Influence of discrete energy spectrum on correlated single-electron tunneling via a mezoscopically small metal granule

D. V. Averin and A. N. Korotkov

Moscow State University
(Submitted 29 December 1989)
Zh. Eksp. Teor. Fiz. **97**, 1661–1673 (May 1990)

The current-voltage characteristic (IVC) of a system of two small series-connected tunnel junctions is calculated under conditions when the energy spectrum of the central electrode of the system is discrete. The form of the IVC depends substantially on the rate of energy relaxation on this electrode. It is shown that at typical real values of the system parameters the global IVC singularities due to correlated single-electron tunneling should be preserved down to very small dimensions of the central electrode. At the same time, the system IVC should have a fine structure that reflects directly the structure of the energy spectrum of the central electrode.

1. INTRODUCTION

It is well known (see, e.g., the reviews, Refs. 1 and 2) that when tunnel-junction dimensions are decreased the Coulomb interaction between the tunneling electrons becomes substantial and makes the tunneling of individual electrons correlated. These correlations lead to singularities on the current-voltage characteristics (IVC) of individual junctions as well as of junction systems. In particular, in a system of two series-connected junctions substantial correlation is possible between tunneling acts through different junctions, as a result of which the IVC become oscillatory. Such oscillations were recently observed in many experiments.[3-7]

The present theory of this correlated one-electron tunneling[1,2] is valid for junctions with electrodes that are not too small, particularly those having a continuous electron energy spectrum. A timely question is how the properties of the tunnel junctions are altered when their sizes are further decreased to a level in which spatial quantization of the energy spectrum becomes substantial. This question is vital, in particular, also in connection with current discussions[1,2,8] of the possibility of using correlated single-electron tunneling to produce molecular-electronics devices.

We consider this question in the present paper for a system of two series-connected tunnel junctions. This is the most important case from the experimental standpoint, since the use of scanning tunnel microscopy permits the production of such systems[5-7] with very small (practically down to atomic) dimensions of the central electrode.

The plan of the paper is the following. In Sec. 2 we obtain a Boltzmann equation for the description of the tunneling dynamics in a system of two junctions, with allowance for the discrete character of the energy spectrum of the central electrode. This equation is used in Sec. 3 to examine the IVC of a system of junctions in a large range of voltages and currents, in which the IVC is determined by the Coulomb correlations between the tunneling electrons. It is shown that in the case of rapid energy relaxation on the central electrode the form of the IVC coincides with the one that follows from the existing theory.[1,2] Some useful analytic expressions for the IVC of the system in this case are derived in Appendix 1. In Sec. 4 we consider the IVC microstructure which is directly connected with discrete character of the energy spectrum. The Conclusion contains estimates of the magnitudes of these effects for experimentally feasible parameter values.

2. FUNDAMENTAL RELATIONS

Consider a system of two tunnel junctions made up of two solid electrodes and a small metallic granule placed between them. We assume that this granule, which serves as the central electrode of the junction system, contains a relatively large number N of atoms, and hence of electrons in the conduction band. The average separation Δ between the levels of the quasiparticle energy spectrum of the granule $\Delta \sim [\nu(0)d^3]^{-1}$, ($\nu(0)$ is the density of states on the Fermi level of the granule material and d is its characteristic dimension) is then much lower than the typical Coulomb energy $E_C \sim e^2/\varepsilon_0 d$ of the granule. Indeed, these two energies become equal in a "granule" with $N \sim 1$, in which case Δ decreases as a function of N much more rapidly ($\propto N^{-1}$) than the Coulomb energy ($\propto N^{-1/3}$). Thus, for $N \gg 1$ the number of states in the granule in the energy interval of order E_C of interest to us is still large enough, and the granule can be regarded as a "massive" metallic sample,[1)] notwithstanding the discrete energy spectrum. In particular, in this case the screening length λ of the electric field is small, $\lambda \sim [e^2\nu(0)/\varepsilon_0]^{-1/2} \ll d$, and the Coulomb energy U of the considered system can be calculated from the usual equations of macroscopic electrostatics:[1,2,9]

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

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

where C_j is the capacitance of the jth junctions, n_j is the number of electrons tunneling through it, $C_\Sigma \equiv C_1 + C_2$, Q_0 is the fractional (in units of e) effective charge on the granule, which can be induced for example by an external magnetic field. The total Hamiltonian of the system is the sum of the Coulomb energy U, of the internal energies H_j of the electrodes and of the granule H_0, and the terms H_{Tj} describing the tunneling:

$$H = H_0 + H_1 + H_2 + U + H_{T1} + H_{T2}, \tag{2a}$$

$$H_0 = \sum_m \varepsilon_m c_m^+ c_m + H', \quad H_{Tj} = \sum_{m,k_j} T_{m,k_j} c_m^+ c_{k_j}. \tag{2b}$$

0038-5646/90/050937-07$03.00

Here $\{\varepsilon_m\}$ is the electron energy spectrum of the granule, H' are the terms describing the electron energy relaxation, and c^+ and c are the electron creation and annihilation operators on the granule (m) and in the external electrodes (k_j).

