PHYSICAL REVIEW B **70**, 024201 (2004)

ac conductivity of a Coulomb glass from computer simulations

S. A. Basylko,[1,]* V. A. Onischouk,[2,†] and A. Rosengren[1]
[1]*Condensed Matter Theory, AlbaNova University Center, Royal Institute of Technology, SE-10691 Stockholm, Sweden*
[2]*Joint Institute of Chemical Physics, RAS, 117 977, Kosygin Street 4, Moscow, Russia*
(Received 27 February 2004; published 2 July 2004)

A method for calculating the photon-induced hopping ac conductivity of a Coulomb glass by computer simulation is proposed. Results obtained by using an effective relaxation algorithm for two three-dimensional models of a Coulomb glass are reported. ac conductance data clearly demonstrate the transition from super-linear to a sub-quadratic power law. We argue that the same qualitative behavior should be expected for compensated semiconductors. It is shown that the transition is driven by the Coulomb energy of sites forming resonant pairs and not by the width of the Coulomb gap.

DOI: 10.1103/PhysRevB.70.024201 PACS number(s): 72.80.Ng, 71.23.−k, 72.20.Ee

I. INTRODUCTION

It is widely accepted now,[1] that the electron-electron interaction plays an important role in forming the properties of disordered systems of localized electrons, so-called Coulomb glasses (CG), of which lightly doped semiconductors at low temperatures are the best known examples. The conductivity in such systems is due to tunneling of electrons between impurity sites accompanied by phonon and/or photon absorption (emission). At zero temperature such systems cannot conduct direct current, but still there is a possibility for photon-assisted ac conductance.

In 1981 Efros and Shklovskii (ES),[2,3] using arguments outlined below, showed that if the Coulomb correlations are taken into account, the phononless hopping ac conductivity spectra should change its frequency dependence from roughly linear at low frequencies to roughly a quadratic power law (predicted earlier by Mott[4]) at higher frequencies where the photon energy is much larger than the Coulomb interaction between the sites forming the "resonant pair" absorbing the photon. Recent experiments[5,6] confirmed this change in the power law, but the crossover from one power to the other is much sharper than that predicted by ES. Although there were many efforts to compute by Monte Carlo simulation the density of states near the Fermi level where it goes to zero[7] (this is called the Coulomb gap, see, e.g., Refs. 8–11), to calculate the dc conductivity of a Coulomb glass,[12–14] and other effects of phonon-assisted hopping,[15] we found no reports on calculation of the phononless hopping ac conductance of it. Another intriguing question is that about the nature of the transition of ac conductivity spectra from ES to Mott power law behavior. According to ES[2,3] and the experimental study of Helgren, Armitage, and Grüner,[6] this transition is driven by the Coulomb energy of the sites forming the resonant pair. However, Lee and Stutzmann[5] claimed that the transition occurs instead when the photon energy equals the double Coulomb gap width. Our calculations corroborate the first scenario. They show the abrupt crossover from super-linear to sub-quadratic dependence on frequency for an amorphous semiconductor model and for the first time predict the same kind of crossover in compensated semiconductors.

We want to stress important distinctions between theoretical (both ours and ES) CG models and the experimental systems in Refs. 5 and 6. These experiments were performed on uncompensated SiB and SiP samples, respectively. In such systems the so-called self-compensation[16] phenomenon is believed to take place. ES results as well as ours were obtained for a compensated amorphous semiconductor with a large Hubbard energy for electrons on the same site, thus with in-practice forbidden double-site occupation. The applicability of the ES and our models to uncompensated samples is still a controversial issue.[17,18] Another distinction is due to the densities of impurities: in order to treat the interaction between sites as the Coulomb interaction of point charges, we had to restrict ourselves to densities much lower than the experimental ones (see Sec. III). One more distinction is due to the fact that ES results as well as ours are obtained in a two-site approximation, which does not take into account simultaneous many-electron hops. The validity of this approximation is still a controversial issue.[19] One can argue that simultaneous hops of two or more electrons have a much lower probability than one-electron hops.[10,14] Another argument for this approximation is that the dc conductivity temperature dependence, predicted in this approximation, $\sigma_{dc} \propto \exp[-(T_0/T)^{1/2}]$, has been observed in many experiments.[20,21] In the following we restrict ourselves to the two-site approximation and consider only one-electron hops in the relaxation process and resonant absorption process.

Here we propose a method for calculating the photon-induced hopping ac conductance of a Coulomb glass by computer simulation and present results for two 3D models of a Coulomb glass at zero temperature. Using a certain observation we were able to develop a much faster relaxation algorithm, which allows to use systems of size up to 5.12×10^5 sites.

