Elementary excitations in disordered systems with localized electrons

S. D. Baranovskiǐ, B. I. Shklovskiǐ, and A. L. Éfros

A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences
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The elementary excitations of a disordered system with localized electronic states is classified by their electric multipole moments. The self-consistent-equation method is used to investigate the energy spectra of charged and dipole excitations for a lattice model. It is shown that because of the interaction of the dipole excitations with one another their state density decreases logarithmically with decreasing energy. This singularity should manifest itself in the low-temperature specific heat and in the high-frequency conductivity. The electronic specific heat of a doped semiconductor is considered. The interaction between charged excitations and dipole excitations (the polaron effect) is investigated, and the exponential decrease of the state density of the charged excitations at very low energies, due to this interaction, is investigated.

The systems dealt with here include amorphous semiconductors as well as doped crystalline semiconductors in which the Fermi level lies in the region of localized states. Strict localization of the electronic states, as shown by Anderson, is a consequence of the scatter of their energies, and the existence of an end-point energy (Fermi level), which separates the empty and filled states, follows in this case not from the Pauli principle but from the condition that the total energy be a minimum with respect to electron transfer from one site to another.

Many papers published in the last decade point to the need for taking into account the electron-electron interaction in such systems. In a number of papers[1,2] an attempt was made to construct a theory "in the image and likeness" of the Fermi-liquid theory, thereby predetermining the end result—a relatively weak renormalization of the state density in the vicinity of the Fermi level. This approach seems inadequate to us. The point is that the Fermi-liquid theory itself is applicable to systems with Coulomb interactions only at sufficiently high density, when the kinetic energy of the electrons is of the order of the energy of the Coulomb interaction or higher. In the system considered here there is no kinetic energy at all. The system is therefore much closer in its properties to a low-density electron gas which, as is well known, constitutes at zero temperature a Wigner crystal, i.e., a state having nothing in common with a Fermi liquid.

Pollak and Knotek[3] have emphasized that because of the electron-electron interaction the state density (SD) should have a dip that separates the empty and filled states. Kurosawa and Sugimoto[4] observed this in a computer experiment. Srinivasan considered a model with a large energy scatter of non-Coulomb character and showed that the Coulomb interaction does not make it possible to use the single-electron approximation in the vicinity of the Fermi level.

Studies by our group[6-10] have shown that, owing to the long-range part of the electron-electron interaction, the SD vanishes on the Fermi level.[1)] This phenomenon, which is genetically connected with Wigner crystalization, was named Coulomb gap. The disorder in the system causes this gap not to be "hard." The system has "soft" (low-energy) elementary excitations that are responsible for the electric and thermal properties of the system, and the Coulomb interaction affects them strongly.

In the present article we propose a general classification of the elementary excitations in the system in question, and consider, for the first time ever, the properties of the dipole excitations responsible for the high-frequency conductivity and specific heat. The analysis is carried out with the aid of a self-consistent equation.

1. CLASSIFICATION OF ELEMENTARY EXCITATIONS. COMPACTNESS PRINCIPLE

The proposed classification is essentially independent of the disordered-system model, but it is nonetheless convenient to describe it with the aid of a concrete example, which we choose to be the lattice model proposed by one of us.[7] In this model, the electrons can occupy only the sites of a periodic lattice, the number of electrons is half the number of the sites, and the charge of a site is half the charge of the electron. Each site is assigned a random energy ϕ_i, uniformly distributed in the interval from $-A$ to A. A site can contain either one electron or none. We have investigated the properties of the ground state, in which the electrons are disposed over the sites in such a way that the total energy of the system

$$H = \sum_i \phi_i n_i + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{r_{ij}} n_i n_j, \tag{1.1}$$

which includes the Coulomb interaction of the charged sites, is minimal. Here n_i are the occupation numbers, equal to $\frac{1}{2}$ for occupied sites and $-\frac{1}{2}$ for empty ones. The single-site energy

$$\varepsilon_i = \phi_i + \sum_j \frac{e^2}{r_{ij}} n_j \tag{1.2}$$

for an occupied site is defined as the work (taken with a minus sign) that must be performed to move an electron from the site to infinity, where the potential is assumed equal to zero. For an empty site this energy is equal to the work necessary to bring an electron from infinity to the empty site. From the charge symmetry of the system it follows (see Ref. 7) that the Fermi level is equal to zero, i.e., in the ground state $n_i = \frac{1}{2}$ at $\varepsilon_i < 0$ and $n_i = -\frac{1}{2}$ at $\varepsilon_i > 0$.

We are interested in the case $\gamma \equiv e^2/a_0 A \ll 1$, where a_0 is the lattice constant. In Ref. 7 (see also Refs. 5 and 11) it was shown that perturbation theory in the Coulomb interaction is not applicable if the energy ε is less than the width $\Delta \equiv A\gamma^{3/2}$ of the Coulomb gap. Moreover, at $\gamma \ll 1$ one can obtain at low energies a universal description of the system (independent of A, a_0, and apparently of the considered model), if Δ is chosen to be the energy unit and $r_0 \equiv e^2/\Delta$ is chosen to be the unit of length.

We proceed now to a classification of the elementary excitations. All of them are transitions of electrons between different sites, i.e., they are described by a change in an even number of occupation numbers relative to the occupation numbers of the ground state. To calculate the probabilities of the transitions it is necessary to know the extent that the wave functions overlap and to identify the interaction that causes the transitions (usually, phonons). We, on the other hand, are interested in the excitation spectrum, a problem that must be solved by using for the total energy the classical expression (1.1), in which it is only implied that the quantum overlap of neighboring states is negligible.

The proposed classification is based on the charge state of the elementary excitation. The excitations are divided into charged, dipole, quadrupole, etc. The simplest positively charged excitation is produced on a site i by removing from it an electron and transferring the electron to another site j, located at a distance much larger than r_0 away from the site i. This produces on the site j a negatively charged excitation. We shall name this a single-site excitation. A more complicated positively charged excitation is produced if we not only remove an electron from the site i, but also permute other electrons with initial and final states located in the immediate vicinity of the site i. We call such excitations polarons. (The concept of electronic polarons was introduced independently in Refs. 7 and 12.) From the charge-conservation law it follows that charged excitations can be produced only in pairs.

The simplest example of dipole excitation is the transfer of an electron from an occupied site to an empty site located at a distance on the order of r_0. We call such excitations paired. A more complicated dipole excitation is produced if several permutations of electrons are carried out in a small region of space (of the order of r_0^3).

