

# Kinetic phenomena and charge discreteness effects in granulated media

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An approach to the problem of charge transport in a system of small metallic particles coupled by tunnel interactions is proposed. The system Hamiltonian is represented in the form  $H = H_0 + H_V + H_T$  where  $H_V$  takes into account electrostatic effects due to charge accumulation in the granules and the discrete character of the charge, and  $H_T$  is the tunnel Hamiltonian. Owing to the electrostatic threshold at low temperatures the conductivity  $\sigma$  is asymptotically proportional to  $T$ . The important role of the specific granule charge fluctuations due to the discrete character of the charge is demonstrated. The fluctuations are manifested by the characteristic oscillations of a number of physical characteristics of granulated media. Some features of the conductivity of  $M-G-M$  and  $S-G-S$  tunnel junctions containing granules of a metal  $G$  in an oxide layer ( $M$  is the normal metal and  $S$  the superconductor) are investigated.

## 1. INTRODUCTION

Minute metallic particles (granules) coupled by weak tunnel interactions are an interesting object of the physics of disordered media. On the one hand, a feature of such systems is the enhancement of the conductivity as a result of "accumulation" of the charge on the granules, causing the probability of the transitions  $M \rightarrow G \rightarrow M'$  ( $M$  and  $G$  stands for metal and granule, respectively) to be much larger than the probability of the through tunneling  $M \rightarrow M'$ . On the other hand, at small dimensions of the granules  $G$ , an important role is assumed by electrostatic effects, which lead to a limitation on the current as a result of formation of a space-charge region<sup>[1-3]</sup>. Since the charge can move from granule to granule only in finite batches, unique "quantization" effects appear, due to the discrete character of the charge<sup>[4]</sup>. Similar phenomena are observed experimentally in tunnel junctions containing metallic inclusions in the oxide layer<sup>[5-8]</sup>. It was shown that the so-called "zero" anomalies of the tunnel current can be due not only to the Kondo effect (scattering by paramagnetic impurities)<sup>[9]</sup>, but also to threshold phenomena connected with the discrete character of the electric charge. In a number of papers<sup>[1, 10-12]</sup>, models were proposed that take into account the activation character of the charge transport in granulated media.

A feature of the considered problem is the need for taking into account fluctuation redistributions of the charge among the granules during the course of current flow. Although there is an electrostatic threshold to the passage of electrons from granule to granule, under certain conditions this threshold can be appreciably lowered. The fact that the electrostatic energy of a granulated system is a quadratic function of the charges, gives rise to the possibility of a unique "degeneracy," wherein several different distributions of the total charge among the granules correspond to the same free energy of the system<sup>[4]</sup>. Since the electric charge is discrete, such a degeneracy can correspond to the smallest value of the free energy. This facilitates appreciably the fluctuation transitions of the electrons within the limits of the separated configurations. It is shown in the present paper that the possibility of removing the energy threshold for the fluctuation redistributions of the charge leads to a number of characteristic oscillatory effects.

A consistent analysis of the indicated questions entails a search for the distribution function  $W(n_1,$

$n_2, \dots)$  of the electrons over the granules. Starting from the equation for the density matrix, a kinetic equation is derived and determines the form of the function  $W$ , and methods of solving this equation are indicated. In a number of cases this permits the kinetic characteristics of the granular medium to be determined. The concrete results pertain mainly to granular systems included in a tunnel junction between two bulky metals.

A more general aspect of the presented analysis touches upon the mechanisms of conductivity in condensed phases of matter, as influenced by the role of electronic correlations. Actually, the model investigated in the present paper turns out to be closely connected with correlation effects in narrow-band substances (the Hubbard problem)<sup>[13]</sup>. The Hamiltonian of a system of granules, neglecting cross-capacitances, takes the form (see<sup>[4]</sup>)

$$H = \sum_{\alpha\beta} \sum_{ij} T_{ij} a_i^{\dagger} a_j + \text{H.c.} + \sum_i \frac{e^2}{2C_i} \left( \sum_{\alpha} a_i^{\dagger} a_i - N_0 \right)^2, \quad (1.1)$$

where  $a_i^{\dagger}(\alpha)$  are the operators for particle production on the granules, and  $\alpha$  is the set of quantum numbers of the electron, including the spin  $\sigma$ . For brevity, we have left out from (1.1) the index  $\alpha$  of the operators  $a_i$  and the tunneling matrix elements  $T_{ij}$ . If we extrapolate (1.1) to the case of individual atoms, then we obtain the Hamiltonian ( $\alpha \equiv \sigma$ )

$$H = \text{const} - \sum_{\sigma} \sum_i (N_0 - 1/2) U_i a_{i\sigma}^{\dagger} a_{i\sigma} + \sum_{\sigma} \sum_{ij} T_{ij} a_{i\sigma}^{\dagger} a_{j\sigma} + \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow},$$

where  $U_i = e^2/C_i$  is the electrostatic energy, and  $\hat{n}_{i\sigma} = a_{i\sigma}^{\dagger} a_{i\sigma}$  is the operator of the number of electrons at the site  $i$ . The index  $\sigma$  takes on two values:  $\uparrow$  or  $\downarrow$ . With the exception of the inessential self-energy term (the second term of (1.2)), the Hamiltonian (1.2) coincides with the Hubbard Hamiltonian<sup>[13-15]</sup>.

The analysis that follows is macroscopic and allows us to study the case when the granules are large, so that they contain a large number of electrons. It can be stated that this is the case of the Hubbard model for particles with spin  $s = \infty$ .

One of us has shown earlier<sup>[4]</sup> that if the particle dimensions are not too small we can neglect the spatial quantization of the spectrum, but we must take into account the discreteness of the charge, since the latter effect begins to manifest itself with decreasing particle dimensions before the quantization effects become significant<sup>1)</sup>. A correct mathematical description was pro-

vided for the charge transport in a system of small particles, and the singularities of the conductivity of tunnel systems containing metal granules in the barrier layer were considered. The case of tunneling was analyzed for both normal (M - G - M) and superconducting (S - G - S) junctions. The conductivity of three-dimensional granulated media was dealt with in another paper by the authors<sup>[16]</sup>.

## 2. HAMILTONIAN OF A SYSTEM OF MINUTE PARTICLES

We consider a system of metallic particles contained between bulky conductors. We assume the tunnel model of transitions between particles, described by the so-called "tunnel Hamiltonian" (see, e.g.,<sup>[17]</sup>). The total Hamiltonian of the system is represented as a sum of three terms:

$$H = H_0 + H_V + H_T, \quad (2.1)$$

where  $H_0$  is the Hamiltonian of the aggregate of noninteracting bodies,  $H_V$  is the electrostatic part of the energy and depends on the applied voltage  $V$ , and  $H_T$  is the tunnel term.

For a correct separation of the term  $H_V$ , we consider a system of three bodies:  $M_1$ ,  $G$ , and  $M_2$  (Fig. 1), where  $M_1$  and  $M_2$  are bulky metals in which the discrete character of the charge plays no role, and  $G$  is a granule whose characteristic electrostatic energy  $e^2/C_i$  can be of the order of the temperature  $T$  or of the difference of the electrochemical potentials  $\mu_1^* - \mu_2^* = eV$  (or else, if the metals  $M_1$  and  $M_2$  are superconducting, of the order of the energy gap  $\Delta$ ). We choose the term  $H_T$  in the form (we neglect the direct tunnel transitions  $M_1 \rightleftharpoons M_2$ )

$$H_T = T_1 \sum_{pq} (a_p^+ c_q + c_q^+ a_p) + T_2 \sum_{kq} (b_k^+ c_q + c_q^+ b_k), \quad (2.2)$$

where  $a_p^+$ ,  $b_k^+$ , and  $c_q^+$  are respectively the operators for the production of electrons in the metal  $M_1$ , in the metal  $M_2$ , and on the granule  $G$ ;  $p$ ,  $k$ , and  $q$  is the set of quantum numbers of the electrons. All three bodies are characterized by Fermi distributions with chemical potentials  $\mu_1$ ,  $\mu_2$ , and  $\mu_3$ , which are not altered by the addition of electrons. The basis for this assumption is the fact that the equilibrium inside of each of the metals ( $M_1$ ,  $M_2$ ,  $G$ ) is established within atomic times that are short in comparison with the times of the tunnel transitions between the granule and the edges, and the metals themselves are still macroscopic.

