Anisotropic hopping conductivity in superlattices and multilayer structures

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The conductivity anisotropy of systems in which the concentration of local centers is periodically modulated in one dimension is discussed under conditions such that the conductivity is related to hopping both in the direction of the periodicity and along the layers. It is shown that when the hopping conductivity of these systems is calculated by reducing its evaluation to a problem of percolation theory, the problem that results is characterized by the presence of two percolation thresholds whose difference determines the anisotropy in the primary exponential behavior of the conductivity. Anisotropies in other hopping-related kinetic coefficients (thermopower and AC conductivity) are discussed as well.

1. INTRODUCTION

Distinctive features of the conductivity of compositional superlattices in the direction of periodicity (in particular, the appearance of negative differential conductivity) can be understood naturally in terms of a model based on hopping transport that associates the conductivity with phonon-assisted tunneling transitions between neighboring layers.[1–5] Analogous processes have been discussed by Belen'kiy[6] for amorphous compositional superlattices, including transitions between conducting channels located within the same layer of the superlattice. Calecki *et al.*[7] have studied the conductivity of the layered semiconducting compound InSe under conditions such that the conductivity along the layers is due to delocalized carriers; in this case, the activation energy of the conductivity is observed to be temperature-dependent in the transverse direction. This is described fairly well by the Mott law, which applies to the regime of conductivity by variable-range hopping. However, a very different situation is encountered in the case of a one-dimensional periodic modulation of the density of hopping centers, where the conductivity is due to a hopping mechanism both along the layers and in the direction of the periodicity.[8] This situation can be realized, e.g., in compensated semiconductor systems whose concentration of majority doping impurities varies periodically; in particular, structures with periodically repeating δ-doped layers and multilayer structures. An analogous situation can occur in compositional doping superlattices, especially at low temperatures under compensation conditions when the Fermi level lies in the region of localized states.

In disordered media with a large scatter in the random probabilities for transitions between localized states, hopping conductivity is usually computed by reducing the equations of balance to a corresponding problem in percolation theory (the problem of bonds for random sites). In this case the primary exponential factor in the expression for the conductivity is determined by the percolation value of the transition rate, which corresponds to the appearance of an infinite cluster of connected bonds. For systems with anisotropic wave functions a unique threshold exists for the appearance of a cluster that is infinitely extended in all directions; accordingly, the anisotropy of such a system can be associated only with anisotropy of the pre-exponential factor, while the primary exponential dependence of the conductivity on the concentration of hopping centers and temperature remains isotropic.[9,10]

In Refs. 11, 12, we discussed anisotropy in the hopping conductivity of quasi-one-dimensional disordered systems, and the way its temperature and frequency dependence changes as the effect of transitions between one-dimensional filaments increases. For the quasi-two-dimensional systems we will discuss here the transition to the three-dimensional case can give rise to features in the variation of the conductivity that are generally different from those we discussed in these references, due to the different topology of the optimal transport paths. For example, although it is in fact impossible to obtain a finite value of the DC conductivity for a system of one-dimensional chains of local centers without taking into account interchain hopping, for quasi-two-dimensional systems the DC conductivity along the layers turns out to be finite even in the absence of transitions between layers.

The specific features of hopping processes in the multilayer structures we discuss here are associated with two factors. The first of these is that the hopping integral between local centers depends not only on the distance between them but also on their relative positions, in particular, which regions these centers are located in (the attenuation length of the wave functions are different for the different regions of the superlattice). The second feature is connected with the nonuniform distribution of local centers in space. Both of these factors are very important, and, generally speaking, will affect the results of the percolation calculation of hopping kinetic coefficients in different ways.

2. THE PROBLEM OF *R*-PERCOLATION FOR A MULTILAYER MEDIUM

In order to explain the basic features of these systems, let us first consider the problem of R-percolation for a

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superlattice consisting of a system of δ-doped parallel layers located at a distance d from one another. In the problem of R-percolation the transition probabilities between localized states do not depend on their energies, but rather are determined only by the spatial position of the sites. For the coordinate dependence of the probability of a tunneling transition W_{ij} between localized states i and j we can write the following standard model expression

$$W_{ij}=W_0\exp\{-2R_{ij}/a\}. \quad (1)$$

Here W_0 is a pre-exponential factor, which for simplicity we assume is constant, $R_{ij}=\sqrt{\rho_{ij}^2+(z_i-z_j)^2}$, $(\boldsymbol{\rho}_i,z_i)$ is the point at which the i-th state is localized, $\boldsymbol{\rho}_i$ is the two-dimensional radius vector of the center in the plane of the layer, z_i is the coordinate of the layer at which the center is found in the direction of periodicity, and a is the radius of the localized state.

