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Correlated Single-Electron Tunneling via Mesoscopic Metal Particles: Effects of the Energy Quantization

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The existing theory of correlated single-electron tunneling in the double normal-metal tunnel junction is extended to the case of an ultrasmall central electrode of the structure. It is shown that the form of the I-V *curve of such a system depends on the energy relaxation rate in this electrode. For realistic values of the relaxation rate, the large-scale shape of the dc* I-V *curve, which is associated with Coulomb correlations, is close to that following from the earlier theory. However, the* I-V *curve should also exhibit small-scale singularities reflecting the structure of the energy spectrum of the central electrode.*

1. INTRODUCTION

The Coulomb interaction of the tunneling electrons in small tunnel junctions gives rise to correlations between different tunneling events (for reviews, see Refs. 1 and 2). These correlations show up in the dc I-V curves of small tunnel junctions and systems of such junctions. Specifically, in the case of two junctions connected in series, the correlation between events of tunneling through different junctions results in the "Coulomb staircase," a periodic modulation of the dc I-V curve of the system reflecting the stepwise increase of an electric charge Q of its middle electrode. Such modulation has been observed in several experiments.[3-7]

Existing "orthodox" theory of correlated single electron tunneling[1,2] assumes that the energy spectra of the junction electrodes are continuous, an assumption that is valid for not too small particles ($\gtrsim$10 nm or so). The energy spectra of the smaller electrodes are discrete due to the quantum-size

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effect. It seems important to extend the orthodox theory to the case of such junctions, particularly in view of possible applications of the single-electron tunneling in molecular electronics.[1,2,8] In the present work we make this extension for the simplest case of the double junction formed by an ultra-small normal metal particle and two bulk external electrodes. This case is realistic from an experimental point of view, since such a system is quite feasible using the scanning tunnel microscope technique.[5-7]

The work is organized as follows. In the next section the master equation is written down, which describes electron tunneling through the junctions with an appropriate account of the energy spectrum discreteness of the middle electrode of the system. Starting from this master equation, we study the large-scale form of the *I-V* curve of the double-junction system (Sec. 3) and its fine structure (Sec. 4). In the last section we discuss some practical consequences of our calculations.

2. MASTER EQUATION

Let us consider the double tunnel junction formed by a small, normal metal particle placed between two bulk external electrodes. We shall assume that although the particle is small, it is still mesoscopic, i.e., contains a large number of atoms ($N \geq 10^2$-10^3). The average spacing Δ between energy levels of such a particle can be estimated as $(\nu(0)r^3)^{-1}$, where $\nu(0)$ is a density of states at the Fermi level of the metal the particle is made of and r is its radius. In the mesoscopic particle, the spacing Δ is much less than the characteristic charging energy $E_C \simeq e^2/4\pi\bar{\varepsilon}\varepsilon_0 r$ (here $\bar{\varepsilon}$ is the dielectric constant of the tunneling barriers), since these energies are comparable for a single atom, and with increasing N, Δ decreases much faster ($\propto N^{-1}$) than the charging energy ($\propto N^{-1/3}$).

The condition $\Delta \ll E_C$ ensures that the penetration depth λ_F of the static electric field $\lambda_F \simeq (e^2\nu(0)/\varepsilon_0)^{-1/2}$ is small in comparison to the particle radius, so that the usual formula[1,2,9] can be used to calculate the electrostatic energy U of the double-junction system

$$U(n_1, n_2) = \frac{Q^2}{2C_\Sigma} - \frac{eV}{C_\Sigma}(C_1 n_2 + C_2 n_1) \tag{1a}$$

where

$$Q = en + Q_0, \qquad n = n_1 - n_2 \tag{1b}$$

Here, C_j is the capacitance of the jth junction, n_j is the number of electrons that have tunneled through it, V is a total voltage across the system, and $C_\Sigma \equiv C_1 + C_2$. (In the considered case of the junctions formed by the small particle, the total capacitance C_Σ should not differ much from the capacitance $4\pi\bar{\varepsilon}\varepsilon_0 r$ of the particle itself). In Eq. (1b), Q_0 is a noninteger part of the electric charge of the particle (measured in the e units) that can be induced, for example, by an external electric field.[1,2]

The total Hamiltonian of the double junction includes the electrostatic energy U, the internal energies of the electrodes H_j, the particle H_p, and the standard tunnel Hamiltonians H_{Tj} $(j=1,2)$

$$H = U + H_1 + H_2 + H_p + H_{T1} + H_{T2} \tag{2}$$

All the parts of the Hamiltonian can be expressed via operators c^+, c of creation and annihilation of electrons in the electrodes k_j, and the particle m. In particular

$$H_{Tj} = \sum_{k_j, m} T_{k_j, m} c^+_{k_j} c_m + h.c. \tag{3}$$

$$H_p = \sum_m \varepsilon_m c^+_m c_m + H' \tag{4}$$

where $\{\varepsilon_m\}$ is the energy spectrum of the particle, and H' describes electron energy relaxation in it.