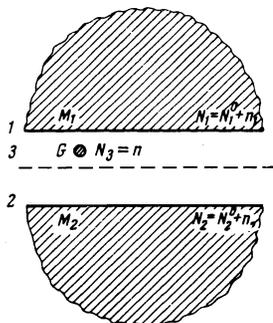


FIG. 1

The only effect that is produced by the electron transitions is the potential-energy shift due to the finite charges of the bodies. The state of the granule is characterized by a distribution function  $W_n$ , which depends in general on the time  $t$  and which shows the probability of finding an integer number of electrons  $n$  on the granule. The possibility of such an abbreviated description will be justified in detail in Sec. 3.

The numbers of electrons on the bodies  $M_1$  and  $M_2$  and in the granule  $G$  will be designated respectively by  $N_1 = N_1^0 + n_1$ ,  $N_2 = N_2^0 + n_2$ ,  $N_3 = n$ , where  $N_i^0$  are the equilibrium numbers of particles corresponding to the neutrality condition. The tunnel Hamiltonian (2.2) describes transitions in which  $n_1$ ,  $n_2$ , and  $n$  change by  $\pm 1$ , so that it suffices to use the expansion of the electrostatic energy

$$E = \frac{e^2}{2} \sum_{k,l=1}^3 \alpha_{kl} (N_k - N_k^0) (N_l - N_l^0) - \sum_k w_k^0 N_k \quad (2.3)$$

in powers of  $n_1^{-1}$  and  $n_2^{-1}$ . We assume that since the bodies  $M_1$  and  $M_2$  are macroscopic, the number of particles on them are large (excessive) in the presence of voltage on them. In formula (2.3),  $w_k^0$  is the work function of the metal  $k$ , namely,  $w_k^0 = W_k - \mu_k$ , where  $W_k$  is the depth of the potential well and  $\mu_k$  is the chemical potential.

Carrying out the indicated expansion, discarding inessential constant terms, and regarding  $n_k$  as operators, we obtain

$$H_V = \hat{N}_1 eV_1 + \hat{N}_2 eV_2 + e^2 \hat{n}^2 / 2C_i + (\alpha_1 eV_1 + \beta_1 eV_2 + w_1) \hat{n}, \quad (2.4)$$

where  $V_{1,2}$  are the potentials on the bodies  $M_1$  and  $M_2$ , and are expressed in terms of their mean charges  $Q_1 = e(\bar{N}_1 - N_1^0)$  and  $Q_2 = e(\bar{N}_2 - N_2^0)$ . The quantities  $\alpha_i$ ,  $\beta_i$ ,  $w_i$ , and  $C_i$  in (2.4) are defined by the formulas

$$\alpha_i = \frac{\alpha_{13}\alpha_{22} - \alpha_{23}\alpha_{12}}{\alpha_{11}\alpha_{22} - \alpha_{12}^2}, \quad \beta_i = \frac{\alpha_{23}\alpha_{11} - \alpha_{13}\alpha_{22}}{\alpha_{11}\alpha_{22} - \alpha_{12}^2}, \quad (2.5)$$

$$w_i = \alpha_i w_1^0 + \beta_i w_2^0 - w_3^0 - e^2 N_3^0 / C_i, \quad C_i = \alpha_{33}^{-1}.$$

If the junction contains many granules whose electrostatic interaction can be neglected<sup>(2)</sup>, then the Hamiltonian  $H_V$  is obtained from (2.4) by summing over the index  $i$ .

The quantities  $\alpha_i$ ,  $\beta_i$ ,  $w_i$ , and  $C_i$  will be regarded as the primary parameters of the particles, and we shall choose some distribution law for them. It can be shown that in the limit of large bodies  $M_{1,2}$  and of small granules (see Appendix 2) we have  $\alpha_i + \beta_i = 1$  and  $\beta_i = d_i/d$ , where  $d$  is the distance between the metals  $M_1$  and  $M_2$  while  $d_i$  is the distance from the surface of the metal to the granule. In this case the tunnel current turns out to depend on the difference of the potentials of two bodies  $V_1$  and  $V_2$ . In the general case, however, strictly speaking, there should be separate sensitivity to  $V_1$  and  $V_2$ , i.e., to the character of the joining of the contact with the external bodies (for example, to grounding of one of its edges). The quantity  $w_i$  has the meaning of the effective work function of the granule (which depends, generally speaking, on its dimensions), and  $C_i$  is the effective granule capacitance.

We note that the foregoing procedure used to separate the term  $H_V$  is applicable also to the ordinary two-body tunnel problem, where it yields the result<sup>[17]</sup>  $H_V = \hat{N}_1 eV_1 + \hat{N}_2 eV_2$ , which can be replaced, by virtue of gauge invariance, by  $\hat{N}_1 eV$ , where  $V$  is the potential

difference between the metals. Thus, the term (2.4) takes correctly into account all the electrostatic effects and expresses them in terms of macroscopic parameters, namely the external potentials  $V_1$  and  $V_2$ .

The complete Hamiltonian of the system (2.1) is a sum of the terms (2.2) and (2.4). The quantity  $H_0$  in (2.1) can describe, generally speaking, normal metals, superconductors, etc.

### 3. DENSITY MATRIX AND KINETIC EQUATION FOR THE DISTRIBUTION FUNCTION $W_n$

We consider a system of three bodies (see Fig. 1) and stipulate that the statistical operator  $\hat{\rho}$  remain stationary after turning on the interaction  $H_T$  (2.2). Let  $|n, \alpha\rangle$  be the eigenfunctions of the Hamiltonian  $H = H_0 + H_V$  (see Sec. 2), and let  $\alpha$  be the set of quantum numbers characterizing, besides the number of particles  $n$  on the granule, the state of the entire system. We put

$$\text{Sp}\{\hat{\rho}\}_n = \sum_{\alpha} \langle n\alpha | \hat{\rho} | n\alpha \rangle = W_n, \quad (3.1)$$

where  $\text{Sp}\{\dots\}_n$  is the trace at a fixed value of  $n$ .

After turning on the term  $H_T$ , the system is described by a density matrix  $\hat{\rho}'$  satisfying the Liouville equation

$$\frac{\partial \hat{\rho}'}{\partial t} = -i[H', \hat{\rho}'], \quad \hat{\rho}'_{t \rightarrow -\infty} = \hat{\rho}, \quad H' = H + H_T. \quad (3.2)$$

The solution of the equation takes the form

$$\hat{\rho}'(t) = e^{-iHt} S(t) \hat{\rho} S^{-1}(t) e^{iHt}, \quad S(t) = T \exp \left\{ -i \int_{-\infty}^t dt' H_T(t') \right\}. \quad (3.3)$$

Putting  $\tilde{\rho}(t) = e^{iHt} \hat{\rho}' e^{-iHt}$ , we obtain, accurate to terms quadratic in  $H_T$ ,

$$\partial \tilde{\rho} / \partial t = -i[H_T(t), \tilde{\rho}] - \int_{-\infty}^t dt' [H_T(t), [H_T(t'), \tilde{\rho}]]. \quad (3.4)$$

Applying the operation  $\text{Sp}\{\dots\}_n$  to this equation, we have

$$\frac{\partial W_n'}{\partial t} = -i \text{Sp}\{[H_T(t), \tilde{\rho}]\}_n - \int_{-\infty}^t dt' \text{Sp}\{[H_T(t), [H_T(t'), \tilde{\rho}]]\}_n. \quad (3.5)$$