Since the characteristic distances between localized centers greatly exceed a, the random scatter of the coordinates of the centers leads to exponentially large scatter in the transition probabilities, allowing us to reduce the problem of calculating the hopping conductivity to the corresponding problem in percolation theory. Once we have chosen a certain value for the transition probability W (or a certain distance R) we will say that there is a bond between pairs of centers i and j when the relation $W_{ij}>W$ (or $R<R_{ij}$) holds; in this case, all of the centers that lie within a sphere of radius R around a given center are connected by bonds with the given center.

Since the distance between centers located in different layers cannot be smaller than d ($R_{ij}\geqslant d$), our problem is characterized by the presence of two percolation thresholds, corresponding to the appearance of a cluster that is infinitely extended in directions parallel to the layer (W_{c1}) and a cluster that is infinitely extended in all directions (W_{c2}). The first threshold, which corresponds to the appearance of percolation parallel to a layer (the circles problem), is reached when the average number of bonds at one center, given by πR^2N_2 where N_2 is the concentration of centers in the layer, equals a critical value $B_c^{(2)}$ (numerical calculations give $B_c^{(2)}\simeq 4.5$).[10,13-15] Accordingly, the percolation radius $R_{c1}=(B_c^{(2)}/\pi)^{1/2}N_2^{-1/2}\simeq 1.2\,N_2^{-1/2}$. It is obvious that for $d>R_{c1}$ there is no transverse percolation in the range $R_{c1}<R<d$, since there are no bonds between centers located in different layers. Transverse percolation must necessarily occur for values of $R>d$ such that percolation can take place in the problem of the circles obtained from the intersection of the spheres by the planes of the neighboring layers. For $R_{c1}\ll d$ the corresponding percolation value R_{c2} exceeds d by an amount on the order of the localization radius, i.e., $R_{c2}\simeq d$. Since the value of the percolation radius determines the primary exponential concentration dependence of the hopping conductivity, this implies that for $d>R_{c1}$ this dependence should differ considerably for conductivity in the different directions.

The existence of two different percolation thresholds essentially distinguishes our problem from both the problem of anisotropic percolation on regular lattices[16] and the problem of percolation along a system of random sites with

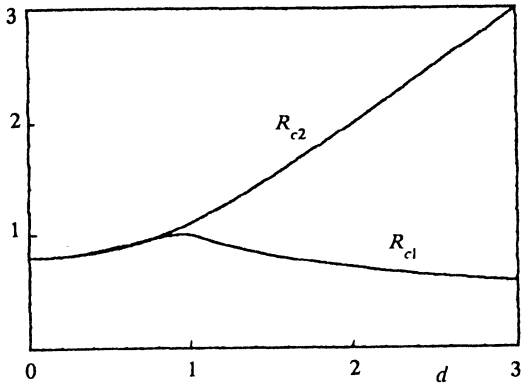

FIG. 1. Dependence of the critical hopping lengths along (R_{c1}) and transverse (R_{c2}) to the layers of the superlattice on the distance between layers containing the centers through which the hopping transport takes place, for the problem of R-percolation [distances are measured in units of $(\pi N)^{-1/3}$].

anisotropic transition probabilities,[9,10] for which the presence of a single percolation threshold (which depends on the anisotropic probabilities of existence of bonds) is characteristic. The anisotropy in the percolation problem for random sites that arises in calculating the conductivity due to hopping between states with anisotropic wave functions appears as anisotropy in the pre-exponential factor of the conductivity. For the system we discuss here, the anisotropy appears in the exponential factor of the hopping conductivity; this is due to the significantly nonuniform distribution of local centers in space, which yields a lower bound on the probability for transitions between centers in different layers. It is clear that when the distance between layers becomes smaller than the percolation threshold R_{c1} along the layer, this anisotropy of the hopping conductivity becomes small.

