Simple Model for Energy and Force Characteristics of Metallic Nanocontacts

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The quantum size oscillations of the energetic properties and the elongation force of the gold slabs and wires, isolated and in a contact with electrodes, are calculated in a free-electron model. A simple relation between the Fermi energy and the square-potential-well depth is used and tested for low-dimensional systems. It is shown that considering the electron subsystem of a slab (or wire) in a contact as open one, the contact acts like a sort of electron pump which sucks or pumps out electrons from the sample. The effect of the contact potential difference on the elastic force oscillations is considered. The calculated amplitudes of force oscillations are in a qualitative agreement with those observed experimentally.

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I. INTRODUCTION

Recent experimental investigations of gold point-contacts by using the atomic force microscope [1] have demonstrated a violation of the Hooke and Ohm laws: a deformation of the contact leads to a coherent step-wise variation of the conductance and force. Theoretical interpretation of these observations for a deformed nanowire was given in the framework of the simple free-electron models [2, 3, 4, 5, 6]. The energetic picture of the nanocontact is more complicated, however, than that adopted in [2, 3, 4, 5, 6]. First, in the previous considerations the contact potential difference \( \Delta \) is not taken into account. Secondly, the metallic contact changes its dimensionality with the variation of sample-cantilever distance. At first, its shape is rather similar to a slab, to resemble a wire at the moment of rupture [3, 10]. Thus, we have to do with a transformation of the 2D open electron systems into the 1D one. The purpose of this work is to investigate the limiting cases of this transition. We study the size dependence of the work function, ionization potential and elastic force of the isolated quantum system. We begin with consideration of the energetic pictures of an isolated sample. The results are exploited later to determine the contact potential difference (CPD) for a sample connected with electrodes.

In order to trace analytically the effect of dimensionality on the energetic and force properties one needs a simple model relating mutually Fermi energy, work function and the potential barrier. We consider a rectangular sample of the volume \( V = a \times a \times L \), where \( L \) denotes the dimension along the \( x \)-axis. The inequalities \( a \gg L \) and \( a \ll L \) correspond to the geometry of a slab and a wire, respectively. The consideration of these two asymptotic limits which simulate the early and late phases of elongation, allows us to trace the evolution of the energy and force characteristics of the 2D and 1D metallic structures.

We have assumed that the energetics of a finite system of bounded electrons can be described by a square potential well of the width \( L \) (slab) or \( a \) (wire) and the depth \( U < 0 \),

\[
-U(E_F) = W(E_F) + E_F,
\]

where \( W \) is the work function and \( E_F \) is the Fermi energy of a finite sample.

Let us remember that the potential well of the depth \( U \) of metal sphere shows the size oscillations [11]. The Fermi energy of metal film also has a size oscillations [12]. Due to an expansion of the chemical potential of spherical cluster of radius \( \mu = \mu_0 + \mu_1/R \) in powers of the inverse radius [12] we paid attention on the inequality

\[
W = W_0 - \frac{\mu_1}{R} < W_0.
\]

The criterion \( W < W_0 \) (subscript zero labels the quantities for a semi-infinite metal) for one-dimensional systems was introduced by us [14] to check the correct size dependence of the \( U(E_F) \) (by means \( E_F(L) \)). In order to determine the size oscillations in the potential well depth \( U(E_F) \) we have employed a simple expression for the \( W_0(E_{F0}) \) provided by [15]. The latter is based on the concept of the image force action [16, 17] and a spontaneous polarization of metallic plasma, to determine the distance, from the classical metal surface at which the image force begins to act (see also [18] and references therein),

\[
W_0(E_{F0}) = \frac{B}{r_s^{3/2} E_{F0}^{1/2}}, \quad (\text{a.u})
\]

where \( B \) is the adjusting parameter and \( r_{s0} \) is the average distance between electrons in the bulk, \( B = 0.3721 \) a.u. and \( r_s = 3.01 \) bohr radius for gold.

Adapting the result for the low-dimensional systems we replace \( W_0(E_{F0}) \) by \( W(E_F) \) and suppose \( W \to W_0 = \)
4.30 eV \[14\] as \( L \to \infty \). In order to get a qualitative agreement of calculated force with the experimental data we have to assume that Au is monovalent \[2, 3, 6\].

