Phase transition in granulated superconductors

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The influence of the Coulomb energy on the superconducting-transition temperature in granulated materials is investigated. A microscopic analysis yields an effective Hamiltonian that depends on the number of particles and on the phase as the variables. A study of the system described by this Hamiltonian shows that at zero temperature there exists a critical value of the Josephson energy, of the order of the Coulomb energy, below which superconductivity is impossible. At nonzero temperature the Coulomb interaction is screened and superconductivity is possible at lower values of the Josephson energy. A sufficiently high temperature enhances the fluctuations and destroys the superconductivity. It is shown that at low values of the Josephson energy the system is an insulator in the region where the superconductivity is destroyed.

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1. INTRODUCTION

Granulated material, meaning a system of metallic grains coated with an insulator layer, is presently the subject of intensive investigation. Depending on the material, the size of the grains can range from several dozen to several tens of thousands angstroms. The small thickness of the insulator enables the electrons to tunnel from one grain to another. This tunneling determines the property of the entire material. At low temperatures, the electron-phonon interaction leads to formation of a superconducting gap in each grain. This, however, does not produce a phase transition in an isolated small grain.[6,7] Only the tunneling of the electrons from one grain to another can cause a phase transition and the onset of superconductivity in the entire sample.[8] Of extreme importance in this case is the Coulomb interaction of the electrons. For one electron to transfer from one grain to a neighboring one, the energy required is of the order of e^2/d, where e is the electron charge and d is the grain dimension. If the grain is small, this energy can become larger than the grain binding energy. According to a qualitative analysis presented in Ref. 9, superconductivity is impossible at this energy ratio. We report below an investigation of the feasibility of a superconducting transition at various relations between the Coulomb energy, the grain binding energy, and the superconducting gap width. The analysis is carried for the case when the grain dimension is smaller than the radius of the Cooper pair. It is assumed at the same time that the distance between the energy levels in each grain is less than the size of the Cooper gap. This imposes a lower bound on the grain size. The partition function is calculated by continual integration with respect to the order parameter. It turns out that the fluctuations of the modulus of the order parameter are small in a wide range of temperatures. Only the phase fluctuations are substantial. The functional of the free energy is derived. It depends on the phases and includes the Coulomb interaction. Averaging with this functional yields the thermodynamic and kinetic quantities. It is shown that even at zero temperature a superconducting transition is possible only when the Josephson energy exceeds the energy needed by the electron to leave the grain. When the coupling constant is decreased, the superconductivity is destroyed and the system goes into the dielectric state. At nonzero temperature, a screening of the Coulomb interaction by the charged excitations is possible. This leads to the existence of superconductivity in the region of lower Josephson energies. When the coupling constants between the grains are large, the transition temperature is close to the value calculated in the BCS approximation. In this region, the Coulomb interaction becomes completely negligible and the superconductivity limit is determined only by the Josephson energy.

2. CHOICE OF MODEL

We consider a system of metal grains. The electrons in each grain interact via phonons. In addition, the electrons are subject to Coulomb repulsion. The insulator film that coats each grain can be described by introducing a potential barrier that prevents free motion of the electrons from one grain to another. The Hamiltonian $\hat{H}$ of such a system is

$$\hat{H}=\int\Big[\frac{1}{2m}\sum_{\alpha}(\nabla\psi_\alpha^+(\mathbf{r})\nabla\psi_\alpha(\mathbf{r})-p_F^2\psi_\alpha^+(\mathbf{r})\psi_\alpha(\mathbf{r}))$$
$$-V_0\sum_{\alpha,\beta}\psi_\alpha^+(\mathbf{r})\psi_\beta^+(\mathbf{r})\psi_\beta(\mathbf{r})\psi_\alpha(\mathbf{r})+\sum_{\alpha}U(\mathbf{r})\psi_\alpha^+(\mathbf{r})\psi_\alpha(\mathbf{r})\Big]d\mathbf{r}$$
$$+\frac{1}{2}\int_{|\mathbf{r}-\mathbf{r}'|>\Lambda}\sum_{\alpha,\beta}D_0(\mathbf{r}-\mathbf{r}')\psi_\alpha^+(\mathbf{r})\psi_\beta^+(\mathbf{r}')\psi_\beta(\mathbf{r}')\psi_\alpha(\mathbf{r})\,d\mathbf{r}\,d\mathbf{r}', \qquad (1)$$

where $D_0=e^2/|\mathbf{r}-\mathbf{r}'|$ and e is the electron charge.

In the Hamiltonian (1), the operators $\psi_\alpha^+(\mathbf{r})$ and $\psi_\alpha(\mathbf{r})$ denote respectively the operators for the creation and annihilation of the electron with spin α at the point $\mathbf{r}$. The first term in (1) describes the electron kinetic energy reckoned from the Fermi energy ε_F. The quantity V_0 describes the interaction via the phonons. In addition, this quantity includes the short-range part of the Coulomb repulsion. The third term in (1) describes the action of the external potential due to the presence of the dielectric coating. The last term in (1) takes into account the long-range part of the Coulomb interaction. The integration is carried out at distances larger than Λ. The quantity Λ is the cutoff parameter that satisfies the relation $p_F^{-1}\ll\Lambda\ll d$, where p_F is the

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Fermi momentum and d is the characteristic dimension of the grain.

The thermodynamic quantities are expressed in the usual manner in terms of sums over the states of the Hamiltonian H. For example, the free energy Ω is given by

$$\beta\Omega=-\ln \operatorname{Sp}\exp(-\beta\hat{H}), \tag{2}$$

where $\beta=1/T$ is the reciprocal temperature.