In a disordered system with randomly located sites, the excitations can be only dipole or charged. The

probability of formation of quadrupole etc. excitations is zero. In the lattice model, however, all these excitations exist. Nonetheless, we shall not consider them in detail. They interact weakly with one another and with electrically active (charged and dipole) excitations, so that they can be disregarded in the analysis that follows. However, as we shall show, the interactions weakens greatly the state density of electrically active interactions with low energy, as a result of which the quadrupole interactions (in those systems in which they exist) make the predominant contribution to the specific heat at sufficiently low temperatures.

Two charged excitations of opposite sign always form a dipole and the proposed classification is meaningless if we do not explain why the length r_0 is the line of demarcation between the charged and dipole excitations. The classification is supplemented by the compactness principle. It states that if the displacement of the dipole $r > r_0$, then the probability of the onset of a dipole excitation decreases sharply with increasing r. We consider paired interactions and introduce the function

$$\mathscr{F}(\omega, \mathbf{r}_1, \mathbf{r}_2) \equiv \mathscr{F}(\omega, r) \quad (r = |\mathbf{r}_1 - \mathbf{r}_2|)$$

such that $\mathscr{F}(\omega, r)d\omega d\mathbf{r}_1 d\mathbf{r}_2$ is the probability that there is an occupied site in the interval $d\mathbf{r}_1$ and an empty site in the interval $d\mathbf{r}_2$, and the energy necessary for the transition of an electron between the sites lies in the interval ω, $\omega + d\omega$. Mathematically, the compactness principle is expressed by the fact that the integral

$$\Phi(\omega) = \int \mathscr{F}(\omega, r)d\mathbf{r} \tag{1.3}$$

has asymptotic convergence on the upper limit at small values of ω (at distances on the order of r_0), so that it is possible to introduce the concept of the density $\Phi(\omega)$ of paired excitations per unit volume.

We note that this statement is quite nontrivial and is valid exclusively because of the presence of Coulomb interaction. In fact, if we set the electron charge e in (1.1) equal to zero, then $\mathscr{F}(\omega, r) \equiv \omega/4A^2 a_0^6$, i.e., it is independent of r. This makes the pair density $\Phi(\omega)$ proportional to the volume, which is absurd.

The compactness principle is arrived at from the concept of the Coulomb gap. It was shown in Ref. 6 that because of the interaction of the charged excitations with one another their state density decreases when the Fermi level is approached at a rate not less than ε^2 in the three-dimensional case and not less than $|\varepsilon|$ in the two-dimensional case. It is this which leads to the compactness principle. With increasing dipole arm the excitation is gradually transformed from dipole to charged. But the density of the charged excitations has a Coulomb gap at low energies. Therefore when the dipole arm is increased the probability of excitation formation decreases sharply.

We examine now in greater detail the case of paired excitations. The work necessary to transfer an electron from an occupied site 2 to an empty site 1 is[6]

$$\omega = \varepsilon_1 - \varepsilon_2 - e^2/r_{12}, \tag{1.4}$$

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where r_{12} is the distance between the sites, and ε_1 and ε_2 are the site energies defined above. In first-order approximation we can represent the function $\mathscr{F}(\omega, r)$ in the form

$$\mathscr{F}^0(\omega, r) = \int_0^\infty d\varepsilon_1 \int_{-\infty}^0 d\varepsilon_2\, g(\varepsilon_1) g(\varepsilon_2) \delta(\varepsilon_1 - \varepsilon_2 - e^2/r - \omega), \quad (1.5)$$

where $g(\varepsilon)$ is the state density of the single-site excitations. Since $g(\varepsilon)$ vanishes as $\varepsilon \to 0$, the main contribution to the integral at $r \approx r_0$ and $\omega = 0$ is made by pairs with $\varepsilon_1 \approx \Delta$ and $|\varepsilon_2| \approx \Delta$, in which a low excitation energy is achieved by exact cancellation of the positive term $\varepsilon_1 - \varepsilon_2$ by the negative term $-e^2/r$. With increasing r, the characteristic values of ε_1 and $|\varepsilon_2|$ decrease so that the Coulomb gap results in a sharp decrease of $\mathscr{F}^0(\omega, r)$. This in fact is the manifestation of the compactness principle, wherein, as will be shown below, the distribution function of any dipole excitation $\mathscr{F}(\omega, \mathscr{P})$ consisting of several electron permutations also decreases sharply when the dipole moment $\mathscr{P}$ exceeds er_0.

We note that at $r > e^2/\omega$ the function $\mathscr{F}^0(\omega, r)$ ceases to depend on r. Therefore the integral (1.3) converges only asymptotically. The main contribution to it is made by pairs with a displacement on the order of r_0. Thus, the compactness principle and the entire proposed classification are meaningful only for excitations with low energy ($e^2/\omega \gg r_0$ or $\omega \ll \Delta$). We note also that in the approximation (1.5) the function $\Phi(\omega)$ obtained with the aid of (1.3) has a nonzero limit as $\omega \to 0$, and in order of magnitude we have $\Phi(0) \approx g_0$.

Thus, the Coulomb gap makes it possible to separate the branch of dipole excitations. It is precisely these excitations which determine the thermodynamics of the system, and also the reaction to an alternating electric field. However, as already stated, the static conductivity is determined by the charged excitations. In fact, pairs with low energy ω are compact formations located at large distances from one another ($R \propto \omega^{-1/3}$). For the static conductivity we need a "pass-through" system of paths from contact to contact, which cannot be organized by compact pairs. The length of the hop in low-temperature static conduction must inevitably be large, and the transition energy must be small. It follows therefore, according to (1.4), that the cancellation of the positive and negative terms, which is inherent in a typical pair, cannot take place in this case and the energies ε_1 and $|\varepsilon_2|$ must be small enough.

Dipole excitations make an important contribution to the dielectric properties of the system. In a weak electric field E, the "soft" pairs become polarized and produce a polarization $P \approx er_0 \Phi(0) eEr_0$. Assuming that $\Phi(0) \approx g_0$, we find that the corresponding contribution to the dielectric constant is of the order of $g_0 e^2 r_0^2 = g_0 e^6/\Delta^2 \approx 1$. It will be shown below that the interaction of the pairs with one another leads to $\Phi(\omega) \sim g_0 [\ln(\Delta/\omega)]^{-1/2}$ as $\omega \to 0$, thus decreasing somewhat their contribution to the dielectric constant.