Figure 1 shows schematically the dependence of the thresholds R_{c1} and R_{c2} on the distance d between layers of the superlattice for a prespecified average bulk concentration $N=N_2/d$ of centers randomly distributed in the layers. For $Nd^3\ll 1$ the average distance between the layers is small compared to the distance between centers in the plane of a layer, and the problem is close to the isotropic three-dimensional problem for which the percolation threshold R_c is of order $N^{-1/3}$. For $Nd^3\gg 1$ the anisotropy is present: the threshold R_{c2} for transverse percolation grows linearly with increasing d, while the d dependence of the percolation threshold R_{c1} along the layer is determined solely by the variation of the density $N_2=Nd$ (for fixed N).

Let us now consider the problem of R-percolation for a doped type-I compositional superlattice made of compensated material (see Fig. 2). Let the forbidden band width in the A-regions of the superlattice be smaller than in the B-regions, and let the hopping centers be randomly distributed in the A-regions (the spatial distribution of the compensating impurities primarily affects the form of the random field and is not important in our problem). Furthermore, let the attenuation length b of the wave func-

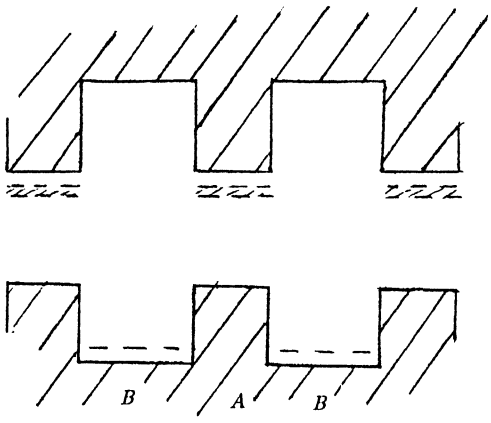

FIG. 2. Energy sketch of a doped compositional superlattice.

tion in the B-regions be much smaller than the corresponding length a in the A-regions. In this case the transition probabilities between centers located in different layers is found to be much smaller than between centers for comparable distances within a single layer. Therefore we can use the following expression for the transition probabilities between superlattice local centers:

$$W_{ij}=W_0\exp\{-2\rho_{ij}/a-2|z_i-z_j|/b\}. \qquad (2)$$

In deriving the approximate expression (2) we assumed $b\ll a$ and that d_A, i.e., the thickness of the A-regions of the superlattice, is on the order of the radius a; the last of these conditions allows us to ignore the dependence of the transition rate on the z-coordinate of the centers lying in the same layer of the superlattice. Since we have $b\ll a$, the primary decay of the transition probabilities between centers located in different layers of the superlattice is connected only with the B-regions, which we take into account by including the second term in the argument of the exponential function (2). The scale transformation $z\to z(b/a)$ leads to a problem of the same type as that of a superlattice of δ-doped layers; in this case, however, the effective distance between A-layers is now equal to $d=d_B(a/b)$, where d_B is the thickness of the B-regions. The transition probability has the form (1), with the sole difference that R_{ij} is replaced by $\rho_{ij}+|z_i-z_j|$. Accordingly, the existence of a bond between pairs of centers i and j is determined by the relation $W_{ij}>W$ (or $R<\rho_{ij}+|z_i-z_j|$). Thus, a given center is found to be connected by bonds to all centers lying within a double cone formed by rotating a square with side $\sqrt{2}R$ around its diagonal, i.e., a double cone inscribed within a sphere of radius R.

The bond concentration in this system depends on W (R) as W decreases (R increases), there is a certain $W=W_{c1}$ ($R=R_{c1}$, the percolation threshold along the bonds) for which an infinite cluster of interlinked bonds appears in the system. In contrast to the problem of spheres, which arises in the theory of percolation along random sites that are randomly distributed on the periodically spaced layers, we now have a problem of double cones with a spatially nonuniform (laminar) distribution. Under conditions where the average distance between local centers in the planes of the layers is smaller than the distance between layers, the threshold value W_{c1} corresponds to the appearance of percolation along the layers alone; as we pointed out above, the threshold value W_{c1} and the value R_{c1} corresponding to it are determined from the problem of the circles defined by intersections of the double cones and the planes of the layers. For $R_{c1}<R<d$ there is no percolation in the direction of periodicity. In this direction percolation appears for $W=W_{c2}$ ($R=R_{c2}$) for layer widths such that $R_{c2}>d$. When $d>R_{c1}$ this threshold corresponds to the condition that percolation take place along the intersections of the double cones with the planes of neighboring layers, i.e., in this case the approximate relation $R_{c2}\simeq d$ holds.