It is known that Coulomb correlations can take place in tunneling only in junctions with small conductivities G_j, $G_j \ll R_Q^{-1}$, where $R_Q \equiv \pi\hbar/2e^2$ (this condition can be written in the form $\hbar\Gamma_j(\varepsilon_m) \ll \Delta$, where

$$\Gamma_j(\varepsilon_m)=2\pi\langle|T_{m,k_j}|^2\rangle\nu_j(0)/\hbar$$

is the probability of tunneling from the level ε_m to the jth electrode). Confining ourselves to this case, we can describe tunneling at not too low temperatures $T \gg \hbar\Gamma_j(\varepsilon_m)$ by using a simple Boltzmann equation for the probability density $\rho_n(\varepsilon_m)$, obtained from the Hamiltonian (2) by perturbation theory in H_{Tj}. If the rate τ_ε^{-1} of electron relaxation on the granule is also low, $\hbar/\tau_\varepsilon \ll \Delta$, this relaxation can also be described with the aid of a Boltzmann equation. (In the opposite case the discrete energy levels are smeared out and the picture of the correlated tunneling is the same as the one that follows from the existing theory[1,2]). Writing the Boltzmann equation in accordance with the usual rules, we obtain

$$\dot{\rho}_n(\varepsilon_m,t)=S_n-S_{n-1}$$
$$+\sum_{j=1,2}\{\Gamma_j(\varepsilon_m)[\sigma_{n-1}g(\varepsilon_m-E_j(n))(1-f_{n-1}(\varepsilon_m))$$
$$-\sigma_n(1-g(\varepsilon_m-E_j(n)))f_n(\varepsilon_m)]\}+F_\varepsilon, \tag{3a}$$

$$S_n=\sum_j\sum_{m'\neq m}\Gamma_j(\varepsilon_{m'})$$
$$\times\{\sigma_{n+1}[1-g(\varepsilon_{m'}-E_j(n+1))]f_{n+1}(\varepsilon_{m'})f_{n+1}(\varepsilon_m)$$
$$-\sigma_n g(\varepsilon_{m'}-E_j(n+1))[1-f_n(\varepsilon_{m'})]f_n(\varepsilon_m)\}, \tag{3b}$$

$$\rho_n(\varepsilon_m)=\sigma_n f_n(\varepsilon_m),\quad \sum_n\sigma_n=1,\quad f_n(\varepsilon_m)\to 1 \text{ for } \varepsilon_m\to\infty, \tag{3c}$$

where $E_j(n)$ is the change of the electrostatic energy (1) due to tunneling of an electron from the granule to the jth electrode, $g(\varepsilon)$ is the Fermi distribution function, σ_n is the probability of the presence of n excess electrons on the granule, $f_n(\varepsilon_m)$ is the electron distribution function in energy at the given n, and the term F_ε describes the energy relaxation.

The matrix elements $T_{m,k}$ can be represented in the usual manner[11]

$$T_{m,k}=\text{const}(U-\varepsilon_m)\exp\{-a\lambda(1+\varkappa^2/2\lambda^2)\}$$
$$\times\int_V dr\,\psi_m(r)\exp\{i\varkappa\rho-z(\lambda^2+\varkappa^2)^{1/2}\},$$
$$k=\{\varkappa,k_z\},\quad r=\{\rho,z\},\quad \lambda=[2m(U-\varepsilon_m)]^{1/2}/\hbar, \tag{4}$$

where the z axis is perpendicular to the barrier, a and U are its thickness and height, V is the granule volume, and $\psi_m(r)$ is the wave function of the state ε_m. In a typical case, a macroscopic ($N \gg 1$) granule has random shape irregularities of at least atomic size, and the motion of the electrons in it can be regarded as ergodic. The energy spectrum $\{\varepsilon_m\}$ is then random (see, e.g., Ref. 12), and the amplitude $|\psi_m(r)|$ of the wave functions of the stationary states, averaged over distances of order k_F^{-1}, is constant over the granule volume.[13] For $\lambda \ll k_F$ the matrix elements $T_{m,kj}$ and hence the tunneling probabilities Γ_j for states with $\varepsilon_m \ll U$ can therefore be regarded as independent of m. We shall assume in what follows that these conditions are met, although the results are qualitatively valid also in the general case.

Equation (3) allows us to find the IVC of this system of two junctions. This IVC has singularities of two types: some connected with the Coulomb correlations between the tunneling electrons, and some connected with the discrete character of the granule energy spectrum. In the present case of a macroscopic granule these two singularity types have substantially different voltage scales, E_C/e and Δ/e respectively ($E_C \equiv e^2/2C_\Sigma \sim e^2/\varepsilon_0 d \gg \Delta$), so that they can be discussed separately.

3. FORM OF IVC IN A LARGE RANGE OF VOLTAGES AND CURRENTS

We consider in this section the IVC oscillations due to Coulomb correlations between the tunneling electrons. To this end it is convenient to subdivide Eq. (3) into two equations—for the probabilities σ_n and for the distribution function $f(\varepsilon_m) = \Sigma_n\sigma_n f_n(\varepsilon_m)$ averaged over n:

$$\dot{\sigma}_n=S_n'-S_{n-1}', \tag{5a}$$

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

$$f(\varepsilon_m)=\sum_j\Gamma_j\sum_n\sigma_n\{g(\varepsilon_m-E_j(n+1))[1-f_n(\varepsilon_m)]$$
$$-[1-g(\varepsilon_m-E_j(n))]f_n(\varepsilon_m)\}+F_\varepsilon. \tag{6}$$

The condition that the granule be macroscopic, $\Delta \ll E_C$, makes it possible to simplify these equations considerably. In this case a small number of electron tunneling acts, which alters substantially the charge en on the granule, cannot alter $f_n(\varepsilon_m)$ in the substantial energy interval $\sim E_C$. This makes it possible, first, to neglect the dependence of $f_n(\varepsilon_m)$ on n. Second, the rate of change of σ_n is in this case considerably higher than the rate of change of $f(\varepsilon_m)$, so that in (6) we can use a stationary distribution σ_n corresponding to the instantaneous value of $f(\varepsilon_m)$. In addition, neglecting the IVC microstructure, we can replace the summation in (3b) by integration, and obtain ultimately the system of equations

$$\frac{\sigma_{n+1}}{\sigma_n}=\sum_j\left\{\frac{G_j}{e^2}\int d\varepsilon[1-g(\varepsilon-E_j(n+1))]f(\varepsilon)\right\}^{-1}$$
$$\times\sum_j\frac{G_j}{e^2}\int d\varepsilon\, g(\varepsilon-E_j(n+1))[1-f(\varepsilon)], \tag{7a}$$

$$f(\varepsilon,t)=\sum_j\Gamma_j\sum_n\sigma_n\{g(\varepsilon-E_j(n+1))[1-f(\varepsilon)]$$
$$-[1-g(\varepsilon-E_j(n))]f(\varepsilon)\}+F_\varepsilon. \tag{7b}$$

where $G_j = ce^2\Gamma_j/\Delta$ and the coefficient c is equal to 2 in the absence of a magnetic field (owing to the double degeneracy of all the states ε_m) and to unity in the presence of a magnetic field $H \gtrsim \nu/\mu$, where μ is the Bohr magneton and ν is the characteristic level width.