The dielectric-constant contribution produced by the dipole excitations influences substantially the state densities of the charged excitations. If an electron is added to an empty site, then it turns out that in the

presence of this electron a large number of pairs located in the vicinity of this site find it convenient to move the electron from one site to another (see Fig. 1). The resultant dipole potential lowers the energy of the new electron. In ionic crystals this phenomenon is called the polaron shift, and we borrow this terminology but bear in mind that the shift is produced not by the lattice but by the electronic transitions. If the configurations and energies of the pairs in the vicinity of all the sites were the same, then the filled states would be separated from the empty ones by a rigid polaron gap that contains no states whatever. However, since the system in question is disordered, there is a probability that the polaron shift is small near certain sites, i.e., the soft pairs that produced polarization are accidentally absent. The average number of pairs participating in the production of the polaron atmosphere is large, so that the probability of such a fluctuation is exponentially small. It follows therefore[7] that the state density $g(\varepsilon)$ of single-site excitations vanishes exponentially as $\varepsilon \to 0$ (see Fig. 2). Inasmuch as, accurate to logarithmic factors, the renormalization of the dielectric constant is of the order of unity, the width of the polaron gap is a priori of the order of the width Δ of the Coulomb gap. The form of $g(\varepsilon)$ was likewise calculated in Ref. 7. The unknown quantity there was the fluctuation probability whereby the vicinty of the occupied site with energy ε does not contain even a single pair such that the system energy decreases as a result of a process consisting of excitation of this pair and simultaneous transfer of the electron from the considered site to a remote hypothetical site having an energy equal to the Fermi energy. As shown in Ref. 7, this probability is proportional to $\exp\{-\beta(\Delta/\varepsilon)^{1/2}\}$, where β is a numerical factor.

In the present paper we prove that the polarization gap has a more abrupt form, namely,

$$g(\varepsilon) \sim \exp\left\{ -\gamma \frac{\Delta}{\varepsilon[\ln(\Delta/\varepsilon)]^{1/4}} \right\}, \quad (1.6)$$

where γ is a numerical factor. The right-hand side of (1.6) is the probability of a fluctuation such that in the vicinity of a site with energy ε there are no pairs that create conditions facilitating the transfer of an electron from this site to a remote site having an energy equal to the Fermi energy, with simultaneous excitation of all the pairs that become polarized in the field of a point charge located on the site in question.

It must be noted that the hard gap (1.6) becomes soft if a transition is made from single-site excitations to polarons.[7] The energy of a polaron on an empty site is defined as the minimum work necessary to transport
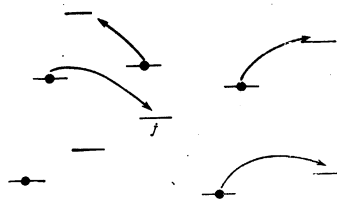

FIG. 1. Formation of "polaron atmosphere" when an electron is placed on site j.
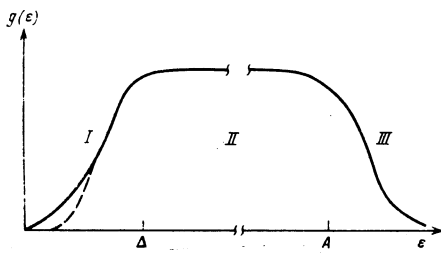
FIG. 2. State density of single-site excitations without allowance for the polaron interaction (solid line) and with the allowance (dashed) in the lattice model at $A \gg e^2/a_0$. The Fermi level is equal to zero. I—Region of Coulomb gap, II—region of very weak change of the state density, where the Coulomb effects play a small role, III—high-energy region, where the state density is again determined by the Coulomb interaction. Attention: note the break on the abscissa axis!

the electron from infinity to this site and simultaneously produce the pair polarization to accommodate the new electron. Analogously, the energy Ψ_i of a polaron corresponding to an occupied site can be defined as the minimum work (with minus sign) which must be performed to transport an electron from site i to infinity, with simultaneous relaxation of the polarization of the pairs in the vicinity of this site. It follows from the definition that $|\Psi_i| \le |\varepsilon_i|$. Repeating the arguments of Ref. 6, one of us[7] found that the state density of the polarons $G(\Psi)$ decreases as a result of their interaction with one another like

$$G(\Psi) = \alpha_3 \Psi^2 / e^6, \tag{1.7}$$

where α_3 is a numerical coefficient. Since the interaction with polaron excitations is included in the polaron shell, this interaction does not lead to further decrease of the state density.

Thus, the density of the charged excitations decreases with energy like the square of energy, and at sufficiently low energies these excitations should be for the most part polarons, while the contribution of the single-site excitations decreases with decreasing energy exponentially. The charged excitations are responsible, for example, for the hopping charge transfer, and relation (1.7) leads to $\sigma \propto \exp[-(T_0/T)^{1/2}]$ (σ is the electric conductance and T_0 is the characteristic temperature), which differs from the known Mott law.

The classification described above is in essence only a more general and more succinct formulation of the ideas that were formulated already by 1976. In that year there was developed the self-consistent equation (SCE) method, which made it possible to take into account the interaction of charged excitations.[7] This was followed by a numerical computer simulation of the system in question,[8,9] as well as of a system with randomly disposed sites[10], and some unexpected results were obtained. Only the SD of single-site excitations were investigated, and in the two-dimensional case the result confirmed splendidly the existing theory: at small ε we have the linear law

$$g(\varepsilon) = \alpha_2 |\varepsilon| / e^4 \tag{1.8}$$

with a coefficient α_2 that follows from the SCE. With

increasing A, the SD tends to a universal function of ε/Δ that also agrees well with the SCE solution. The unexpected fact was that a similar picture was observed also in the three-dimensional case. At small ε the following law holds

$$g(\varepsilon) = \alpha_3 \varepsilon^2 / e^6, \tag{1.9}$$

wherein the constant α_3 and the entire energy dependence of the SD are well described by an SCE that takes into account only the interaction of the charged excitations. No signs of interaction between charged and dipole excitations, which should lead to a polaron gap and to the exponential decrease (1.6), were observed. To be sure, the boundary effects that increased with decreasing energy made it possible to investigate only the region $\varepsilon \gtrsim 0.15\Delta$, but in this region the SD decreased by approximately a factor of 20 and in essence it is precisely this region which is of greatest interest for modern experiments.