Coulomb correlations of the electron tunneling can take place only in the junctions with small tunnel conductances G_j, $G_j \ll R_Q^{-1}$, $R_Q \equiv \pi\hbar/2e^2$. This condition implies that the electron tunneling rates $\Gamma_j(\varepsilon_m)$, $\Gamma_j(\varepsilon_m) = 2\pi\langle|T_{k_j,m}|\rangle^2 N_j(0)/\hbar$, are small enough, $\hbar\Gamma_j(\varepsilon_m) \ll \Delta$. Hence, at not too low temperatures, $T \gg \hbar\Gamma_j$, one can treat the terms H_{Tj} in Hamiltonian (2) as a perturbation and describe the tunneling by a simple master equation for the probaiblity density $\rho_n(\varepsilon_m)$.[1,2] If the energy relaxation rate τ_ε^{-1} is also small, $\hbar/\tau_\varepsilon \ll \Delta$, this equation takes the form

$$\begin{aligned} \dot{\rho}_n(\varepsilon_m, t) = S_n - S_{n-1} + \sum_{j=1,2} (\Gamma_j(\varepsilon_m)[\sigma_{n-1} g(\varepsilon_m - E_j(n))(1 - f_{n-1}(\varepsilon_m)) \\ - \sigma_n(1 - g(\varepsilon_m - E_j(n)))f_n(\varepsilon_m)]) + F_\varepsilon \end{aligned} \tag{5a}$$

$$\begin{aligned} S_n = \sum_j \sum_{m'=m} \Gamma_j(\varepsilon_{m'})\{\sigma_{n+1}[1 - g(\varepsilon_{m'} - E_j(n+1))] f_{n+1}(\varepsilon_{m'}) f_{n+1}(\varepsilon_m) \\ - \sigma_n g(\varepsilon_{m'} - E_j(n+1))(1 - f_n(\varepsilon_{m'})) f_n(\varepsilon_m)\} \end{aligned} \tag{5b}$$

$$\rho_n(\varepsilon_m) = \sigma_n f_n(\varepsilon_m), \quad \sum_n \sigma_n = 1, \quad f_n(\varepsilon_m) \to 1 \quad \text{when } \varepsilon_m \to -\infty \tag{5c}$$

where $g(\varepsilon)$ is the Fermi distribution function, $E_j(n)$ are the changes of electrostatic energy (1) at electron tunneling from the particle to the jth external electrode, σ_n and $f_n(\varepsilon_m)$ are the probabilities to find exactly n "excess" electrons on the particle and the corresponding energy distribution functions, respectively, and the term F_ε describes the energy relaxation.

The matrix elements $T_{k_j,m}$ can be calculated according to the usual formula[10]

$$T_{k,m} = \text{const}(U - \varepsilon_m)$$

$$\times \exp\left\{-a\lambda\left(1+\frac{\kappa^2}{2\lambda^2}\right)\right\} \int_V dr\,\psi_m(r) \exp\{i\boldsymbol{\kappa}\boldsymbol{\rho} - z(\lambda^2+\kappa^2)^{1/2}\}, \tag{6}$$

$$k = \{\boldsymbol{\kappa}, k_z\}, \qquad r = \{\rho, z\}, \qquad \lambda = [2m(U-\varepsilon_m)]^{1/2}/\hbar$$

where the z axis is perpendicular to the tunnel barrier plane, a and U are its width and height, respectively, $\psi_m(r)$ is a wavefunction of the mth energy level, and integration in the first of Eqs. (6) is carried out over the volume of the particle. Since the mesoscopic particle has inevitable irregularities of shape at least on the atomic level, the electron motion in it can be treated as ergodic. In this case the energy spectrum $\{\varepsilon_m\}$ of the particle is random (see, e.g., Ref. 11), and the amplitude $|\psi_m(r)|$ of the wave functions averaged over distances of the order of k_F^{-1} (k_F is the Fermi-level wavenumber) is constant.[12] As a consequence, under the assumptions $\lambda \ll k_F$ and $\varepsilon_m \ll U$, the matrix elements $T_{k_j,m}$ and, hence, the tunneling rates Γ_j, are independent of m. In what follows we shall accept these assumptions, although the main results should remain qualitatively valid also in the case of m-dependent Γ_j.

