Electrical resistivity of thin potassium films: Evidence of a weak quantum confinement

Vladimir V. Gridin
Department of Physics, University of the Witwatersrand, Johannesburg, South Africa

W. R. Datars
Department of Physics, McMaster University, Hamilton, Ontario, Canada LBS 4M1
(Received 24 October 1990)

A study of the temperature dependence of the electrical resistivity due to the electron-phonon umklapp scattering process $[\rho(T)\sim(T/\Theta)^n\exp(-\Theta/T)]$ of thin potassium films shows that the thickness dependence of the parameter Θ, when n is a fixed constant, is closely related to the sample thickness dependence of the energy difference, Δe between the adjacent electronic subbands, which is caused by the weak quantum confinement of a free-electron gas, i.e., whenever the thermal energy k_BT, the level broadening $\hbar/t$, and Δe are interrelated as $k_BT>\Delta e \gg \hbar/t$. It is suggested that this type of confinement may cause the recently reported anomalous behavior in the temperature coefficient of the resistivity of thicker samples of alkali metals that have been studied at much lower temperatures than the 4.2–17.4-K range of the present study.

In an infinite crystal, the electron energy is known to be a multivalued function of the crystal quasimomentum **k**. For a thin metallic film with an infinite extension in the XY plane and confinement in the Z direction, the energy spectrum in the case of perfectly flat surfaces is given by a particle in the box model for independent electrons as

$$E(k,n)=\hbar^2k_{xy}^2/2m+e_0n^2\ , \quad n=1,2,\ldots, \tag{1}$$

so that the in-plane momentum is specified by a continuous quantum number k_{xy} and n is a discrete subband index. The zero-point energy e_0 is given by $e_0=(\hbar\pi)^2/2md^2$, where d is the film thickness and m is an effective mass. The energy difference between adjacent subbands for a given k_{xy} value is

$$\Delta e=e_0(2n+1)\ , \quad n=1,2,\ldots, \tag{2}$$

which, for the subbands in the vicinity of the Fermi energy, is written as

$$\Delta e_F=v_F(\hbar\pi/d)\ . \tag{3}$$

For the weak-quantization case, we assume that $n=n_F\gg 1$ and the Fermi velocity v_F is specified in terms of the Fermi momentum $k_F=\hbar\pi n_F/d$ by the substitution $v_F=k_F/m$. We note that this model is equivalent to the assumption that electronic states are represented by standing waves, which correspond to the elastic reflection at the film boundaries.[1] The subband splitting might be washed out by the level broadening due to the diffusive reflection (rough surface), impurity scattering, and/or the background scattering mechanism (e.g., electron-phonon, electron-electron, lattice imperfections) which, in the relaxation-time approximation, is of the order of $\hbar/t$. Here t is the relaxation time of the bulk specimen.[2] The temperature smearing of the discrete nature usually occurs whenever $k_BT\gg\Delta e_F$, where k_B is the Boltzmann constant. Trivedi and Ashcroft[2] have recently shown that, in the case of a strong quantization (i.e., $k_BT\ll\Delta e$), the perfect reflectivity from the film boundaries does not result in the recovery of the bulk electrical resistivity as one might expect for the thin-film resistivity from the models which are based on the Fuchs-Sondhiemer[3] analysis. Unfortunately, the treatment of Trivedi and Ashcroft[2] is limited to the $T=0$ K case, which makes it difficult to compare it with the data of the temperature dependence of the electrical resistivity in the limit $k_BT>\Delta e_F\gg\hbar/t$. Let us, at this point, compare the characteristic energies k_BT, Δe, and $\hbar/t$ for the recently reported study of the temperature dependence of the electrical resistivity of thin potassium films with the thickness d in the range 3.4–67 μm and for the temperature range $T=4.2$–17.4 K.[4] For the film thicknesses of the order of tens of μm (which is comparable with the mean free path of pure bulk potassium at helium temperatures), the quantum numbers of the levels in the Z direction in the perfectly specular case are still as big as several tens of thousands. However, for $d=10$ μm and $T=5$ K, the thermal energy $k_BT=0.43$ meV is only slightly larger than the separation between the adjacent levels that is of the order of 0.18 meV for this case. Here we have used, in Eq. (3), that, for potassium, the free-electron value of the Fermi velocity at $T=5$ K is $v_F=0.86\times10^6$ m/sec. Using the data of Ekin and Maxfield[5] for electrical resistivity of bulk potassium samples, ρ_B, and the free-electron relation

$$t=0.22/\rho_B(r/a)^3\times10^{-10}\ ,$$

we estimate the $\hbar/t$ term to be 0.002 meV at $T=5$ K [here r/a is a ratio of the free-electron sphere radius to the Bohr radius, which is equal to 4.86 at $T=5$ K; ρ_B is expressed in $\mu\Omega$ (Ref. 6)].