The relaxation term F_ε can have a complicated energy dependence; we confine ourselves to a qualitative discussion of the energy relaxation, assuming $F_\varepsilon = [g(\varepsilon) - f(\varepsilon)]/\tau_\varepsilon$ and $\tau_\varepsilon = \text{const}$. If the rate of energy relaxation is not small, $\tau_\varepsilon^{-1} \gg \Gamma_j$ (i.e., $\tau_\varepsilon^{-1} \gg \tau_j^{-1}\Delta/E_C, \tau_j^{-1} \equiv G_j/C_\Sigma$) the system of electrons on the granule is in equilibrium and Eq. (7a) coincides with the corresponding equation of the "orthodox" theory.[1,2] At a low relaxation rate the electrons can "overheat" and affect the IVC of the junction system. The results of a numerical calculation of the IVC with the aid of Eqs. (7) for the case $\tau_\varepsilon^{-1} = 0$ and $T = 0$ are shown in Fig. 1.

It is seen from Fig. 1a that if the junction conductivities do not differ too much, the electron overheating in the granule weakens the Coulomb correlations between the tunneling acts and smoothes out the associated IVC oscillations. If the conductivities differ greatly (Fig. 1b) the tunneling through a well-conducting junction plays the role of a relaxation mechanism and the overheating effects are weakened.

The IVC of the junctions differ also in their asymptotic values at $\tau_\varepsilon = 0$ and $\tau_\varepsilon = \infty$. At voltages $V \gtrsim (E_C/e) \times (G_1/G_2)$ (we have assumed $G_1 > G_2$) both curves become asymptotically linear.

$$I = (V - V_{of})/R_\Sigma, \quad R_\Sigma \equiv (G_1^{-1} + G_2^{-1})^{-1},$$

but have different values of V_{of}: $V_{of} = e/C_\Sigma$ for $\tau_\varepsilon = 0$ and $V_{of} = e/2C_\Sigma$ for (Fig. 1). This result can be obtained analytically from Eqs. (7) (see Appendix 2) but its simple qualitative interpretation is more illustrative. Namely, in the case of low relaxation the energy lost by the electron to overcome the Coulomb barrier in tunneling through the first junction is not scattered. Therefore energy is lost in the tunneling only when the Coulomb barrier is overcome in the second junction, corresponding to half as large a shift of the IVC asymptote.

When the relaxation rate is finite, the IVC of the junctions lie between the IVC corresponding to the limiting cases $\tau_\varepsilon = 0$ and $\tau_\varepsilon = \infty$. In particular (see Appendix 2),

$$V_{of} = (e/2C_\Sigma)\{1 + [1 + (\Delta\tau_\varepsilon/ce^2R_\Sigma)]^{-1}\}. \tag{8}$$

It must be noted, however, that the result (8) is the consequence of the assumed relaxation model, with an energy-independent τ_ε. In a more realistic case the relaxation rate increases with increase of energy, so that at sufficiently high voltages the shift V_{of} of the linear asymptote of the IVC tends to e/C_Σ.

4. MICROSTRUCTURE OF IVC

We consider now the IVC microstructure which reflects directly the structure of the quasiparticle energy spectrum of the granule. Since the energy spectrum is discrete, the current flowing through the system increases with the voltage discretely (in steps of the order of $e\Gamma_j$) if the Fermi level coincides in one of the outer electrodes with the next level ε_m in the granule. The dependence of the differential conductivity of the system on the voltage should therefore contain resonant peaks whose voltage positions correspond to discrete energy levels of the granule. The IVC microstructure should be most strongly pronounced when the rate of energy relaxation is high $\tau_\varepsilon^{-1} \gtrsim \Gamma_j$ (but the spectrum is still discrete, $\hbar/\tau_\varepsilon \ll \Delta$ so that at low temperatures $T \ll \Delta$ the electron distribution in energy, in both the outer electrodes and in the granule, has an abrupt (on the Δ scale) boundary.

At $V = V_0$, where

$$V_0 \equiv (-1)^{j+1}[C_\Sigma\varepsilon_m/e + ep + Q_0 - e/2]/C_{j'},$$

with $j, j' = 1, 2, j \neq j'$, the Fermi level of the jth electrode coincides with the level ε_m in the granule (with allowance for the shift of its Fermi level upon tunneling of the electron), when the granule contains p "excess" electrons, i.e., $\varepsilon_m = E_j(p)$. This equality leads to the presence, at $V \approx V_0$, of a system conductivity peak whose form is determined by Eq. (6):

$$G(V) = bc\Gamma_j(\varepsilon_m)e^2(C_{j'}/C_\Sigma)g'(e(V - V_0)C_{j'}/C_\Sigma)$$
$$g'(z) = (1/4T)\mathrm{ch}^{-2}(z/2T), \quad b \equiv (\partial I/\partial I_{j,p})|_{V=V_0}, \tag{9}$$

where the constant b can be obtained from Eq. (7a) in which $f(\varepsilon) = g(\varepsilon)$; $I_{j,p}$ is the current flowing through the jth junction at $n = p$.

