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Effect of Electron Overheating on the Tunneling Current of a Molecular Transistor

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Abstract—The effect of overheating of the electron subsystem on the Coulomb blockade in a structure (molecular transistor) based on a metal cluster containing a finite number of atoms has been theoretically studied. The electron energy spectrum in such quantum grains of cylindrical and spherical shape has been calculated. An increase in the electron subsystem temperature in the cluster leads to vanishing of the current gap and pronounced smoothening of the quantum and Coulomb steps on the current–voltage characteristic of the structure, in agreement with experimental observations.

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Island films containing small metal clusters (grains) are promising objects in nanotechnology [1–3]. In tunnel structures, the charging of such grains leads to the so-called Coulomb blockade of the current, which is manifested by steps (Coulomb ladder) appearing on a quite low background of thermal fluctuations on the current–voltage (*I–V*) characteristic (see, e.g., [4]). The elementary tunnel structure can be schematically modeled by a sandwich [1, 3] comprising a massive gold film (emitter) covered by a thin dielectric film with a permittivity of $\kappa \approx 3$, on which monatomic-height islands (disk-shaped clusters) [1] or almost spheroidal clusters [3] of gold are formed; the role of the third electrode in this molecular transistor structure is played by the point of a tunneling microscope.

Recently, we have studied [5] some features of the experimental *I–V* curves of such cluster structures. However, the fact that the steps on the *I–V* curves of a grain consisting of several dozen atoms (i.e., a system with a quantized spectrum) are significantly smoothened remained unclear. This feature is characteristic of such molecular structures [6]. In addition, Wang et al. [1] reported that, as the temperature was increased from 5 to 300 K, the current gap of a structure based on a disk-shaped cluster with a diameter of $2R \approx 4$ nm virtually completely vanished [1, Fig. 2].

This Letter shows that the smoothening of *I–V* curves of small metal clusters can be explained by overheating of the electron subsystem of the grain, which is caused by the relaxation of conduction electrons.

Let us consider monolayer disk-shaped island grains with diameters within $2R = 1$–8.5 nm, which corresponds to the number of atoms in these clusters from $N_0 \cong 14$ up to about 10^3. By the same token, for spherical gold grains with the diameters within $2R \cong 1.4$–2.8 nm, we have $N_0 \cong 100$–600. Note that the clusters of these dimensions obey the condition $L \gg R$, where L is the electron mean free path in the bulk metal. Calculations of the electron energy spectra for the cylindrical and spherical potential wells of finite depth give different values of distances between the lowest unoccupied (LU) and highest occupied (HO) energy levels, $\Delta\varepsilon_p = \varepsilon^{LU} - \varepsilon^{HO}$, in magic clusters with the indicated dimensions (Fig. 1). In non-magic clusters, these energies coincide ($\varepsilon^{LU} = \varepsilon^{HO}$).

The energy of a charged grain can be defined as $\tilde{E}_C = e^2/C$, where e is the elementary charge and C is

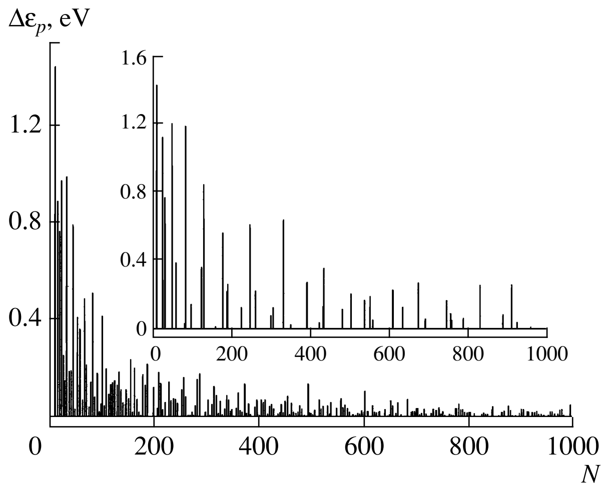

Fig. 1. Electron energy spectra of neutral Au_{N_0} clusters of cylindrical and (inset) spherical shapes.