From these estimates one sees that the study of Ref. 4 corresponds to the case $k_BT>\Delta e_F\gg\hbar/t$. It was found[4]

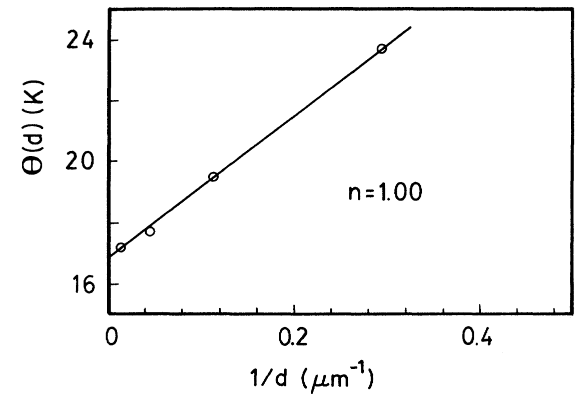

FIG. 1. The linear increase of Θ (fixed $n=1.00$) as a function of the reciprocal of the film thickness $(\mu\ \mathrm{m})/d$, see Eq. (5).

that, for $T=4.2-17.4$ K, the temperature dependence of the electrical resistivity of thin potassium films can be expressed in the form

$$\rho_S(T)=\rho_S(0)+C_S(T/\Theta)^n\exp(-\Theta/t)\ , \tag{4}$$

where the subscript S indicates the size-effect case. We quote here[4] that, for a fixed value of the power $n=1$, which was found for the bulk case by van Kempen *et al.*,[7] the parameter Θ shows a linear increase with the increase of the reciprocal thickness $1/d$. This observation is presented in Fig. 1 and the empirical thickness dependence of Θ is given by

$$\Theta=\Theta(\infty)+A/d\ , \tag{5}$$

where $\Theta(\infty)=16.8$ K and $A=(2.3\pm0.1)\times10^{-5}$ K m. The slope of this increase has been found[4] to be insensitive to a wide range of the fixed value of the power-law parameter n. Since the value of Θ in the bulk limit $(d\to\infty)$ has been found to agree quite satisfactorily with the reported values of $\Theta=20-23$ K,[5,7] we may decompose the second term on the right-hand side of Eq. (4) and get, with the help of Eq. (5), the following:

$$\begin{aligned}\rho_s(T)&=\rho_s(0)+C_s(T/\Theta)\exp[-\Theta(\infty)/T]\exp(-A/dT)\\&\simeq\rho_s(0)+C_s\Delta\rho_B(T)\exp(-A/dT)\ .\end{aligned} \tag{6}$$

Here, $\Delta\rho_B(T)$ stands for the temperature-dependent term of the background scattering mechanism for a bulk specimen, which, in the present case, is due to electron-phonon umklapp process.[8,4] The most surprising result is that the A/d term which appears in the exponent of Eq. (6) is, in fact, closely related to the subband energy gap Δe_F, which, at the Fermi level, is specified by Eq. (3). Dividing Δe_F of Eq. (3) by the Boltzmann constant k_B (thus expressing it in terms of K), we find that the ratio of $\Delta e_F/k_B$ to the A/d term [see Eq. (5)] is quite close to unity:

$$\begin{aligned}(\Delta e_F/k_B)/(A/d)&=(v_F\hbar\pi/k_B\times2.3\times10^{-5})\\&=0.89\pm0.05\ ,\end{aligned} \tag{7}$$

where $v_F=0.86\times10^6$ m/sec is the free-electron value of the Fermi velocity at $T=5$ K. This observation, together with the approximate relation that is given by the second line of Eq. (6) brings us to suggest that, under the conditions of the weak quantum confinement (i.e., $k_BT>\Delta e_F\gg\hbar/t$) of the conducting electrons, the principal temperature-dependent part,

$$\Delta\rho_s(T)=\rho_s(T)-\rho_s(0)$$

of the electrical resistivity of potassium may be expressed in terms of its ideal bulk counterpart, $\Delta\rho_B(T)$, in the following form:

$$\Delta\rho_s(t)\sim\Delta\rho_B(T)\exp(-v_F\hbar\pi/dk_BT)\ . \tag{8}$$

This result, however, is capable of giving the observed anomalous behavior of the temperature coefficient of the electrical resistivity $\partial\rho/\partial T$ that has been reported by Pratt[9] and Lee *et al.*[10] for potassium and Yu *et al.*[11] for Li and Rb. This anomalous behavior has been discussed by Kaveh and Wiser,[12] Haerle, Pratt, and Schroeder,[13] and van Vucht *et al.*[14] It has been pointed out by Haerle, Pratt, and Schroeder[13] that the explanation of their results requires the appearance of a multiplicator of the form $\exp(-\Delta\varepsilon/T)$ at the lowest temperatures to fit their data on the temperature dependence of the electron-electron scattering T^2 terms which is a feature of bulk potassium samples at $T<2$ K. They have used the model developed by Gantmakher and Kulesko[15] for the scattering of electrons by dislocations in metals. In fact, this model relies on the assumption that, due to the dislocations, there are electronic levels which are elevated by some characteristic energy $\Delta\varepsilon$ above the energy of the Fermi level. As we have found, the exponential term does show up for the above-listed weak quantum conditions and its characteristic energy $\Delta\varepsilon$ is equal to the value of the level splitting Δe_F due to the quasiconfinement of the conducting electrons in thin potassium films. Since the Δe_F that is given by Eq. (3) is proportional to the reciprocal of the length of the smallest sample axis, we suggest that the confinement of the discussed type may show up in a study of thick potassium samples at very low temperatures. We note however, that our suggested form presented by Eq. (8), which postulates that the exponential term due to the weak electronic confinement is a common feature of the temperature dependence of the electrical resistivity of simple metals, should be given a more rigorous theoretical justification. Indeed, despite our experimental evidence that Eq. (8) holds for the electron-phonon umklapp scattering process in potassium films, we found no theoretical treatments of the case $k_BT>\Delta e_F\gg\hbar/\tau$ that would support the form of Eq. (8) when, e.g., $\Delta\rho_B(T)\sim T^2$, that is, a case for the bulky electron-electron scattering events. The experimental extension of the study of the implication of the weak quantum confinement upon the electrical resistivity of thin potassium films for different scattering processes is welcome and encouraged.

The experimental part of our research was supported by the Natural Sciences and Engineering Council of Canada.

[1]B. A. Tavger and V. Ya Demikhovski, Fiz. Tverd. Tela Leningrad **5**, 644 (1963) [Sov. Phys. Solid State **5**, 469 (1963)].

[2]Nandini Trivedi and N. W. Ashcroft, Phys. Rev. B **38**, 12 298 (1988).

[3]C. R. Tellier and A. J. Tosser, *Size Effects in Thin Films* (Elsevier Scientific, Amsterdam, 1982).

[4]Temperature dependence of the electrical resistivity of these potassium films in terms of the electron-phonon umklapp scattering mechsnism is discussed by V. V. Gridin and W. R. Datars, Phys. Rev. B **40**, 5967 (1989).

[5]J. W. Ekin and B. W. Maxfield, Phys. Rev. B **4**, 4215 (1971).

[6]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt-Saunders International Editions, Philadelphia, 1976), Chap. 1.

[7]H. van Kempen, J. S. Lass, J. H. Ribot, and J. A. Wyder, Phys. Rev. Lett. **37**, 1574 (1976); H. Van Kempen, J. H. Ribot, and P. Wyder, J. Phys. F **11**, 597 (1981).

[8]R. J. M. van Vucht, H. van Kempen, and P. Wyder, Rep. Prog. Phys. **48**, 853 (1985).

[9]W. P. Pratt, Jr., Can. J. Phys. **60**, 703 (1982).

[10]C. W. Lee, M. L. Haerle, V. Heinen, J. Bass, W. P. Pratt, Jr., R. A. Rowlands, and P. A. Schroeder, Phys. Rev. B **25**, 1411 (1982).

[11]Z. Z. Yu, M. Haerle, J. W. Zwart, J. Bass, W. P. Pratt, Jr., and P. A. Schroeder, Phys. Lett. **97A**, 61 (1983); Phys. Rev. Lett. **52**, 368 (1984).

[12]M. Kaveh and N. Wiser, Adv. Phys. **33**, 257 (1984); see p. 354 for the deviation from the T^2 law.

[13]M. L. Haerle, W. P. Pratt, Jr., and P. A. Schroeder, J. Phys. F **13**, L243 (1983).

[14]R. J. M. van Vucht, G. F. A. Van Der Walle, H. Van Kempen, and P. Wyder, J. Phys. F **12**, L217 (1984).

[15]V. F. Gantmakher and G. I. Kulesko, Zh. Eksp. Teor. Fiz **67**, 2335 (1974) [Sov. Phys. JETP **40**, 1158 (1975)].