Since  $H_T$  does not conserve the number of electrons on the granule, the first term vanishes. We shall calculate the second term by using the explicit form of the Hamiltonian  $H_T$  (2.2). Introducing the notation

$$\langle a_p^+(t) a_p(t') \rangle_n = \text{Sp} \{ a_p^+(t) a_p(t') \hat{\rho} \}_n / \text{Sp} \{ \hat{\rho} \}_n \quad (3.6)$$

and analogously for  $b^+$  and  $c^+$ , we get

$$\begin{aligned} \partial W_n' / \partial t &= \hat{L} \{ W_n \} = F_{n+1} - F_n \\ F_n &= W_n \{ P(n, q | n-1, p) + P(n, q | n-1, k) \} \\ &\quad - W_{n-1} \{ P(n-1, p | n, q) + P(n-1, k | n, q) \}, \end{aligned} \quad (3.7)$$

where

$$\begin{aligned} P(n, q | n-1, p) &= 4T_1^2 \text{Re} \sum_{pq} \int_{-\infty}^t dt' \langle a_p(t) a_p^+(t') \rangle_n \langle c_q^+(t) c_q(t') \rangle_n, \\ P(n, q | n-1, k) &= 4T_2^2 \text{Re} \sum_{kq} \int_{-\infty}^t dt' \langle b_k(t) b_k^+(t') \rangle_n \langle c_q^+(t) c_q(t') \rangle_n, \\ P(n-1, p | n, q) &= 4T_1^2 \text{Re} \sum_{pq} \int_{-\infty}^t dt' \langle a_p^+(t) a_p(t') \rangle_{n-1} \langle c_q(t) c_q^+(t') \rangle_{n-1}, \\ P(n-1, k | n, q) &= 4T_2^2 \text{Re} \sum_{kq} \int_{-\infty}^t dt' \langle b_k^+(t) b_k(t') \rangle_{n-1} \langle c_q(t) c_q^+(t') \rangle_{n-1}. \end{aligned} \quad (3.8)$$

The mean values of the product of the Fermi opera-

tors in the foregoing formulas can be regarded as not differing from the corresponding mean values at fixed chemical potentials of the bodies. The difference between these mean values, which represents the difference between the canonical and grand canonical ensembles, is small when the particle number is large. For the metals  $M_1$  and  $M_2$  this certainly takes place, and for the granule  $G$  we shall assume that this condition is also satisfied, inasmuch, as already noted, the spatial-quantization interval  $\epsilon_F / \bar{N}_i$  ( $\bar{N}_i$  is the average number of particles in the granule) is assumed to be small, i.e.,  $\bar{N}_i \gg 1$ . Nonetheless, a change of  $N_i$  by  $\pm 1$  leads to the appearance of a noticeable electrostatic energy and is not a negligibly small effect<sup>[4]</sup>.

The quantities  $P$  in (3.7) have the obvious meaning of the probabilities of transitions accompanied by change in the number of particles in the granule. The index  $p$  (the quantum number of the electron) fixes the body  $M_1$ , the index  $k$  the body  $M_2$ , and  $q$  respectively the granule  $G$ . Thus, for example,  $P(n, q | n-1, p)$  is the probability of a transition connected with a decrease in the number of electrons in the granule by unity ( $n \rightarrow n-1$ ), and appears when an electron tunnels between the granule  $G$  and the edge of  $M_1$  ( $q \rightarrow p$ ). The meaning of the remaining quantities  $P(\dots | \dots)$  is the same. Formula (3.7) is the kinetic equation for the system in question.

The integrals (3.8) are easy to calculate. We have

$$P(n, q | n-1, p) = \lambda_1 f(E_n - E_{n-1} - eV_1), \quad P(n, q | n-1, k) = \lambda_2 f(E_n - E_{n-1} - eV_2), \quad (3.9)$$

$$P(n-1, p | n, q) = \lambda_1 f(E_{n-1} - E_n + eV_1), \quad P(n-1, k | n, q) = \lambda_2 f(E_{n-1} - E_n + eV_2);$$

$$f(x) = x / (1 - e^{-\beta x}),$$

$$\lambda_1 = 4\pi T_1^2 N_1(0) N_i(0), \quad \lambda_2 = 4\pi T_2^2 N_2(0) N_i(0), \quad (3.10)$$

where  $N_{1,2}(0)$  and  $N_i(0)$  are the state densities at the Fermi level for the corresponding metals and  $E_n$  is the energy of the granule as a function of the number of electrons:

$$E_n = e^2 n^2 / 2C_1 + (\alpha e V_1 + \beta e V_2 + w_i) n. \quad (3.11)$$

In the equilibrium state we should have  $\partial W_n' / \partial t = 0$ .

Using (3.9), we see that if the potentials of the edges of the junction coincide ( $V_1 = V_2 = V$ ), then the solution of (3.7) is the Gibbs distribution function

$$W_n = Z^{-1} \exp [-\beta (E_n - neV)]. \quad (3.12)$$

At  $V_1 \neq V_2$  the distribution function does not take the form (3.12). In this case, although the stationarity condition  $\partial W_n' / \partial t = 0$  holds as before, the distribution over the number of particles is formed kinetically as a result of equality of the numbers of the direct and reverse transitions, and can differ appreciably from a Gibbs distribution.

### 4. TUNNEL CURRENT IN M-G-M SYSTEM

We proceed to the calculation of the tunnel current. In the tunnel-Hamiltonian method, the current is defined as the mean value

$$I = e \langle \dot{N}_1 \rangle = -e \langle \dot{N}_2 \rangle. \quad (4.1)$$

The indicated mean values coincide, since the Hamiltonian (2.2) conserves the total number of particles  $N_1 + N_2$ . Using the formula  $\langle \dot{N}_2 \rangle = \text{Tr} \{ \dot{N}_2 \rho' \}$  and abbreviating the description of the system by introducing the number of particles  $n$  in the granules (see Secs. 2 and 3), we obtain

$$I=e \sum_{n=-\infty}^{+\infty} W_n \{P(n, k|n+1, q) - P(n, q|n-1, k)\}. \quad (4.2)$$

The probabilities  $P(i|f)$  are given by formulas (3.8), while  $W_n$  satisfies the stationary condition (see (3.7)):

$$\tilde{L}\{W_n\}=0. \quad (4.3)$$

We assume that the system is connected to some external sources that maintain constant values of the potentials  $V_1$  and  $V_2$  (actually, constant values of the mean charges on the bodies  $M_1$  and  $M_2$ ). The change of the charges during tunneling is a slow process, and therefore the external sources needed to ensure the stationary condition can be disregarded in the calculation. For the same reason, each of the metals is characterized by an equilibrium (Fermi) distribution of the electrons with respect to their energies. The chemical potentials of this distribution (reckoned from the bottom of the band) remain unchanged, since the characteristic changes in the number of particles are of the order of unity and are small in comparison with their mean values  $\bar{N}_k \gg 1$ . In the case considered by us, the current in the system of granules is the sum of the currents flowing through the individual granules.

It is impossible to solve (4.3) in general form. We consider therefore limiting cases, in which it is possible to obtain the form of the distribution function  $W_n$ . These cases are the following: a) the case of small voltages ( $V_1 - V_2 \rightarrow 0$ ), when  $W_n$  deviates little from a Gibbs distribution; b) the case of an asymmetrical junction when, for example,  $T_1 \gg T_2$ , i.e., the  $M_1 \rightleftharpoons G$  and  $G \rightleftharpoons M_2$  tunneling probabilities differ strongly.

In the latter case, the resistance is determined mainly by the second process, and the distribution function  $W_n$  is close to Gibbsian for the pair of bodies ( $M_1 + G$ ), since the electron executes transitions between these bodies much more frequently, and goes over to the body  $M_2$  relatively rarely (violating equilibrium, i.e., producing a current).

## A. Linear Response

Equation (4.3) has a "first integral"

$$F_n = C \quad (4.4)$$

(see (3.7)), where  $C$  is a constant obtained from the normalization condition  $\Sigma W_n = 1$ . Under the condition  $|v| \ll 1$ , where

$$v = \beta e(V_1 - V_2), \quad (4.5)$$

we solve Eq. (4.4) by successive approximations, putting

$$\begin{aligned} W_n &= W_n^0 + v W_n^1 + \dots, \\ W_n^0 &= Z_0^{-1} \exp(-\beta \epsilon_n^0), \quad \epsilon_n^0 = e^2 n^2 / 2C_i + \tilde{w}_i n, \\ \tilde{w}_i &= w_i + (\alpha_i + \beta_i - 1) V_i. \end{aligned} \quad (4.6)$$

We note that in the macroscopic limit (see Sec. 2 and Appendix 2), when  $\alpha_i + \beta_i = 1$ , we have  $\tilde{w}_i = w_i$  and the dependence on  $V_i$  drops out.