It is seen from (9) that the width ν of the conductivity peak is determined by the temperature, $\nu \sim T$, i.e., by the "broadening" of the Fermi levels of the outer electrodes. At low temperatures $T \ll \hbar(\Gamma_j, \tau_\varepsilon^{-1})$, at which the Boltzmann equation (3) and expression (9) are not valid, the conductivity-peak width is determined by the natural width of the energy levels of the granules, $\nu \sim \hbar(\tau_\varepsilon^{-1} + \Gamma_1 + \Gamma_2)$ (Refs. 14 and 15). As ν increases to Δ/n_0, where n_0 is the characteristic width of the σ_n distribution, the conductivity peaks be-

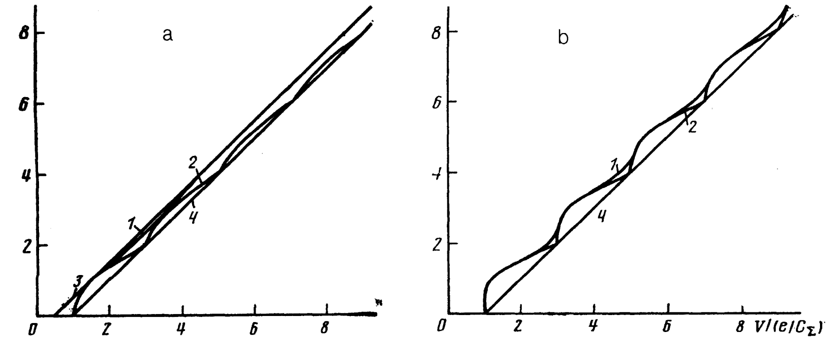

FIG. 1. IVC of a system of two series connected junctions ($C_1 = C_2, Q_0 = 0$) for not too strongly ($a, G_1 = 10G_2$) and strongly ($b, G_1 = 100G_2$) differing conductivities of the junctions in the two limiting cases: in the absence of energy relaxation on the central electrode of the system ($\tau_\varepsilon = \infty$, curve 1) and for fast (compared with $\Gamma_j = G_j\Delta/ce^2$) relaxation ($\tau_\varepsilon = 0$, curve 2). The asymptotes of these curves are respectively 3—$I = (V - e/2C_\Sigma)/R_\Sigma$ and 4—$I = (V - e/C_\Sigma)/R_\Sigma$.

gin to overlap and the amplitudes of the corresponding IVC singularities decrease.

The constant b in (9) can be obtained explicitly in certain cases. For $G_1 \gg G_2$ we have $b=1$ for $j=2$, $p=\text{Int}[C_2V - Q_0 + e/2)/e]$ (Int is the integer part) and $b=0$ for other values of p and j. If, to be specific, the barrier is removed initially in the first junction, i.e., $(e/2-Q_0)/C_1 > (e/2+Q_0)/C_2$, then in the voltage region directly above the Coulomb-blockade interval,

$$V>(e/2+Q_0)/C_2, \quad V<\min\{(e/2-Q_0)/C_1,\ (3e/2+Q_0)/C_2\},$$

only two probabilities, σ_0 and σ_1, differ from zero:

$$\sigma_0=\frac{-G_2E_2}{G_1E_1-G_2E_2}, \quad \sigma_1=\frac{G_1E_1}{G_1E_1-G_2E_2},$$
$$E_1=e(VC_2-e/2-Q_0)/C_\Sigma, \quad E_2=-e(VC_1+e/2+Q_0)/C_\Sigma. \tag{10}$$

Here $b=\sigma_0^2\equiv b_1$ and $b=\sigma_1^2\equiv b_2$ respectively for the conductivity peaks connected with the resonance in the first junctions ($\varepsilon_m=E_1$) and the second junction ($\varepsilon_m=E_2$).

In the most interesting case of substantially different junction conductivities, $G_1\gg G_2$, when the conductivity peaks are due to resonances in only one junction (j) and at one value of p, their aggregate corresponds exactly to the energy spectrum of the granule.

For granules of irregular shape, the energy spectrum is described by the theory of random matrices. In particular, the statistics (over different granules or over different levels in one granule) of the intervals between the nearest levels, and consequently, in our case—the statistics of the voltage intervals δV between the nearest conductivity peaks is described with good accuracy by distributions of the type (see, e.g., Ref. 12):

$$f_\beta(x)=A_\beta x^\beta \exp(-B_\beta x^2), \quad x=\delta V/\langle\delta V\rangle, \quad \langle\delta V\rangle=cC_\Sigma\Delta/2C_1e. \tag{11}$$

Here

$$\beta=1;\ 2;\ 4, \quad A_\beta=\pi/2;\ 32/\pi^2;\ (64/9\pi)^3, \quad B_\beta=\pi/4;\ 4/\pi;\ 64/9\pi$$

respectively for the following cases:

—weak spin-orbit interaction H_{so} in the granule and weak magnetic field H: $|\langle H_{so}\rangle|, \mu H\ll\Delta$ (orthogonal ensemble of random matrices);

—strong spin-orbit interaction and strong magnetic field (unitary ensemble);

—strong spin-orbit interaction and weak magnetic field (symplectic ensemble).

A characteristic feature of the distribution (11) is the repulsion of the energy levels: $f_\beta(x)\to 0$ as $x\to 0$. In the case of a strong magnetic field and weak spin-orbit interaction the energy levels of the granule break up (along the spin direction) into two uncorrelated subsystems, so that there is no level repulsion, and the statistics of the intervals between the levels can be described by the distribution $\tilde{f}_2(x)$—see Eq. (16b) below.