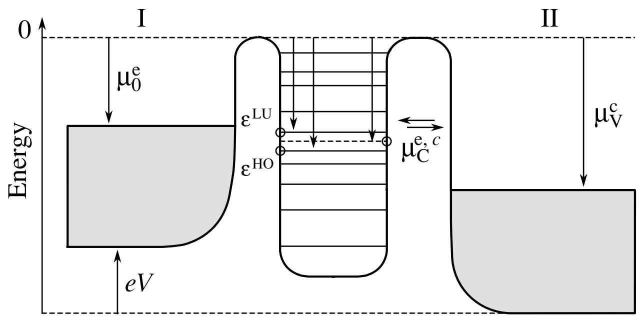

Fig. 2. Energy diagram of a cluster structure for a direct branch of the *I–V* curve: (I) emitter; (II) collector (see the text for explanations).

the electric capacitance of a separate spherical grain in vacuum (for a disk, the capacitance can be we estimated by considering it an oblate spheroid on the same volume). However, the results of calculations [5] showed that this approximation does not provide correct evaluation of the width of the current gap, especially in the case of a disk, which has about half of its surface in contact with the dielectric film (for such grains, we will introduce an effective capacitance by replacing $C \Rightarrow (1 + \kappa)C/2$. It should be noted that the capacitance is highly sensitive to the grain shape, so that even small deviations from the sphericity significantly change the C value. The mutual capacitances cannot be taken into account in a simple way, and this factor will be ignored. In these terms, we have $\tilde{E}_C \cong 1.6$–0.21 and 1.82–1.06 eV for the disks and spheres, respectively, with the dimensions indicated above.

A deformation of the phonon spectrum of a grain leads to weakening of the electron–phonon interaction, so that $\vartheta_F/R \cong \omega_D$, where ϑ_F is the velocity of electrons at the Fermi level and ω_D is the Debye frequency. This interaction can be suppressed to such an extent that the electron–electron collisions become the main mechanism of dissipation of the energy introduced in to a particle (a current of $I \sim 1$ pA is carried by $\sim 10^6$ electrons per unit time). This energy leads to overheating of the electron subsystem, which obeys the Fermi statistics with a certain effective (elevated) temperature, while the temperature of the ion subsystem changes rather insignificantly [7]. As the bias voltage V is increased, the fraction of conduction electrons involved into relaxation in the grain significantly increases due to a growth in the flux of electrons tunneling toward one of the electrodes (Fig. 2). This fraction includes all electrons occurring within an energy interval of $e\eta V$ below the Fermi level (ηV is the fraction or part of the bias voltage drop per grain). This leads to the appearance of loss channels, which are related to the generation of holes at the occupied levels, followed by their recombination with the excitation of plasma oscillations, the energy released upon the recombination of holes can be radiated as a light quantum of transferred to the ion subsystem (or the substrate) via excitation of the vibrational degrees of freedom. Even i the case of a significant overheating of the electron subsystem, the grain does not exhibit fragmentation, which is confirmed by the *I–V* curves reproduced in the course of cyclic variations of the bias voltage [1, 3]. Apparently, the heat is removed via the dielectric film, which is in a direct contact with the grains (especially, with disk-shaped ones). Thus, the dielectric film can be considered as both sink and source of phonons. Semrau et al. [8] considered such a phonon reservoir as a necessary condition for the momentum and energy conservation in the course of electron tunneling between various energy levels of grains in a system comprising a chain of grains (of various dimensions) packed in a DNA molecule, which was grafted to a substrate.

The energy pumped by the conduction electrons into the grains of island films was estimated at ~0.2–0.3 eV per electron [9–11]. Therefore, the experiments [1, 3] correspond, in the entire range of grain dimensions R and reasonable values of effective temperatures $T_{\rm eff}^g$ in the region of the current gap, to a regimes with

$$\tilde{E}_C > \Delta\varepsilon_F \geq k_B T^{e,c,g},$$

where $\Delta\varepsilon_F$ is a difference between the discrete energy levels near the Fermi energy in the grain and k_B is the Boltzmann constant. For a comparison with the experimental results [1, 3], our calculations were performed for the emitter (T^e) and collector (T^c) temperatures equal to the thermostat temperature ($T^e = T = 5$, 30, and 300 K) and for $T_{\rm eff}^g = T^e = 3000$ K.