For  $W_n^1$  we obtain the equation

$$-W_n^1 + W_{n-1}^1 \exp[\beta(\epsilon_{n-1}^0 - \epsilon_n^0)] = \frac{C_1}{\lambda_1 + \lambda_2} f^{-1}(\epsilon_n^0 - \epsilon_{n-1}^0) - \frac{\beta_i(\lambda_1 + \lambda_2) - \lambda_2}{\lambda_1 + \lambda_2} W_n^0,$$

where  $C_1$  is a constant. This equation has a solution satisfying the condition  $\Sigma W_n^1 = 0$  only at  $C_1 = 0$ , in the form

$$W_n^1 = \frac{\beta_i(\lambda_1 + \lambda_2) - \lambda_2}{\lambda_1 + \lambda_2} (n - \bar{n}) W_n^0,$$

$$\bar{n} = \sum_{n=-\infty}^{+\infty} n W_n^0. \quad (4.7)$$

Substituting (4.7) in (4.2) and using (3.9), we obtain

$$I = ev \frac{\lambda_1 \lambda_2}{\lambda_1 + \lambda_2} \sum_{n=-\infty}^{+\infty} f(\epsilon_n^0 - \epsilon_{n-1}^0) W_n^0, \quad (4.8)$$

where  $f(x)$  is defined in (3.9).

We introduce the quantities

$$R_1 = [4\pi e^2 N_i(0) N_i(0) T_1^2]^{-1}, \quad R_2 = [4\pi e^2 N_2(0) N_i(0) T_2^2]^{-1},$$

which have the meaning<sup>[17]</sup> of the resistances of the tunnel junctions  $M_1 - G$  and  $M_2 - G$ , without allowance for the effects due to discreteness of the charge. Then formula (4.8) shows that the resistance of the junction  $M_1 - G - M_2$  is proportional to the sum of the resistances  $R_1$  and  $R_2$ , and its value is

$$R = \gamma(R_1 + R_2) \quad (4.9)$$

with a certain proportionality coefficient  $\gamma > 1$ . The quantity  $\gamma^{-1}$  describes the decrease produced in the current by the effect of "quantization" of the charge, and is given by

$$\gamma^{-1} = \varphi(\xi, 0),$$

$$\begin{aligned} \varphi(\xi, \eta) &= \sum_{n=-\infty}^{+\infty} \exp[-\lambda(n - \xi + 1/2)^2] \frac{\lambda(n - \xi + 1/2) - \eta/2}{\text{sh}[\lambda(n - \xi + 1/2) - \eta/2]} \\ &\times \left\{ e^{\lambda/4} \sum_{n=-\infty}^{+\infty} \exp[-\lambda(n - \xi)^2] \right\}^{-1}, \end{aligned} \quad (4.10)$$

$$\lambda = \beta e^2 / 2C_i, \quad \xi = \tilde{w}_i C_i / e^2. \quad (4.11)$$

At high temperatures ( $\lambda \rightarrow 0$ ) we have  $\gamma = 1$ . At low temperatures ( $\lambda \gg 1$ ), to the contrary, an important role is played in the sums (4.10) (depending on the value of  $\xi$ ) by either one or two terms corresponding to  $n \sim \xi$ . In this case, the resistance oscillates when the parameter  $\xi$  is varied:

$$\gamma = \text{sh} \{2\lambda(\xi - E[\xi] - 1/2)\} / \lambda(\xi - E[\xi] - 1/2) \quad (4.12)$$

( $E[x]$  is the integer part of  $x$ ). A similar effect was noted earlier<sup>[4]</sup>.

At high temperatures ( $\lambda \ll 1$ ), the oscillating increment  $R_{\text{osc}}$  to the resistance  $R = R_1 + R_2$  is exponentially small:

$$R_{\text{osc}} = \gamma_{\text{osc}}(R_1 + R_2), \quad \gamma_{\text{osc}} = 2\pi(\pi/\lambda)^{1/2} \exp(-\pi^2/\lambda) \cos 2\pi\xi. \quad (4.13)$$

According to (4.11), (4.6), and (2.5), the quantity  $\xi$  is determined by the parameters of the granule and in the macroscopic limit, when  $\alpha_i + \beta_i = 1$ , it takes the form<sup>3)</sup>

$$\xi = C_i [\alpha_i(w_1^0 - w_2^0) + w_2^0 - w_3^0] / e^2. \quad (4.14)$$

As seen from (4.12) and (4.13), the resistance of the junction depends on the ratio of the work function of the granule  $w_3^0$  to the work functions of the junction edges  $w_1^0$  and  $w_2^0$ . If the bodies  $M_1$  and  $M_2$  and the granule  $G$  are made of identical metals ( $w_1^0 = w_2^0 = w_3^0$ ), then  $\xi = 0$  and the resistance depends exponentially on the temperature:

$$R = (R_1 + R_2) \frac{2C_i T}{e^2} \exp\left(\frac{e^2}{2C_i T}\right), \quad (4.15)$$

$$T \ll \frac{e^2}{C_i}.$$

On the other hand, if  $w_3^0 - w_{1,2}^0 \neq 0$  (different materials of the edges and granules), then, generally speaking,  $e^2/C_i |w_{1,2}^0 - w_3^0| \ll 1$ , i.e.,  $\xi$  is a large quantity. When

the periodicity in  $\xi$  is taken into account, the averaging over the parameters of the granules reduces in this case to integration with respect to  $\xi$  from 0 to 1. After averaging, the oscillations in (4.12) vanish, and the temperature dependence of the resistance takes the form<sup>4)</sup>

$$R = 4\pi^{-2} (R_1 + R_2) e^2 / C_i T, \quad T \ll e^2 / C_i. \quad (4.16)$$

At low temperatures, the resistance increases in comparison with its value at high temperatures. A "zero anomaly" of this type is observed in experiment<sup>[5,6]</sup>. We note that at a granule diameter  $r \sim 10^3 \text{ \AA}$ , the temperatures satisfying the condition  $\lambda > 1$  amount to  $\lesssim 10^6 \text{ K}$  (if the dielectric constant is  $\epsilon \approx 10$ ).

The temperature dependences of the reduced conductivity  $\sigma = (R_1 + R_2) / R$ , calculated in accordance with formula (4.10), are shown in Fig. 2. As seen from the figure,  $\sigma$  increases with increasing  $T$  and becomes equal at  $T \gg e^2 / C_i$  to the value of the conductivity without allowance for the electrostatic barriers. As already noted, at  $w_1^0 = w_2^0 = w_3^0$  the dependence of  $\sigma$  on  $T$  as  $T \rightarrow 0$  is exponential (curve b), but if averaging is carried out with respect to the parameters  $w_1^0$  (curve a), then  $\sigma \sim T$ .

## B. Current-voltage Characteristic of Asymmetrical M-G-M Junction

Let us find the  $I(V)$  dependence at  $R_1 \gg R_2$ . In this case, according to (4.16), the scale of the junction resistance is  $R \sim R_2$ , i.e., it is determined by transitions in the  $G - M_2$  "bottleneck". We seek the solution of (4.4) in the form

$$W_n = W_n^0 + \epsilon W_n^1 + \dots, \quad \epsilon = \lambda_2 / \lambda_1 \ll 1,$$

where  $\lambda_1$  is obtained from (3.10). For  $w_n^0$  we obtain the equation ( $\epsilon_n^{1,2} = E_n - nV_{1,2}$ )

$$W_{n-1}^0 \exp[\beta(\epsilon_{n-1}^1 - \epsilon_n^1)] - W_n^0 = C f^{-1}(\epsilon_n^1 - \epsilon_{n-1}^1),$$

the solution of which

$$W_n^0 = Z^{-1} \exp(-\beta \epsilon_n^1) \quad (4.17)$$

corresponds to a Gibbs distribution for the bodies  $M_1 + G$ . The constant  $C$  should be set equal to zero, for otherwise the function  $W_n$  increases as  $n \rightarrow \infty$ , i.e., the corresponding distribution cannot be normalized to unity. This confirms the hypothesis advanced above concerning the equilibrium between the bodies  $M_1$  and  $G$ .