In the opposite limiting case, when the distance between layers is small compared to the percolation length of a bond, we are led to the problem of a uniform distribution of centers in space. For this case the percolation problem reduces to the problem of randomly distributed double cones, for which the percolation threshold $W_c(R_c)$ is characterized by the appearance of a cluster that is infinitely extended in all directions. Consequently, as the ratio R_c/d increases the thresholds R_{c1} and R_{c2} approach one another and the threshold for three-dimensional percolation. The latter is determined by the bond criterion, according to which the percolation threshold corresponds to a specific value of the average number of bonds in a calculation for a single center $B_c^{(3)}$. For the problem of spheres the average number of bonds at a center equals $(4\pi/3)NR^3$, where N is the average bulk concentration of centers, so that the percolation radius is given by $R_c=AN^{-1/3}$, where $A=(3B_c^{(3)}/4\pi)=1.13$ for $B_c=3$. For our problem of double cones with a random distribution of centers that is uniform in space, the average number of bonds at a center equals $(2\pi/3)NR^3$. Since the average number of bonds at a center $B_c^{(3)}$ depends weakly on the shape of the figure,[9,11] we can use the same value of $B_c^{(3)}$ for the problem of double cones as we did for the problem of spheres; as a result, we obtain the previous expression for R_c with a somewhat different value of the constant $A=1.42$. In this case all the basic conclusions obtained previously for a system of δ-doped layers (existence of two percolation thresholds and large anisotropy for $d>R_{c2}$) remain valid for hopping conductivity along isoenergetic centers in the superlattices as well.

3. THE PROBLEM OF R-ε-PERCOLATION FOR A LAYERED DISTRIBUTION OF LOCAL CENTERS

As a rule, the problem of R-percolation cannot be applied directly to describe impurity hopping conductivity, since a significant role is played by the scatter in the energy of localized states connected with broadening of the impurity levels into an impurity band. This broadening, which is primarily caused by the classical shifts in levels, is determined in superlattices by two factors. First of all, there is the positional disorder, which leads to fluctuations in energy levels because the position of energy levels of impurities in the superlattice depends on the position of impuri-

ties in the superlattice layers.[1,17] Secondly, the classical broadening of the levels is caused by the random field created in compensated materials, primarily by charged impurities. Thus, the actual impurity spectrum in a superlattice consists of a range of localized-state energies (i.e., a band) with a width on the order of several rydbergs. Accordingly, in the region of low temperatures where the conductivity along impurities is significant, the width of this band is found to be smaller than kT, and inclusion of the dependence of the transition probabilities between localized states on their energies becomes important. When the Fermi level is located in the region of the quasicontinuous spectrum of localized states, while the width of the impurity band exceeds the characteristic hopping energy, a regime of variable-range hopping conductivity results. A temperature-dependent hopping length R_h is characteristic of this regime, so that we may expect the relation between R_h and d to vary as well, i.e., the anisotropy will be temperature-dependent.

In the variable-range hopping regime the hopping conductivity is calculated by solving the balance equation, which contains the transition rates $\Gamma_{ij}=W_{ij}f_i(1-f_j)$, where both the transition probabilities W_{ij} and the equilibrium occupation numbers f_i depend on the energy ε_i of the states.[10,13,14] For the case under discussion here, i.e., a compositional superlattice, the primary exponential dependence of the transition rates on the center coordinates and on their energies has the form

$$\Gamma_{ij}=\Gamma_0\exp(-\eta_{ij}) \tag{3}$$

where

$$\eta_{ij}=2(\rho_{ij}/a+|z_i-z_j|/b)+\varepsilon_{ij}/kT, \tag{3a}$$

and $\varepsilon_{ij}=(\varepsilon_i-\varepsilon_j)\theta(\varepsilon_i-\varepsilon_j)$ [here $\varepsilon_{ij}>kT$]. In this case the conductivity can be calculated as in the case of R-percolation by reducing the problem to the corresponding bond problem. The presence of a bond between centers is indicated by the relation $\Gamma_{ij}>\Gamma$ (or $\eta_{ij}<\eta$); for $d>b\eta$ there are no bonds between centers located in different layers.