This paper is organized as follows: in Sec. II we introduce the models considered and describe the relaxation algorithm and the method of ac conductance calculation. In Sec. III we discuss finite-size effects and present results for ac conductance for two CG models. The paper concludes in Sec. IV with a summary of results.

II. MODELS AND ALGORITHMS

Here we consider two CG models: the amorphous semiconductor (AS) and the classical impurity band (CIB) model.

These models describe an ensemble of localized electrons on the donor sites with different origin of the disorder. In the AS model the random potential on each donor site originates from the violation of short-range order in an amorphous semiconductor, while in the CIB model the random potential is generated by randomly distributed charged acceptors.

A. Relaxation algorithm

The energy of the models considered is represented by the Hamiltonian

$$H = \sum_{i=1}^{N} \varphi_i(1-n_i) + \frac{1}{2}\sum_{i\neq j} \frac{(1-n_i)(1-n_j)}{r_{ij}}, \qquad (1)$$

where $n_i = 0, 1$ is the occupation number of donor site i,[22] r_{ij} are distances between sites i and j, and N the number of sites. For the AS model, the random energy φ_i on site i is uniformly distributed in the interval $[-A, A]$, and for the CIB model $\varphi_i = \Sigma_\alpha 1/r_{i\alpha}$, where $\alpha = 1, 2, \ldots, KN$ denotes acceptor sites and $K = 0.5$ is the compensation. The sites are randomly distributed in a sample of length L, $N = L\times L\times L$, and periodic boundary conditions[10] for calculating r_{ij} were used. Hereafter we use dimensionless units taking as length, energy and frequency scale $n^{-1/3}$, $e^2 n^{1/3}/\kappa$, and $e^2 n^{1/3}/\kappa\hbar$ correspondingly, where n is donor density, e is electron charge, and κ is the dielectric constant. In the ground state the energy (1) fulfills a set of stability conditions with respect to one-, two-, three-, etc., electron hops within the system. It means that after performing such hops, the energy of the whole system should not decrease. The simplest condition, corresponding to one-electron hops within the system, has the following form

$$\varepsilon_i - \varepsilon_j - 1/r_{ij} \geqslant 0, \quad n_i = 0, \quad n_j = 1, \qquad (2)$$

where $\varepsilon_k = \delta H/\delta n_k$. As we discussed in the Introduction, we assume, that the state satisfying (2) gives a good approximation to the true ground state of the model (1). The state of model (1) in which condition (2) is fulfilled will further on be referred to as the ground state.

We constructed the ground state relaxation algorithm as follows. We start from a random distribution of occupation numbers n_i satisfying the electroneutrality condition $\Sigma_i n_i = KN$ and calculate the initial set $\{\varepsilon_i\}_{i=1,\ldots,N}$. Then, according to Ref. 8, we first satisfy the inequality (2) without the term $1/r_{ij}$ (subroutine M1). Considering the efficiency of the algorithm proposed by Möbius, Richter, and Drittler,[10] we noted that most of the transitions needed to satisfy condition (2) happens for sites that are close to each other. That is why we then, in subroutine M21, search for an energetically favorable transition (i.e., a transition of an electron from site j to site i for which $\delta H = \varepsilon_i - \varepsilon_j - 1/r_{ij} < 0$) among all pairs with $r_{ij} < r^* = 2$ (note that the number of such "close" neighbor pairs $\sim N$). If the search is successful we perform the transition and, after a recalculation of $\{\varepsilon_i\}$, return to the entrance point of the M1 subroutine. After the exit of subroutine M21 after having exhausted all transitions we, in the M22 subroutine, search for transitions among pairs of "far" neighbors with $r_{ij} \geqslant r^*$. It is important to note that on entering subroutine M22 the transition of an electron on a distance $r_{ij} \geqslant r^*$ is favorable only if the energy on sites i and j satisfy the condition $|\varepsilon_{i,j} - \mu| \leqslant 1/r^*$,[10,23] where the Fermi level μ is defined as

$$\mu = \frac{1}{2}[\min_i\{\varepsilon_i : n_i = 0\} + \max_i\{\varepsilon_i : n_i = 1\}]. \qquad (3)$$

Thus the number of pairs considered by M22 is proportional to N as well. So, instead of searching an energetically favorable transition among $\sim N^2$ pairs, we need to consider only $\sim N$ pairs of sites in either subroutine M21 and M22. Since the number of transitions also increases linearly with N the whole relaxation time is proportional to N^2 and the time for one sample (calculation of the initial state $\{\varepsilon_i\}$ and relaxation to the ground state) in our case increases exactly proportional to N^2 so that for $N = 5.12\times 10^5$ and $A = 4.2$ the time needed per sample is $t_{samp} \approx 1.8\times 10^5$ sec on an Intel P4 2.4 GHz processor.