The remainder of the calculation is trivial. Calculating  $W_n^1$  and substituting the corresponding expression in the formula for the current (4.2), we get

$$I = 2 \frac{V_1 - V_2}{v R_2} \text{sh} \left( \frac{v}{2} \right) \varphi(\xi', v), \quad (4.18)$$

$$\xi' = \xi - \beta v / 2\lambda, \quad (4.19)$$

where  $\varphi(\xi, \eta)$ ,  $\lambda$ ,  $\xi$ , and  $v$  are defined by formula (4.10), (4.11), and (4.5). As  $v \rightarrow 0$ , this expression coincides with (4.10). It is seen from (4.10) and (4.18) that  $I$  is periodic in  $\xi'$  with a period  $\Delta \xi' = 1$ . Since  $\xi'$  depends on  $v$ , Eqs. (4.18) describes the nonmonotonic dependence of  $I$  on  $v$ .

To obtain the current-voltage characteristic (CVC) of the tunnel junction it is necessary to average (4.18) over the parameters of the granule. The result of the averaging depends significantly on the ratio of the work

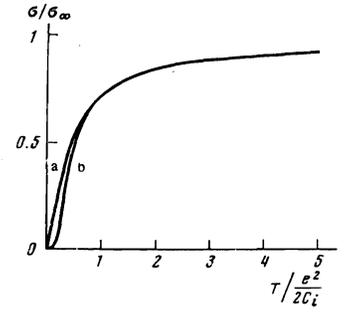


FIG. 2. Temperature dependence of the conductivity of a tunnel junction M-G-M at  $V = 0$ : a—conductivity averaged over the work functions  $w_1^0$  of the granules, b—case of constant  $w_1^0$  ( $w_1^0 = w_2^0 = w_3^0$ ).

functions of the granule  $w_3^0$  and of the edges of the junction  $w_1^0$  and  $w_2^0$ .

1) If  $C_i |w_3^0 - w_{1,2}^0| / e^2 \gg 1$ , then the averaging reduces to integration with respect to  $\xi'$  from zero to unity (see footnote 4). Averaging causes the oscillations of  $I(V)$  to vanish, and the CVC becomes monotonic. The differential conductivity  $\sigma(V) = d\bar{I}/dV$  ( $\bar{I}(V)$  is the averaged current) at  $T = 0$  ( $\lambda = \infty$ ) takes the form

$$\sigma = \sigma_\infty \begin{cases} u, & u < 1 \\ 1, & u > 1 \end{cases}, \quad u = \frac{VC_i}{e}, \quad (4.20)$$

where  $\sigma_\infty$  is the conductivity of the junction without allowance for the effects of the discreteness of the charge. At high temperatures ( $\lambda \rightarrow 0$ ) we have  $\sigma(V) = \sigma_\infty$ .

2) If the granule and the edges of the junction are made of identical metals ( $w_1^0 = w_2^0 = w_3^0$ ), then  $\xi = 0$  (see (4.14)), and when averaging (4.15) it is necessary to take into account the explicit dependence of the parameter  $\xi'$  (4.19) on the position of the granule in the junction. As seen from (4.19), this dependence is determined by the parameter  $\beta_1$ , which takes the following form in the considered model (Appendix 2)

$$\beta_1 = x/d, \quad (4.21)$$

where  $x$  is the distance between the granule and the surface of the nearest large body ( $M_1$ ), and  $d$  is the distance between the metals  $M_1$  and  $M_2$ . Depending on the position of the granule, a change takes place also in the resistance  $R_2$ . We shall henceforth assume that  $R_2(x)$  takes the form

$$R_2(x) = R_0 e^{\alpha(d/2-x)}. \quad (4.22)$$

If the granules are distributed uniformly with respect to the coordinate  $x$ , then it is necessary, in order to calculate the average current  $I(V)$ , to substitute (4.21) and (4.22) in (4.18) and (4.19), and to integrate the result with respect to  $x$  from 0 to  $d/2$ . With this method of averaging,  $\bar{I}(V)$  retains both the characteristic threshold due to the activation mechanism of the conductivity and the oscillations due to the discreteness of the charge. At high temperatures ( $\lambda \rightarrow 0$ ), the CVC is mainly ohmic ( $\sigma(V) \sim \sigma_\infty$ ), and the characteristic oscillatory increments to the differential conductivity  $\sigma(V)$  are exponentially small ( $\sigma_{osc}(V) \sim \exp(-\pi^2/\lambda)$ ).

At low temperatures ( $\lambda \gg 1$ ), the threshold character of the conductivity and of the oscillations of the CVC manifest themselves most strongly. If  $T = 0$  ( $\lambda = \infty$ ), we obtain for  $\sigma(V)$  the expression ( $\kappa d \gg 1$ )

$$\sigma(V) = \sigma_\infty (\Omega_0 + \Omega_{osc}), \quad (4.23)$$

$$\Omega_0 = \theta(u-1), \quad (4.24)$$

$$\Omega_{osc} \approx \frac{1}{2} \begin{cases} u^{-1} \exp[\kappa d(1-u^{-1})/2] \theta(u^{-1/2}), & u < 1 \\ -1 + \frac{\kappa d(1+2E[u/2])}{u^2} \exp\left[\frac{\kappa d(1-2D)}{2u}\right] \theta\left(D - \frac{1}{2}\right), & 1 < u < \kappa d \\ \kappa d u^{-1} [\theta(D^{-1/2}) - D], & \frac{1}{\kappa d} \ll u \end{cases} \quad (4.25)$$

$$D = \frac{u}{2} - E \left[ \frac{u}{2} \right], \quad \theta(x) = \begin{cases} 1, & x > 0 \\ 0, & x < 0 \end{cases} \quad (4.26)$$

The quantities  $u$  and  $\kappa$  are defined in (4.20) and (4.22).

Figure 3 and 4 show plots of  $\sigma/\sigma_\infty$  against  $u$  for both considered methods of averaging, calculated numerically for a number of values of the parameter  $\lambda$ . From the experimental point of view it appears that the relations shown in Fig. 3 are more probable (or, at least, relations of Fig. 4 with weak oscillations), since the granule parameters<sup>[5]</sup> are subject to a large scatter.

### C. Capacitance Oscillations

In the case of an asymmetrical junction, it is easy to calculate the change produced in the charge by the redistribution of the electrons among the edges of the junction and the granules. The charge on the granule

$$\Delta Q = e \sum_{n=-\infty}^{+\infty} W_n (n - N_s^0) \quad (4.27)$$

can be obtained by substituting in place of  $W_n$  the distribution function  $W_n^0$  (formula (4.17)). The quantity  $\Delta Q$  contains an increment  $\Delta Q_{osc}$  that oscillates with the voltage. We find the oscillating part of the capacitance  $\Delta C_{osc} = d(\Delta Q_{osc})/dV$ . We have

$$\Delta C_{osc} \approx \begin{cases} \frac{1}{2} C_i \beta_i \lambda / ch^2 (\xi' - E[\xi']^{-1/2}) \lambda, & \lambda \gg 1 \\ -C \lambda^{-1} \beta_i 4\pi^2 \exp(-\pi^2/\lambda) \cos 2\pi \xi', & \lambda \ll 1 \end{cases} \quad (4.28)$$

where  $\xi'(V)$  is determined by (4.19) and (4.11). These oscillations of the capacitance in an asymmetrical junction were observed experimentally<sup>[8]</sup>.