Solving Eq. (5) for the stationary probability density $\rho_n(\varepsilon_n)$, one can find the dc I-V curve of the double junction. The form of the curve on the voltage scale E_C/e ($E_C \equiv e^2/2C_\Sigma$) is determined by the Coulomb correlations between tunneling electrons, while on the voltage scale Δ/e it reflects the structure of the particle energy spectrum. Since in our case of mesoscopic particles $E_C \gg \Delta$, we can discuss features of these two characteristic scales separately.

3. GLOBAL STRUCTURE OF THE *I-V* CURVE

We begin with a discussion of the large-scale features arising due to the Coulomb correlations. In order to calculate them, it is convenient to transform the master equation (5) into two equations, one for the probabilities σ_n and another for the energy distribution function $f(\varepsilon_m)$ averaged over σ_n, $f(\varepsilon) \equiv \sum_n \sigma_n f_n(\varepsilon_m)$

$$\dot{\sigma}_n = S'_n - S'_{n-1} \tag{7a}$$

$$S'_n = \sum_{j,m} \Gamma_j\{\sigma_{n+1}[1 - g(\varepsilon_m - E_j(n+1))]f_{n+1}(\varepsilon_m)$$

$$- \sigma_n g(\varepsilon_m - E_j(n+1))(1 - f_n(\varepsilon_m))\} \tag{7b}$$

$$\dot{f}(\varepsilon_m) = \sum_j \Gamma_j \sum_n \sigma_n \{g(\varepsilon_m - E_j(n+1))(1 - f_n(\varepsilon_m))$$
$$- (1 - g(\varepsilon_m - E_j(n)))f_n(\varepsilon_m)\} + F_\varepsilon \qquad (8)$$

Further calculations are simplified considerably by the condition $E_C \gg \Delta$. First of all, it allows one to neglect the n-dependence of the distribution functions $f_n(\varepsilon_m)$, since particular events of electron tunneling cannot influence $f_n(\varepsilon_m)$ considerably while changing n significantly in this case. (The distribution functions can be changed only by the large number, $\simeq E_C/\Delta$, of the tunneling events.) Then the probabilities σ_n reach their stationary values much faster than $f(\varepsilon_m)$, so that solving Eq. (8) for $f(\varepsilon_m)$, one can use the stationary values of σ_n corresponding to the instantaneous distribution function $f(\varepsilon_m)$. Lastly, the summation over m in Eq. (7b) can be replaced by an integration. As a result of these transformations, we arrive at the following set of equations

$$\sigma_{n+1}/\sigma_n = \left[\sum_j (G_j/e^2) \int d\varepsilon (1 - g(\varepsilon - E_j(n+1))f(\varepsilon)\right]^{-1}$$
$$\times \sum_j (G_j/e^2) \int d\varepsilon\, g(\varepsilon - E_j(n+1))(1 - f(\varepsilon)), \qquad (9a)$$

$$\dot{f}(\varepsilon, t) = \sum_j \Gamma_j \sum_n \sigma_n [g(\varepsilon - E_j(n+1))(1 - f(\varepsilon))$$
$$- (1 - g(\varepsilon - E_j(n)))f(\varepsilon)] + F_\varepsilon \qquad (9b)$$

where $G_j = ce^2\Gamma_j/\Delta$, and $c = 2$ for small magnetic field H ($H \lesssim \nu/\mu$, where ν is the width of the energy levels and μ is the Bohr magneton) when all energy levels are twofold degenerate, and $c = 1$ otherwise. Equations in (9) are reasonably simple and can be readily solved numerically. Before that, one needs to specify the energy relaxation term F_ε, which generally depends on the energy in a complicated way. We shall adopt the simplest model that makes possible a qualitative discussion of the energy relaxation: $F_\varepsilon = [g(\varepsilon) - f(\varepsilon)]/\tau_\varepsilon$, $\tau_\varepsilon = \text{const}$.