According to a simple model [5, 12], let us represent the emitter and collector as the electron reservoirs with continuous spectra described by the Fermi distribution functions

$$f(\varepsilon^{e,c} - \mu_0^{e,c}) = \{1 + \exp[\varepsilon^{e,c} - \mu_0^{e,c})/k_B T^{e,c}\}^{-1}, \quad (1)$$

where $\mu_0^{e,c} = W_0^{e,c}$ is the chemical potentials of conduction electrons in a semi-infinite metal cluster and $W_0^{e,c}$ is the work function for the semi-infinite metal (for Au, $W_0 = 5.13$ eV; here and below, the energies in all cases are measured fro the vacuum level). In the quantum case, the chemical potential μ^g of electrons in the grain is determined from the condition of normalization at $T_{\rm eff}^g$ as

$$\sum_{p=1}^{\infty} \{1 + \exp[(\varepsilon_p - \mu^g)/k_B T_{\rm eff}^g]\}^{-1} = N. \quad (2)$$

where summation is carried out over all single-particle states and N is the total (average) number of conduction electrons in the grain (including both valence and excess electrons). The spectrum of states is assumed to be known and, hence, Eq. (2) can be used to determine the chemical potential of neutral Au_{N_0} grains as a function of the temperature. For $T^{g}_{eff} = 0$, the Fermi level of a non-magic cluster coincides with a discrete ε^{HO} level in the cluster. For a magic cluster, the Fermi energy is in a gap between terms. As expected, the temperature dependence of the Fermi energy occurs in the middle between the ε^{LU} and ε^{HO} terms.

The current passing through a quantum grain (with limitations imposed on the Coulomb instability [5, 12]) is determined as

$$I^e \equiv -e\sum_{n_{min}}^{n_{max}} P_n(\overrightarrow{\omega^e_n} - \overleftarrow{\omega^e_n}) = -e\sum_{n_{min}}^{n_{max}} P_n(\overrightarrow{\omega^c_n} - \overleftarrow{\omega^c_n}) \equiv I^c, \quad (3)$$

where P_n (probability that n excess electrons occur on the average in the island) is determined by solving a determinant equation in the stationary case. In fact, we calculate the reduced current defined as $\tilde{I} \equiv I/(eP_0\Gamma^{e,c})$, where $\Gamma^{e,c}$) are the tunneling rates; the quantities $P_{n\neq 0}/P_0$ are calculated from recurrent relations as $P_{n+1} = P_n\omega^{in}_n/\omega^{out}_{n+1}$. The total fluxes of electrons from the terminal electrodes to and from the grain,

$$\omega^{in}_n = \overrightarrow{\omega^e_n} + \overleftarrow{\omega^c_n}, \quad \omega^{out}_n = \overrightarrow{\omega^e_n} + \overleftarrow{\omega^c_n}$$

are expressed vie the corresponding partial fluxes and rates as

$$\overrightarrow{\omega^e_n} = 2\Gamma^e\sum_p f(\overrightarrow{\varepsilon^e} - \mu^e_V)[1 - f(\overrightarrow{\varepsilon^e} - \overrightarrow{\mu^e_C})], \quad (4)$$

$$\overleftarrow{\omega^c_n} = 2\Gamma^c\sum_p f(\overleftarrow{\varepsilon^c} - \mu^c_V)[1 - f(\overleftarrow{\varepsilon^c} - \overleftarrow{\mu^c_C})], \quad (5)$$

$$\overleftarrow{\omega^e_n} = 2\Gamma^e\sum_p [1 - f(\overleftarrow{\varepsilon^e} - \mu^e_V)]f(\overleftarrow{\varepsilon^e} - \overleftarrow{\mu^e_C})], \quad (6)$$

$$\overrightarrow{\omega^c_n} = 2\Gamma^c\sum_p [1 - f(\overrightarrow{\varepsilon^c} - \mu^c_V)]f(\overrightarrow{\varepsilon^c} - \overrightarrow{\mu^c_C})]. \quad (7)$$

For the direct I–V branch, assuming $V = 0$ and considering electron transitions from the emitter to grain and in the reverse direction, we can determine the energy resonances involved in the charge transfer process. As a result of the emitter ionization and the electron transfer to or from a grain containing n excess electrons, we obtain

$$\overleftrightarrow{\varepsilon^e} = \varepsilon'_p + \tilde{E}_C(n \mp 1/2) - e\eta^+V, \quad (8)$$

where the upper and lower arrows on the left-hand side correspond to the upper and lower signs on the right-hand side, respectively. By the same token, we obtain relations for the electron transitions between the grain and the collector,

$$\overleftrightarrow{\varepsilon^c} = \varepsilon'_p + \tilde{E}_C(n \mp 1/2) + e(1 - \eta^+)V, \quad (9)$$