If the conductivity peaks are narrow enough, $\nu\ll\Delta$, and $\Gamma_j(\varepsilon_m)=\langle\Gamma_j\rangle=\text{const}$, the correlation function of the system conductivity

$$\langle\tilde{G}(V')\tilde{G}(V'+V)\rangle=\langle\tilde{G}\tilde{G}(V)\rangle, \quad \tilde{G}=G-\langle G\rangle,$$

is expressed in the present case ($G_1\gg G_2$) for $V\gg\nu/e$ directly in terms of the correlation function $R(x)$ of the levels in the granule:

$$\langle\tilde{G}\tilde{G}(V)\rangle=\langle G\rangle^2[R(x)-1], \quad \langle G\rangle=G_2C_1/C_\Sigma, \quad x=V/\langle\delta V\rangle. \tag{12}$$

The level repulsion is manifested in the characteristic oscillations of $R(x)$ (see, e.g., Ref. 16):

$$1-R(x)=\begin{cases} s^2(x)+\dfrac{ds}{dx}\displaystyle\int_x^\infty s(t)\,dt, & \beta=1,\\ s^2(x), & \beta=2,\\ s^2(2x)-\dfrac{ds(2x)}{dx}\displaystyle\int_0^\infty s(2t)\,dt, & \beta=4,\end{cases} \tag{13}$$
$$s(x)\equiv\sin(\pi x)/\pi x.$$

The correlation function $\langle\tilde{G}\tilde{G}(V)\rangle$ for small V, $V\sim\nu/e\ll\Delta/e$, is determined by the form of an individual conductivity peak. At $T\gg\hbar(\Gamma_j,\tau_\varepsilon^{-1})$ and $T\ll\Delta$ we have

$$\langle GG(V)\rangle=\langle G\rangle^2c\frac{\Delta}{16T}\int_{-\infty}^{+\infty}dx\,\text{ch}^{-2}(x)\,\text{ch}^{-2}\left(x+\frac{eVC_1}{2TC_\Sigma}\right), \tag{14a}$$

and in particular

$$\langle G^2\rangle=\langle G\rangle^2c(\Delta/12T). \tag{14b}$$

At $T=0$ and $\tau_\varepsilon^{-1}=0$ the conductivity peak has a Lorentz shape of width $\hbar(\Gamma_1+\Gamma_2)$. This result, obtained for an isolated level,[15] can be used also in the present case of tunneling through a granule, inasmuch as in this case the tunneling takes place simultaneously though a large number of levels, and in spite of the presence of Coulomb correlations on the whole, the tunneling through each individual level can be regarded as uncorrelated with the tunneling through other levels. We obtain then for $\langle GG(V)\rangle$

$$\langle GG(V)\rangle=\left(\frac{2ce^2\langle G\rangle}{\pi\hbar G_1}\right)^2\int_{-\infty}^{+\infty}\frac{dV'}{\langle\delta V\rangle}\left\{\left[1+\left(\frac{4e^3V'C_1}{\Delta\hbar G_1C_\Sigma}\right)^2\right]\right.$$
$$\left.\times\left[1+\left(\frac{4e^3(V-V')C_1}{\Delta\hbar G_1C_\Sigma}\right)^2\right]\right\}^{-2}$$
$$=\langle G\rangle^2c\frac{e^2}{\pi\hbar G_1}\left[1+\left(\frac{2e^3V}{\Delta\hbar G_1}\right)^2\right]^{-2}. \tag{15}$$

Equations (12)–(15) and the analogous equations (17) and (18) below describe mesoscopic fluctuations of the conductivity (see, e.g., Ref. 15 and the citations there) in this system of two tunnel junctions.

If the conductivity peaks are connected with resonances at several (j,p) values, their aggregate corresponds to several superimposed sections of the granule energy spectrum. Since these spectrum sections are separated in energy by a value of order E_C ($\gg\Delta$), they are statistically independent and the level repulsion vanishes in this case. For example, for $G_1\sim G_2$ and voltages close to the Coulomb-blockade threshold, two (j,p) pairs are significant. It can then be approximately assumed that the conductivity peaks connected with the resonances in the first junction randomly divide a part (C_1/C_2) of the intervals between the nearest conductivity peaks connected with the resonance in the second junction (we assume, to be definite, that $C_1 < C_2$). In this approximation we obtain the following distribution of the voltage intervals between neighboring peaks:

$$u(x)=[(C_2-C_1)/C_\Sigma]f_\beta(x)+(2C_1/C_\Sigma)\bar{f}_\beta(x), \quad x=2eVC_2/c\Delta C_\Sigma, \tag{16a}$$

$$\bar{f}_\beta(x)=\int_x^\infty dy\frac{f_\beta(y)}{y}=\begin{cases}(\pi/2)\,\mathrm{erfc}(\sqrt{\pi}x/2), & \beta=1,\\ (4/\pi)\exp[-(4/\pi)x^2], & \beta=2,\\ (B_4/2)\exp(-B_4x^2)(1+B_4x^2), & \beta=4.\end{cases} \tag{16b}$$

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

Superposition of two uncorrelated systems of conductivity peaks also weakens the characteristic oscillations of the correlation function $\langle \tilde{G}\tilde{G}(V)\rangle$, which for $V \gg \nu/e$ and $\nu \ll \Delta$ in this case takes the form

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

$$\gamma_j=\left(\frac{b_jG_jC_1C_2}{\langle G\rangle C_jC_\Sigma}\right)^2, \tag{17a}$$

$$\langle G\rangle=\sum_{j=1,2}b_jG_jC_1C_2/C_jC_\Sigma. \tag{17b}$$

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

$$\langle GG(V)\rangle=\langle G\rangle^2\frac{c\Delta}{16T}\sum_{j=1,2}\gamma_j\int dx\,\mathrm{ch}^{-2}(x)\,\mathrm{ch}^{-2}\left(x+\frac{eVC_1C_2}{2TC_jC_\Sigma}\right), \tag{18a}$$

In particular,

$$\langle G^2\rangle=\langle G\rangle^2c(\gamma_1+\gamma_2)(\Delta/12T). \tag{18b}$$

In the absence of a magnetic field all the states of the energy spectrum of the granule are at least doubly degenerate owing to the presence of symmetry with respect to time reversal. The magnetic field, by breaking this symmetry, lifts the degeneracy:

$$\varepsilon_m\to\varepsilon_m\pm(1-\rho)\mu H. \tag{19}$$

For weak spin-orbit interaction, the parameter ρ that depends in general on the level number m is on the order of[12] $|\langle H_{so}\rangle|^2/\Delta^2$, and in the limit $|\langle H_{so}\rangle| \gg \Delta$ we have $\rho \to 1$ (the level splitting in this limit can be roughly estimated at $(\mu H)^2/|\langle H_{so}\rangle|$). The magnetic field should thus split each conductivity peak into two, separated in voltage by

$$\delta V=2(C_\Sigma/C_j)(1-\rho)\mu H/e,$$

with the centers of these two peaks shifted by a value on the order of $(C_\Sigma/C_j)\cdot(2\rho-\rho^2)(\mu H)^2/e\Delta$. This behavior in a magnetic field can serve as the distinguishing feature of system of conductivity peaks connected with the discrete character of the energy spectrum of the granule.