Figure 1 shows results of the numerical solution of Eqs. (9) in the two limiting cases of negligibly small- and large-energy relaxation rate. If the relaxation rate is large, $\tau_\varepsilon^{-1} \gg \Gamma_j$, i.e., $\tau_\varepsilon^{-1} \gg \tau_j^{-1}\Delta/E_C$, where $\tau_j^{-1} \equiv G_j/C_\Sigma$, the current flow through the system does not disturb the equilibrium of electrons in the particle, and Eq. (9a) coincides with the corresponding equation of the orthodox theory.[1,2] Thus in this case the energy spectrum discreteness (or, it is better to say, the finite density of energy states) does not influence the large-scale form of the dc I-V curve.

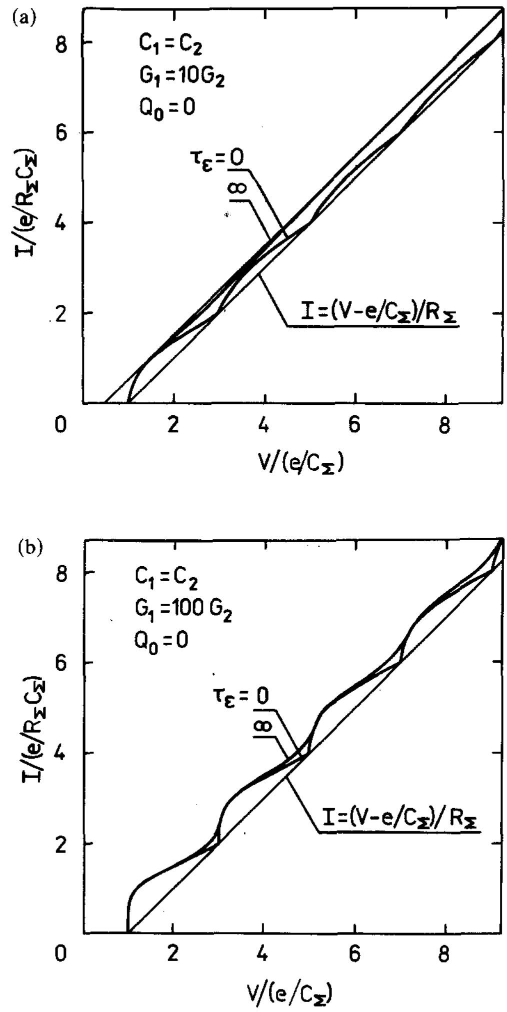

Fig. 1. Large-scale form of the *I-V* curve of two junctions connected in series at large ($\tau_\varepsilon^{-1} \gg \tau_j^{-1}\Delta/E_C$) and small energy relaxation rates for the middle electrode of this system for (a) not very strong and (b) very strong difference between junction conductances.

When the relaxation rate is small, $\tau_\varepsilon^{-1} \ll \Gamma_j$, the electron distribution function can fall into non-equilibrium. Such an "overheating" suppresses the Coulomb correlations and, hence, the large-scale singularities of the *I-V* curve (Fig. 1a). In the case of strong inequality of the junction conductances, however, the tunneling through the junction with the higher conductance serves as an energy relaxation mechanism, and this effect vanishes (Fig. 1b).

At large voltages, $B \geqslant (e/C_\Sigma)(G_1/G_2)$ (for definiteness we shall assume that $G_1 > G_2$), the *I-V* curve reaches its linear asymptote, $I=(V-V_{of})/R_\Sigma$, $R_\Sigma = (G_1^{-1}+G_2^{-1})^{-1}$, with the voltage offset V_{of} depending on τ_ε[13] (see also Fig. 1)

$$V_{of} = (e/2C_\Sigma)(1+[1+\Delta\tau_\varepsilon/ce^2R_\Sigma]^{-1}) \tag{10}$$

Thus, for $\tau_\varepsilon^{-1} \ll \Gamma_j$ $(\Delta\tau_\varepsilon/ce^2R_\Sigma \gg 1)$, the offset is two times smaller than in the usual case when $\Delta\tau_\varepsilon/ce^2R_\Sigma \ll 1$ (large energy level density or relaxation rate).