Thus, numerical simulation has shown that for some reason the hard polaron gap is much narrower than the Coulomb gap, although *a priori* they should be of the same order. The desire to explain this circumstance stimulated in fact the present study. Its purpose is to investigate the interaction between dipole excitations which, as is clear beforehand, should make these excitations harder and weaken thereby the polaron gap. In the present article we give only the asymptotic results, which are valid in the limit of very low energies. They state that because of the interaction the DS of the dipole excitations has a logarithmic singularity that should manifest itself in the specific heat and in the dielectric properties of the system. It leads also to a logarithmic factor in (1.6), which weakens the polaron gap. It is so far impossible to say whether this is sufficient to explain the results of the simulation, since expression (1.6) is valid only in the limit of very small ε. More detailed calculations are needed to investigate the region $\varepsilon > 0.1\Delta$.

2. THE SCE METHOD AND INTERACTION OF ISING DIPOLES

The SCE for the DS was first proposed in Ref. 7. Its derivation calls for a number of assumptions which have not yet been substantiated. This method, however, is the simplest mathematical expression of the ideas described in the preceding section and, in addition, in all the hitherto considered cases it yields very good quantitative agreement with the results of computer simulation.[8-10] We present a detailed derivation of the SCE, for the purpose of discussing some its aspects to which no attention was paid before.

The SCE can be written in the case $A \gg 1$ and is based on an expression obtained in Ref. 7 for the distribution function $\rho(\varepsilon_1, \ldots, \varepsilon_N)$:

$$\rho = (2A)^{-N} \prod_{\substack{\varepsilon_i < 0 \\ \varepsilon_j > 0}} \theta(\Delta_i^j) \cdots \prod_{\substack{\varepsilon_{i_1} \cdots \varepsilon_{i_p} < 0 \\ \varepsilon_{j_1} \cdots \varepsilon_{j_p} > 0}} \theta(\Delta_{i_1 \cdots i_p}^{j_1 \cdots j_p}) \cdots, \tag{2.1}$$

where $\theta(x) = 1$ at $x > 0$ and $\theta(x) = 0$ at $x < 0$; $\Delta_{\cdots}^{\cdots}$ is the work that must be performed to transfer p electrons from the sites $i_1 \ldots i_p$ that are occupied in the ground

state to the empty sites $j_1 \ldots j_p$:

$$\Delta^{j_1 \ldots j_p}_{i_1 \ldots i_p} = \sum_{k=1}^{p} \varepsilon_{j_k} - \sum_{k=1}^{p} \varepsilon_{i_k} - E^{j_1 \ldots j_p}_{i_1 \ldots i_p}; \tag{2.2}$$

$E^{\ldots}_{\ldots}$ is the electrostatic interaction energy of a system consisting of p positive charges at the points $i_1 \ldots i_p$ and p negative charges at the points $j_1 \ldots j_p$. The inequality corresponding to the transfer of a single charge is of the form

$$\Delta_i^j = \varepsilon_j - \varepsilon_i - e^2/r_{ij} > 0. \tag{2.3}$$

We shall give below a more complete derivation of (2.1) than in Ref. 7.

To each set $\{\phi_i\}$ there corresponds a set of occupation numbers, which describes the ground state, and a set of single-site energies $\{\varepsilon_i\}$. The function (2.1) describes the distribution of ε_i averaged over all the sets $\{\phi_i\}$. The product of the θ functions reflects the fact that in the ground state the transfer of any number of electrons from site to site increases the total energy of the system. The sets $\{\varepsilon_i\}$ which do not satisfy these conditions are forbidden. To derive (2.1) it suffices to prove that in N-dimensional energy space the probability density of all the allowed sets $\{\varepsilon_i\}$ is the same and is equal to $(2A)^{-N}$. We consider the allowed N-dimensional interval from $\{\varepsilon_i\}$ to $\{\varepsilon_i + \delta\varepsilon_i\}$, and find the probability of observing the system within this interval. It must be assumed here that not one of the energies ε_i reverses sign within the interval, since the function ρ is not defined at the points $\varepsilon_i = 0$, where the occupation numbers n_i change jumpwise.[2] The considered interval of sets $\{\varepsilon_i\}$ corresponds to an interval of sets of $\{\phi_i\}$ from $\{\phi\}$ to $\{\phi_i + \delta\phi_i\}$. Therefore the sought probability is equal to the probability of finding the set $\{\phi_i\}$ in this interval. By definition, the latter probability is equal to

$$W = (2A)^{-N} \prod_i \delta\phi_i.$$

Since the occupation numbers do not change their values, we have according to (1.2) $\delta\phi_i = \delta\varepsilon_i$ and

$$W = (2A)^{-N} \prod_i \delta\varepsilon_i,$$

q.e.d.

The limitations described by the θ functions play a role in the low-energy region and are responsible for the Coulomb gap. At high energies, the values of ε_i are bounded by the additional conditions

$$|\phi_i| = \left| \varepsilon_i - \sum_j n_j e^2/r_{ij} \right| \leqslant A.$$

However, the characteristic values of the electrostatic potential are small compared with A. Thus, between the region of the Coulomb gap ($\varepsilon \leqslant \Delta$), where the values of ε_i are strongly correlated, and the region where the additional conditions are important ($\varepsilon_i \approx A$), there lies a broad region in which the probability of finding energy in the interval $d\varepsilon_i$ depends little on the energies of the surrounding sites and equals, with good accuracy, $d\varepsilon_i/2A$. Therefore the state density $g(\varepsilon)$ in the region $\Delta \ll \varepsilon \ll A$ is close to $g_0 = (2Aa_0^3)^{-1}$ (see Fig. 2, as well

as Fig. 7 of Ref. 9).

When we speak of interaction of single-site excitations, we bear in mind that only $\theta(\Delta_i^j)$ is taken into account in (2.1), i.e., the only conditions considered are those that require a minimum of the total energy with respect to all possible permutations of one electron. We now derive the SCE corresponding to this approximation. By definition we have

$$g(\varepsilon) = \frac{1}{a_0^3} \int \rho(\varepsilon_1 \ldots \varepsilon_i \ldots \varepsilon_N) \delta(\varepsilon_i - \varepsilon) \prod_j d\varepsilon_j = g_0 W_\varepsilon, \tag{2.4}$$

where W_ε can be interpreted as the probability that the energies of all the neighbors of the site i assume values such that the conditions (2.3) are satisfied. The idea of the derivation of the SCE is that W_ε is represented in the form of a product of probabilities, each of which is expressed in terms of the sought function $g(\varepsilon)$. Assume, for the sake of argument, that $\varepsilon > 0$ and let the origin be at the site i. A certain site j, located at a distance r_j from the site i and having a negative energy ε_j, must satisfy the condition $\varepsilon - \varepsilon_j - e^2/r_j > 0$. The probability of this event (considered independently of the energies of the other sites) is

$$1 - a_0^3 \int_{\varepsilon - e^2/r_j}^{0} g(\varepsilon') d\varepsilon' \quad \text{and} \quad r_j < e^2/\varepsilon,$$

and is equal to unity at $r_j > e^2/\varepsilon$.