Another advantage of such a division of pairs into "close" and "far" neighbors is an effective usage of memory: we do not store all $N(N-1)/2$ values of $1/r_{ij}$, but only those for close neighbors, and for far neighbors calculate $1/r_{ij}$ directly from the coordinates x_i, y_i, z_i and x_j, y_j, z_j of sites i and j.

B. Calculation of ac conductance

To calculate the ac conductance of the Coulomb glass we follow the arguments of ES.[2,3] First note that the absorption of a photon at frequency ω occurs on rare in space pairs, which means that we can consider the quantum overlap for only the two sites forming the resonant pair. The Hamiltonian of the interaction can then be written as (we omit constant terms here)

$$H = \Phi_1 n_1 + \Phi_2 n_2 + \frac{n_1 n_2}{r_{12}} + I(r_{12})(a_1 a_2^\dagger + a_2 a_1^\dagger), \qquad (4)$$

where $\Phi_k = \varphi_k + \Sigma_{i\neq 1,2} n_i/r_{ik}$, a_i, $a_i^\dagger$ are creation and annihilation operators of an electron on site i, and $I(r_{12}) = I_0 \exp(-r_{12}/\xi)$. The overlap integral of the wave functions of the electron on sites 1 and 2, ξ is the localization length. To take into account the Coulomb correlation of sites, ES considered all possibilities of occupying the sites, i.e., the number of electrons on a pair is equal to 0, 1, or 2. But photon absorption is possible only if a pair has one electron. In our case Coulomb correlation is taken into account by satisfying condition (2), and we need to consider only the case with one electron on a pair. In this case there are two levels with energies $E_1^\pm$:

$$E_1^\pm = \frac{1}{2}(\Phi_1 + \Phi_2) \pm \frac{1}{2}\Gamma, \ \Gamma = \sqrt{(\Phi_1 - \Phi_2)^2 + 4I^2(r_{12})}. \qquad (5)$$

The energy of the external field $\vec{\mathcal{E}} = \vec{\mathcal{E}}_0 \cos(\omega t)$, absorbed by pair per unit time, Q, is equal to

$$Q = 2\pi\frac{\omega}{4}|\vec{\mathcal{E}}_0\langle - |\vec{r}| + \rangle|^2\delta(\omega - \Gamma)\exp\left(\frac{\Omega - E_1^- + \mu}{T}\right) \times \left[1 - \exp\left(-\frac{\omega}{T}\right)\right], \tag{6}$$

where $\langle -|\vec{r}|+\rangle$ is the matrix element corresponding to the electron's transition from the state with energy E_1^- to that with energy E_1^+, $\exp(-\Omega/T)=\exp[-(E_1^- - \mu)/T]+\exp[-(E_1^+ - \mu)/T]$, and the temperature scale is $e^2 n^{1/3}/\kappa k_B$. It can be shown that $|\vec{\mathcal{E}}_0\langle -|\vec{r}|+\rangle|^2=(\vec{E}_0\vec{r}_{12})^2 I^2(r_{12})/\Gamma^2$ and that the conductivity can be found as

$$\sigma(\omega) = \frac{2\sum Q}{\mathcal{E}_0^2}. \tag{7}$$

ES[2,3] have calculated the sum in (7) at $T=0$ (in their case the definition of Ω had two more terms: 1 and $\exp[-(\Phi_1+\Phi_2+1/r_{12}-\mu)/T]$) and got the following expression for the conductivity at frequency ω (in natural units):

$$\sigma_{ES}(\omega) = \frac{1}{3}\pi^2 e^2 g^2 \xi\omega r_\omega^4\left[\hbar\omega + \frac{e^2}{\kappa r_\omega}\right], \quad r_\omega = \xi \ln\frac{2I_0}{\hbar\omega}, \tag{8}$$

where g is the density of states in the noninteracting system. For sites inside the Coulomb gap, ES argued[2,3] that one should divide the result (8) with a term proportional to r_ω^4, since the DOS in the region of the Coulomb gap $g \sim \varepsilon^2$ and $\varepsilon \sim e^2/\kappa r_\omega$.