4. FINE STRUCTURE OF THE *I-V* CURVE

We shall now discuss the small-scale singularities of the *I-V* curve, which is directly related to the discreteness of the energy spectrum of the particle. In fact, due to such discreteness, the current flowing through the junctions should increase stepwise with increasing voltage. Each step is located at voltages at which the Fermi level in one of the external electrodes coincides with one of the energy levels of the particle. The fine structure of the *I-V* curve should be most pronounced at medium-energy relaxation rates, $\Delta/\hbar \gg \tau_\varepsilon^{-1} \gg \Gamma_j$, when the particle energy spectrum is still discrete, but on the other hand electrons in the particle are in thermodynamic equilibrium. In this case, at low temperatures, $T \ll \Delta$, the energy distribution function has a sharp edge on the energy scale of Δ. Below we shall discuss exactly this range of the relaxation rates.

Let us consider the situation when the Fermi level in one of the external electrodes approaches the energy level ε_m of the particle with p excess electrons in it (taking into account the shift of the particle Fermi level due to electron tunneling), that is, $\varepsilon_m = E_j(p)$. Such coincidence takes place at $V=V_0$, where $V_0 = (-1)^{j+1}[C_\Sigma\varepsilon_m/e + ep + Q_0 - e/2]/C_{j'}$, $j, j' = 1, 2$, $j \neq j'$, and results in the differential conductance peak centered to V_0. According to Eq. (8) the shape of the peak is given by the expression

$$G(V) = bc\Gamma_j(\varepsilon_m)e^2(C_{j'}/C_\Sigma)g'(e(V-V_0)C_{j'}/C_\Sigma)$$

$$g'(z) = (1/4T)ch^{-2}(z/2T), \qquad b = (\partial I/\partial I_{j,p})|_{V=V_0}, \tag{11}$$

where the constant b can be determined from Eqs. (7) (in the case considered $(\tau_\varepsilon^{-1} \gg \Gamma_j)$ these equations coincide with those of the orthodox theory); $I_{j,p}$ is a current flowing through the jth junction at $n=p$.

Equation (11) shows that the width of the peak is determined by the temperature smoothing of the edges of the energy distribution functions in the external electrodes, $\nu \simeq T$. This is true only for not-too-low temperatures

$T \gg \hbar/\tau_\varepsilon, \Gamma_j$. At $T \lesssim \hbar/\tau_\varepsilon, \Gamma_j$ the master equation (5) and Eq. (11) are not valid, and ν is determined by the width of the energy level itself, $\nu \simeq \hbar(\tau_\varepsilon^{-1}+\Gamma_1+\Gamma_2)$[14,15].

The constant b in Eq. (11) can be calculated from Eqs. (7) explicitly in several cases. When $G_1 \gg G_2$, $b=1$ for $j=2$, $p=\mathrm{Int}[(C_2V-Q_0+e/2)/e]$, and $b=0$ for any other values of (j,p). In the voltage region just above the Coulomb blockade threshold, in which the Coulomb blockade is lifted only in one of the junctions (say, the first one), $(e/2+Q_0)/C_2<V<\min\{(e/2-Q_0)/C_1, (3e/2+Q_0)/C_2\}$ there are only two nonvanishing probabilities σ_0, σ_1

$$\sigma_0=\frac{-G_2E_2}{G_1E_1-G_2E_2}, \qquad \sigma_1=\frac{G_1E_1}{G_1E_1-G_2E_2}$$

$$E_1=e(VC_2-e/2-Q_0)/C_\Sigma, \qquad E_2=-e(VC_1+e/2+Q_0)/C_\Sigma \tag{12}$$

and $b=\sigma_0^2\equiv b_1$, $b=\sigma_1^2\equiv b_2$ for the conductance peak arising due to the "resonance" in the first ($\varepsilon_m=E_1$) and the second ($\varepsilon_m=E_2$) junction, respectively.

If one of the junction conductances is much greater than another, say $G_1 \gg G_2$ (this case is most frequently encountered in experiments), the conductance peaks arise due to resonances at one set of (j,p) values. In this case the system of peaks reflects directly the structure of the particle energy spectrum.

The energy spectrum of the particle with irregular shape can be described (see, e.g., Ref. 11) by the random matrix theory, and for $G_1 \gg G_2$ the well-known results of this theory immediately yield the distribution of the voltage intervals δV between nearest-neighbor conductance peaks

$$f_\beta(x)=A_\beta x^\beta \exp(-B_\beta x^2), \qquad x=\delta V/\langle\delta V\rangle, \qquad \langle\delta V\rangle=cC_\Sigma\Delta/2C_1e \tag{13}$$

Here $\beta=1, 2, 4$, $A_\beta=\pi/2$, $32/\pi^2$, $(64/9\pi)^3$, $B_\beta=\pi/4$, $4/\pi$, $64/9\pi$ for the following cases, respectively: 1) weak spin-orbit interaction H_{so} in the particle and weak magnetic field H, $|\langle H_{so}\rangle|$, $\mu H \ll \Delta$ (orthogonal ensemble of the random matrices); 2) strong spin-orbit interaction and strong magnetic field (unitary ensemble); 3) strong spin-orbit interaction and weak magnetic field (symplectic ensemble).