The approximation used in Ref. 7 was

$$W_\varepsilon = \prod_{r_j < e^2/\varepsilon} \left(1 - a_0^3 \int_{\varepsilon - e^2/r_j}^{0} g(\varepsilon') d\varepsilon' \right), \tag{2.5}$$

where the product was taken over all the sites. Subsequently,[8] however, this was found to be incorrect. In fact, in this calculation it is necessary to take into account only the probabilities of independent events. Assume that a situation has been reached wherein each site satisfies inequalities that connect it with sites located only to one side of the site. This is sufficient to interrelate the energies of all the sites of the system by the required inequalities. It follows therefore that expression (2.5) takes each inequality into account twice and it suffices to include in the products only the terms with $j > i$.[3]

Substituting (2.5) in (2.4), taking the logarithm, and recognizing that the integral in (2.5) is small compared with unity, we obtain

$$\ln[g(\varepsilon)/g_0] = - \sum_{\substack{j > i \\ r_j < e^2/\varepsilon}} a_0^3 \int_{\varepsilon - e^2/r_j}^{0} g(\varepsilon') d\varepsilon'. \tag{2.6}$$

The expression in the right-hand side represents (with a minus sign) the average number of sites for which the inequalities that relate them to the site i are not satisfied. Therefore the right-hand side, in accordance with the Poisson distribution, represents the logarithm of the probability of the total absence of such sites. At low energies, an important role is played by large distances. We can therefore neglect the lattice structure and replace the summation by integration. After integrating with respect to r_j we get

$$g(\varepsilon) = g_0 \exp\left\{ -\frac{e^{2d} v_d}{2} \int_0^\varepsilon \frac{g(\varepsilon') d\varepsilon'}{(\varepsilon + \varepsilon')^d} \right\}, \tag{2.7}$$

where v_d is the volume of a sphere of unit radius in d-dimensional space, and $d = 2$ or 3 is the number of dimensions of the space. This equation was uased in Refs. 8 and 9, but for the reasons indicated above it differs from the equation proposed in Ref. 7 by a factor of 2 in the argument of the exponential. At low energies we obtain the laws (1.8) and (1.9), with $\alpha_2 = 2/\pi$ and $\alpha_3 = 3/\pi$. As shown in Refs. 9 and 10, these values agree well with the results of computer experiments. According to (2.7), the state density $g(\varepsilon) \to g_0$ as $\varepsilon/\Delta \to \infty$. We see therefore that the SCE is valid only under the condition $\gamma = e^2/a_0 A \ll 1$. At low energies, however, the solution does not depend on A at all. A hypothesis was advanced in Ref. 6 that the state density of the charged states is universal at low energies. It was shown in Refs. 9 and 10 that, within the limits of the experimental computer accuracy, the state density at low energies is independent not only of A, but also of the model of the disordered system. All this suggests that at low energies the solution of the SCE (2.7) is valid not only at $\gamma \ll 1$ and not only for the considered lattice model.

It is of interest further to generalize the SCE to the case when the interaction in (1.1) is given not by e^2/r_{ij} but by $l^{n-1} e^2/r_{ij}^n$, where $n \geq 1$. It is easy to verify that the argument of the exponential in (2.7) is of the form

$$\frac{v_d}{2} (e^2 l^{n-1})^{d/n} \int_0^A \frac{g(\varepsilon') d\varepsilon'}{(\varepsilon + \varepsilon')^{d/n}}.$$

The value $d/n = 1$ is in this case critical. If the potential decreases just barely more rapidly than r^{-d}, then the SD does not vanish. For example, in an MIS (metal-in-sulator-semiconductor) structure the interaction between electrons localized on the surface of the semiconductor takes the form e^2/r_{ij} if $r_{ij} \ll l$, where l is the distance from the surface of the semiconductor to the surface of the metal, and the form $e^2 l^2/r_{ij}^3$ at $r_{ij} \gg l$. (The image forces transform the charges into dipoles.) In the latter case $d = 2$, the ratio is $d/n = \frac{2}{3} < 1$, and at $\varepsilon < e^2/l$ the state density stops decreasing with ε and $g(0) \neq 0$.

In this article we study the interaction of dipole excitations in three-dimensional space, and this corresponds to the critical situation $d/n = 1$. In the model (1.1) the dipoles have different arms and different orientations in space, therefore the SCE formalism must be modified. We shall do this in the next section and consider for the time being the model of identical Ising dipoles on a lattice, whose Hamiltonian is obtained from (1.1) by replacing e^2/r_{ij} by $e^2 l^2/r_{ij}^3$, where l is the displacement of the dipole. In this case we have in place of (2.7)

$$g(\varepsilon) = g_0 \exp\left\{ -\frac{2\pi}{3} e^2 l^2 \int_0^A \frac{g(\varepsilon') d\varepsilon'}{(\varepsilon + \varepsilon')} \right\}. \tag{2.8}$$

At small ε the solution takes the form

$$g(\varepsilon) = \frac{3}{2\pi e^2 l^2} \frac{1}{\ln(tA/\varepsilon)}, \tag{2.9}$$

where t is a numerical factor. At high energies, g tends to g_0. Defining the gap width Δ by the condition $g(\Delta) = g_0$, we get

$$\Delta = tA \exp\{-3/2\pi e^2 l^2 g_0\}.$$

Thus, in the critical case $g(\varepsilon)$ goes to zero logarithmically, and the width of the gap, just as in the BCS theory, depends exponentially on the coupling constant.

