

Free-electron model for mesoscopic force fluctuations in nanowires

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When two metal electrodes are separated, a nanometer-sized wire (nanowire) is formed just before the contact breaks. The electrical conduction measured during this retraction process shows signs of quantized conductance in units of $G_0=2e^2/h$. Recent experiments show that the force acting on the wire during separation fluctuates, which has been interpreted as being due to atomic rearrangements. In this paper we use a simple free-electron model, for two simple geometries, and show that the electronic contribution to the force fluctuations is comparable to the experimentally found values, about 2 nN. [S0163-1829(98)01816-5]

I. INTRODUCTION

The electrical conductance through a narrow constriction with a diameter of the order of the electron wavelength is quantized in units of $G_0=2e^2/h$.^{1,2} Such conductance quantization is observed at low temperatures in semiconductor devices containing a two-dimensional electron gas.^{3,4} Similar effects are possible^{5,6} at room temperature in metallic wires with a diameter of the order of 1 nm (nanowires) and are observed using scanning tunneling microscopy,⁷⁻¹³ mechanically controlled break junctions,^{14,15} or, as recently shown,^{16,17} just plain macroscopic wires. These techniques use the same basic principle: By pressing two metal pieces together a metallic contact is formed that can be stretched to a nanowire by the subsequent separation of the electrodes. The conductance in such a system is found to decrease in abrupt steps with a height of about $2e^2/h$, just before the contact breaks.

In a recent experiment by Rubio, Agrait, and Vieira,¹⁸ following earlier attempts,¹⁹⁻²² the force and the conductance were simultaneously measured during elongation, from formation to rupture, of a gold nanowire. They show that the stepwise variation of the conductance is always accompanied by an abrupt change of the force. One interpretation^{10,18,23-25} is that the structural transformations of the nanowire, involving elastic and yielding stages, cause the stepwise variation of the conductance.

In this paper we study the electronic contribution to the observed force fluctuations using a simple free-electron approach neglecting all atomic structures of the wire: a jellium model (see also three other recent reports²⁶⁻²⁸). In metals the electronic contribution to the binding energy is significant (metallic binding) and one might suspect that the quantized electronic energy levels in the nanowire would be reflected in the binding energy. When a conductance mode closes it should produce a sharp change in the electronic binding energy and subsequently the force. The quantized energy levels are of the order of electron volts and the wire elongation of the order of nanometers, giving a change in force of the order of nanonewtons, the same as observed in the experiments. Considering this, we develop in this paper a simple free-electron model. The calculations show force fluctuations of the same size as in the experiments.

II. MODEL

We use a free-electron model neglecting all atomic structure in the wire, a jellium model. Further, cylindrical nanowires of length L and with two different cross sections are studied: first with a circular cross section (see Fig. 1) and then with a square cross section. Under the assumed ideal plastic deformation, the volume V of the wire will be constant during elongation. We are interested in the tensile forces acting on the wire during elongation. In general the

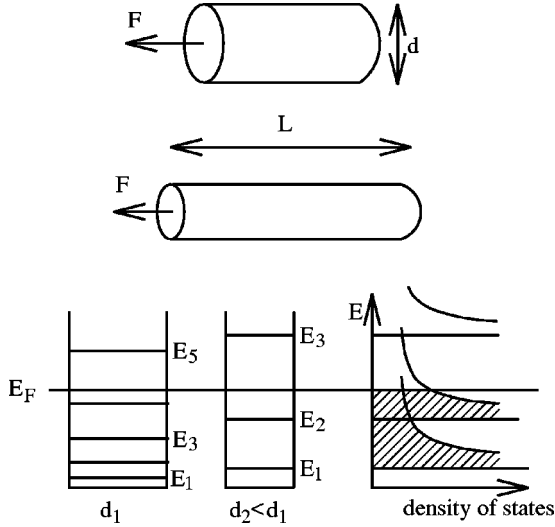


FIG. 1. Model of the nanowire. The pulling force F is acting on a nanowire of length L and width d . When the wire is elongated more and more transverse quantized modes are pushed above the Fermi level and closed. The corresponding one-dimensional densities of states are filled up to the Fermi level.

force is the derivative of the energy with respect to distance; however, our system is open and we should instead consider the thermodynamic potential Ω . The Fermi energy E_F in metals is much higher than the thermal energy and we can approximate the chemical potential by E_F and the thermodynamic potential is found to be $\Omega = E - E_F N$, where E is the energy and N the number of electrons. The force is then $F = -d\Omega/dL$.