Let us denote by Γ_{c1} (η_{c1}) the value corresponding to the percolation threshold along a layer; under conditions such that the hopping length in a layer corresponds to this threshold, i.e., $R_{c1}<d$, the threshold value is determined by the solution of the two-dimensional R–ε-percolation problem at random sites, $\eta_{c1}=(T_2/T)^{1/3}$, where the temperature satisfies $T_2=A_2/(\rho_2 a^2)$; here ρ_2 is the density of local centers in the layer and A_2 is a constant of order unity ($A_2\simeq 5.7$; see Ref. 15), which is related to the critical number of bonds at a single center in the calculation for the two-dimensional problem. The threshold value of the parameter η corresponding to percolation in the direction of periodicity of the superlattice for $d\gg R_{c1}$ is

$$\eta_{c2}\simeq(2d/a)=(2d_B/b). \tag{4}$$

Accordingly, for $d>R_{c1}$ we have $\eta_{c2}>\eta_{c1}$, and the anisotropy of the hopping conductivity turns out to be exponentially large.

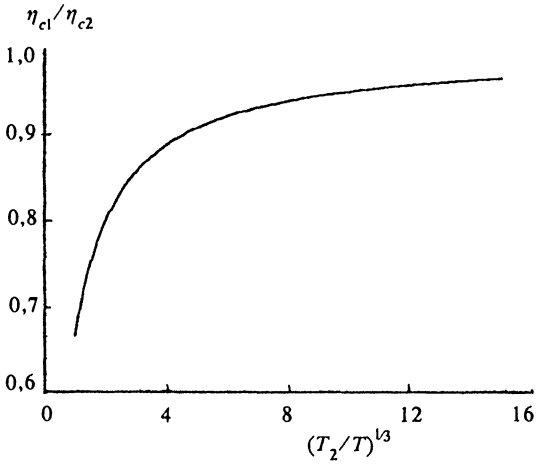

FIG. 3. Temperature dependence of the ratio η_{c1}/η_{c2} that characterizes the anisotropy of the hopping conductivity.

We note that anisotropy was obtained in the exponential factor of the conductivity for quasi-one-dimensional systems by Bonch-Bruevich *et al.*[12] for weak transverse linking of filaments in the temperature range above and immediately adjacent to the range where the conductivity is isotropic and described by the Mott law. The nature of this anisotropy, which is probably the same as that discussed in this paper, could be related to the appearance of optimal hopping paths in this range that extend infinitely along the filaments. This corresponds to the appearance of a system of infinite clusters of connected bonds that are not connected to one another but extend along filaments of the cluster under conditions such that there still is no transverse percolation.

4. DISCUSSION OF RESULTS

Because the relation between R_{c1} and d is temperature-dependent, the hopping length may become considerably larger than d as the temperature decreases, so that Eq. (4) cannot be applied. For large hopping lengths we are led to the problem for random sites with a uniform distribution of sites in space. In this case there exists a single threshold for the appearance of a cluster that is infinite in all directions, i.e., $\eta_c=(T_0/T)^{1/4}$, where $T_0=A_3/(k\rho_3 a^3)$; here ρ_3 is the spatially averaged density of states, $\rho_3=\rho_2/d$, and A_3 is a constant ($A_3\simeq 17.6$).

Thus, in the regime of variable-range hopping conductivity the anisotropy depends on the ratio of the hopping length to the period of the superlattice and, consequently, on temperature. In Fig. 3 we show schematically the temperature dependence of the ratio η_{c1}/η_{c2}, which characterizes the anisotropy of the argument of the exponential function in the expression for the hopping conductivity. It is clear that the anisotropy decreases with decreasing temperature as the hopping between layers begins to dominate.

The conductivity anisotropy discussed above is primarily connected with the differing transition rates between localized states due to their dependence on the distance

between centers. In this case the anisotropy of the thermoelectric power should be considerably weaker. Actually, the topology of optimal paths is such that for $d > R_{c1}$ a significant number of these paths are located in the conducting A-layers of the structure, so that the Peltier heat (the average energy of the electrons that give the primary contribution to the conductivity) is the same for the longitudinal and transverse transports, whereas in the variable-range hopping conductivity regime it is close to the hopping energy along the layers $kT(T_2/T)^{1/3}$.