It is convenient to use in the calculation by means of (2) the method of integration over an auxiliary field.[10] This method was used to describe isolated grains.[7] The integration in Ref. 7, however, was carried out only over static fields. To consider the influence of Coulomb interaction, account must be taken of the time-dependent fields. The usual transformations yield

$$\beta\Omega=-\ln\int \exp(-\beta\mathcal{F}[\Delta,\Delta^*])D\Delta(\mathbf{r},\tau)D\Delta^*(\mathbf{r},\tau). \tag{3}$$

The free-energy functional that enters in (3) is written in the form

$$\beta\mathcal{F}[\Delta,\Delta^*]=-\ln \operatorname{Sp}\exp\left(-\int_0^\beta \hat{H}_0(\tau)\,d\tau\right)T_\tau \times\exp\left(-\int_0^\beta \hat{H}_{int}(\tau)\,d\tau\right)\exp\left(-\int_0^\beta\frac{|\Delta(\mathbf{r},\tau)|^2\,d\mathbf{r}\,d\tau}{V_0}\right), \tag{4}$$

where

$$\hat{H}_0(\tau)=\sum_\alpha\psi_\alpha^+(\mathbf{r})\left(-\frac{1}{2m}\frac{\partial^2}{\partial\mathbf{r}^2}-\varepsilon_F+U(\mathbf{r})\right)\psi_\alpha(\mathbf{r})\,d\mathbf{r} +\int[\Delta(\mathbf{r},\tau)\psi_\uparrow^+(\mathbf{r})\psi_\downarrow^+(\mathbf{r})+\Delta^*(\mathbf{r},\tau)\psi_\downarrow(\mathbf{r})\psi_\uparrow(\mathbf{r})]\,d\mathbf{r}, \tag{5}$$

T_τ denotes the chronological operator.

The term $\hat{H}_{\text{int}}$ in (4) describes the long-range part of the Coulomb interaction and is equal to

$$\hat{H}_{int}=\frac{1}{2}\sum_{\alpha,\beta}\int_{|\mathbf{r}-\mathbf{r}'|>\Lambda} D_0(\mathbf{r}-\mathbf{r}')\psi_\alpha^+(\mathbf{r},\tau)\psi_\beta^+(\mathbf{r}',\tau)\psi_\beta(\mathbf{r}',\tau)\psi_\alpha(\mathbf{r},\tau)\,d\tau\,d\mathbf{r}\,d\mathbf{r}', \tag{6}$$

where

$$\psi_\alpha(\mathbf{r},\tau)=\exp\left(\int_0^\beta\hat{H}_0(\tau)\,d\tau\right)\psi_\alpha(\mathbf{r})\exp\left(-\int_0^\beta\hat{H}_0(\tau)\,d\tau\right).$$

The continual integration in (3) will be carried out separately with respect to the modulus and with respect to the phase of the external field Δ. The dependence of the free-energy functional $\mathcal{F}[\Delta,\Delta^*]$ on the modulus $|\Delta|$ is a strong one in a wide range of temperatures and the integration can be carried out by the saddle-point method. We use for the calculations the Green's-function formalism employed in superconductivity theory. Introducing the normal and anomalous Green's functions $G(\mathbf{r},\mathbf{r}',\tau)$ and $F(\mathbf{r},\mathbf{r}',\tau)$ defined by

$$\begin{aligned}G(\mathbf{r},\mathbf{r}',\tau)&=\langle T_\tau\psi_\uparrow(\mathbf{r},0)\psi_\uparrow^+(\mathbf{r}',\tau)\rangle,\\ F(\mathbf{r},\mathbf{r}',\tau)&=\langle T_\tau\psi_\downarrow(\mathbf{r},0)\psi_\uparrow(\mathbf{r}',\tau)\rangle\end{aligned} \tag{7}$$

and neglecting the term H_{int} in the Hamiltonian, we obtain the equations

$$\begin{gathered}(\partial/\partial\tau-\nabla_r^2/2m-\varepsilon_F+U(\mathbf{r}))G(\mathbf{r},\mathbf{r}',\tau)\\ -\Delta(\mathbf{r},\tau)F^*(\mathbf{r},\mathbf{r}',\tau)=\delta(\mathbf{r}-\mathbf{r}')\delta(\tau),\\ (-\partial/\partial\tau-\nabla_r^2/2m-\varepsilon_F+U(\mathbf{r}))F^*(\mathbf{r},\mathbf{r}',\tau)+\Delta^*(\mathbf{r},\tau)G(\mathbf{r},\mathbf{r}',\tau)=0.\end{gathered} \tag{8}$$

Assuming the dependence of $|\Delta|$ on τ to be slow and minimizing $\mathcal{F}[\Delta,\Delta^*]$ with respect to $|\Delta|$, we obtain a third equation

$$\Delta(\mathbf{r})=V_0F(\mathbf{r},\mathbf{r}'). \tag{9}$$

Equations (8) and (9) constitute the system of Gor'kov's equations. The only difference in the case of a homogeneous bulk superconductor lies in the presence of the external potential $U(\mathbf{r})$. If this potential is large enough, then the solution of the system (8) and (9) reduces to a solution of the corresponding system in the isolated grain. Integration with respect to the momenta is then replaced by summation over discrete levels. If the grain is not perfectly spherical, the distance ΔE between the levels is determined by the formula

$$\Delta E=1/VN(0), \tag{10}$$

where $N(0)$ is the density of the states on the Fermi surface and V is the volume of the granule.

At sufficiently low value of ΔE, the summation over the levels can be replaced by integration, and we arrive at the usual BCS formula

$$\Delta_0=V_0N(0)T\sum_{\varepsilon_n}\int\frac{\Delta_0\,d\xi}{\varepsilon_n^2+\xi^2+\Delta_0^2}. \tag{11}$$

Replacement of the sum over the discrete levels by an integral is legitimate if

$$\Delta E=1/VN(0)\ll T_{c0}, \tag{12}$$

where T_{c0} is the temperature at which the solution first appears in (11).

The appearance of a nonzero solution in (11) does not mean a phase transition in the isolated granule, since allowance for the fluctuations smears out the singularity. The fluctuations of the modulus of the order parameter turned out to be small under the following condition

$$|T_{c0}-T|/T_{c0}\gg(T_{c0}N(0)V)^{-1/2}. \tag{13}$$

These fluctuations were considered in Refs. 6 and 7. The condition (13) shows that when the inequality (12) is satisfied there exists an extensive region of temperatures, in which the fluctuations of the modulus of the order parameter are small. The analysis that follows will pertain precisely to this region. Equation (11) was derived without allowance for the long-range part of the Coulomb interaction and without allowance for the jumps from grain to grain. This neglect is legitimate if the frequency T_{ij} of the jumps from one grain to another exceeds the Coulomb energy E_c that appears when the number of electrons in the grain is decreased by unity:

$$S_{ij}T_{ij}\gg E_c\sim e^2/d, \tag{13a}$$

where S_{ij} is the area of the contact between the i-th and j-th grains. The square of the jump frequency $|T_{ij}|^2$ is proportional to the coefficient of the penetrability through the barrier. This coefficient is assumed to be small. When condition (13a) is satisfied, the system of granules in the absence of superconducting pairing would be a normal metal. In the opposite limiting case, a Mott transition would take place and the system would become an insulator. The condition (13a) is hereafter assumed satisfied throughout.