A. Nanowire with a circular cross section

In a wire with a circular cross section, using the adiabatic approximation,^{29,30} the transverse motion of the electrons gives rise to quantized modes n of energy,

$$E_n^c = E_{jl} = \frac{\hbar^2}{2mR^2} \beta_{jl}^2 = \frac{\hbar^2 \pi}{2mV} L \beta_{jl}^2, \quad (1)$$

where β_{jl} are roots to Bessel functions, i.e., $\beta_{jl} = 2.4048, 3.8317, \dots$, and the degeneracy is twofold (not counting spin) unless $j=0$. The third equality in Eq. (1) is valid for a wire of constant volume. A mode is considered to be open if $E_F > E_n^c$. The number of electrons in the wire is

$$N = \sum_n N_n = \sum_n \int_{E_n^c}^{E_F} LD(\varepsilon - E_n^c) d\varepsilon \quad (2)$$

$$= \sum_n 2L \sqrt{\frac{2m}{\pi^2 \hbar^2}} \sqrt{E_F - E_n^c}, \quad (3)$$

where $D(\varepsilon)$ is the one-dimensional density of states

$$D(\varepsilon) = \sqrt{\frac{2m}{\pi^2 \hbar^2}} \frac{1}{\sqrt{\varepsilon}}. \quad (4)$$

The total electronic energy of the wire is the integral of the energy times the density of states up to the Fermi energy and summed over all open modes,

$$E = \sum_n \int_{E_n^c}^{E_F} L \varepsilon D(\varepsilon - E_n^c) d\varepsilon \quad (5)$$

$$= \sum_n \left\{ \frac{2}{3} L \sqrt{\frac{2m}{\pi^2 \hbar^2}} (E_F - E_n^c)^{3/2} + E_n^c N_n \right\}. \quad (6)$$

The thermodynamic potential is then

$$\Omega = E - E_F N = - \sum_n \frac{4}{3} L \sqrt{\frac{2m}{\pi^2 \hbar^2}} (E_F - E_n^c)^{3/2} \quad (7)$$

and the derivative gives the force

$$F = - \frac{d\Omega}{dL} = \sum_n \sqrt{\frac{2m}{\pi^2 \hbar^2}} \left\{ \frac{4}{3} (E_F - E_n^c)^{3/2} - 2(E_F - E_n^c)^{1/2} E_n^c \right\}. \quad (8)$$

B. Nanowire with a square cross section

Using the same approach as above, the transverse motion of the electrons in the wire with a square cross section gives rise to quantized modes n of energy

$$E_n^q = \frac{\hbar^2 \pi^2}{2md^2} n^2 = \frac{\hbar^2 \pi^2}{2mV} n^2 L, \quad (9)$$

where $n^2 = l^2 + m^2$, $l = 1, 2, \dots$, and $m = 1, 2, \dots$, i.e., $n^2 = 2, 5, 8, 10, \dots$, and the degeneracy is twofold (not counting spin) unless l and m are equal. The second equality in Eq. (9) is valid for a wire of constant volume. Replacing E_n^c with E_n^q in Sec. II A gives the appropriate expressions for the wire with a square cross section.

III. FORCE FLUCTUATIONS

Figure 2 shows a plot of the force during elongation in a wire with a circular cross section, according to Eq. (8). Also the conductance of the wire is shown. The wire volume is taken to be 3 nm^3 . The number of modes that contribute to the conductance is taken from Eq. (1). Whenever a mode closes the conductance jumps one quantum unit and an abrupt change in the force appears. The peak-to-peak amplitude of the force fluctuations between two modes is about 2 nN. Figure 3 shows the corresponding force and conductance for a wire with a square cross section.

IV. DISCUSSION

The force from our calculations shown in Figs. 2 and 3 agrees both qualitatively and quantitatively with experiments.¹⁸ The only significant effect of the geometry of the cross section is on the degeneracy of the modes.