A similar model was investigated by Kirkpatrick and Varma.[14] This model is obtained from our model by putting $A = 0$ and assuming that the interaction takes the form $b_{ij} e^2 l^2 / r_{ij}^3$, where b_{ij} is a random quantity that assumes with equal probability the value $+1$ or -1. It is this quantity which produces a level scatter of the order of $\tilde{A} = e^2 l^2 / a_0^3$. It seems to us, that, just as in the case of a Coulomb gap, the solution (2.9) should be independent of the model as $\varepsilon \to 0$. It is only necessary to recognize that because of the random sign of b_{ij} it is necessary to introduce into the argument of the exponential in (2.8) the factor $\frac{1}{2}$ (at fixed i we have $b_{ij} > 0$ for only half of the sites j) and replace A by $\tilde{A}$. We then obtain

$$g(\varepsilon) = \frac{3}{\pi e^2 l^2} \frac{1}{\ln(t\tilde{A}/\varepsilon)}. \tag{2.10}$$

Kirkpatrick and Varma[14] advance arguments that in their model $g(0) = 0$; these arguments are very close to the derivation of the Coulomb gap, given in Ref. 6. They have also reported numerical simulation by the Monte Carlo method, which confirms the vanishing of $g(0)$, but claim that the obtained function $g(\varepsilon)$ is $\sqrt{\varepsilon}$. It seems to us that the accuracy of their numerical experiment does not make it possible to distinguish between $\sqrt{\varepsilon}$ and expression (2.10).

3. INTERACTION OF DIPOLE EXCITATIONS

We begin with paired excitations that represent the transfer of one electron over a distance r with an expenditure of energy ω: the SCE in this case must be written for the function $\mathscr{F}(\omega, r)$ introduced in Sec. 1. The interaction is described by the condition that the energy be a minimum with respect to simultaneous permutation of two electrons. As will be shown, the important role is played here by interaction over large distances between compact pairs, whose potential can be regarded as a dipole potential. We consider a pair with excitation energy ω and displacement $\mathbf{r}$, located at the origin. The parameters ω' and $\mathbf{r}'$ of any pair located at a distance $\mathbf{s}$ from the considered pair should satisfy the inequality

$$\omega + \omega' - U > 0, \tag{3.1}$$

where U is the energy of the dipole-dipole interaction

$$-U = \frac{e^2 r r'}{s^3} f(\vartheta_1, \vartheta_2, \vartheta_3), \quad f = \sin\vartheta_1 \sin\vartheta_2 \cos\vartheta_3 - 2\cos\vartheta_1 \cos\vartheta_2. \tag{3.2}$$

Here ϑ_1 is the angle between $\mathbf{r}$ and $\mathbf{s}$, ϑ_2 is the angle between $\mathbf{r}'$ and $\mathbf{s}$, and ϑ_3 is the angle between the planes $(\mathbf{r}, \mathbf{s})$ and $(\mathbf{r}', \mathbf{s})$. The condition (3.1) follows from the fact that the increment of the total energy of the system due to simultaneous excitation of two pairs is positive.

The average number of pairs whose parameters ω' and $\mathbf{r}'$ and do not satisfy the inequalities (3.1) that connect them with the considered pair can be expressed by the formula

$$G(\omega, r) = \int \mathscr{F}(\omega', r') \theta(U - \omega - \omega') d\omega' dr' ds, \tag{3.3}$$

and the probability that not a single pair which does not satisfy the inequalities (3.1) is located in the vicintiy of the considered pair is equal to $\exp\{-G(\omega, r)\}$. Let $\mathscr{F}^{-1}(\omega, r)$ be the function obtained by substituting in (1.5) the state density $g(\varepsilon)$ which is the solution of the SCE (2.7). It is easily seen that the $\mathscr{F}^{-1}(\omega, r)$ obtained in this manner depends only on the argument $x = \omega + e^2/r$. In the spirit of the SCE, it must be assumed that if one takes into account only the interaction of single-site excitations, [i.e., if only $\theta(\Delta_i^j)$ are retained in (2.1)], then the distribution function $\mathscr{F}(\omega, r)$ should coincide with $\mathscr{F}^1(\omega, r)$. Then the SCE for the interacting pairs must be written in the form

$$\mathscr{F}(\omega, r) = \mathscr{F}^1(\omega, r)\exp\{-G(\omega, r)\}. \tag{3.4}$$

Just as in the derivation of (2.7), in order not to take into account each inequality twice, the integration with respect to s in (3.3) should be carried out only over half the space. Integrating with respect to s, we obtain

$$G(\omega, r) = \alpha r e^2 \iint \frac{\mathscr{F}(\omega', r')}{\omega + \omega'} r'^3 \, dr' \, d\omega', \tag{3.5}$$

where

$$\alpha = -\frac{\pi}{3}\int f(\vartheta_1, \vartheta_2, \vartheta_3)\, d\cos\vartheta_1 \, d\cos\vartheta_2 \, d\vartheta_3, \tag{3.6}$$

and the integration limits are determined by the inequalities

$$0 < \vartheta_1 < \pi, \quad 0 < \vartheta_2 < \pi, \quad 0 < \vartheta_3 < 2\pi, \quad f(\vartheta_1, \vartheta_2, \vartheta_3) < 0.$$

The integral (3.6) was calculated with a computer by the Monte Carlo method. We obtained $\alpha = 9.1$. Defining the function $\varphi(\omega, r)$ by the equation

$$\mathscr{F}(\omega, r) = \mathscr{F}^1(\omega, r)\exp\{-rr_0^{-1}\varphi(\omega, r)\}, \tag{3.7}$$

we find that it must satisfy the equation

$$\varphi(\omega, r) = \alpha r_0 e^2 \iint \frac{\mathscr{F}^1(\omega', r')}{\omega + \omega'} \exp\left\{-\frac{r'}{r_0}\varphi(\omega', r')\right\} r'^3 \, d\omega' \, dr'. \tag{3.8}$$

At $\omega \gg \Delta$ the Coulomb interaction is insignificant, and the function $\mathscr{F}(\omega, r)$ should coincide with $\mathscr{F}^1(\omega, r)$, while the function $\varphi(\omega, r)$ should tend to zero. As a result, the integral (3.8) diverges with respect to ω at the upper limit. This divergence is due to the fact that values of s of the order of r and smaller become significant and the dipole-dipole approximation becomes meaningless.

As seen from (3.1)–(3.3), at small ω we have $s \approx (rr'e^2/\omega')^{1/3}$. Because of the exponential in (3.8), the significant values are $r' \approx r_0/\varphi$. As we shall see later, $\varphi \approx |\ln(\Delta/\omega)|^{1/4}$, and we can assume $r' \approx r_0$ with logarithmic accuracy. Then s becomes comparable with r at $\omega' \approx e^2 r_0/r^2$.