Equation (11) determines the modulus of the order parameter, but not its phase. Therefore the dependence of the functional $\mathscr{F}[\Delta, \Delta^*]$ on the phase is a slow one. In the next section this functional will be reduced to a simpler form.

3. DERIVATION OF THE FREE-ENERGY FUNCTIONAL

To simplify the functional $\mathscr{F}[\Delta, \Delta^*]$ defined in (4), in which the modulus of the order parameter is fixed, we neglect for the time being the coupling between the grains. Only slow changes of the phase will be of importance hereafter. We therefore expand the functional $\mathscr{F}[\Delta, \Delta^*]$ (4) in terms of the deviations of the phase:

$$\beta\mathscr{F}[\Delta,\Delta^*]=\beta\mathscr{F}_0+{}^1/_2\int_0^\beta\int d\tau\, d\tau'\, d\mathbf{r}\, d\mathbf{r}'\, \Phi(\mathbf{r},\mathbf{r}',\tau-\tau')\varphi(\mathbf{r},\tau)\varphi(\mathbf{r}',\tau'), \quad (14)$$

where

$$\Phi(\mathbf{r},\mathbf{r}',\tau-\tau')=\delta^2\mathscr{F}/\delta\varphi(\mathbf{r},\tau)\delta\varphi(\mathbf{r}',\tau').$$

$\mathscr{F}_0$ in (14) is the equilibrium part. There are no terms linear in φ, since the expansion is about the equilibrium position. Carrying out the functional differentiation in (4), we obtain

$$\begin{aligned}{}^1/_2\Phi(\mathbf{r},\mathbf{r}',\tau-\tau')&=\Delta_0^*F(0,0)\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau')\\&-\langle T_\tau\psi_\downarrow(\mathbf{r},\tau)\psi_\uparrow(\mathbf{r},\tau)\psi_\uparrow^+(\mathbf{r}',\tau')\psi_\downarrow^+(\mathbf{r}',\tau')\rangle|\Delta|^2\\&-\langle T_\tau\psi_\downarrow(\mathbf{r},\tau)\psi_\uparrow(\mathbf{r},\tau)\psi_\downarrow(\mathbf{r}',\tau')\psi_\uparrow(\mathbf{r}',\tau')\rangle\Delta^{*2}.\end{aligned} \quad (15)$$

The averaging in (15) is over the states of a Hamiltonian that includes a term $\hat{H}_{\text{int}}$ that describes the long-range Coulomb part. Only connective graphs are taken into account.

We calculate the mean values in (15) by perturbation theory, expanding the powers of H_{int}. The main contribution can be taken into account in the random-phase approximation. The quantities Φ are determined from the following equations:

$$\Phi_\omega(\mathbf{r}^i,\mathbf{r}'^j)=\delta_{ij}\Pi_{1\omega}(\mathbf{r}^i,\mathbf{r}'^j)+4\sum_m\Pi_{2\omega}(\mathbf{r}^i,\mathbf{r}_1^i)D(\mathbf{r}_1^i-\mathbf{r}_2^m)\tilde{\Pi}_{2\omega}(\mathbf{r}_2^m,\mathbf{r}'^j)\,d\mathbf{r}_2^m\,d\mathbf{r}_1^i,$$

$$\tilde{\Pi}_{2\omega}(\mathbf{r}^i,\mathbf{r}'^j)=\Pi_{2\omega}(\mathbf{r}^i,\mathbf{r}'^j)\delta_{ij}-\sum_m\Pi_{3\omega}(\mathbf{r}^i,\mathbf{r}_1^i)D(\mathbf{r}_1^i-\mathbf{r}_2^m)\tilde{\Pi}_{2\omega}(\mathbf{r}_2^m,\mathbf{r}'^j)\,d\mathbf{r}_1^i\,d\mathbf{r}_2^m. \quad (16)$$

The indices of the variables in (16) designate the number of the grain. The integration is over each grain. The quantity $\Phi_\omega(\mathbf{r},\mathbf{r}')$ denotes the Fourier component of $\Phi(\mathbf{r},\mathbf{r}',\tau-\tau')$ with respect to time. The functions $\Pi_{\alpha\omega}(\alpha=1,2,3)$ are equal to

$$\begin{aligned}\Pi_{1\omega}(\mathbf{r},\mathbf{r}')&=-\Delta^*F(0,0)\delta(\mathbf{r}-\mathbf{r}')-T|\Delta|^2\sum_\varepsilon G_\varepsilon(\mathbf{r},\mathbf{r}')G_{-\varepsilon+\omega}(\mathbf{r},\mathbf{r}')\\&-T\Delta^{*2}\sum_\varepsilon F_\varepsilon(\mathbf{r},\mathbf{r}')F_{-\varepsilon+\omega}(\mathbf{r},\mathbf{r}'),\\\Pi_{2\omega}(\mathbf{r},\mathbf{r}')&=-T\sum_\varepsilon G_\varepsilon(\mathbf{r},\mathbf{r}')F_{\varepsilon+\omega}(\mathbf{r}',\mathbf{r}),\\\Pi_{3\omega}(\mathbf{r},\mathbf{r}')&=T\sum_\varepsilon[G_\varepsilon(\mathbf{r},\mathbf{r}')G_{\varepsilon+\omega}(\mathbf{r}',\mathbf{r})+F_\varepsilon(\mathbf{r},\mathbf{r}')F^*_{\varepsilon+\omega}(\mathbf{r}',\mathbf{r})],\end{aligned} \quad (16a)$$

where $G_\varepsilon(\mathbf{r},\mathbf{r}')$ and $F_\varepsilon(\mathbf{r},\mathbf{r}')$ are determined by Eqs. (8). If the potential barrier is high enough, the functions $G_\varepsilon(\mathbf{r},\mathbf{r}')$ and $F_\varepsilon(\mathbf{r},\mathbf{r}')$ differ substantially from zero only when both arguments, $\mathbf{r}$ and $\mathbf{r}'$, are located in the same grain. This circumstance is reflected in Eqs. (16).