In a computer simulation we can consider pairs of empty (i) and filled (j) sites and calculate from Eq. (5) the resonance frequency for each pair $\omega_{ij}=\Gamma$, where $\Gamma^2=(\varepsilon_i-\varepsilon_j-1/r_{ij})^2+4I^2(r_{ij})$ with ε_i and ε_j defined as in (2), and its contribution to the conductivity $Q_{ij}\sim x_{ij}^2 I^2(r_{ij})/\omega_{ij}$, where the field is supposed to be in the x direction and x_{ij} is the distance between the sites in this direction calculated using periodic boundary conditions. Dividing the ω axes into small bins and summing up the contributions Q_{ij} of every pair with its resonance frequency within a given bin, we get a histogram $\sigma(\omega)$, which could be considered as proportional to the ac conductance of our sample.

III. RESULTS

The length of a sample considerably influences the density of states (DOS), $g(\varepsilon)=1/N\cdot\Sigma_i\delta(\varepsilon-\varepsilon_i)$, measured in computer simulations, especially in the vicinity of the Fermi level. Particularly, it follows from (2) that for a D-dimensional sample the DOS will be exactly 0 ("hard gap") in the segment $\varepsilon\in[\mu-1/\sqrt{D}L,\mu+1/\sqrt{D}L]$, since in a sample of length L there are no sites on a distance more than $\sqrt{D}L/2$ (note that this is the case for periodic boundary conditions). Another phenomenon noticed in Ref. 24 is a slightly increased DOS with respect to its thermodynamical limit, $\lim_{L\to\infty}g(\varepsilon,L)$, in a region next to the "hard gap" described above. We estimated the whole region of finite size effects to be

$$|\varepsilon_i - \mu| \leqslant 2/L. \tag{9}$$

Thus we considered only sites with energies outside this region during the calculations of the ac conductance.

A. AS model

Before proceeding to a discussion of results, we present what parameters we have taken for the model. The parameter I_0 is determined by the Bohr energy of the localized electron, which gives us in dimensionless units $I_0=1/\xi$. Near the metal-insulator transition ξ diverges and we cannot take the localization length large (compared to the mean distance between sites) if we want to use formula (1) for the energy of the system. In that case one should replace the Coulomb interaction of point charges with that of smeared ones[10] and take into account hybridization and polarization. So we used $\xi=0.8$. Then, according to McMillan's scaling theory[25] of the metal-insulator transition, when the density of impurities is approaching the critical density n_c the localization length diverges as (in dimensionless units)

$$\xi = \left(\frac{n}{n_c}\right)^{1/3}\left(1 - \frac{n}{n_c}\right)^{-\nu}, \tag{10}$$

where $\nu\approx 1$. The value $\xi=0.8$ corresponds to $n/n_c\approx 0.23$. Note that this is much less than the smallest values used in experiments, 0.85[5] and 0.39.[6] Finally, we can evaluate A from the impurity band width data $\Delta E_b\approx 0.1$ eV[21] and get $2A=\Delta E_b/(e^2 n^{1/3}/\kappa)=\Delta E_b/(e^2 n_c^{1/3}/\kappa)(n_c/n)^{1/3}$, which gives $A\approx 2.6(n_c/n)^{1/3}$. Thus we get $A=4.2$. For this value of A we found the Coulomb gap width to be $\Delta\approx 0.2$.

Figure 1 presents results for samples with $L=70$ (crosses) and 80 (stars). One can see the convergence of results with increasing L (inset shows a comparison with results for samples with smaller sizes). Solid lines are minimum squares fit of low- and high-frequency data to the formula $\ln\sigma=\gamma_1+\gamma\ln\omega$ for $L=80$. We obtained $\gamma=1.42$ for $\omega\leqslant 2\cdot 10^{-2}$ and $\gamma=1.93$ for data with $\omega\geqslant 5\times 10^{-2}$.

Together with the ac conductance we calculated the number of sites *outside* the Coulomb gap to the total number of sites participating in the ac conductance at given frequency ω, to clarify the nature of the transition from ES to Mott power-law behavior for the ac conductivity. Our calculations show that sites with energy ε_i lying outside the Coulomb gap begin to contribute to ac conductance for $\omega\gtrsim 0.5$ which means that all data presented in Fig. 1 are related to sites inside the Coulomb gap. The dotted line in Fig. 1 is the ES formula (8) modified for sites inside the Coulomb gap, namely

$$\sigma_{ESCG}\sim\omega\left[\omega + \frac{1}{\xi\ln(2I_0/\omega)}\right]. \tag{11}$$

One can see that our data show larger exponents and a more abrupt transition to sub-quadratic dependence than that predicted by (11). We believe that this is due to the assumption, made by ES when deriving (8), about a uniform spatial distribution of sites with different energies, whereas "cluster-