### II. BASIC RELATIONS FOR AN ISOLATED SAMPLE

For the 2D and 1D metallic systems the allowed energy levels (the electron kinetic energies) form a quasi-continuum \[21\],

\[
E_{n_{xy}} = E_{n_x} + E_{n_y} + E_{n_z} = \frac{\hbar^2}{2m} (k_{n_x}^2 + k_{n_y}^2 + k_{n_z}^2).
\]

For an infinitely deep potential well of the width \( a \) (the shape is a cube) this expression reduces to

\[
E_{n_{xyz}} = \frac{\hbar^2 \pi^2 n_{xyz}^2}{2ma^2} + \frac{\hbar^2 \pi^2 n_{yz}^2}{2mn_a^2} + \frac{\hbar^2 \pi^2 n_{xz}^2}{2mn_a^2} + \frac{\hbar^2 \pi^2 n_{yz}^2}{2mn_a^2} + \frac{\hbar^2 \pi^2 n_{xz}^2}{2mn_a^2} + \frac{\hbar^2 \pi^2 n_{yz}^2}{2mn_a^2}.
\]

In the equation (5) for a slab

\[
D(E) = \frac{m}{\pi L \hbar^2} nE,
\]

and

\[
D(E) = \frac{L}{V} \sqrt{\frac{2m}{\pi \hbar^2}} \sum_{n_y, n_z} (E - E_{n_y} - E_{n_z})^{-1/2}
\]

for the finite slab (or wire, respectively).

In the equation (4) for a slab \( n_E \) is the integer part of number,

\[
n_E = \left[ \frac{Lk + 2 \arcsin(k/k_0)}{\pi} \right],
\]

where \( \hbar k = \sqrt{2mE} \).

In the equation (6) for a wire the plus sign in the limit of summation indicates that \( n_y \) and \( n_z \) run from 1 to the maximum value for which the expression under the square root is positive.

Subsequently, the total number of electrons in a slab is given by

\[
N = \frac{a^2 m}{\pi \hbar^2} \sum_{n_z=1}^{n_F} (E_F - E_{n_z}),
\]

where \( n_F \) is the number of highest occupied subband, \( n_F \) equals \( n_E \) in Eq. (7) with the change \( k \to k_F \).

The total number of electrons in a wire is given by

\[
N = 2L \sqrt{\frac{2m}{\pi \hbar^2}} \sum_{n_y, n_z} (E_F - E_{n_y} - E_{n_z})^{1/2}.
\]

Here \( N = \overline{n}V \), where \( \overline{n} = 3/4\pi r_0^2 \) is the electron density in the bulk of semi-infinite metal.

Discussing the effect of dimensionality and charging, it is useful to analyze ionization potential which is a well defined quantity for an arbitrary size of a sample \[21, 22\]. The ionization potential which is defined as a work needed to remove an electron from a neutral metallic sample, can be expressed as

\[
IP = W + \frac{e^2}{2C},
\]

where \( C \) is the capacitance of the sample. It should be mentioned that \( W_0 \) and \( IP \) are the experimentally measured quantities, while \( W \) has mainly a methodical sense because it can be measured only in the limit of \( C \to \infty \). An extended thin slab or wire of infinite length has an infinite capacitance, \( C \to \infty \), therefore \( IP \to W \). Equation (10) can be interpreted as the effect of charging on the work function of the neutral finite sample. It should be noted that the size correction in \( W(L) \) which is similar to that of the spherical cluster (see Eq. (9)) competes with the term \( e^2/2C \). Since it is impossible to derive an analytical expression for the rectangular sample, in order to estimate the monotonic size dependence of \( IP \) we apply the well-known formula for the capacitance \( C \) of the disk of the width \( L \) and of the needle of the length \( L \) \[23\].

Solving the set of Eqs. (4) and (5) or (6), by using (10) and adapted Eq. (3), one can determine the Fermi energy \( E_F \) and subband energies for a given \( L \) under condition \( V = \) constant. Consequently, one gets the work function \( W \), as function of the Fermi energy of finite sample.