A characteristic feature of the distributions (13) is the "repulsion" of peaks, $f_\beta(x)\to 0$ for $x\to 0$. For weak spin-orbit interaction and strong magnetic field, there is no correlation between the energy levels for opposite spins, so that the repulsion of the level and, accordingly, that of the conductance peak, vanishes. The nearest-neighbor spacing distribution in this case is given approximately by $f_2(x)$—see Eq. (18b) below.

The repulsion of energy levels results also in characteristic oscillations of two-level correlation function $R(x)$[16]

$$1-R(x)=\begin{cases} s^2(x)+(ds/dx)\displaystyle\int_x^\infty s(t)\,dt, & \beta=1, \\ s^2(x), & \beta=2, \\ s^2(2x)-(ds(2x)/dx)\displaystyle\int_0^x s(2t)\,dt, & \beta=4, \end{cases} \tag{14}$$

$$s(x)=\sin(\pi x)/\pi x$$

If the conductance peaks are sufficiently narrow, $\nu \ll \Delta$, and $\Gamma_j(\varepsilon_m)=\langle\Gamma_j\rangle=\text{const}$, the conductance correlation function $\langle\tilde{G}(V')\tilde{G}(V'+V)\rangle=\langle\tilde{G}\tilde{G}(V)\rangle$, $\tilde{G}\equiv G-\langle G\rangle$, for $V\gg\nu/e$ is directly related to $R(x)$:

$$\langle\tilde{G}\tilde{G}(V)\rangle=\langle G\rangle^2(R(x)-1), \qquad \langle G\rangle=G_2C_1/C_\Sigma, \qquad x\equiv V/\langle\delta V\rangle \tag{15}$$

For $V\simeq\nu/e$ the conductance correlation function is determined by the shape of the peaks. Namely, for $T\gg\hbar(\Gamma_j,\tau_\varepsilon^{-1})$, $T\ll\Delta$

$$\langle GG(V)\rangle=\langle G\rangle^2 c(\Delta/16T)\int_{-\infty}^{+\infty} dx\, ch^{-1}(x)ch^{-2}\left(x+\frac{eVC_1}{2TC_\Sigma}\right) \tag{16a}$$

and, in particular,

$$\langle G^2\rangle=\langle G\rangle^2 c(\Delta/12T). \tag{16b}$$

One can calculate $\langle GG(V)\rangle$ at $V\simeq\nu/e$ and also for $T=0$ and $\tau_\varepsilon^{-1}=0$ by making use of a result[15] obtained for tunneling via an isolated energy level, according to which the conductance peak associated with this level has the Lorentzian shape with a width $\hbar(\Gamma_1+\Gamma_2)$. This result can be applied to the case considered of tunneling via the metallic particle (in spite of the Coulomb correlations between different tunneling events in this case), since the condition $E_C\gg\Delta$ ensures that a large number of energy levels participate in the tunneling, and the tunneling events via any two given levels can be treated as uncorrelated. In this way we obtain

$$\begin{aligned}\langle GG(V)\rangle&=\left(\frac{2ce^2}{\pi\hbar}\frac{\langle G\rangle}{G_1}\right)^2\int_{-\infty}^{+\infty}\frac{dV'}{\langle\delta V\rangle}\left[\left(\frac{4e^3V'C_1}{\Delta\hbar G_1C_\Sigma}\right)^2\right)\\ &\quad\times\left(1+\left(\frac{4e^3(V-V')C_1}{\Delta\hbar G_1C_\Sigma}\right)^2\right)\Bigg]^{-2}\\ &=\langle G\rangle^2 c\frac{e^2}{\pi\hbar G_1}\left[1+\left(\frac{2e^3V}{\Delta\hbar G_1}\right)^2\right]^{-2}\end{aligned} \tag{17}$$

Equations (14)-(17) and similarly Eqs. (19) and (20) below describe the mesoscopic conductance fluctuations (see, e.g., Ref. 15 and references therein) in the system of two tunnel junctions considered.