At the same time, under conditions such that the DC conductivity is anisotropic those characteristics that are determined by the total transition rate (i.e., that also depend on the spatial distribution of centers) are generally anisotropic as well. Among these is the AC conductivity, for which the characteristic hopping length R_h at frequency ω is determined by the condition $\omega\tau(R_h) \simeq 1$, where $\tau(R_h)$ is the relaxation time for the corresponding finite clusters.[13] Since the length R_h decreases with increasing frequency, the transverse AC conductivity becomes small compared to the conductivity along the layers when $R_h < d$. Since the transition rates between layers are exponentially small under these conditions, even for electric fields normal to the layers, the primary contribution to the conductivity comes from pairs of centers located within a single layer. An approach based on the pairwise approximation gives the following expression for the conductivity along the layers in the quasi-two-dimensional approximation (neglecting electron-electron interactions):

$$\mathrm{Re}\,\sigma_1(\omega) = A\omega \ln^3(1/\omega\tau_0) \qquad (5)$$

where A is a known constant that depends on temperature and the density of states, and τ_0 is the pre-exponential factor in the expression for the relaxation time in a pair.[11] For the transverse conductivity the potential difference between sites of localized centers and the projection of the change in dipole moment of pairs onto the direction of current contains not the moment arm of the pair $R_h = (a/2)\ln(1/\omega\tau_0)$, but rather the layer thickness w. Accordingly, for the real part of the transverse conductivity we obtain

$$\mathrm{Re}\,\sigma_t(\omega) \simeq A(2w/a)^3\omega \ln(1/\omega\tau_0). \qquad (6)$$

It is clear that although the power-law character of the frequency dependence of the transverse conductivity remains almost the same as in the longitudinal case, its value is smaller by a factor of $(a/2w)^3 \ln^3(1/\omega\tau_0) = (R_h/w)^3$.

Thus, our investigation of the anisotropy of hopping transport phenomena in systems with one-dimensional periodic modulation in the concentration of hopping centers (in particular, in doping compositional superlattices at low temperatures) makes it possible to obtain information about the topology of the paths of charge carriers and about a number of properties of the system connected with disorder (such as the radius of localization, the energy and length of hopping, and the density of states in the impurity band).

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[1] M. Hermann, *Semiconductor Superlattices* (Russ. translation). Mir, Moscow, 1989.
[2] L. Esaki and L. L. Chang, Phys. Rev. Lett. **33**, 495 (1974).
[3] R. Tsu and G. Döhler, Phys. Rev. B **12**, 680 (1975).
[4] R. F. Kazarinov and R. A. Suris, Fiz. Tverd. Tela (Leningrad) **14**, 120 (1972).
[5] M. Saglam, M. Pollak, and L. R. Friedman, J. Phys.: Condensed Matter **1**, 5441 (1989).
[6] G. L. Belen'kiĭ, JETP Lett. **47**, 584 (1988).
[7] D. Calecki, J. F. Palmier, and A. Chomette, J. Phys. C **17**, 5017 (1984).
[8] V. N. Rud'ko, Fiz. Tverd. Tela (Leningrad) **30**, 3470 (1978) [Sov. Phys. Solid State **20**, 2006 (1978)].
[9] B. I. Shklovskiĭ, Fiz. Tekh. Poluprovodn. **11**, 2142 (1977) [Sov. Phys. Semicond. **11**, 1257 (1977)].
[10] B. I. Shklovskiĭ and A. L. Éfros, *Electronic Theory of Doped Semiconductors* (Springer-Verlag, New York, 1984); Nauka, Moscow, 1979.
[11] V. N. Prigodin, A. N. Sanuikhin Fiz. Tverdogo Tela (St. Petersburg) **26**, 1344 (1984) [Solid State Phys. **26**, 817 (1984)].
[12] E. P. Nakhmedov, V. N. Prigodin, A. N. Sanukhin, Fiz. Tverdogo Tela (St. Petersburg) **31**, 31 (1989).
[13] I. P. Zvyagin, *Kinetic Phenomena in Disordered Semiconductors* (in Russian). Moscow Univ. Publ., Moscow, 1984.
[14] I. L. Bonch-Bruevich, I. P. Zvyagin, R. Kaiper *et al.*, *Electronic Theory of Disordered Semiconductors* (in Russian). Nauka, Moscow, 1981.
[15] G. E. Pike and C. H. Seager, Phys. Rev. B **10**, 1421 (1974).
[16] S. Redner and H. E. Stanley, J. Phys. A **12**, 1267 (1979).
[17] G. Bastard, Phys. Rev. B **24**, 4714 (1981).

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