',m'} \left[\frac{1}{(B+B_1)^2} + \frac{1}{(B+B_2)^2} \right] , \tag{18}$$

where $B = |\mathbf{K}-\mathbf{K}'|^2$, $B_1 = k_0^2 + (m-m')^2\pi^2/d^2$, $B_2 = k_0^2 + (m+m')^2\pi^2/d^2$ and N_c is the concentration of impurity atoms. Transforming the sum over $\mathbf{K}',m'$ into an integral, we get

$$\Gamma_{\mathbf{K},m} = \frac{4\pi q_0^2 N_c}{d} \int_{1/2(\pi/d)^2}^{E_F} \int_{K'_{\min}}^{K'_{\max}} \int_0^{2\pi} \left[\frac{1}{(B+C_1+C_2)^2} + \frac{1}{(B+C_1-C_2)^2} \right] \delta(E_{\mathbf{K}',m'} - E_{\mathbf{K},m}) \times \left[\delta\left\{ \left[E' - \frac{K'^2}{2} - \frac{1}{2}\left[\frac{\pi}{d}\right] \right]^2 \right\} + \delta\left\{ \left[E' - \frac{K'^2}{2} - \frac{1}{2}\left[\frac{2\pi}{d}\right]^2 \right] \right\} + \cdots + \right] d\phi' K' dK' dE' , \tag{19}$$

where

$$C_1 = k_0^2 + (2E - K^2) + (2E' - K'^2) ,$$

$$C_2 = 2(2E - K^2)^{1/2}(2E' - K'^2)^{1/2} ,$$

and ϕ' is the angle between $\mathbf{K}'$ and $\mathbf{K}$. The values of $K'_{\max}$ and $K'_{\min}$ are fixed using Eq. (2):

$$K'_{\max} = \left[2\left(E' - \frac{\pi^2}{2d^2} \right) \right]^{1/2} , \quad K'_{\min} = [2(E'-I)]^{1/2} ,$$

where I is the maximum integer square multiple of $\pi^2/2d^2$ that can be contained in E'.

On evaluation of the integral, one gets

$$\Gamma_{K,m}=\frac{4\pi^2 q_0^2 N_c}{d}\sum_{m'=1}^{m'_{\max}}\left[\frac{P}{(P^2-4QR)^{3/2}}+\frac{S}{(S^2-4QR)^{3/2}}\right], \tag{20}$$

where

$$P=k_0^2+4E_F+2mm'\pi^2/d^2,\ Q=2E_F-(m\pi/d)^2,$$

$$R=2E_F-(m'\pi/d)^2,\ S=k_0^2+4E_F-2mm'\pi^2/d^2,$$

and $(m+m')$ must always be even. Using Eqs. (20) and (17) and after averaging over z, one can obtain the following expression for the conductivity

$$\sigma_{aa}=\frac{1}{8\pi^3 N_c q_0^2}\sum_{m=1}^{m_{\max}} G^{-1}[2E_F-(m\pi/d)^2], \tag{21}$$

where

$$G=\sum_{m'=1}^{m'_{\max}}\left[\frac{P}{(P^2-4QR)^{3/2}}+\frac{S}{(S^2-4QR)^{3/2}}\right].$$

The QSE on the electrical conductivity σ_{aa} is studied using Eq. (21). The dependence of σ_{aa} on d is shown in Fig. 1 for aluminum and copper thin films. As seen from Fig. 1, σ_{aa} oscillates with thickness d, and the amplitude of oscillation decreases with an increase of thickness and becomes constant at large thickness. The de Broglie wavelengths for aluminum and copper films are found to be 6.72 and 8.78 a.u., respectively. From Fig. 1 the periods of oscillation for aluminum and copper thin films are found to be 3.56 and 4.32 a.u., respectively. These features are in agreement with the results of Sandomirskii who has studied the QSE in semimetal films solving the Boltzmann transfer equation with a δ potential for the randomly distributed static impurity.

The existence of the QSE in thin metal films is explicitly brought out using the Kubo formalism by considering the case of scattering of electrons by a randomly distributed static impurity with screened Coulomb potential. In the present calculation we have shown that in the damping approximation the Kubo formalism leads to oscillatory behavior of the conductivity. This is because the density of states exhibits quantum character, i.e., a steplike function with respect to the energy of thin films. In order to simplify the calculation, we have used a one-electron approximation. However, the problem may be studied by replacing the Fermi-Dirac distribution by the many-body electron density operator.