### 5. TUNNEL CURRENT IN S-G-S JUNCTION

In this section we calculate the current in a tunnel junction whose electrodes are identical superconductors (S), and the granule (G) is in the normal state. In this junction, the main contribution to the current is made by quasiparticle tunneling. The Cooper-pair current (the Josephson current) is a quantity of second order in the transparency  $T_{1,2}^2$  of the junctions, and the contribution of this current will be neglected. In this case, just as before, the calculation of the current calls for solution of the system of equations (4.2) and (4.3), with (3.7) and (3.8) taken into account. The only difference is that the mean values of the operators pertaining to the edges of the junction, which enter in (3.8), must be calculated over the states of the superconductor. Taking this into account, we obtain for  $P(i|f)$

$$P(n, q|n-1, p) = \lambda_1 \tilde{f}(E_n - E_{n-1} - eV_1), \quad P(n, q|n-1, k) = \lambda_2 \tilde{f}(E_n - E_{n-1} - eV_2), \\ P(n-1, p|n, q) = \lambda_1 \tilde{f}(E_{n-1} - E_n + eV_1), \quad P(n-1, k|n, q) = \lambda_2 \tilde{f}(E_{n-1} - E_n + eV_2),$$

$$\tilde{f}(E) = e^{\beta E/2} \Phi(\beta E/2) = \int_{-\infty}^{+\infty} dx \frac{|x| \theta(|x| - \Delta)}{(x^2 - \Delta^2)^{1/2}} n_F(x - E) [1 - n_F(x)], \quad (5.1)$$

where  $n_F(x) = (1 + e^{\beta x})^{-1}$  is the Fermi distribution function and  $\Delta$  is the energy gap. The quantities  $\lambda_{1,2}$  and  $E_n$  are defined in (3.10) and (3.11). The function  $\tilde{f}(E)$  coincides with  $f(E)$  (3.9) at  $\Delta = 0$ .

The subsequent calculations are analogous to those in Sec. 4. The kinetic equation (4.3) was solved for the same limiting cases ( $V \rightarrow 0$  and  $T_2/T_1 \rightarrow 0$ ) that were considered in Sec. 4.

#### A. Linear Response

Solving the kinetic equation (4.3) and substituting its solution together with the quantities (5.1) in the formula for the current (4.2) we obtain

$$I = ev \frac{\lambda_1 \lambda_2}{\lambda_1 + \lambda_2} \sum_{n=-\infty}^{+\infty} \tilde{f}(e_{n-1} - e_n^0) W_n^0. \quad (5.2)$$

The quantities  $v$ ,  $W_n^0$ , and  $\epsilon_n^0$  are defined in (4.5) and (4.6), while  $\tilde{f}(E)$  is defined in (5.1). If we introduce the resistances  $R_1$  and  $R_2$  of the tunnel junctions  $M_1 - G$  and  $M_2 - G$  (see Sec. 4), then we can obtain from (5.2) expressions analogous to (4.9) and (4.10):

$$R = \tilde{\gamma}(R_1 + R_2), \quad (5.3)$$

$$\tilde{\gamma}^{-1} = \tilde{\Phi}(\xi, 0), \quad (5.4)$$

$$\tilde{\Phi}(\xi, \eta) = \sum_{n=-\infty}^{+\infty} \exp[-\lambda(n - \xi + 1/2)^2] \Phi[\lambda(n - \xi + 1/2) - \eta/2]$$

$$\times \left\{ e^{\lambda/4} \sum_{n=-\infty}^{+\infty} \exp[-\lambda(n - \xi)^2] \right\}^{-1}, \quad (5.5)$$

where the quantities  $\lambda$ ,  $\xi$ , and  $\Phi(x)$  are defined in (4.11) and (5.1).

At  $\Delta = 0$ , expressions (5.4) and (5.5) coincide with the analogous results (formula (4.10)) for the M-G-M junction. The quantity  $\tilde{\gamma}$  is an oscillating function of the parameter  $\xi$  with period  $\Delta \xi = 1$ . At  $\lambda \ll 1$  (high temperatures), the oscillating part of  $\tilde{\gamma}$  is exponentially small

$$\sigma^i = \tilde{\gamma}^{-1} = \sigma_\infty + \sigma_{osc}, \quad (5.6)$$

$$\sigma_{osc} \approx \begin{cases} (2\pi\beta\Delta)^{1/2} e^{-\beta\Delta/2}, & \beta\Delta \gg 1 \\ 1 + \beta\Delta/2, & \lambda \ll \beta\Delta \ll 1, \\ 1, & \beta\Delta \ll \lambda \end{cases} \quad (5.7)$$

$$\sigma_{osc} \approx 2\pi e^{-\pi^2/\lambda} \begin{cases} \frac{(\beta\Delta)^{1/2}}{ch(\beta\Delta/2)} \frac{\exp[(\pi^2 - (\beta\Delta)^2)/4\lambda]}{[\pi^2 + (\beta\Delta)^2]^{1/2}} \sin\left(\frac{\pi\beta\Delta}{2\lambda} + \varphi\right) \sin 2\pi\xi, & \beta\Delta \gg \lambda \\ -\beta\Delta (\pi\lambda)^{-1/2} e^{\pi^2/4\lambda} \cos 2\pi\xi, & e^{-\pi^2/4\lambda} \ll \beta\Delta \ll \lambda, \\ (\pi/\lambda)^{1/2} \cos 2\pi\xi, & \beta\Delta \ll e^{-\pi^2/4\lambda} \end{cases} \quad (5.8)$$

$\varphi = (\pi/2) \tan^{-1}(\pi/\beta\Delta)$ . With decreasing temperature, the amplitude of the oscillations increases, and at  $\lambda \gg 1$  we obtain for  $\sigma^S$

$$\sigma^S = \Phi[\lambda(\xi - E[\xi']^{-1/2})/2] / 2 \text{ch} \lambda(\xi - E[\xi']^{-1/2}), \quad (5.9)$$

$\Phi(x)$  is defined in (5.1).

To obtain the total resistance of the S-G-S junction it is necessary to average formulas (5.6)–(5.9) over the granule parameters. If  $w_1^0 = w_2^0 = w_3^0$ , then according to (4.14) the temperature dependence of the resistance  $R$  is determined by the expressions (5.6)–(5.9), in which we should put  $\xi = 0$ . On the other hand, if  $C_{i1}|w_3^0 - w_2^0|/e^2 \gg 1$ , then the averaging, just as in Sec. 4,

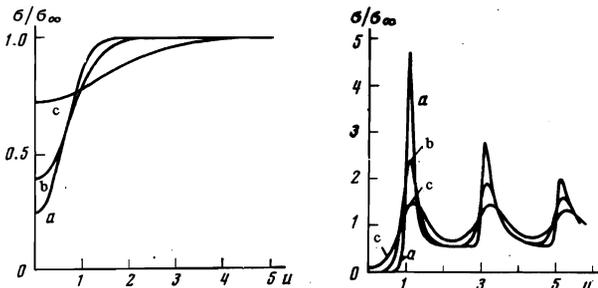


FIG. 3

FIG. 4

FIG. 3. Derivative of the current-voltage characteristic  $\sigma = dI/dV$  of an M-G-M junction, averaged over the values of the work function  $w_i^0$ : a)  $\lambda = 5$ , b)  $\lambda = 3$ , c)  $\lambda = 1$ , where  $\lambda = e^2/C_i T$  and  $u = VC_i/e$  is the reduced voltage.

FIG. 4. The  $dI/dV - V$  characteristic of monometallic M-G-M junctions ( $w_1^0 = w_2^0 = w_3^0$ ): a)  $\lambda = 30$ ,  $\kappa d = 20$ ; b)  $\lambda = 10$ ,  $\kappa d = 20$ ; c)  $\lambda = 5$ ,  $\kappa d = 20$

reduces to integration of (5.6)–(5.9) with respect to  $\xi$  between 0 and 1. At  $\lambda \ll 1$  we obtain

$$\sigma = \bar{R}^{-1} = \sigma_{\infty} \sigma_{\infty}^{\prime}, \quad (5.10)$$

$\sigma_{\infty}$  is the conductivity of the M-G-M junction without allowance for the discreteness of the charge, and  $\sigma_{\infty}^{\prime}$  is given by formula (5.7).