5. ESTIMATES AND CONCLUSIONS

We now estimate the magnitude of the effects considered above for realistic values of the parameters. The discreteness interval Δ of the energy spectrum is approximately 10^{-4} eV (1K) for metallic particles with diameter 100 Å, and increases to 0.1 eV when the diameter is decreased to 10 Å (Ref. 12). The characteristic Coulomb energy E_C in this diameter interval (assuming for the tunnel layers a dielectric constant $\bar{\varepsilon} \sim 5$) charges from 0.05 to 0.5 eV. The energy relaxation time τ_ε in a metallic granule should not differ substantially at high energies $\varepsilon \gg \Delta$ from the time of energy relaxation in a bulky sample, which in turn can be estimated (with allowance for electron-electron scattering only) at $\hbar\varepsilon_F/\varepsilon^2$, where ε_F is the Fermi energy of the metal.

It follows from these estimates that for $\varepsilon \sim E_C$ the energy levels in the granule are broadened, $\hbar/\tau_\varepsilon \gtrsim \Delta$, and the corresponding microstructure of the IVC is smoothed out. In addition, since $G_j^{-1} \gg R_Q$ the condition $\hbar/\tau_\varepsilon \gtrsim \Delta$ means that $\tau_j \gg (\Delta/E_C)\tau_\varepsilon$, and overheating of the electrons in the granule is even less likely to occur. Thus, at characteristic voltages $V \sim E_C/e$ the IVC of the system of two junctions should be well described by the "orthodox" theory,[12] in which no account is taken of the discrete nature of the electron spectrum. A similar conclusion will in all probability be valid also for more complicated systems. This means that the orthodox theory should remain valid for tunnel junctions measuring all the way to 1–2 nm.

In the present case of a two-junction system, the only deviation from this theory is the presence of microstructure of the IVC (peaks of differential conductivities) at voltages close to the threshold V_t of the Coulomb barrier, $V - V_t \sim \Delta/e$. In this voltage region the tunneling in the granule takes place in states close to the Fermi surface, whose broadening is quite small. Indeed, the discrete character of the electron and phonon spectra of the granule can on the average only increase the relaxation time of these levels compared with the time of inelastic relaxation in bulky samples at the same energies, namely 10^{-11}–10^{-12} s at $\varepsilon \sim 10$ K, and increases approximately like ε^{-2} as ε decreases (Ref. 17). It follows from this estimate that for states near the Fermi surface we have $\hbar/\tau_\varepsilon \lesssim \Delta$ and the conductivity peaks associated with them should be observable. Such an observation would permit, to our knowledge for the first time ever, a direct verification of the applicability of the random-matrix theory to the description of the spectrum of minute metallic particles.

One more important parameter that determines the statistics of the conductivity peaks and of their behavior in a magnetic field is the magnitude of the spin-orbit interaction $\langle H_{so}\rangle$. It can be estimated from the change δg of the g-factor of the metal conduction electrons compared with the g-factor of the free electron. Since the matrix elements of the spin-orbit interaction operator $\langle H_{so}'\rangle$ in bulky samples differ from zero only for states belonging to different bands of the energy spectrum, and since[18] $\langle H_{so}'\rangle \sim \delta g E$, where E is the characteristic interband energy interval, we have

$$\langle H_{so}\rangle\sim\langle H_{so}'\rangle^2/E\sim\delta g^2E.$$

It can be concluded from this estimate that, for example at $\Delta = 10$ K, for granules of aluminum and gold ($\delta g = -5\cdot 10^{-3}$ and $\delta g = 0.1$, Ref. 19), the respective conditions of weak ($|\langle H_{so}\rangle| \ll \Delta$) and strong ($|\langle H_{so}\rangle| \gg \Delta$) spin-orbit interaction should be met. This means that a magnetic field influences the conductivity peaks in the cases of tunneling through granules of these two metals differently. In particular, for aluminum granules the splitting of the conductivity peaks in a magnetic field $H \sim 10$ T should be $\delta V \sim 100\ \mu$V, while for gold granules the splitting in such fields it should be practically unobservable, $\delta V \lesssim 1\ \mu$V.

The authors thank K. K. Likharev and Yu. V. Nazarov for helpful discussion in the course of this work.

APPENDIX 1

In this Appendix we write down some useful equations for the IVC of a system of two tunnel junctions at zero temperature in the case of rapid energy relaxation of the electrons. The equations used to calculate the IVC of this system (see Refs. 1 and 2) simplify greatly in the case of low temperature ($T \ll e^2/C_\Sigma$). There is then no need for numerical methods to find the IVC.

The expressions for the probability of tunneling through the left- and right-side junctions in the presence of n additional electrons in the middle electrode assume at $T = 0$ the form

$$\Gamma^+(n) = G_1 e^{-2} E_1(n+1), \quad \Gamma^-(n) = -G_2 e^{-2} E_2(n). \qquad (A1.1)$$

Here

$$E_j(n) = (-1)^{j+1} \frac{eVC_1C_2}{C_jC_\Sigma} + \frac{e^2}{C_\Sigma}\left(\frac{1}{2} - \frac{Q_0}{e} - n\right), \quad j = 1, 2. \qquad (A1.2)$$