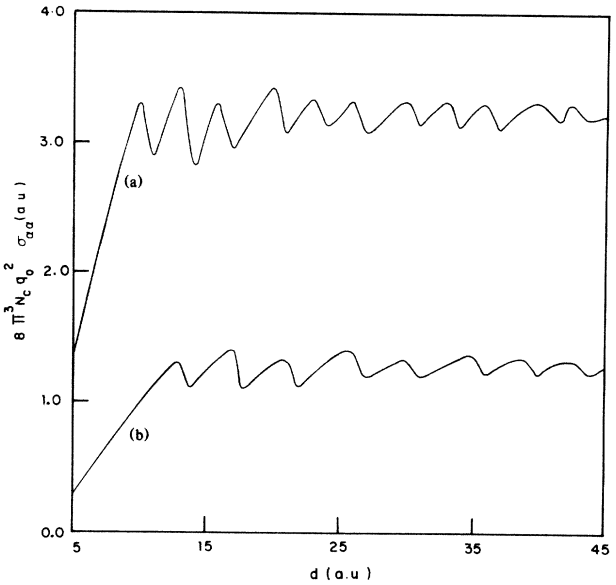

FIG. 1. Conductivity as a function of thickness for (a) aluminum and (b) copper thin films.

Financial support from the Department of Science and Technology, Government of India, and also from the University Grants Commission COSIST program is gratefully acknowledged.

[1]M. I. Elinson, V. A. Volkov, V. N. Lutskii, and T. N. Pinsker, Thin Solid Films **12**, 383 (1972).

[2]D. C. Larson, in *Physics of Thin Films,* edited by M. H. Francombe and R. W. Hoffman (Academic, New York, 1971), Vol. 6, p. 71.

[3]V. B. Sandomirskii, Zh. Eksp. Teor. Fiz. **52**, 158 (1967) [Sov. Phys. JETP **25**, 101 (1967)]; Radio Eng. Electron. Phys. (USSR) **7**, 1831 (1962).

[4]G. Dorda, in *Festkorperprobleme,* edited by H. J. Queisser (Pergamon, Vieweg, 1973), Vol. 13, p. 215.

[5]V. Benzak, J. Phys. Chem. Solids **27**, 821 (1966).

[6]Yu. F. Ogrin *et al.*, Zh. Eksp. Teor. Fiz. **53**, 1218 (1968) [Sov. Phys. JETP **26**, 714 (1968)].

[7]I. O. Kulik, Pis'ma Zh. Eksp. Teor. Fiz. **5**, 423 (1967) [JETP Lett. **5**, 345 (1967)].

[8]R. C. Fivaz and P. F. Schmid, in *Physics and Chemistry of Materials with Layered Structure—Optical and Electrical Properties,* edited by P. A. Lee (Reidel, Boston, 1976), Vol. 4, p. 343.

[9]V. K. Arora and F. G. Awad, Phys. Rev. B **23**, 5570 (1981).

[10]K. Stackulec, Physica B **95**, 197 (1978).

[11]C. R. Tellier and A. J. Tosser, *Size Effect in Thin Films* (Elsevier, Amsterdam, 1982), p. 142.

[12]V. N. Lutskii and T. N. Pinsker, Thin Solid Films **66**, 55 (1980).

[13]N. Garcia, Y. H. Kao, and M. Strongin, Phys. Rev. B **5**, 2020 (1972).

[14]H. Hoffman and G. Fischer, Thin Solid Films **36**, 25 (1976).

[15]F. K. Schulte, Phys. Status Solidi B **79**, 149 (1977).

[16]M. Huberman and G. V. Chester, Adv. Phys. **24**, 489 (1975).

[17]R. Kubo, J. Phys. Soc. Jpn. **12**, 579 (1967).

[18]W. Jones and N. H. March, *Theoretical Solid State Physics* (Wiley, New York, 1973), Vol. 2, p. 750.

[19]A. A. Abrikosov, *Introduction to the Theory of Normal Metals* (Academic, New York, 1972), p. 57.

[20]Geetha Ajoy, V. Devanathan, and G. Govindaraj, Phys. Rev. B **28**, 6852 (1983).