Alternative mechanism of quantum-size-effect observation in normal-metal tunneling

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A mechanism based on an energy-dependent transit lifetime is proposed to account for the appearance of conductance peaks, rather than conductance steps, in observation of quantum size effects in normal-metal tunnel junctions. This allows these results to be viewed consistently with tunneling results involving electronic subbands, and may permit additional band-structure parameters to be inferred.

Recent tunneling experiments[1-3] have clearly revealed equally spaced peaks in the conductance dJ/dV arising from energy-resolved electron standing-wave states across the thickness $t \approx 200$–1000 Å of one of the normal electrodes. Assuming a geometrically perfect film of width t in the z direction, with an infinite work function, the z dependence of the wave function $\psi = \phi_n(z)\chi_{\parallel}(x, y)$ is given by

$$\phi_n(z) = (1/\sqrt{t}) \sin(n\pi z/t), \quad n = 1, 2, \ldots, N, \tag{1}$$

corresponding to discrete values of wave vector $k_z = n\pi/t$ and z kinetic energy

$$E_{nz} = (\hbar^2/2m_z)(n\pi/t)^2 . \tag{2}$$

The total energy of the electron is

$$E = E_{nz} + E_{\parallel}, \tag{3}$$

where the transverse energy $E_{\parallel}$ can take any positive value. The splitting

$$\Delta E_{n,n-1} = \frac{\partial E_n}{\partial k}\frac{\partial k}{\partial n} = \frac{\hbar v_z(E_n)\pi}{t} , \tag{4}$$

where v_z is the group velocity in the z direction, can easily be large enough for observation, e.g., $\Delta E = 164$ meV for $t = 250$ Å and $v_z = 2\times 10^8$ cm/sec. The experimentally observed splittings in dJ/dV show both an accurate dependence on reciprocal film thickness t^{-1} and the expected magnitude, to leave no doubt that they arise from the box-quantization effect.

The experimental films are a mosaic of oriented single grains of closely similar thickness, whose transverse dimensions are estimated as several hundred angstroms.[1] The individual grain thicknesses are assumed to be integer multiples $t = Na$ of an appropriate lattice spacing, where the integer N is estimated to vary as much as 10% from grain to grain. The importance, in observing this effect of certain "commensurate" states whose energy E_{nz} is independent of the integer N and the resulting modification of Eq. (4) are clearly explained in Ref. 1.

The present paper is an attempt to reconcile the observed spectrum, which consists of equally spaced *peaks* in dJ/dV, and the equally spaced *steps* in dJ/dV which, contrary to the impression left by Ref. 1, are predicted from the theory of normal-metal tunneling into a perfect thin film. Our comment raises no question about the band-structure parameters inferred from the experiments,[1-3] but may shed some light on the experimentally noted importance of small grain size in the thin films. It also suggests an unexpected sensitivity to an aspect of the E-k relation in observing the effect.

The tunnel current at $T = 0$ into the nth z-quantized state can be obtained, following BenDaniel and Duke,[4] from the standard expression[5] simply by inserting a δ function to relate total and transverse energy, according to Eq. (3):

$$J_n(V) = \frac{2e}{h}\int_{\mu_F - eV}^{\mu_F} dE \int_0^E \rho_{\parallel}(E_{\parallel})\, D(E, E_{\parallel}) \times \delta(E - E_{\parallel} - E_{nz})\, dE_{\parallel} , \tag{5}$$

where energy E is measured from the bottom of the conduction band in the thin electrode. Here $\rho_{\parallel}(E_{\parallel})$ is the density of states per unit energy for motion parallel to the junction, which appears in converting from an integral over parallel wave vector $d^2k_{\parallel}$, and $D(E, E_{\parallel})$ is the barrier-penetration factor. Restricting to the free-electron case, $\rho_{\parallel}(E_{\parallel})$ (in two dimensions) is constant for $E > E_{nz}$, and $D(E, E_{\parallel})$ becomes a function $D(E - E_{\parallel})$ only of the perpendicular component of energy $E_{nz} = E - E_{\parallel}$, and hence is *constant*, for a given z quantum number n, apart from a possible weak dependence on bias voltage V, which we shall neglect. Because of the two-dimensional nature of the final state, expressed by Eq. (3), the integration over parallel energy is limited to one value, $E_{\parallel} = E - E_{nz}$. While the angle of tunneling does increase with $E_{\parallel}$, in this case the total energy E must increase by the same amount, maintaining the barrier-penetration factor $D(E - E_{\parallel})$ *independent* of $E_{\parallel}$ for a given quantum state n. Thus,

$$J_n(V) = \frac{2e}{h} D(E_{nz}) \int_{\mu_F - eV}^{\mu_F} \rho_{\parallel}(E - E_{nz})\, dE , \tag{6}$$

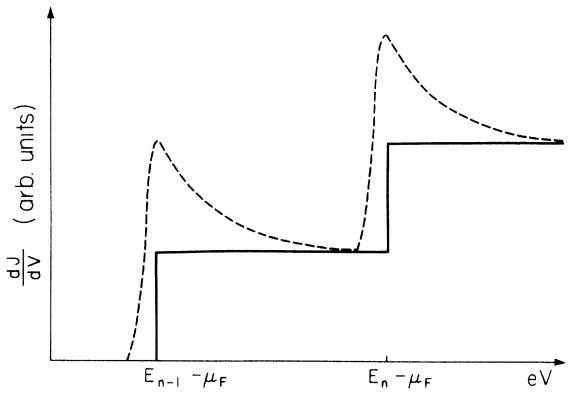

FIG. 1. Expected tunnel conductance dJ/dV for two adjacent z-quantized levels. Solid curve: perfect thin film (infinite grain size). Dashed curve: *schematic* indication for small grain size, differing thickness $t = Na$ for adjacent grains, *and* rapid variation of group velocity $(1/\hbar)\, dE_{\parallel}/dk$ (for motion parallel to the junction) with transverse energy $E_{\parallel}$ at the *bottom* of the band in question (see text).

corresponding to conductance, with positive bias V applied to the thin electrode,

$$\frac{dJ_n}{dV} = \frac{2e}{h} D(E_{nz})\rho_{\parallel}, \quad eV \geq E_{nz} - \mu_F, \qquad \frac{dJ_n}{dV} = 0, \quad eV < E_{nz} - \mu_F. \tag{7}$$

dJ/dV is thus expected to be the sum of equally spaced steps, as indicated in Fig. 1, in the solid curve.