We are interested in values $r \approx r_0/\varphi$, so that accurate to logarithmic and numerical factors we can set the upper limit of the integration equal to Δ. At large values of ω' the dipole-dipole approximation is violated, and the logarithmic divergence is eliminated if the energy U is correctly expressed.

As already mentioned, the function $\mathscr{F}^1$ depends only on $\omega + e^2/r$ and in the integration region the term ω can be neglected. Because of the logarithmic factor in φ we can assume that $r' \ll r_0$, and substituting $g(\varepsilon) = g_0$ in the integral (1.5) we obtain

$$\mathscr{F}^1(0, r) = g_0^2 e^2/r. \tag{3.9}$$

Thus, the function $\varphi(\omega, r_0)$ must satisfy the equation

$$\varphi(\omega, r_0) = \frac{\alpha}{4r_0^3}\int_0^\Delta \frac{d\omega'}{\omega + \omega'}\int_0^\infty r'^2 \exp\left\{-\frac{r'}{r_0}\varphi(\omega', r')\right\} dr'. \tag{3.10}$$

In view of the weak dependence of $\varphi(\omega, r)$ on r, we replace in the argument of the exponential $\varphi(\omega', r')$ by $\varphi(\omega', r_0)$. We then obtain

$$\varphi(\omega, r_0) = \frac{\alpha}{2}\int_0^\Delta \frac{d\omega'}{(\omega + \omega')\varphi^3(\omega', r_0)}. \tag{3.11}$$

The solution of (3.11) at $\omega \ll \Delta$ (accurate to the number under the logarithm sign) is of the form

$$\varphi(\omega, r_0) = 2^{1/4}\alpha^{1/4}\ln^{1/4}(\Delta/\omega). \tag{3.12}$$

It should be borne in mind that the calculation of $\mathscr{F}(\omega, r)$ is a problem in itself. A pair with arm $r \gg r_0/\varphi$ polarizes not only several neighboring soft pairs, but produces a macroscopic polarization in a large volume. By a method similar to that described in the appendix, it can be shown that in this case the argument of the exponential takes the form $-rr_0^{-1}\varphi(\omega, r)$, where the function φ depends on ω and r logarithmically. The exponential dependence at large r is in essence the consequence of the polaron effect, due to the polarization of the pairs with $r' \approx r_0/\varphi(\omega, r_0)$. Just as in the case of charged excitations, it is possible to introduce "dipole plarons," for which the distribution functions $\mathscr{F}(\omega, r)$ do not decrease like $\exp\{-r\varphi/r_0\}$ at $r \gg r_0/\varphi$, but decrease in power-law fashion because of their interaction with one another, and it is this which is the manifestation of the compactness principle.

So far we have dealt in this section only with paired excitations, but up to formula (3.9) all the results are valid for any dipole excitation, provided that r and r' are taken to mean the arms of the dipoles produced as a result of several electronic transitions. In particular, the exponential decrease described by (3.5) takes place for all dipole excitations having a low excitation energy ω and a displacement r.

It can be verified, however, that the "gross" contribution to the state density $\Phi(\omega)$ is determined precisely by the paired excitations. It can be shown that the functions $\mathscr{F}^1(0, r)$ for excitations that cover four or more sites and have a dipole-moment displacement r, is less than expression (3.9) at $r \ll r_0$ since it does not contain the large factor e^2/r. Thus, it is precisely paired excitations that make the main contribution to the thermodynamics of this system, and also make up the shell of the polaron. The Appendix contains a derivation of Eq. (1.6), that describes the state density of single-site excitations with account taken of the polaron gap.

4. SPECIFIC HEAT

In conclusion we wish to discuss the phenomena in which the above-predicted anomaly of the state density of the dipole excitations can manifest itself. First to be discussed is the low-temperature specific heat. It was emphasized in Ref. 6 that the state density (1.9) cannot be used to calculate the specific heat, since the

dipole excitations make a much larger contribution, proportional to T. Nonetheless, in Ref. 13 the specific heat was calculated with the aid of the law (1.9) and the result $C_v \propto T^3$ obtained there is in our opinion in error.

The logarithmic singularity of the function $\mathcal{F}(\omega, r)$ at low energies leads to the specific heat anomaly. The state density that enters in the linear law for the specific heat is determined by the function $\Phi(\omega)$ [see 1.3)]. Substituting (3.7) and (3.9) in (1.3) and neglecting the weak dependence of φ on r, we obtain

$$\Phi(\omega) = 2\pi g_0/\varphi^2. \tag{4.1}$$

Since φ is a weak function of the frequency, the dependence of the specific heat on the temperature takes the form $C_v \propto T\Phi(T)$.

The asymptotic $\varphi(\omega)$ dependence is determined at low frequencies by (3.12) [we note that this equation is valid only at very low frequencies and for comparison with experiment it would be necessary to solve (3.11) numerically], and therefore C_v has a low-temperature anomaly:

$$C_v \infty T/|\ln T|^{\frac{1}{2}}. \tag{4.2}$$

We discuss now the role of quantum effects. The overlap of the wave functions of two states localized at a distance r from each other leads to a quantum "repulsion" of the levels by an amount $I = E_0 \exp\{-2r/a\}$, where E_0 is to the order of the binding energy and a is the characteristic dimension of the wave function. If the replusion energy of the two states separated by a distance r_0 is small compared with the gap width Δ, then classical theory can be used in a wide range of lengths and energies.

Nonetheless, at sufficiently low energy ω (and accordingly at low temperatures) quantum effects become significant. An important role in the integral of (3.5) is played by lengths $r \approx r_0/\varphi$, which decrease with energy ω. At sufficiently low energy, I exceeds ω, after which the classical theory no longer holds. [The condition for its applicability is $r_0/\varphi \gg r_q \equiv \frac{1}{2} a \ln(E_0/\omega)$.] We note here only that the logarithmic anomalies in the specific heat are preserved also in the quantum region.

We recall that in the lattice model there exist quadrupole etc. excitations which in fact do not participate in the interaction and therefore have a state density that does not depend on energy (although it is small at high values of A). These excitations yield a strictly linear specific heat at sufficiently low temperature. However, as already emphasized, in a disordered system with randomly distributed centers there are no such excitations.

We discuss now the electronic specific heat of a doped semiconductor under the assumption that the electronic states are localized and that compensating impurities are present. For the sake of argument we assume that the donor density N_D exceeds the acceptor density N_A. According to the foregoing, the specific heat is determined by the electron transfer within donor pairs consisting of empty and filled donors. It is first necessary to calculate the function $\mathcal{F}^1(\omega, r)$, which determines the number of pairs with a displacement r smaller than the average distance between the donors and with low excitation energy under the assumption that these pairs do not interact with one another.