The elastic elongation force acting on a finite sample is given by \( F = -dE_t/dL \), where \( E_t \) is the total energy of the sample. Neglecting the temperature effects in the adiabatic approximation, from the virial theorem we have \( E_t = -K \), which means that the energy of a bound electron-ion system is negative \[14, 24\]. (The virial and stress theorems for electron–ion mixture of solid metal representation was formulated in Ref. \[23\]). Consequently,

\[
F = \frac{dK}{dL}.
\]

In expression (11) we take into account the potential energy of the electron-ion system. This gives a plus sign in front of \( K \) (an opposite sign appears in \[2, 3, 6\], where the simplest model of nanowire has been exploited). The plus sign follows from the application of the virial theorem.
The total kinetic energy of electrons are given by the expressions

\[
K = \frac{a^2 h^2}{2\pi^2 2m} \sum_{k_{ax}} \int_0^{\sqrt{k_{ax}^2 - k_{ax}^2}} (\kappa^2 + k_{ax}^2)2\pi\kappa dk = \frac{a^2 m}{2h^2} \sum_{n_z=1}^{n_F} (E_F - E_{n_z}^2),
\]

and

\[
K = \frac{2L}{3} \sqrt{\frac{2m}{\pi^2 h^2}} \sum_{n_x,n_z}^+ (E_F - E_{n_y} - E_{n_z})^{1/2} \times (E_F + 2E_{n_y} + 2E_{n_z}),
\]

for an isolated slab and wire, respectively.

### III. THE EFFECT OF A "POINT" CONTACT

When a sample is inserted in a contact between the reservoirs then its electron subsystem has to be considered as an open one, under the condition \( W(L) = W_0 \), where \( W \) and \( W_0 \) are the work functions for an isolated sample and reservoirs, respectively. Due to the contact potential difference, \( \delta\phi \), the electroneutrality of the sample breaks down and \( \delta N \) electrons from the electron subsystem are transferred into reservoirs. In order to determined \( \delta\phi \) one can imagine a simple energetic cycle in which electronic charges are transferred from a sample to infinity and then into electrodes. By expressing the ionization potential, \( IP \), of the sample charged by \( +e\delta N \) in a form similar to that for the charged spherical metal clusters [27] we have,

\[
IP = E_{N-\delta N-\Delta} - E_{N-\delta N} = W\Delta + \frac{e^2}{2C}[(\delta N + \Delta)^2 - \delta N^2],
\]

where \(-e\Delta\) is the part of the electronic charge which leaves the charged sample. The electron affinity of this electronic charge, transferred to reservoirs is \( EA = W_0\Delta \). The equilibrium condition \( IP - EA = 0 \) leads to

\[
W_0 - W - \frac{e^2}{2C}(2\delta N + \Delta) = 0.
\]

It should be noted that the magnitude of \( \Delta \) can be infinitesimally small (non-integer), because in a contact (open system), the residual electron can be transferred only partially (i.e., there is a finite probability that it could be found both in a sample and in a reservoir). Thus, we can ascribe to \( \delta N \) a continuous value. We also suppose that \( C \) of rectangular sample appearing in Eq. (14) corresponds to the total capacitance \( C_v \) of both contacts. This is justified by the fact that near the two faces of a sample the surplus positive charges have a similar surface distribution both in the case of real ionization of an isolated sample and for a sample in contact. (This situation is similar to that encountered in single-electron devices [28]). Then, assuming that \( C_v = e\delta N/\delta\phi \), \( \delta N \ll N \) and \( \Delta \to 0 \), Eq. (15) one gets

\[
\delta\phi = (W_0 - W)/e > 0.
\]

For example, the energy spectrum \( E_{n_1x} \) of the remaining electrons in the slab, \( N_1 = N - \delta N \), can be found by solving [14] for a square potential well of depth

\[
U_1 = U - e\delta\phi,
\]

where \( U \) corresponds to the isolated slab. Comparing [17] with [14] and using [18] we find that in equilibrium between a sample and the electrodes, the magnitude of \( E_{F1} \) is equal to that of an isolated sample (\( E_F \)). The value of \( \delta N \) can be calculated using Eq. (5) with the change \( N \to N_1 \) and \( k_{ax} \to k_{n_1} \). The total kinetic energy, \( K_1 \), of the remaining electrons is defined as earlier (Eq. (12)) with the changed energy spectrum and the number of electrons. The same approach was used for the nanowire. In the case of an open system the elastic force is determined by a surplus pressure, relative to the reservoirs, multiplied by the area of the contact:

\[
F_1 = -\frac{d\Omega}{dL},
\]

where the size-dependent part of the grand potential \( \Omega \) is given by

\[
\Omega = E_1 + W_0N_1,
\]

and \( E_1 = -K_1 \).