The probability distribution of the charge states σ_n has a lower limit

$$n_{min} = -l = -[-E_2(1)C_\Sigma/e^2]$$

and an upper limit

$$n_{max} = k = [E_1(0)C_\Sigma/e^2]$$

(where $[\cdots]$ is the integer part); in these limits the following relation is valid

$$\sigma_{n+1}/\sigma_n = G_1E_1(n+1)[-G_2E_2(n+1)]^{-1}.$$

The average current in the stationary state can be calculated from the equation

$$I = \frac{e}{C_\Sigma}\left\{\left[G_1A + G_1\frac{G_1}{G_2}\frac{A(A-1)}{B} + \ldots + G_1\left(\frac{G_1}{G_2}\right)^{k-1} \times \frac{A(A-1)\ldots(A+1-k)}{B(B+1)\ldots(B-2+k)}\right] + \begin{bmatrix} k \to l \\ A \leftrightarrow B-1 \\ G_1 \leftrightarrow G_2 \end{bmatrix}\right\}\left\{1 + \left[\frac{G_1}{G_2}\frac{A}{B} + \left(\frac{G_1}{G_2}\right)^2 \times \frac{A(A-1)}{B(B+1)} + \cdots + \left(\frac{G_1}{G_2}\right)^k \frac{A(A-1)(A+1-k)}{B(B+1)(B-1+k)}\right] + \begin{bmatrix} k \to l \\ A \leftrightarrow B-1 \\ G_1 \leftrightarrow G_2 \end{bmatrix}\right\}^{-1}, \qquad (A1.3)$$

where

$$A = (VC_2 - Q_0)/e - 1/2, \quad B = (VC_1 + Q_0)/e + 1/2,$$

$$|Q_0| < e/2, \quad V > 0.$$

The equation given is suitable for $VC_\Sigma/e \lesssim 10$ so long as the number of terms in the numerator and denominator is not too large. It is convenient to investigate with the aid of (A1.3) the shapes of the steps on the IVC and the dependence of the differential conductivity on the voltage.

For high voltage ($VC_\Sigma/e \gtrsim 10$) it is convenient to use another equation obtained by summing expressions (A1.1) with weights σ_n and taking into account the equality of the currents through both junctions in the stationary case:

$$I = R_\Sigma^{-1}(V - eC_\Sigma^{-1}[1 - \sigma_k(1-\{A\}) - \sigma_{-l}(1-\{B\})]), \qquad (A1.4)$$

where $\{\cdots\}$ is the fractional part. Expression (A1.4) can also be obtained from (A2.2) if $f(\varepsilon) = g(\varepsilon) = -\theta(\varepsilon - \varepsilon_F)$. It is easy to trace with the aid of (A1.4) the suppression of the oscillations on the IVC with increase of voltage, by estimating σ_k and σ_{-l}. In particular, if $G_1 \gg G_2$, we have for $1 \ll VC_\Sigma/e \ll G_1/G_2$

$$I \approx R_\Sigma^{-1}\left(V + \left[(1-\{A\})\left(1 + \frac{G_2}{G_1}\frac{VC_\Sigma}{e\{A\}}\right)^{-1} - 1\right]\frac{e}{C_\Sigma}\right). \qquad (A1.5)$$

For $VC_\Sigma/e \gg G_1/G_2$ the oscillations of the IVC are determined, apart from a constant coefficient, by the expression

$$(IR_\Sigma - V + e/C_\Sigma)C_\Sigma/e \approx \{A\}(1-\{A\})\exp(-VC_\Sigma G_2 e^{-1} G_1^{-1}). \qquad (A1.6)$$

The oscillations are thus suppressed at $V \sim eG_1/C_\Sigma C_2$. In the case of equal junction conductivities ($G_1 = G_2$), only a few first oscillations are observed, since they likewise damp exponentially at $V \gtrsim e/C_\Sigma$:

$$(IR_\Sigma - V + e/C_\Sigma)C_\Sigma/e \approx [\{A\}(1-\{A\}) + \{B\}(1-\{B\})]2^{-VC_\Sigma/e}. \qquad (A1.7)$$

APPENDIX 2

In this Appendix we calculate the shift of the linear asymptote of the IVC for a system of two tunnel junctions.

In the stationary state, the currents through the left- and right-hand junctions are equal:

$$I = (-1)^j G_j e^{-1} \sum_n \sigma_n \int \{[1 - g(\varepsilon - E_j(n))] f(\varepsilon) - g(\varepsilon - E_j(n+1))[1 - f(\varepsilon)]\} d\varepsilon$$

$$= (-1)^{j+1} G_j e^{-1} \sum_n \sigma_n \left\{E_j(n+1) + \int f(\varepsilon)[g(\varepsilon - E_j(n)) - g(\varepsilon - E_j(n+1))] d\varepsilon\right\}, \qquad (A2.1)$$

where $E_j(n)$ is given by Eq. (A1.2), and $j = 1$ or 2. From (A2.1) it follows that

$$IR_\Sigma=V-e^{-1}\sum_n\sigma_n\int f(\varepsilon)\sum_j(-1)^{-j}\{g(\varepsilon-E_j(n))$$

$$-g(\varepsilon-E_j(n+1))\}d\varepsilon. \qquad \text{(A2.2)}$$

For higher voltages ($V\gg e(G_1+G_2)^2/G_1G_2C_\Sigma, V\gg T/e$), when the characteristic width n_0 of the probability distribution σ_n is large enough, we can obtain from (A2.2) a simple equation for shift of the linear asymptote of the IVC V_{of}:

$$V_{of}=eC_\Sigma^{-1}\sum_n\sigma_n[f(E_2(n))-f(E_1(n))]. \qquad \text{(A2.3)}$$

If the relaxation in the granule is fast, then $f(\varepsilon)$ is the Fermi function. In this case, for those n for which σ_n differs substantially from zero (i.e., $n\approx\langle n\rangle = V(C_2G_1-C_1G_2)e^{-1}(G_1+G_2)^{-1}-Q_0/e$) the expression in the square brackets in (A2.3) is practically equal to unity at high voltages. We thus obtain for V_{of} the usual result $V_{of}=e/C_\Sigma$.