The scatter of the levels in the system is due to the random fields of the charged donors and acceptors, whose electrostatic potential changes little over distances $r < N_D^{-1/3}$. Therefore the pair excitation energy can be represented in the form $\omega = e\mathbf{E} \cdot \mathbf{r}$, where $\mathbf{E}$ is the random electric field at the location of the pair. Then, for example at $N_D = 2N_A$, we have

$$\mathcal{F}^1(\omega, r) = \frac{N_A^2}{4\pi} \int_0^\infty F(E)\delta(\omega - eEr)\,dE\,d\Omega_E, \tag{4.3}$$

where $F(E)$ is the distribution function of the electric fields and the averaging is over the angles between the field $\mathbf{E}$ and the vector $\mathbf{r}$. The integral with respect to the angles can be calculated, and

$$\mathcal{F}^1(\omega, r) = \frac{N_A^2}{2er} \int_{\omega/er}^\infty \frac{F(E)}{E}\,dE. \tag{4.4}$$

The function $F(E)$ is bell-shaped with one characteristic scale $E_N = eN^{2/3}/\varkappa$, i.e., $F(E) = E_N^{-1}\Psi(E/E_N)$ ($\varkappa$ is the dielectric constant of the lattice). In the case of interest to us $\omega \ll erE_N$, so that we can put $\omega = 0$ in (4.4). We obtain ultimately

$$\mathcal{F}^1(0, r) = \frac{N_A^2}{2(e^2 N^{1/3}/\varkappa)^2} C \frac{e^2}{\varkappa r}, \tag{4.5}$$

where

$$C = \int_0^\infty dx\, \Psi(x)/x$$

is a dimensionless constant. We note now that (4.5) coincides with (3.9) if we put

$$g_0^2 = N_A^2 C \varkappa^2/2(e^2 N^{1/3})^2.$$

Therefore the theory that describes pair interaction does not differ from that developed above and leads to the same behavior of the specific heat.

At the maximum impurity concentration that leads itself to the classical treatment, the width of the Coulomb gap can reach 5 meV in silicon and 1.5 meV in germanium. Therefore the anomalies of the specific heat should be sought at helium temperatures and below. We know of experiments of this type with silicon,[15] but unfortunately the samples used there were uncompensated and practically all the donors were neutral.

The function $\mathcal{F}(\omega, r)$ plays an important role in the theory of high-frequency electric conductivity. In particular under relatively low-frequency relaxation conditions[16] this function, taken at $\omega = kT$, determines the temperature dependence of the electric conductivity. The singularities indicated in the present paper for low energies can become important in this case.

APPENDIX

DERIVIATION OF EQUATION (1.6)

Just as in the derivation of the SCE, we assume that the fluctuations of the density of the paired excitations

are described by Poisson statistics and that the method of optimal fluctuations is applicable (see Ref. 17). The probability of the fluctuation is proportional to $\exp(-\Omega\{\xi\})$, where

$$\Omega\{\xi\} = -\int \left[(\xi^0+\xi')\ln\frac{\xi^0}{\xi^0+\xi'} + \xi' \right] ds\, dr\, d\omega, \qquad (A.1)$$

and $\xi(r, \omega, s) = \xi^0 + \xi'$ is the density of pairs with displacement r and with excitation energy ω per unit volume, $\xi^0 \equiv \mathcal{F}(\omega, r)$, and $\xi'(\omega, r, s)$ is the deviation from the mean value and depends on the spatial coordinate s. It is required to find the probability that an empty electron level with energy $\varepsilon \ll \Delta$ is present at the origin. The necessary condition for the existence of such a level in the ground state is that the process consisting of the landing of the electron on this level with simultaneous polarization of pairs in its vicinity is energywise not favored, i.e., $\varepsilon - E > 0$, where

$$E = \int \left(\frac{e^2 rs}{s^3} - \omega \right) \theta \left(\frac{e^2 rs}{s^3} - \omega \right) \xi\, dr\, ds\, d\omega \qquad (A.2)$$

is the energy gain resulting from the polarization of the pairs and the field of a point charge located at the origin. The average value of E is of the order of Δ and greatly exceeds ε, and we shall therefore be interested in low-probability fluctuations, in which $\xi \ll \xi^0$. To determine the argument of the exponential we must put $E = \varepsilon$, which leads to the following variational problem: find the function ξ that minimizes $\Omega\{\xi\}$ under the condition that $E\{\xi\} = \varepsilon$. Introducing the Lagrange multiplier λ, we obtain from the condition $\delta(\Omega + \lambda E) = 0$, that

$$\xi = \mathcal{F}(\omega, r)\exp\{-\lambda x\theta(x)\}, \qquad (A.3)$$

where $x = e^2 r \cdot s/s^3 - \omega$. Substituting (A.3) in (A.2) and equating (A.2) to ε, we get

$$\lambda = \frac{\varepsilon_0}{\varepsilon^2}, \qquad \varepsilon_0^{1/3} = \frac{16\pi^{1/3}}{15}\int_0^\infty \mathcal{F}(\lambda^{-1}, r)\, r^{7/3}\, dr. \qquad (A.4)$$

In the derivation of (A.4) we have neglected the logarithmic dependence of $\mathcal{F}(\omega, r)$ on ω, putting $\omega = 1/\lambda$ in the argument of this function. Substituting (A.3) in (A.1) and making an analogous replacement, we obtain (1.6), with $\gamma = 1.5$.

[1] In this most important problem our point of view does not agree with that of Pollak and Knotek,[11] who admit of a non-zero state density on the Fermi level.

[2] For this reason, the state density $g(\varepsilon)$ need not necessarily be an analytic function at small ε, as is stated in Refs. 11 and 13. An example of non-analytic behavior is provided by Eq. (1.8).

[3] In essence, this rule is only empirical. For example, in the region $|\varepsilon| \gg \Delta$, where $g(\varepsilon)$ differs little from g_0, we can use perturbation theory and show that the first correction to g_0, which follows from the SCE, is undervalued by a factor of two. However, at low energies the result of the SCE agrees well with the computer experiment. In the limit as $\varepsilon/\Delta \to \infty$ it, of course, also yields $g(\varepsilon) = g_0$ and therefore approximates $g(\varepsilon)$ well at all energies.

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