Force fluctuations are also seen in molecular-dynamics simulations^{10,21,23,25} and the jumps in conductance are interpreted as due to atomic rearrangements. However, because of the experimental-like conditions in these simulations, it is

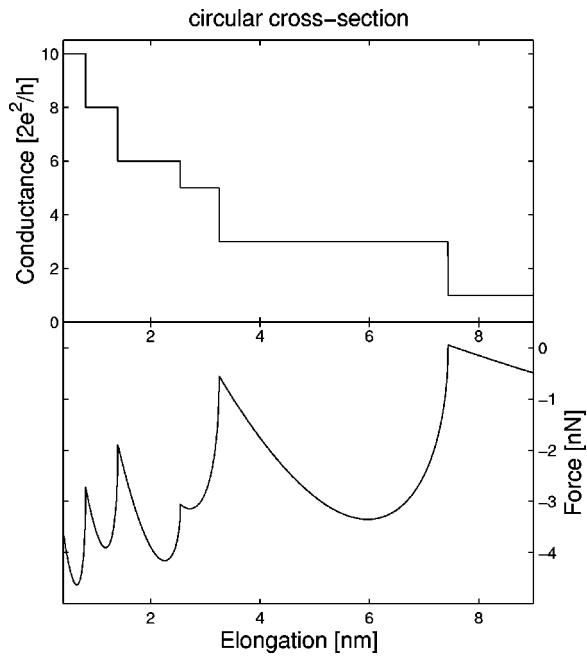


FIG. 2. Calculated conductance and force as a function of elongation for gold nanowires with a circular cross section and constant volume 3 nm^3 . Whenever a mode closes the conductance jumps one quantum unit and an abrupt change in the force appears. The peak-to-peak amplitude of the force fluctuations between two modes is about 2 nN .

difficult to separate the different contributions to the binding energy. Our interpretation is more or less the reverse: The electronic contribution to the binding energy is so large that the change of the quantized energy levels in the wire, with a corresponding quantized conductance, causes the force fluctuations. These force fluctuations might then give rise to atomic rearrangements, but not necessarily. Although this is a bit like the story about the chicken and the egg, our simple model shows that the electronic contribution must be considered seriously because it constitutes a significant part of the metallic binding energy in these nanowires.

One electronic contribution to the binding energy, which is neglected in the present model, is the Coulomb interaction. In metallic binding the electrostatic energy could be of the

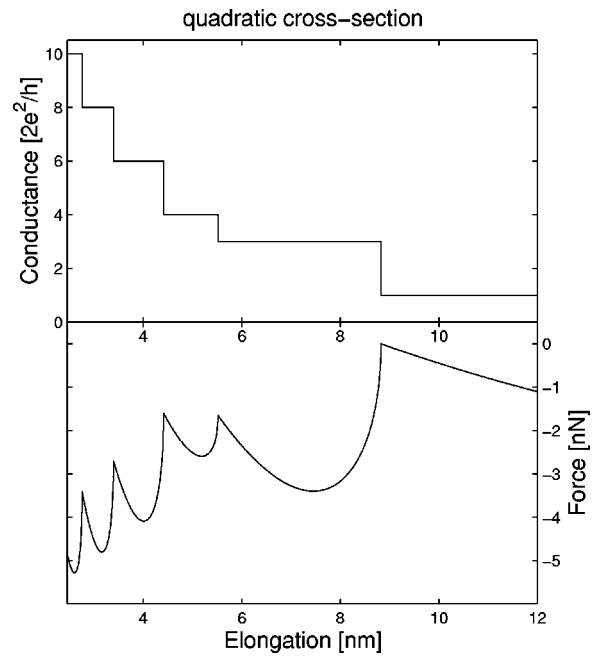


FIG. 3. Calculated conductance and force as a function of elongation for gold nanowires with a square cross section and constant volume 3 nm^3 .

same order as the kinetic one and a natural extension of the present model would be to include the electrostatic energy, which would change the electronic energy. The force fluctuations would, however, still be present.

V. CONCLUSION

We have shown, using a simple free-electron model, that the electronic contribution to the force fluctuations is comparable to the experimentally found values. This could be of importance to understand the mechanism of formation of metallic nanowires as well as in the wider context of nanomechanics.

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