At low temperatures ( $\lambda \gg 1$ ) after integration of (5.10) with respect to  $\xi$  we obtain

$$\sigma \approx \sigma_{\infty} \begin{cases} (2\pi\beta\Delta)^{1/2} e^{-\beta\Delta}, & \beta\Delta \gg \lambda \\ (2\pi\beta\Delta)^{1/2} \beta\Delta e^{-\beta\Delta/2\lambda}, & 1 \ll \beta\Delta \ll \lambda \\ 1/\lambda^{-1}(\pi^2/4 + \beta\Delta), & \beta\Delta \ll 1 \end{cases} \quad (5.11)$$

In contrast to the M-G-M junction, the temperature dependence of  $\sigma = 1/\bar{R}$  is exponential (at  $\beta\Delta \gg 1$ ), this being due to the presence of the energy gap  $\Delta$  in the spectrum of the quasiparticles on the edges of the S-G-S junction. Figure 5 shows the temperature dependences of  $\sigma/\sigma_{\infty}$ , calculated numerically for a number of values of the parameters  $\delta = C_1\Delta/e^2$ .

## B. Current-voltage Characteristics of Asymmetrical S-G-S Junction

If  $R_1 \ll R_2$ , then Eqs. (4.3) and (3.7) can be solved by perturbation theory with respect to the small parameter  $\epsilon = \lambda_2/\lambda_1$ . After calculations that are perfectly analogous to those of Sec. 4, and after substituting  $W_n$  in formula (4.2) for the current, with allowance for (5.1), we obtain

$$I = 2 \frac{V_1 - V_2}{vR_2} \text{sh} \left( \frac{v}{2} \right) \bar{\varphi}(\xi', v), \quad (5.12)$$

where  $\xi'$  and  $\bar{\varphi}(\xi, \eta)$  are defined in (4.19) and (5.4). Since  $I(\xi')$  is periodic and  $\xi'$  is linear in the voltage (see (5.4) and (4.19)), expression (5.12) reflects the nonmonotonic character of  $I(V)$ . Just as in Sec. 4, the form of the current-voltage characteristic of the junction depends essentially on the method of averaging over the granule parameters.

1)  $C_1 |w_{1,2}^0 - w_3^0|/e^2 \gg 1$ . The oscillating increment to the current vanishes upon integration with respect to  $\xi'$ . At  $T = 0$  we obtain for the differential conductivity  $\sigma(V) = dI/dV$  the expression

$$\sigma(V) = \sigma_{\infty} \Omega, \quad (5.13)$$

$$\Omega = \theta(u - \delta) \{ [(u^2 - \delta^2)^{1/2} - ((u-1)^2 - \delta^2)^{1/2}] \theta(u-1-\delta) + (u^2 - \delta^2)^{1/2} \theta(\delta+1-u) \}, \quad (5.14)$$

$$\delta = \Delta C_1 / e^2, \quad (5.15)$$

$u$  is defined by (4.26). Equations (5.13) and (5.14) coin-

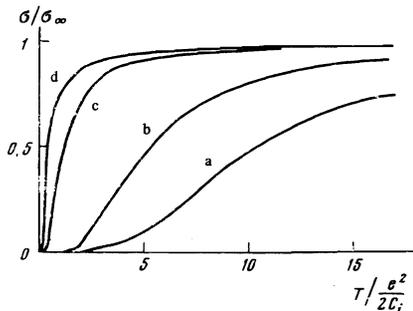


FIG. 5. Temperature dependence of the conductivity of a mono-metallic ( $w_1^0 = w_2^0 = w_3^0$ ) S-G-S tunnel junction: a)  $\delta = 10$ , b)  $\delta = 5$ , c)  $\delta = 1$ , d)  $\delta = 0.1$ , where  $\delta = \Delta C_1 / e^2$  is a parameter. Averaging over the granule parameters  $W_1^0$  does not lead to a noticeable change in the shape of the curves.

cide at  $\Delta = 0$  with expression (4.20) obtained for the M-G-M junction. The conductivity has a threshold  $u = \delta$ , which is typical of S-N junctions<sup>[17]</sup>. At high temperatures ( $\lambda \rightarrow 0$ ), the CVC coincides with the well-known result

$$I(V) = \sigma_{\infty} (e^{\beta e V} - 1) \bar{f}(-eV) \quad (5.16)$$

where  $\bar{f}$  is given by (5.1). Figure 6 shows plots of the CVC calculated numerically for a number of values of the parameter  $\delta$  at finite temperature.

2)  $w_1^0 = w_2^0 = w_3^0$ . The parameter  $\xi'$ , which enters in the expression for the current (5.12), is determined by the relations (4.19) (at  $\xi = 0$ ) and (4.21), and depends on the distance  $x$  between the granule and the nearest edge. When averaging over the different positions of the granule, it is necessary to take into account the dependence of  $R_2$  in (5.12) on  $x$  (see (4.22)). If the granules are uniformly distributed over the thickness of the junction, the averaging reduces to integration of (5.12) with respect to  $x$  from 0 to  $d/2$  ( $d$  is the thickness of the S-G-S junction). With this method of averaging, the CVC retains the characteristic oscillations. At high temperatures ( $\lambda \ll 1$ ), the oscillating increment to the current is exponentially small ( $\sim \exp[-\text{const}/\lambda]$ ), and the CVC is determined mainly by (5.16). With decreasing temperature, the contribution of the oscillations increases. Even in the case  $T = 0$  ( $\lambda = \infty$ ), the expression for the differential conductivity  $\sigma^S(V)$  is quite cumbersome. However, if  $|eV| \gg \Delta + e^2 C_1$ , this expression simplifies to

$$\sigma^S(V) = \sigma(V) (1 + \delta^2 / 2u^2), \quad (5.17)$$

where  $u$  and  $\delta$  are defined in (4.20) and (5.15), while  $\sigma(V)$  is the conductivity of the M-G-M junction.

## 6. CONCLUSIONS

The main results of the present paper reduces to the following:

1. The description of the tunnel charge transport in granulated systems differs from the usual scheme used to calculate the current in tunnel structures. In these systems, the relaxation process connected with the redistribution of the charge among the granules is quite slow, so that the distribution function of the charge on the granules should be determined by solving the corresponding kinetic equation, and does not coincide in the general case with the Gibbs function.

2. Owing to the smallness of the granules, the system is on the whole sensitive to the discreteness of the electric charge, so that the tunnel current and the charges on the granules are nonmonotonic functions of the voltage.

3. The form of the current-voltage characteristic

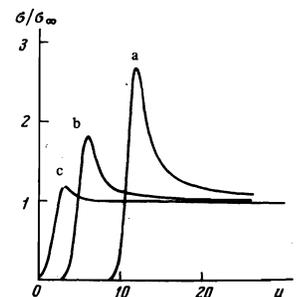


FIG. 6. Derivative of the CVC of an S-G-S junction for the case  $\delta = 5$  and for parameter values  $\delta = 5$  (a), 2 (b), and 0.5 (c).

and the temperature dependence of the resistance of a M-G-M junction depends significantly on the relation between the work functions  $w_1^0$  and  $w_2^0$  of the edges of the junction and  $w_3^0$  of the granules. If  $C_1 |w_{1,2}^0 - w_3^0|/e^2 \gg 1$ , then after averaging over the parameters of the granules the current-voltage characteristic does not contain the characteristic electrostatic threshold, and is monotonic (Fig. 3). The temperature dependence of the conductivity  $\sigma(V=0)$  at sufficiently low temperatures ( $T \ll e^2/C_1$ ) is linear (see (4.16)), i.e.,  $\sigma$  increases with increasing  $T$ . On the other hand, if  $C_1 |w_{1,2}^0 - w_3^0|/e^2 \ll 1$ , then the current voltage characteristic contains a characteristic electrostatic threshold ( $e/C_1$ ) and oscillations whose period is equal to double the threshold voltage (Fig. 4). The temperature dependence of the conductivity  $\sigma(V=0)$  is exponential in this case:  $\sigma(V=0) \sim \exp(-e^2/C_1 T)$ .