### IV. RESULTS AND DISCUSSION

The calculations were performed for the set of isolated Au slabs and wires and then for the ones in contact with reservoirs. It allowed us to calculate the contact potential which is needed for the force characteristics. Assuming the ideal plastic deformation in the experiments [1, 10], the volume of deformed sample will be constant. All considered samples have a same volume, \( V = 4 \text{ nm}^3 \), and the number of electrons \( N = 236 \). The linear sizes of a sample vary in the range: \( \sqrt{\pi}r_s < L < 13a_0 \) for slabs, and \( L_0/10 > a > \sqrt{\pi}r_s \) for wires. Here, \( a_0 = \hbar^2/m_e^2 \) is the Bohr radius. The calculated energy and force characteristics corresponding to a slab and wire, respectively, are displayed in the left and right parts of Figs.1 and 2. The wire length, \( L = L_0 + \Delta L \), has been increased by about seven times. Figure 1(a) shows the density of states, \( D(E_F) \), of an isolated sample (curve 1) and the one contacted with reservoirs (curve 2) versus the length of wire. For a better demonstration the curves
We consider the isolation function of an isolated sample, ionization potential, and contact potential difference (curves 1) and sample in a contact (curves 2). (b) The work function, ionization potential, and contact potential difference of the isolated slab and wire. For comparison, the values of the Fermi level of a isolated sample and a sample in a contact with reservoirs. (c) Total kinetic energy $K$ of the electrons (left axis) and elongation force $F$ (right axis) of the isolated sample. (d) Total kinetic energy $K$ of the electrons (left axis) and elongation force $F$ (right axis) of the isolated sample.

The peaks of the $D(E_F)$ for a wire with square cross-section are more intensive for the additionally degenerate subband, $k_n = k_{n_z}$.

The size correction in $W(L)$ competes with the $e^2/2C$ term in expression $IP$ for the ionization potential. The magnitude of $IP(L)$ depends on the shape of a sample: $IP(L) > W_0$ for $L < 43\lambda_{F0}$ and $IP(L) < W_0$ for $L > 43\lambda_{F0}$. Note that the positions of the local minima in the $IP(L)$ and $W(L)$ curves correspond to the positions of the peaks in the density of states of an isolated sample (Fig.1(a)). In the case $a = L$, we deal with a metal cube whose ionization potential must have a value similar to that of a sphere. It should be noted that the ionization potential depends only on the geometry of the sample and is independent of the direction of electron emission.

The CPD leads to a noticeable negative shift of the potential well depth (in utmost point by about 0.5 eV) and this in turn leads (Fig.1(a)) to a shift in the density of states (curve 2) to a zone of the bigger cross section.

Figure 1(c) shows the size dependence of the number of electrons $\delta N$ that have left the sample. An increase of the cross-section or a decrease in the wire length leads to the breaks appearing in the monotonic components of $\delta\phi(L)$ and $\delta N(L)$ which correspond to a spherical sample. For $a$ of the size close to the atomic diameter the $\delta N$ makes up 20% of the initial number of electrons. The account for the contact potential leads to a dependence of $\delta N$ on the hierarchy of energy levels in an isolated sample. This aligns specific breaks in the $\delta N(L)$ curve which positions of the peaks in the density of states $D(E_F)$ of both isolated and contacted samples. The results show that some part of the electrons is spilled out from the slab (wire) in a contact and the contact can be thought of as a sort of the “electron pump” which pulls out the electron liquid from or draws it into reservoirs. The dipole layers formed in a vicinity of both contacts must stimulate additional longitudinal deformation of the wire and contribute to a change in its shape and in the electron density. The above results allow to calculate size dependence of the effective capacitance $C_e = e\delta N/\delta\phi$ for a sample in contact.