When the conductance peaks arise from resonances at several (j, p) values, they are related to the energy levels from several various parts of the particle energy spectrum, the energy difference between these parts being of the order of E_C ($E_C \gg \Delta$). Hence, the conductance peaks with different (j, p) values can be regarded as uncorrelated, and the "repulsion" of conductance peaks vanishes.

In particular, if the junction conductances are of the same order of magnitude, $G_1 \simeq G_2$, then, in the voltage region just above the Coulomb blockade threshold, the peaks arise at two sets of (j, p) values—see Eq. (12). If, for definiteness, $C_1 < C_2$, one can assume that the peaks related to resonances in the first junction divide at random part (C_1/C_2) of the intervals between nearest-neighbor peaks related to resonances in the second junction. Adopting this assumption, we arrive at the following expression for total nearest-neighbor spacing distribution

$$u(x) = ((C_2 - C_1)/C_\Sigma)f_\beta(x) + (2C_1/C_\Sigma)\tilde{f}_\beta(x), \qquad x = 2eVC_2/c\Delta C_\Sigma \tag{18a}$$

$$\tilde{f}_\beta(x) \equiv \int_x dy(f_\beta(y)/y)$$
$$= \begin{cases} (\pi/2)\,\mathrm{erfc}(\sqrt{\pi}\,x/2), & \beta = 1, \\ (4/\pi)\exp\{-(4/\pi)x^2\}, & \beta = 2, \\ (B_4/2)\exp\{-B_4x^2\}(1 + B_4x^2), & \beta = 4. \end{cases} \tag{18b}$$

The average interval is $c\Delta/2e$.

Besides this effect, the coexistence of the two uncorrelated subsystems of the conductance peaks makes the characteristic oscillations of $\langle\tilde{G}\tilde{G}(V)\rangle$ less pronounced. For $V \gg \nu/e$ ($\nu \ll \Delta$) it has the form

$$\langle\tilde{G}\tilde{G}(V)\rangle = \langle G\rangle^2 \sum_{j=1,2} \gamma_j \left[R\left(\frac{2eVC_1C_2}{c\Delta C_jC_\Sigma}\right) - 1\right], \qquad \gamma_j \equiv \left(\frac{b_jG_jC_1C_2}{\langle G\rangle C_jC_\Sigma}\right)^2 \tag{19}$$
$$\langle G\rangle = \sum_{j=1,2} b_jG_jC_1C_2/C_jC_\Sigma$$

For $V \simeq T/e$, $\Delta \gg T \gg \hbar(\Gamma_j, \tau_\varepsilon^{-1})$

$$\langle GG(V)\rangle = \langle G\rangle^2 \frac{c\Delta}{16T} \sum_{j=1,2} \gamma_j \int dx\, ch^{-2}(x) ch^{-2}\left(x + \frac{eVC_1C_2}{2TC_jC_\Sigma}\right) \tag{20a}$$

in particular,

$$\langle G^2\rangle = \langle G\rangle^2 c(\gamma_1 + \gamma_2)(\Delta/12T) \tag{20b}$$

In small magnetic fields, $H \ll \nu/\mu$, the energy levels are at least twofold degenerate due to time-reversal symmetry of electron motion in the particle. Strong magnetic fields break this symmetry and lift the degeneracy

$$\varepsilon_m \to \varepsilon_m \pm (1 - \rho)\mu H \tag{21}$$

At small spin-orbit interactions, $|\langle H_{so}\rangle| \ll \Delta$, the parameter ρ, which is generally dependent on m, can be estimated[11] as $|\langle H_{so}\rangle|^2/\Delta^2$. In the opposite limit, $|\langle H_{so}\rangle| \gg \Delta$, $\rho \to 1$. (The field-induced splitting of energy levels in this limit can be crudely estimated as $(\mu H)^2/|\langle H_{so}\rangle|$. Thus each conductance peak should be split by a magnetic field. The voltage interval between the arising two peaks and the shift of the middle of this interval equal $\delta V = 2(C_\Sigma/C_j)(1-\rho)\mu H/e$ and $(C_\Sigma/C_j)(2\rho-\rho^2)(\mu H)^2/e\Delta$, respectively.