Evidently the energy and angular dependences of the barrier-penetration factor[6] cannot provide the selection of final states with $E_{\parallel} = 0$, which is suggested by the experimental peaks at $eV = E_{nz} - \mu_F$. It is perhaps worth pointing out that this analysis has been verified experimentally in the closely related case of a quantized surface accumulation layer on a semiconductor.[7] Steps rather than peaks in dJ/dV are observed in this experiment.

An alternative mechanism for the enhancement of the $E_{\parallel} = 0$ states in the conductance may result from an increasing lifetime energy broadening of states of increasing $E_{\parallel}$ arising from the transit time of the electron across the small grain. The lifetime of the particular state with $E_{\parallel} = 0$ is determined presumably by the time required for the electron to tunnel back into the counterelectrode through the oxide barrier.[4] In any case, an electron with finite $E_{\parallel}$, will have a shorter lifetime, for it will transit across the individual grain of thickness Na and flow out into an adjacent grain,[8] limiting the time $\tau(E_{\parallel})$ during which its energy $E_{nz} + E_{\parallel}$ can be measured, and leading to an additional lifetime energy width $\Gamma(E_{\parallel}) = h/2\tau(E_{\parallel})$. In this case the individual states $E_{nz} + E_{\parallel}$ must be represented by normalized spectral functions $g(E' - E_{\parallel}, \Gamma)$ whose widths $\Gamma \propto \partial E_{\parallel}/\partial k_{\parallel}$ increase with $E_{\parallel}$, and the conductance (7) must be generalized to

$$\frac{dJ_n(V')}{dV} = \frac{2e}{h} D(E_{nz})\rho_{\parallel} \int_0^{\infty} g(eV' - E_{\parallel}, \Gamma)\, dE_{\parallel}, \tag{8}$$

where we have set $eV' \equiv eV - E_{nz} + \mu_F$, measuring bias voltage from the threshold for tunneling into the nth band. If the $E_{\parallel} = 0$ lifetime is sufficiently long, and if $\Gamma(E_{\parallel})$ increases sufficiently rapidly with $E_{\parallel}$, an appreciable pileup of spectral density may occur at $E_{\parallel} = 0$ from the tails of states of higher transverse energy $E_{\parallel}$. This effect is indicated only schematically in Fig. 1 by the dashed curve.

While a quantitative fit to the experiments is not possible, the orders of magnitude involved and the qualitative features of $g(E' - E_{\parallel}, \Gamma)$ and $\Gamma(E_{\parallel})$ suggest that this effect may be strong enough to impart the observed peak character to the expected step structure in the conductance.

First, near the bottom of the bands in question, which lie in the second and third zones of Pb[1] and Mg,[2] respectively, $\Gamma(E_{\parallel})$ increases rapidly from zero to about a hundred millivolts for $E_{\parallel}$ of the order of half the interband gap, also of the order of a few hundred millivolts, for nearly-free-electron-like bands.[9] For $E_{\parallel}$ large enough that the band-edge perturbation is unimportant, v_g is of order 10^8 cm/sec, and for grain diameter D several hundred angstroms,[1] τ is several times 10^{-14} sec. Second, the expected spectral distribution $g(E - E_{\parallel}, \Gamma)$ for a fixed transit lifetime τ is of the form $\sin[(E' - E_{\parallel})\tau/2\hbar]/(E' - E_{\parallel})$, which falls off fairly slowly with $E' - E_{\parallel}$. A fit to the data would require a more detailed knowledge of the functional dependence of τ on $E_{\parallel}$ than is available; however it does appear that an enhancement factor of 2, as suggested in Fig. 1, and sufficient to provide the observed peaks in dJ/dV, is not unreasonable on this mechanism.

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[4]D. J. BenDaniel and C. B. Duke, Phys. Rev. 160, 679 (1967).

[5]C. B. Duke, *Tunneling in Solids* (Academic, New York, 1969), p. 55.

[6]John Lambe and R. C. Jaklevic (private communication) have suggested an additional possibility, that of dependence of D on bias voltage V, through the change in barrier shape. This mechanism, which is to be discussed by the above authors in a forthcoming publication involves bias dependence of D in Eq. (7) and the appearance of a second term in Eq. (7) arising from V dependence of D in Eq. (6).

[7]D. C. Tsui, Phys. Rev. B 4, 4438 (1971).

[8]The films are conducting, so that it is not reasonable to assume that reflection back into an individual grain is likely at the grain boundary. If one does assume that the electron wave function has a node also at the grain boundaries, the degeneracy of the electron states is completely removed and $E_{\parallel}$ does not go to zero but assumes a minimum value depending on the transverse dimensions of the grain. In this case sharp energy peaks would not be observed, but would be washed out by the spread in transverse energies from grain to grain.

[9]This behavior is clearly indicated at the bottom of the relevant band in the case of Mg; see Fig. 3 of Ref. 2.