The kinetic energy of electrons and the elongation force of the isolated sample are displayed in Fig. 1(d). Their counterparts for the sample after junction with reservoirs as well as the potential $\Omega$ are shown in Fig. 2. Total electron kinetic energy $K(L)$ and $K_1(L)$ of metal cube is similar to that for semi-infinite metal, $(3/5)N\varepsilon E_{F0}$. The amplitudes of the force oscillations $F(L)$ of the isolated slab are several times bigger than those measured for the nanowires $[1, 10]$. It is stipulated by a large value of the cross-section $a \times a$. Nevertheless, dividing the first amplitude of the force by the number of atoms in the slab one gets 0.4 nN. This value is smaller than the force.
acting on a single atom (1.6 nN) in the point contact experiments \[1\]. That can be explained by the different dimensionality of a slab and wire. For the reduced dimensionality the amplitudes of force oscillations range from 1 to 3 nN, which is similar to the measured values. The character of the dependence \( F(L) \) changes significantly at the wire contact because of depletion of the electron subsystem and due to a shift of the spectrum. It should be mentioned that in our approach the size dependencies \( K(L) \) and \( K_1(L) \) are qualitatively different from those obtained for an infinitely deep potential well \[2, 3, 6\]. The second term appearing in \( \Omega \) only partly smoothes out the oscillations in \( \Omega(L) \). This is in contrast to the infinitely deep potential well where the oscillations in \( \Omega(L) \) disappear. As a result, the amplitudes of force \( F_1(L) \) and their shape change significantly. In our more advanced model, the attempt to take into account the contact potential difference leads to the appearance of spikes in the elastic force versus length which result from the fact that for the isolated and contacted wires the densities of states are not in line. It is possible that in experiment these peaks could not be recognized in the background of the thermal fluctuations of the wire-form which occur during deformation. With the exception of the interval \( 54 < L/\lambda_{K0} < 62 \) (Fig.2(b)) the calculated amplitudes of force agree with the experimental values. The difference may be stipulated by not self-consistent determination of the CPD for the wires of diameter that is comparable with the atomic dimensions and by the assumption that \( L \) varies continuously. Another reason might be that the elastic force is sensitive to the applied voltage as suggested in Refs. \[33, 34\].

The choice of the simplified form of the wire and a non self-consistent treatment of the size oscillations in the potential \( U \) are the disadvantages of the model. The free-electron model is not able to describe in detail the effect of the atomic rearrangements on the magnitude of force in the atomic contact \[10\]. On the other hand the obtained qualitative results are quite general and should not depend on the symmetry of the problem. It should be also noted that for the stabilized jellium model the bulk modulus \[22, 23\], work function, surface energy, and surface stress \[21, 22\] for gold can be described only when its valency is assumed to be three. The transition from monovalent to trivalent gold can lead to increased, by several times, amplitude of oscillations of the elastic force, in analogy with that observed for the trivalent Al \[13\]. These problems can be connected with an unusual size dependence of \( IP(R) \) that is measured for spherical gold particles: for the very large as well as for the atomic-size particles \( IP > W_0 \), whereas \( IP < W_0 \) for particles of the intermediate \( R \). \[37, 38\]. In our view, this could be explained by a reduced valency of some metals which is connected with the decreasing of the size of a sample (it reminds the metal-nonmetal transition). A similar transition was observed \[22\] for mercury clusters and interpreted in Ref. \[12\]. The behavior of the ionization potential measured for aluminium clusters (compare Fig.28(a) in \[13\]) also points to this effect. Therefore, the assumption of monovalency of Au in the low-dimensional structures is quite reasonable.

Finally we would like point to the connection between our results and the famous phenomenon of the quantization of shear modulus for polycrystalline samples that was experimentally observed for 57 elements at the room temperature \[42\]. Possibly this effect may be explained by a quantization of the force characteristics of the 2D electron liquids in a inter-crystallite space of the polycrystal.

The results of this paper can be summarized as follows. The evolution of the size dependence of the ionization potential during transformation of the shape of a sample from a slab toward a wire is investigated. The size dependence of the CPD is calculated in a simple manner. It is shown that the infinite potential well model of a slab \[8\] overestimates the value of the CPD by many times. For the first time, the effect of the CPD on the size oscillations of force was demonstrated. The magnitude of the calculated CPD is significant for the wires of subatomic diameters only. An analytical expression for the dependence of the potential well depth on the Fermi energy for the semi-infinite metallic system is tested for low-dimensional structures. [Remarks: Notwithstanding that expressions derived in \[15\] successfully describe character of the behavior of the electron work function (of positive value) for semi-infinite metals and ionization potentials for metallic clusters, in our view, a usage of the classical electrostatics concept (image force) to define the quantum characteristic - work function - is not whole appropriate because it cannot unequivocally determine the sign of the emitted particle. For example, the positron work function is positive for sodium and
negative for aluminum in spite of identical action of the image force on this particle [43].

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