To calculate V_{of} in the case of a finite relaxation rate we must find the explicit forms of $f(\varepsilon)$ and σ_n from (7b) we have for high voltages (and accordingly strong "smearing" of σ_n) the following equation for the stationary value of $f(\varepsilon)$:

$$f(\varepsilon)=\sum_j[G_j/(G_1+G_2)]\sum_n\sigma_n g(\varepsilon-E_j(n+1))$$

$$+e^2c[\Delta(G_1+G_2)]^{-1}F_\varepsilon. \qquad \text{(A2.4)}$$

We describe the energy relaxation in the τ-approximation, i.e.,

$$F_\varepsilon=-[f(\varepsilon)-g(\varepsilon)]/\tau_\varepsilon.$$

To calculate V_{of} we must solve the system of equations (A2.4) and (7a) for high voltages. We can confine ourselves here to the case $T=0$; obviously, the shift of V_f does not depend on the temperature. Changing to a continuous variable n, we obtain for σ_n an expression having the same form as in the case of rapid relaxation:

$$\sigma_n=(2\pi\delta^2)^{-1/2}\exp[-(n-\langle n\rangle)^2/2\delta^2], \quad |n-\langle n\rangle|\ll\delta^2,$$

$$\delta^2=VG_1G_2C_\Sigma[e(G_1+G_2)^2]^{-1}\gg1. \qquad \text{(A2.5)}$$

In this case the function $f(\varepsilon)$ takes the form shown in Fig. 2. At $\varepsilon=\varepsilon_F$ the function $f(\varepsilon)$ jumps from the value

$$f(-0)=1-G_2(G_1+G_2)^{-1}[1+e^2cR_\Sigma/\Delta\tau_\varepsilon]^{-1}$$

to the value

$$f(+0)=G_1(G_1+G_2)^{-1}[1+e^2cR_\Sigma/\Delta\tau_\varepsilon]^{-1}.$$

There are two other regions where $f(\varepsilon)$ varies, located near ε_1 and ε_2, where

$$\varepsilon_i=(-1)^iVeG_i/(G_1+G_2).$$

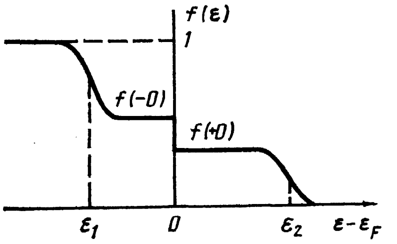

FIG. 2. Electron energy distribution function in a granule at $T=0$ and high voltages.

Near ε_1 and ε_2 (namely for $|\varepsilon-\varepsilon_i|\ll\delta^2e^2/C_\Sigma$), the function $f(\varepsilon)$ is given by the expression

$$f(\varepsilon)=\frac{1-(-1)^i}{2}+\frac{G_1G_2}{G_i(G_1+G_2)}\frac{1}{1+e^2cR_\Sigma/\Delta\tau}\left[\frac{(-1)^i}{2}\right.$$

$$\left.+(2\pi\delta^2)^{-1/2}\int_0^{\varepsilon_i-\varepsilon}\exp\left(-\frac{x^2}{2\delta^2}\right)dx\right]. \qquad \text{(A2.6)}$$

Substituting (A2.5) and (A2.6) in (A2.3) we obtain the final expression (8) for the shift of the linear asymptote of the IVC.

[1)]The condition that the granule be "microscopic" should be satisfied at $N\sim 100$–1000. Other effects become important for granules with fewer atoms. In particular, they are unstable to mechanical decay when their electric charge is increased (for lead, e.g., $N\approx 80$ is the stability limit at a charge $4e$)—see Ref. 10 and the citations therein.

[1] K. K. Likharev, IBM J. Res. Dev. **32**, 144 (1988).
[2] D. V. Averin and K. K. Likharev, in *Quantum Effects in Small Disordered Systems*, B. Al'tshuler, P. Lee, and R. Webb, eds., Elsevier (in press).
[3] L. S. Kuz'min and K. K. Likharev, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 389 (1987) [JETP Lett. **45**, 495 (1987)].
[4] T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. **59**, 109 (1987).
[5] P. J. M. Van Bentum, R. T. M. Smokers, and H. van Kampen, *ibid.* **60**, 1543 (1988).
[6] K. A. McGreer, J.-C. Wan, N. Anand, and A. M. Goldman, Phys. Rev. B **39**, 12260 (1989).
[7] R. Wilkins, E. Ben-Jacob, and R. C. Jaklevic, Phys. Rev. Lett. **63**, 801 (1989).
[8] R. Gilmanshin and P. I. Lazarev, J. Mol. Electr. **4** (Suppl.), S83 (1988).
[9] I. O. Kulik and R. I. Shekhter, Zh. Eksp. Teor. Fiz. **68**, 623 (1975) [Sov. Phys. JETP **41**, 308 (1975)].
[10] C. Baladron, J. M. Lopez, M. P. Inigues, and J. A. Alonso, Z. Phys. D **11**, 323 (1989).
[11] E. S. Burstein and R. S. Lundkvist, eds., *Tunneling Phenomena in Solids*, Plenum, 1969, Ch. 1.
[12] W. P. Halperin, Rev. Mod. Phys. **58**, 533 (1986).
[13] M. V. Berry, J. Phys. A **10**, 2083 (1977).
[14] A. D. Stone and P. A. Lee, Phys. Rev. Lett. **54**, 1196 (1985).
[15] A. I. Larkin and K. A. Matveev, Zh. Eksp. Teor. Fiz. **93**, 1030 (1987) [Sov. Phys. JETP **66**, 580 (1987)].
[16] C. E. Porter, *Statistical Theories of Spectra: Fluctuations*, Academic, 1967.
[17] G. Bergman, Phys. Rep. **107**, 1 (1984).
[18] R. J. Elliott, Phys. Rev. **96**, 266 (1954).
[19] J. Buttet, R. Car, and C. W. Myles, Phys. Rev. B **26**, 2414 (1982).

Translated by J. G. Adashko