The condition that the probability of electron jumps from grain to grain be small is written in the form

$$T_{ij}\ll\varepsilon_F p_F^2, \quad (17)$$

where T_{ij} is the jump frequency referred to unit area. This frequency can in principle be expressed in terms of the potential $U(\mathbf{r})$. We shall not need, however, the explicit form of this relation.

The inequality (17) is the upper bound of the jump frequency. It is assumed as before that the lower bound (13a) is satisfied. The condition (13a) allows us to neglect the influence of the Coulomb interaction on the form of the Green's functions in (16) and (16a).

In the considered approximation $d\ll\xi_0$, where ξ_0 is the dimension of the Cooper pair, a substantial contribution to the physical quantities is made only by configurations in which $\varphi(r^i)$ is constant over the entire volume of the grain. The lowest nonzero harmonic has an energy $(\xi_0/d)^2$ times larger than the characteristic energy of the superconductor. Therefore the contribution of the nonzero harmonics can be disregarded. The free-energy functional (14) contains only the quantity Φ_{ij}:

$$\Phi_{ij\omega}=\int\Phi_\omega(\mathbf{r}^i,\mathbf{r}'^j)\,d\mathbf{r}^i\,d\mathbf{r}'^j. \quad (18)$$

To solve Eqs. (16) we use the fact that the characteristic distance over which the functions $\Pi_{2\omega}(\mathbf{r},\mathbf{r}')$ and $\Pi_{3\omega'}(\mathbf{r},\mathbf{r}')$ decrease is of the order of p_F^{-1}. This distance is much smaller than the distances over which $D(\mathbf{r}-\mathbf{r}')$ changes. We can therefore integrate in (16) directly with respect to the rapidly varying functions. The integration yields

$$\Phi_{ij\omega}=\omega^2\left[\delta_{ij}a_1V_i-4a_2\sum_m\int D(\mathbf{r}^i-\mathbf{r}_2^m)n_j(\mathbf{r}_2^m)\,d\mathbf{r}_2^m\,d\mathbf{r}^i\right],$$

$$n_j(\mathbf{r}^i)=a_2\delta_{ij}-a_3\sum_m\int D(\mathbf{r}^i-\mathbf{r}_2^m)n_j(\mathbf{r}_2^m)\,d\mathbf{r}_2^m, \quad (19)$$

where

$$\begin{aligned}\omega^2a_1&=\int\Pi_{1\omega}(\mathbf{r}^i,\mathbf{r}'^i)\,d\mathbf{r}'^i,\quad \omega a_2=\int\Pi_{2\omega}(\mathbf{r}^i,\mathbf{r}'^i)\,d\mathbf{r}'^i,\\a_3&=\int\Pi_{3\omega}(\mathbf{r}^i,\mathbf{r}'^i)\,d\mathbf{r}'^i,\quad \omega n_j(\mathbf{r}^i)=\int\tilde{\Pi}_{2\omega}(\mathbf{r}^i,\mathbf{r}'^j)\,d\mathbf{r}'^j.\end{aligned} \quad (20)$$

We solve the second equation of (19) by successive approximations. The zeroth approximation $n_j^{(0)}(\mathbf{r}^i)$ is taken to be the solution of this equation without the left-hand side:

$$a_2\delta_{ij}=a_3\sum_m\int D(\mathbf{r}^i-\mathbf{r}_2^m)n_j^{(0)}(\mathbf{r}_2^m)\,d\mathbf{r}_2^m. \quad (21)$$

It is difficult to solve (21) in explicit form. We note, however, that this equation describes the classical electro-static problem. The solution $n_j^{(0)}$ is the distribution of the charges over different granules that produce a potential a_2/e^2a_3 part of the grains and a zero potential at all the remaining ones. Therefore the integral of $n_j^{(0)}(r^i)$ over the grain can be expressed in terms of the capacitance matrix C_{ij}:

$$\int n_j^{(0)}(\mathbf{r}^i)\,d\mathbf{r}^i=\frac{a_2}{a_3e^2}C_{ij}. \tag{22}$$

Calculating the first correction to $n_j^{(0)}(\mathbf{r}_i)$, substituting the obtained solution in (19), and using (22) we reduce Eq. (19) for $\Phi_{ij\omega}$ to the form

$$\Phi_{ij\omega}=\omega^2\left[V_i\delta_{ij}\frac{dN}{d\mu}\lambda(T)(1-\lambda(T))+\lambda^2(T)\frac{C_{ij}}{(2e)^2}\right], \tag{23}$$

where N is the density of the Cooper pairs, and μ is the energy of the pair on the Fermi surface. The function $\lambda(T)$ determines the relative number of superconducting pairs:

$$\lambda(T)=\pi T\sum_{\varepsilon}\Delta^2/(\varepsilon^2+\Delta^2)^{3/2}.$$

In the derivation of (23) we used the inequality $v_F C_{ij}/e^2V_ip_F^2\ll 1$. This inequality ensures smallness of the corrections when Eqs. (19) are solved by successive approximations. In addition, it was assumed that the frequencies ω are much smaller than $T_{c0}(\omega\ll T_{c0})$. At zero temperature all the electrons are paired and $\lambda(0)=1$.

In this case $\Phi_{ij\omega}$ is inversely proportional to the Coulomb energy. At nonzero temperature, the "normal" electrons screen the Coulomb energy and increase Φ_{ij} strongly. This screening is possible if condition (13a) is satisfied, when the "normal" electrons can flow over freely from grain to grain.

Expressions (14), (18), and (23) determine the functional of the phase in the zeroth approximation in $T_{ij}/\varepsilon_F p_F^2$. Allowance for the next-order approximation leads to the appearance of Josephson terms. By means of the usual calculations[11] we obtain

$$\mathscr{F}[\varphi]=\sum_{i,j}\int_0^\beta[\dot\varphi_i\dot\varphi_j/2B_{ij}+J_{ij}(1-\cos(\varphi_i-\varphi_j))]\,d\tau. \tag{24}$$

In (24), B_{ij} is the matrix inverse to $\Phi_{ij\omega}/\omega^2$. The Josephson energy J_{ij} differs from zero only for the nearest neighbors and is expressed in terms of the junction resistance R_{ij} in the normal state:

$$J_{ij}=\frac{\pi}{8e^2R_{ij}}\Delta_0\,\mathrm{th}\frac{\Delta_0}{2T}. \tag{25}$$

The order of magnitude of J_{ij} is described by the relation

$$J_{ij}\sim T_{ij}S_{ij}\left(\frac{\Delta_0}{\varepsilon_F}\frac{T_{ij}}{p_F{}^2\varepsilon_F}\right). \tag{25a}$$

The factor in the parentheses in (25a) is much less than unity and therefore the ratio of the Josephson and Coulomb energies can be arbitrary even if condition (13a) is satisfied.