5. ESTIMATES AND CONCLUSIONS

Now we shall discuss experimentally accessible values of the system parameters and evaluate the effects considered above. The most important parameters are the average level spacing Δ, charging energy E_C, and energy relaxation rate τ_ε^{-1}. Δ is not very sensitive to the material of the particle (at least for metallic particles) and ranges typically from $\simeq 10^{-4}$ eV (1 K) to $\simeq 0.1$ eV for particle diameters in the experimentally feasible interval 100–10 A.[11] Within this interval the charging energy E_C changes from 0.05 eV to 0.5 eV (for the typical value of the dielectric constant of the tunnel barriers, $\bar{\varepsilon} \simeq 5$).

To our knowledge, there are no experimental or thoretical data concerning the electron energy relaxation rate τ_ε^{-1} in small particles. For relatively large energies, $\varepsilon \gg \Delta$, it seems reasonable to use as an estimate of τ_ε^{-1} the bulk value of the relaxation rate, which in its turn can be estimated as $\varepsilon^2/\hbar\varepsilon_F$, where ε_F is the Fermi energy of the metal.

From these estimates it follows that the energy levels with $\varepsilon_m \simeq E_C$ should be considerably broadened ($\hbar/\tau_\varepsilon \gtrsim \Delta$) so that the spectrum of the particle in this energy range is quasicontinuous and the fine structure of the I-V curve is washed out. Since the conditions $G_j^{-1} \gg R_Q$, $\hbar/\tau_\varepsilon \gtrsim \Delta$ imply that $\tau_j \gg (\Delta/E_C)\tau_\varepsilon$, overheating effects also cannot take place, and the I-V curve of the junctions for voltages $V \simeq E_C/e$ should coincide with that derived from the orthodox theory.[1,2]

Hence not only the general concept of correlated single-electron tunneling, but also the quantitative picture of such tunneling as it is given by the orthodox theory, should be valid even for ultrasmall metal particles containing $\simeq 10^2$ atoms. This conclusion should apparently hold not only for the system considered in the present work, but also for more complex multijunctions systems. (Although for smaller particles the theory[1,2] ceases to be quantitatively correct, the general concept of correlated tunneling should still remain intact.)

In our present case of the double-junction system, the only difference with the predictions of the orthodox theory should be the fine structure of the I-V curve in the voltage region just above the Coulomb blockade threshold V_t ($V - V_t \simeq \Delta/e$). In fact, at such voltages the energy levels lying not far from the Fermi level alone do participate in the tunneling. Although the direct estimate of τ_ε^{-1} for these levels from the bulk value is not correct, it gives an upper bound on τ_ε^{-1}, since the discreteness of the electron and, perhaps, phonon spectra can, on an average, only make the relaxation rate smaller than the bulk value at the same energy ε. The bulk inelastic relaxation rate equals 10^{11}-10^{12} $\sec^{-1}$ at $\varepsilon \simeq 10$ K and increases as ε^2 with increasing ε.[17]

From this estimate one can conclude that the conductance peaks related to the energy levels near the Fermi level should be observable. Such observation can be carried out, however, only in a double-junction system whose I-V curve varies smoothly near Coulomb blockade threshold. In the most realistic case of strongly unequal junctions conductances, $G_1 \gg G_2$, this requirement implies that the capacitance of a poorly conducting junction is smaller than that of a better conducting one, $C_1 < C_2$. The observation of the conductance peaks would make it possible (for the first time, to our knowledge) to check directly the applicability of the random-matrix theory to small particles.

As we discussed above, the behavior of the conductance peaks in the magnetic field depends strongly on the magnitude of the spin-orbit interaction, which can be estimated from the bulk value of the shift δg of the g factor of the conduction electrons. Since the matrix elements of the spin-orbit interaction H'_{so} in the bulk samples are nonvanishing only for states in the different energy bands and[18] $|\langle H'_{so}\rangle| \simeq \delta g E$ (where E is characteristic intraband energy interval), then $H_{so} \simeq |\langle H'_{so}\rangle|^2/E \simeq (\delta g)^2 E$. From this estimate it follows, for example, that for $\Delta \simeq 10$ K the conditions of weak ($|\langle H_{so}\rangle| \ll \Delta$) and strong ($|\langle H_{so}\rangle| \gg \Delta$) spin-orbit interaction should be met in aluminum and gold particles ($\delta g = -5 \times 10^{-3}$ and $\delta g = 0.1$ [19]), respectively. It means, in particular, that according To Eq. (15) a magnetic field of 10 T should cause the splitting of the conductance peaks by $\delta V \simeq 100$ μV in the first case, while in the second case such a splitting should be practically unobservable, $\delta V \lesssim 1$ μV.

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