4. The current-voltage characteristic of a S-G-S junction has a threshold character, and the temperature dependence of  $\sigma(V=0)$  is exponential regardless of the relation between the work functions of the granules and the edges of the junction. The effective threshold of the current-voltage characteristic is determined by the sum of the "superconducting" ( $\Delta$ ) threshold and the electrostatic threshold ( $e^2/C_1$ ) (see Fig. 5). If  $C_1 |w_{1,2}^0 - w_3^0|/e^2 \ll 1$ , then  $\sigma(V)$  contains oscillations with a period  $2e/C_1$ , which vanish in the opposite limiting case.

In conclusion, the authors are grateful to É. A. Kel'man for help with the computer calculation of the plots.

## APPENDIX 1

We consider the potential produced by a point charge  $q$  located in a dielectric (with dielectric constant  $\epsilon$ ) between two metallic bodies (Fig. 7). To find the potential  $\varphi$  at the point  $(r, z)$  ( $r$  is the radius vector in the plane perpendicular to the  $z$  axis) it is necessary to take into account the images produced by the point charge (shown by crosses). We obtain

$$\varphi(r, z) = \frac{q}{\epsilon} \sum_{n=-\infty}^{+\infty} [((z-z_n^-)^2 + r^2)^{-1/2} - ((z-z_n^+)^2 + r^2)^{-1/2}],$$

$z_n^\pm = 2nd \pm z$ . Using the Poisson summation formula, we get

$$\varphi(r, z) = \frac{2iq}{d\epsilon} \sum_{n=-\infty}^{+\infty} \exp(i\alpha_n z) \times \sin(\alpha_n x) K_0(|\alpha_n| r),$$

where  $\alpha_n = -\pi n/d$  and  $K_0(x)$  is a modified Bessel function.

If  $\pi r/d \gg 1$ , then  $|\alpha_n| r \gg 1$  (for  $n \neq 0$ ), and we can use the asymptotic form of the Bessel function. Omitting the exponentially small terms ( $\sim \exp(-2\pi r/d)$ ), we obtain ultimately

$$\varphi(r, z) \approx \frac{2q}{\epsilon r} \left(\frac{2r}{d}\right)^{1/2} \sin\left(\frac{\pi z}{d}\right) \sin\left(\frac{\pi x}{d}\right) \exp\left(-\frac{\pi r}{d}\right), \quad \frac{\pi r}{d} \gg 1.$$

It is seen from this formula that the electrostatic interaction of the granules is screened by the metallic edges and decreases exponentially with increasing distance  $r$  between them.

## APPENDIX 2

If the granule dimensions are small enough, then we can neglect the influence of the granule on the distribution of the charge on the surfaces of the large bodies  $M_1$  and  $M_2$ . To calculate the potential coefficients  $\alpha_{ijk}$  of the considered three-body system, we can then use the "spherical capacitor" model shown in Fig. 8. In this model, when calculating  $\alpha_{11}$ ,  $\alpha_{12}$ ,  $\alpha_{13}$ ,  $\alpha_{22}$ , and  $\alpha_{23}$ , the granule  $G$  is regarded as a point charge. The matrix  $\alpha_{ijk}$  takes the form

$$\alpha_{ik} \approx \frac{1}{\epsilon} \times \begin{pmatrix} (a_1+a_2+d)^{-1} & (a_1+a_2+d)^{-1} & (a_1+a_2+d)^{-1} \\ (a_1+a_2+d)^{-1} & a_2^{-1} + (a_1+a_2+d)^{-1} - (a_2+d)^{-1} & (a_2+d)^{-1} - (a_2+d)^{-1} + (a_1+a_2+d)^{-1} \\ (a_1+a_2+d)^{-1} & (a_2+d)^{-1} - (a_2+d)^{-1} + (a_1+a_2+d)^{-1} & r_0^{-1} \end{pmatrix},$$

where  $r_0$  is the characteristic dimension of the granule  $G$ . In the limit  $d/a_{1,2} \ll 1$  we obtain, for  $\alpha_{ijk}$ , accurate to terms  $\sim d/(a_{1,2})^2$ ,

$$\alpha_{ik} \approx \frac{1}{\epsilon} \begin{pmatrix} \alpha_0 - d\alpha_0^2 & \alpha_0 - d\alpha_0^2 & \alpha_0 - d\alpha_0^2 \\ \alpha_0 - d\alpha_0^2 & \alpha_0 + d\alpha_1^2 & \alpha_0 + d_1\alpha_1^2 - d_2\alpha_0^2 \\ \alpha_0 - d\alpha_0^2 & \alpha_0 + d_1\alpha_1^2 - d_2\alpha_0^2 & r_0^{-1} \end{pmatrix},$$

$$\alpha_0 = (a_1+a_2)^{-1}, \quad \alpha_1^2 = a_2^{-2} - (a_1+a_2)^{-2}.$$

Using this matrix, we easily obtain

$$\lim_{a_{1,2} \rightarrow \infty} \beta_i = \lim_{a_{1,2} \rightarrow \infty} \frac{\alpha_{23}\alpha_{11} - \alpha_{13}\alpha_{12}}{\alpha_{11}\alpha_{22} - \alpha_{12}^2} = \frac{d_1}{d},$$

$$\lim_{a_{1,2} \rightarrow \infty} \alpha_i = \lim_{a_{1,2} \rightarrow \infty} \frac{\alpha_{13}\alpha_{22} - \alpha_{23}\alpha_{12}}{\alpha_{11}\alpha_{22} - \alpha_{12}^2} = \frac{d_2}{d},$$

Thus, the quantities  $\alpha_i$  and  $\beta_i$  do not depend on the dielectric constant  $\epsilon$ , nor on the dimensions of  $M_1$  and  $M_2$ , and are determined by the position of the granule in the junction.

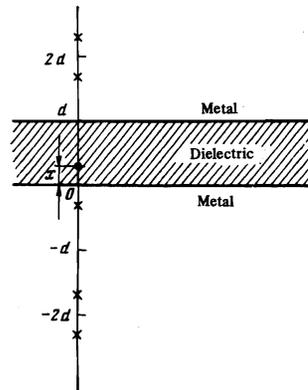


FIG. 7

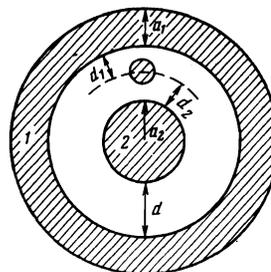


FIG. 8

<sup>1</sup>Actually, the characteristic electrostatic energy is  $U_1 \sim e^2/\epsilon r$ , and the distance between energy levels is  $U_2 \sim \epsilon F/N \sim \epsilon F a^3/r^3$ . At not too small values of  $r$ , we have  $U_1 \gg U_2$  ( $r$  is the particle radius,  $N$  is the total number of electrons in the particle,  $a$  is the interatomic distance, and  $\epsilon$  is the dielectric constant; in order of magnitude, we have  $e^2/a \sim \epsilon F$ ).

<sup>2</sup>As shown in Appendix 1, the electrostatic interaction of the granules  $g_i$  and  $g_j$  is screened by the edges of the junction, and its order of magnitude, at  $r_{ij} \gg d$ , is  $(e^2/\epsilon r_{ij})(r_{ij}/d)^{1/2} \exp(-\pi r_{ij}/d)$ , where  $r_{ij}$  is the distance between granules and  $d$  is the distance between the edges of  $M_1$  and  $M_2$ . The mutual electrostatic influence of the granules can therefore be neglected at  $r_{ij} \gg d$ .

<sup>3</sup>In expression (4.14) we have omitted the integer  $N_0^3$ . Since the quantity  $\gamma$  (4.10) is periodic in  $\xi$  with a period equal to unity, it is possible to add to  $\xi$  any arbitrary integer.

<sup>4</sup>The calculation of the conductivity reduces to the calculation of the integral  $\int_0^1 \gamma^{-1}(\xi) d\xi$  at a fixed  $C_i$ . Formula (4.16) pertains to this case.

The next step, generally speaking, is averaging over  $C_i$ , using some distribution function with respect to the granule parameters. As a result we obtain an expression of the type (4.16), where the role of  $C_i$  will be assumed by the mean values of the capacitance.

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