Equations (3) and (24) enable us to calculate the thermodynamic quantities. It is more convenient to replace the continual integration with the functional $\mathscr{F}[\varphi]$ (24) by summation with respect to the eigenstates of the effective Hamiltonian $\hat H_{\mathrm{eff}}$:[12]

$$\hat H_{\mathrm{eff}}=\sum_{ij}[{}^1/_2B_{ij}\hat\rho_i\hat\rho_j+J_{ij}(1-\cos(\varphi_i-\varphi_j))], \tag{26}$$

where the operator $\hat\rho_i$ is given by

$$\hat\rho_i=-i\partial/\partial\varphi_i. \tag{26a}$$

The operators $\hat\rho_i$ are the operators of the number of the Cooper pairs in the i-th grain. The eigenvalues of the operators $\hat\rho_i$ are integers. The first term in the Hamiltonian (26) describes, with allowance for the screening by the normal electrons, the electrostatic energy that results from pair production on the grains. With the aid of the Hamiltonian (26) we shall investigate in the next section the thermodynamics and the response to an electromagnetic field.

4. PHASE TRANSITION

In the preceding section, the distribution of the quantities B_{ij} and J_{ij} was assumed arbitrary. We assume henceforth that the system is regular and the quantities depend only on the difference $|i-j|$. To investigate the properties of the system we use the self-consistent-field method. Replacing the interaction in (26) by the average field

$$\cos(\varphi_i-\varphi_j)\to\langle\cos\varphi_i\rangle\cos\varphi_j \tag{27}$$

and determining the phase-transition point from the condition that a nonzero value of $\langle\cos\varphi\rangle$ appear, we obtain the equation

$$1={}^1/_2J\int_0^\beta\Pi(\tau)\,d\tau, \tag{28}$$

where

$$\Pi(\tau)=\langle\exp(i\varphi)\exp(\tau\hat H_{\mathrm{eff}}^{(0)})\exp(-i\varphi)\exp(-\tau\hat H_{\mathrm{eff}}^{(0)})\rangle_0,\qquad J=\sum_j J_{ij}. \tag{28a}$$

In Eqs. (28), the averaging is over the states of the free Hamiltonian $\hat H_{\mathrm{eff}}^{(0)}$:

$$\hat H_{\mathrm{eff}}^{(0)}=\sum_{ij}{}^1/_2B_{ij}\hat\rho_i\hat\rho_j. \tag{29}$$

The eigenfunctions of the Hamiltonian (29) can be written in the form

$$\psi_{n_1n_2\ldots n_N}(\varphi_1,\varphi_2,\ldots,\varphi_N)=\exp(i\varphi_1n_1+i\varphi_2n_2+\ldots+i\varphi_Nn_N). \tag{30}$$

The numbers $1,2,\ldots,N$ in (30) label the grains. The function $\psi_{n1\ldots nN}$ should be periodic with a period 2π. From this condition it follows that $n_1,n_2,\ldots,n_N$ are integers. The wave function (30) describes a state in which n_i particles are located on the i-th grain.

Using (26a) and (28)–(30), we reduce the correlator $\Pi(\tau)$ to the form

$$\Pi(\tau)=Z^{-1}\sum_{n_1,n_2,\ldots,n_N}\exp\left[-\frac{\tau B_{11}}{2}-\tau\sum_j B_{1j}n_j-\frac{1}{2T}\sum_{i,j}B_{ij}n_in_j\right], \tag{31}$$

where Z is the partition function

$$Z=\sum_{n_1,n_2,\ldots,n_N}\exp\left[-\frac{1}{2T}\sum_{i,j}B_{ij}n_in_j\right]. \tag{31a}$$

Expression (31) is valid at not very high temperatures: $T \ll T_{c0}$. Near T_{c0} only the term with zero energy is significant in (23) and (24). In this case all the formulas become classical and the correlator $\Pi(\tau)$ is equal to unity.

At zero temperature, only the state with uncharged grains makes a contribution (all $n_i=0$). In this case the correlator $\Pi(\tau)$ is equal to

$$\Pi(\tau)=\exp(-\tau B_{11}/2). \tag{32}$$

The correlator $\Pi(\tau)$ is defined on the imaginary-time axis. In real time, the correlator is described by an exponential of an imaginary argument. This means that the phase of an isolated granule rotates, on the average, with angular velocity $B_{11}/2$ equal to the energy of a state with one pair.

Calculating the integral of the correlator $\Pi(\tau)$ (31), we reduce (28) to the form

$$1=Z^{-1}\frac{J}{2}\sum_{n_1,n_2,\ldots,n_N}\frac{B_{11}}{(B_{11}/2)^2-(\sum_j B_{1j}n_j)^2}\exp\left(-\sum_{i,j}\frac{B_{ij}n_in_j}{2T}\right). \tag{33}$$

At zero temperature we obtain the value of J_c at which superconductivity appears:

$$J_c={}^1/_2B_{11}(0). \tag{34}$$

At $J<J_c$ no superconductivity is possible. This conclusion was arrived at qualitatively in Ref. 9.

Equation (34) can be interpreted as a Mott transition for the Cooper pairs. The Josephson energy assumes the role of the width of the band. If this width is less than the Coulomb energy, the superconductivity is destroyed and, as will be shown in the next section, the system becomes an insulator. The unpaired electrons, which appear at nonzero temperature, can then flow freely from grain to grain, since the condition (13a) is assumed satisfied.