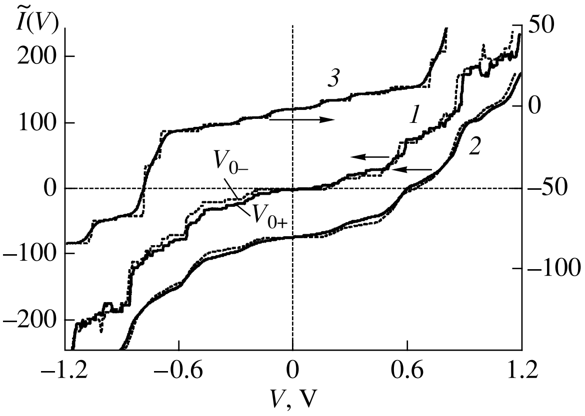

Fig. 3. Calculated I–V curves for magic Au_{230} disk-shaped and Au_{256} spherical clusters: (*1*) Au_{230}, $T^{e,c,g} = 5$ K (dotted curve), $T^{e,c} = 5$ K, $T^{g}_{eff} = 3000$ K (solid curve); (*2*) Au_{230}, $T^{e,c,g} = 300$ K (dotted curve), $T^{e,c} = 300$ K, $T^{g}_{eff} = 3000$ K (solid curve); (*3*) Au_{256}, $T^{e,c,g} = 300$ K (dashed curve), $T^{e,c} = 300$ K, $T^{g}_{eff} = 3000$ K (solid curve).

where $\varepsilon'_p = \varepsilon_p - e\delta\phi$ and η^+ is the bias voltage fraction on the direct I–V branch. Prior to the field application, a contact potential difference $\delta\phi = (\mu^g - \mu^e)/e$ exists between the grain and emitter, which leads to charging of the grain with the residual charge $Q^{eff}_0 = C\delta\phi$. The value of this effective charge is fractional, which is related to the fact that the electron wave function in structures with permeable barriers can be redistributed between the electrodes, thus influencing the energy pattern. We have calculated the ε_p and μ^g spectra for a separate grain in the absence of charging and external fields. It is also assumed that neither en external field nor the Coulomb blockade lift up the degeneracy. With allowance for the applied voltage and the charging of grains, the electron spectra of the grain and collector shift as

$$\mu^e_V \equiv \mu^e_0, \quad \overleftrightarrow{\mu^e_C} = \mu^g - e\delta\phi + \tilde{E}_C(n \mp 1/2) - e\eta^+V,$$

The reverse branch of the I–V curve can be readily calculated by merely switching the polarity to $V = 0$ on the left electrode (now it is the collector) and $V > 0$ on the right electrode (now it is the emitter). Accordingly, the new voltage fraction is $\eta^- = 1 - \eta^+$.

Figure 3 shows the I–V curves calculated as described above the current gap width $\Delta V_g = V_{0+} + |V_{0-}|$ is determined by the threshold voltage V_{0+}/V_{0-}- corresponding to a zero collector/emitter current on the direct/reverse branch of the I–V curve. In the low-temperature limit ($k_B T_{eff}^g \ll \Delta\varepsilon_F$), the gap width is given by an analytical expression:

$$\Delta V_g = \left(\frac{1}{2e}\tilde{E}_C + \frac{1}{e}\Delta\varepsilon\right)\left[\frac{1}{2-\eta^+} + \frac{1}{2-\eta^-}\right], \qquad (10)$$

where $\Delta\varepsilon \equiv \varepsilon^{HO} - \mu^g \geq 0$. The ΔV_g values

calculated using this formula agree well with the experimental data for the transistor structures on spherical and disk-shaped clusters.

In the case of $k_B T_{eff}^g \geq \Delta\varepsilon_F$, where a part of the spectrum greater than $\delta\varepsilon_F$ is involved in the charge transfer, the *I–V* curve ad the current gap width can be calculated only using numerical methods.

Calculations show that the *I–V* curve smoothening clearly depends on the electron subsystem temperature in the grains. However, to provide fitting to the measured *I–V* curves, it is necessary to assume that electrons in the emitter ad collector are also heated to a certain temperature $T_{eff}^{e,c}$, which is higher than the thermostat temperature. In order to demonstrate this situation, we have performed calculations for $T_{eff}^{e,c}$ = 300 K and T_{eff}^g = 3000 K. As can be seen, only the aforementioned assumption allows the *I–V* curve smoothening in metal cluster structures at low thermostat temperatures to be explained. As the bias voltage increases, the current passage proceeds on the background of the growing electron gas temperature.

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