Equation (33) contains the quantities B_{ij}, which are proportional at low temperatures to the elements of the inverse of the capacitance matrix. If the grains are close-packed so that the distance between them is much less than the dimension d of the grains themselves, then the order of magnitude of the element B_{11} is given by[1]

$$B_{11}\sim(e^2/d)\,(a/d\varepsilon_0),$$

where ε_0 is the dielectric constant of the medium between the grains. The quantity B_{11} can be both larger and smaller than T_{z0}. We consider first the case of small $G_{ij}(B_{ij} \ll T_{c0})$. At low temperatures $T \ll B_{11}$, and an appreciable contribution to (33) is made, besides the ground state, only by the lowest excited states. The lowest excited state in the considered system is a dipole consisting of charges +1 and −1 located on neighboring grains. The position of the dipole in space can be arbitrary. Taking the contribution of one dipole into account, we obtain from (33)

$$1=\frac{2J}{B_{11}}\left[1+\sum_{i,j}\left(\frac{1}{1-4((B_{1j}-B_{1,j+1})/B_{11})^2}-1\right)e^{-E_1/T}\right], \tag{35}$$

where $E_1=B_{11}-B_{12}$ is the dipole-excitation energy.

In (35), the summation over i is carried out over the nearest neighbors with respect to j. The quantities B_{ij} were taken at zero temperatures, where they are inversely proportional to the capacitance matrix. The summation in (35), for the case of close packing of the granules, can be carried only numerically. At large distances, the function B_{ij} decreases by $|i-j|^{-1}$, so that the sum converges. The entire factor preceding the exponential in (35) is of the order of unity. In the considered case of close packing, all the terms in the sum preceding the exponential in (35) are positive, and therefore the entire factor is positive. This means that for superconductivity to set in when the temperature is increased it is necessary to have lower values of J. This effect has a simple explanation: a low temperature "shakes up" the phase little, but leads to excitations that screen the Coulomb interaction. The weakening of the Coulomb interaction facilitates the penetration of the Cooper pairs from grain to grain and this, in turn facilitates the onset of superconductivity.

If the distance between the grains is much less than the grain dimension, then the off-diagonal quantities B_{ij} with $i \neq j$ are much less than the diagonal B_{ij}. In this case the lowest excitation is the appearance of a charged Cooper pair on one of the grains. For such a system we get from (33)

$$1=\frac{2J}{B_{11}}\left[1-\frac{8}{3}e^{-B_{11}/2T}\right]. \tag{36}$$

In a system consisting of widely spaced granules, the screening is ineffective and therefore a rise in temperature leads to a weakening in the superconductivity (36). A diagonal Hamiltonian with $B_{ij}=0$ at $i=j$ and B_{ii} independent of temperature was postulated in Ref. 13. To investigate the Hamiltonian, the approximation (27) was made, after which the self-consistency equation was derived. The equation obtained in Ref. 13 differs from (36), although the employed approximations are the same. On the basis of the equation introduced in Ref. 13 it was concluded that in the case of a diagonal Hamiltonian the superconductivity becomes stronger when the temperature is raised. This statement is apparently the consequence of a computational error.

In (36) the off-diagonal terms were completely discarded. However, the B_{ij} decreased slowly as $|i-j|\to\infty$. As a result, the contribution of the single-particle excitations in (33) is formally divergent. The summation of the diverging terms can be carried out by the Debye-Hückel method. Estimates of this contribution showed that it contains exponentials with higher arguments than those written out in (36).

At high temperatures $T \gg B_{11}$ the correlation function that enters in Eq. (28) for T_c can be replaced by unity. We then obtain the classical formula

$$J/2T_c=1. \tag{37}$$

At sufficiently high jump probability, the temperature of the transition is close to T_{c0}. In this case Eq. (37) is also applicable if the temperature-dependent expression (25) is used for J. Combining the formulas in (35) for the case of close packing with formula (37), we can qualitatively draw the entire phase diagram. Such a diagram is shown in the figure. In the considered region $B_{11} \ll T_{c0}$ the quantity $J_{\min}$ is of the same order as J_c. In the region between $J_{\min}$ and J_c, two temperature phase transitions take place.

The phase diagram is much more strongly influenced by the screening in the case of strong Coulomb interaction $B_{11}(0) \gg T_{c0}$. At low temperatures, only single-electron excitations above the superconducting gap are significant. Excitation of pairs above the Coulomb gap requires a high energy. Using (34), in which the quantities J and B_{11} must be taken at finite temperature T, we obtain at low temperatures

$$J(0)=\frac{B_{11}(0)}{2}-V\frac{dN}{d\mu}\sum_j B_{1j}(0)B_{j1}(0)\left(\frac{2\pi\Delta_0}{T_c}\right)^{1/2}e^{-\Delta_0/T_c}. \qquad (38)$$

Expression (38) is valid so long as the second term is much less than the first. The quantities B_{1j} and $J(0)$ are determined respectively by Eqs. (23), (25), and (28a) taken at zero temperature. Expression (38) shows that this region at low temperatures the appearance of superconductivity is facilitated when the temperature is raised. At $T_c \gtrsim T_{c1}$ the classical formula (37) is applicable, with J assumed temperature-dependent. The quantity T_{c1} determines the temperature of the transition from the quantum region to the classical region, and is obtained from the relation

$$B_{11}(T_{c1})\sim T_{c1}. \qquad (39)$$

In order of magnitude we have

$$T_{c1}\sim\Delta_0[\ln(\Delta_0 V dN/d\mu)]^{-1}. \qquad (40)$$

The quantity $J_{\min}(0)$, which determines the smallest jump amplitude at which superconductivity is possible, is of the order of T_{c1}. In the considered region $B_{11}(0) \gg T_{c0}$ the value of $J_{\min}(0) \sim T_{c1}$ is much less than that of $J_c(0) \sim B_{11}(0)$. Therefore in the case of $B_{11}(0) \gg T_{c0}$ two phase transitions can take place in a very wide range of J.

All the calculations are valid at sufficiently low jump probabilities, such that the inequality (17) is satisfied. From this inequality we obtain the value $J_{\lim}$, above which the developed theory is not valid:

$$J_{\lim}=(Sp_0^2)\Delta_0, \qquad (41)$$

where S is the junction area.

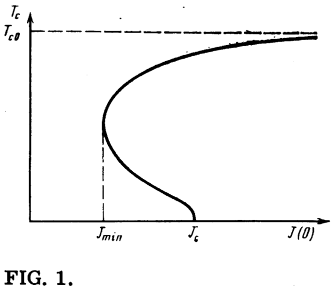

FIG. 1.

We have investigated the phase transitions so far within the framework of the self-consistent field (27). This analysis does not prove the existence of the transition. To check on the statements made, we calculate $\langle\varphi^2\rangle$ in the superconducting region at $T=0$. Simple calculation yields for $J \gg J_c = B_{11}/2$

$$\langle\varphi^2\rangle\sim(J_c/J)^{1/2}. \qquad (42)$$

Expansion at small J yields an expression for the free energy Ω:

$$\Omega=\Omega_0-\frac{J^2}{J_c^2}\left(1+\gamma\frac{J^2}{J_c^2}+\ldots\right), \qquad (43)$$

where γ is a number of the order of unity.

Formulas (43) and (44) shows that the self-consistent-field approximation gives the correct order of magnitude of the critical Josephson energy J_c.

In the superconducting region at $J \gg J_c$, owing to the smallness of the fluctuations of the phase, we obtain the usual formula of the current through the junction:

$$j=-\frac{(2e)^2}{c^2}J_{i,i+1}aA, \qquad (44)$$

where A is the vector potential and c is the speed of light.

In the opposite limiting case $J \ll J_c$ the fluctuations of the phase are strong and the coherence is violated. The properties of the incoherent state will be investigated in the next section.

5. INCOHERENT STATE

We consider the properties of the incoherent state in the limit of small Josephson energies $J \ll J_c$. In this limit, all the averagings can be carried out over the states of the free Hamiltonian $H_{\text{eff}}^{(0)}$ (29). At $B_{11} \ll T_{c0}$ the lowest contribution, in the low-temperature region, is made by the dipole excitations described in the preceding section. Taking the contribution of only one dipole into account, we obtain for the specific heat C

$$C=\frac{mE_1^2}{T^2V}e^{-E_1/T}, \qquad (45)$$

where m is the number of nearest neighbors and E_1 is the energy of the dipole (35a). Only at large values of the Coulomb energy, $B_{11} \gg T_{c0}$, is a substantial contribution made by single-electron excitations above the superconducting gap. Expression (45) shows that a gap exists in the spectrum of the collective excitations in the incoherent state.

To calculate the response to an external electric field, we note that the current j through the junction is equal to the sum of the Josephson current $j^{(1)}$ and the normal current $j^{(2)}$;

$$j=j^{(1)}+j^{(2)}. \qquad (46)$$

The current $j^{(1)}$ in (46) is expressed in terms of the phases of the neighboring grains with the aid of the following expression:

$$j_{1,2}^{(1)}=2eJ_{1,2}\langle\sin(\varphi_2-\varphi_1-2eAa)\rangle. \tag{47}$$

The angle brackets in (47) denote averaging over the states of the effective Hamiltonian in (26), expressed with account taken of the external field A. As before, only phase fluctuations are considered.

The normal current $j^{(2)}$ is expressed in terms of the state densities v_1 and v_2 of neighboring granules[11]:

$$j_{1,2}^{(2)}=\frac{V_{1,2}}{R_{1,2}}\frac{1}{2T}\int_0^\infty\frac{d\varepsilon}{\mathrm{ch}^2(\varepsilon/2T)}\langle\langle\nu_1(\varepsilon)\nu_2(\varepsilon)\rangle\rangle, \tag{48}$$

where $V_{1,2}$ is the difference between the potentials on the barrier

$$\nu_i(\varepsilon)=\begin{cases}\dfrac{\varepsilon}{(\varepsilon^2-\Delta_i^2)^{1/2}}, & \varepsilon>\Delta,\\ 0, & \varepsilon<\Delta.\end{cases} \tag{49}$$

The state densities $v_i(\varepsilon)$ depend only on the modulus of the order parameter and do not depend on the phase. The double angle brackets denote averaging over the fluctuations of the modulus. This averaging is necessary, for otherwise the integral in (48) diverges. Also important is the smearing of the state density on account of the inhomogeneities. The current $j^{(2)}$ varies little at the point of the phase transition described in the preceding section. At low temperatures this current is exponentially small in Δ_0/T, in both the superconducting and in the coherent state. The behavior of the current $j^{(1)}$ is more interesting. Taking into account only the response linear in A and carrying out the corresponding expansions, we reduce expression (47) at $J\ll J_c$ to the form

$$j_\omega^{(1)}=Q_\omega^{(1)}A_\omega, \tag{50}$$

where $Q(\omega)$ is expressed in terms of the retarded Green's function of the currents $K(\omega)$:

$$Q_\omega^{(1)}=2e^2aJ_{1,2}^2(K(\omega)-K(0)). \tag{51}$$

The function $K(\omega)$ is an analytic continuation of the Matsubara Green's function, and after transformations similar to those made in the preceding section, it is expressed in the form

$$K(\omega)=Z^{-1}\sum_{n_1,n_2,\ldots,n_N}\frac{(B_{11}+B_{22}-2B_{12})\exp(-\Sigma_{i,j}B_{ij}n_in_j/2T)}{{}^1/_4(B_{11}+B_{22}-2B_{12})^2-(\Sigma_j(B_{2j}n_j-B_{1j}n_j)+\omega+i\delta)^2}. \tag{52}$$

Expression (52) was written for an arbitrary ratio of B_{11} and B_{22}. This form gets around the difficulty connected with the vanishing of the denominator in (52) at $n_1=0, n_2=\pm1; n_1=\pm1, n_2=0$ and $B_{11}=B_{22}$, $\omega=0$. It can be verified that the terms that diverge as $B_{11}\to B_{22}$ cancel each other. It is therefore convenient to put $B_{11}=B_{22}$ already in the final answer. At zero temperature, a substantial contribution is made to (52) only by the ground state. Substituting (52) in (51) we obtain

$$Q^{(1)}(\omega)=4e^2aJ_{1,2}^2\frac{\omega^2}{\tilde{E}_1(\tilde{E}_1^2-(\omega+i\delta)^2)}, \tag{53}$$

where $\tilde{E}_1=(B_{11}+B_{22}-2B_{12})/2$ is the energy of the transition of a Cooper pair from the first grain to the second.

The form of the response $Q^{(1)}(\omega)$ (53) is typical for insulators. At low frequencies $Q^{(1)}(\omega)$ is proportional to the square of the frequency. Separating the imaginary part of (53), we can easily write down an expression for the conductivity $\sigma^{(1)}(\omega)$:

$$\sigma^{(1)}(\omega)=\frac{2e^2aJ_{1,2}}{\tilde{E}_1}\delta(\omega-\tilde{E}_1). \tag{54}$$

Expressions (53) and (54) show that there exists a resonance corresponding to a transition of a Cooper pair from one grain to a neighboring one. At nonzero temperature, other resonances arise and correspond to higher excitations. A highly peaked resonance is possible only in the case of identical grains packed to form an ideal lattice. Inhomogeneities lead to smearing out of these resonances.

The expression for the conductivity at arbitrary temperature, determined by the imaginary part of (52), can be reduced to a more convenient form. After making simple transformations in (52) we obtain

$$\sigma^{(1)}(\omega)=2e^2aJ_{1,2}^2Z^{-1}\frac{(1-\exp(-\omega/2T))}{\omega}\sum_{n_1\ldots n_N}\delta\Big(E_1-\sum_j(B_{2j}n_j-B_{1j}n_j)-\omega\Big)\exp\Big(-\sum_{i,j}\frac{B_{ij}n_in_j}{2T}\Big). \tag{55}$$

Using (55) and letting the frequency ω go to zero, we obtain a formula for the conductivity at zero frequency:

$$\sigma^{(1)}(0)=\frac{e^2aJ_{1,2}^2}{T}Z^{-1}\sum_{n_1\ldots n_N}\overline{\delta\Big(E_1-\sum_j(B_{2j}n_j-B_{1j}n_j)\Big)}\exp\Big(-\sum_{i,j}\frac{B_{ij}n_in_j}{2T}\Big). \tag{56}$$

The bar over the delta function in (56) denotes averaging over the inhomogeneities. The explicit form of $\sigma^{(1)}(0)$ in (56) is difficult to obtain. Expression (56) shows, however, that the conductivity at zero frequency differs from zero at nonzero temperatures. At temperatures much lower than the characteristic Coulomb energies, the conductivity is exponentially small.

Only kinetic quantities obtained in this section are applicable at $T\ll T_{c0}$. The expression for the conductivity (56) describes the contribution made by the motion of the Cooper pairs. Also important is the contribution of the single-electron current (48), (49). Carrying out the calculations in (48) with allowance for the fluctuations of the modulus of the order parameter we obtain at $T\ll T_{c0}$

$$\sigma^{(2)}(0)=\frac{1}{2R}e^{-\Delta_0/T}\ln\left(\frac{1}{T_{c0}N(0)V}\right). \tag{57}$$

If the inhomogeneities in the system are strong, then the logarithm obtained by calculating the integral in (48) is cut off not by the temperature fluctuations, but by the fluctuations due to the inhomogeneities.

The total conductivity is equal to the sum of $\sigma^{(1)}$ and $\sigma^{(2)}$. Depending on whether the Coulomb energy is larger or smaller than the temperature T_{c0}, the predominant contribution at low temperatures is that of $\sigma^{(2)}$ or $\sigma^{(1)}$, respectively.

6. CONCLUSION

In the preceding sections we have investigated the influence of the fluctuations of the phase on the temperature of the superconducting transition. The analysis was carried out at sufficiently large amplitudes of the jumps of the electrons from grain to grain, such that the inequality (13a) was satisfied. When this inequality is satisfied, the system in the absence of superconducting pairing would be a metal. At zero temperature, there exists a critical value of the Josephson energy J_c, equal in order of magnitude to the Coulomb energy, below which superconductivity is impossible. In the region of low Josephson energies the system is an insulator. The role of the dielectric gap is played by the Coulomb energy or by the superconducting gap, depending on which of these quantities is smaller. If the Coulomb energy is larger than the superconducting gap, the main contribution is made by normal electrons. At nonzero temperatures, superconductivity is possible also at $J<J_c$. This restoration of the superconductivity is due to the screening of the Coulomb interaction at finite temperatures. This screening is produced either by low-lying collective interactions, or by normal electrons. The latter mechanism is more effective if the Coulomb energy is larger than the Cooper gap. Of course, screening by normal electrons is possible only when the condition (13a) is satisfied, when these electrons can flow over freely from grain to grain. The superconducting pairs can in this case be trapped, since the critical value J_c is proportional to the square of the amplitude $|T_{ij}|^2$ and can be both larger and smaller than the Coulomb energy if (13a) is satisfied.

The existence of a critical value J_c was qualitatively predicted in Ref. 8. In the same paper are given estimates of the characteristic parameters of granulated aluminum. Aluminum has a critical temperature $T_{c0}=2$ K, and a gap $\Delta_0=3\times10^{-4}$ eV. The characteristic dimension of the grains is $d=30$ Å, the thickness of the insulator is 5 Å, and the dielectric constant of the insulator between granules is $\varepsilon_0=8.5$. An estimate yields for the Josephson energy $J_{ij}=5.4\times10^{-5}$ eV, and for the Coulomb energy $E_c=1.5\times10^{-2}$ eV. These estimates show that the Coulomb energy is much larger than the superconducting gap. At this ratio of the parameter at finite temperatures, screening by normal electrons is essential. The critical temperature is calculated from formulas (38)–(40). The lowest energy $J_{\min}=mJ_{ij}$ at which superconductivity is possible is of the order of Δ_0. Taking the number of neighbors to be $m=8$, we obtain the estimate $J=mJ_{ij}\approx4.3\cdot10^{-3}$ eV. This is of the same order of or even larger than $J_{\min}$. Therefore superconductivity is possible in this system, as was in fact observed in experiment. It should be noted at the same time that the theory developed is applicable to grains with dimension $d=30$ Å only qualitatively, since the conditions (12) and (13) are not satisfied for such small granules. This leads to strong fluctuations of the modulus of the order parameter.

If the proposed picture is indeed applicable to granulated aluminum, then when the temperature is sufficiently lowered a transition should take place from the superconducting to the insulating state. Measurements at low temperatures would therefore be of interest. However, to the author's knowledge, no such measurements were performed. It is also of interest to investigate the frequency dependence of the conductivity in the incoherent state, and in particular the observation of resonance corresponding to the energies of the transitions of Cooper pairs from one grain to another.

All the results were obtained for a regular lattice of identical grains. The influence of the scatter of the dimensions and positions of the grains on the properties of the system is the subject of further study.

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