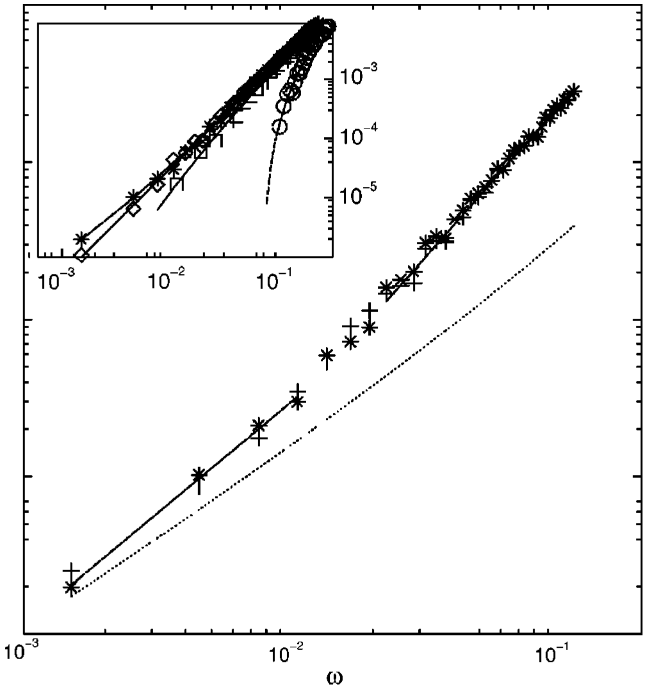

FIG. 1. $\sigma(\omega)$ for the AS model; sample lengths are $L=70$ (crosses) and 80 (stars), averaging were performed over $N_s=22$ and 23 samples respectively. Solid lines are fits with exponents $\gamma=1.42$ and 1.93. A fit to ES formula (11) is given by the dotted line. Inset shows $\sigma(\omega)$ for different $L=10$ (circles), 20 (squares), 50 (diamonds), and 80 (stars). Solid lines are just a guide to the eye. Here we use dimensionless units taking as conductance and frequency scale $e^6 n^{2/3}\xi^5/\hbar\kappa^2(\Delta E_b)^2$ and $e^2 n^{1/3}/\kappa\hbar$, where n, is donor density; e, electron charge; κ, dielectric constant; ξ, localization length; and ΔE_b, impurity band width.

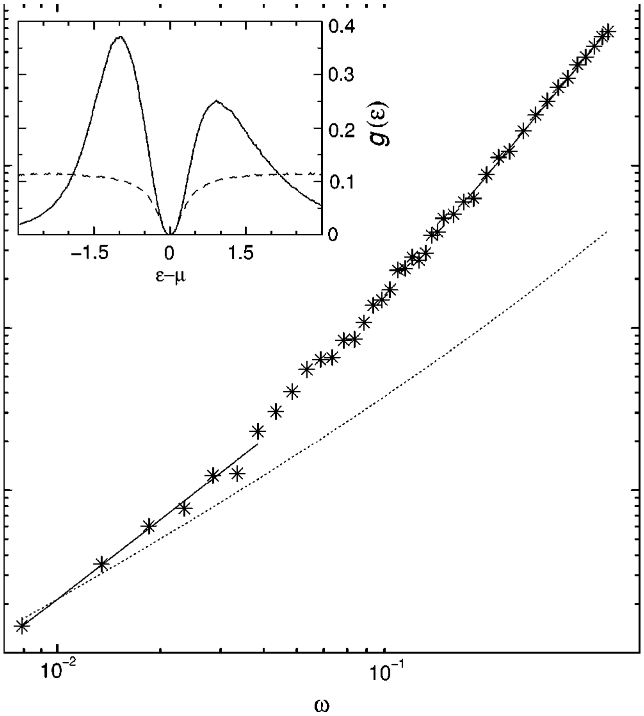

FIG. 2. $\sigma(\omega)$ for the CIB model; sample length is $L=80$ (stars), averaging were performed over $N_s=4$ samples. Solid lines are fits with exponents $\gamma=1.64$ and 2.53. The dotted line represents ES formula (11). Inset shows DOS for the AS (dotted line) and CIB (solid line) models. Here we use dimensionless units taking as conductance, frequency and DOS scale $e^6 n^{2/3}\xi^5/\hbar\kappa^2(\Delta E_b)^2$, $e^2 n^{1/3}/\kappa\hbar$, and $e^2 n^{1/3}/\Delta E_b \kappa$, where n is donor density; e, electron charge; κ, dielectric constant; ξ, localization length; and ΔE_b, impurity band width.

ing” of sites with energies from the same side of the Fermi level in computer simulations has been reported[9,12] and supported by recent analytical results.[26]

B. CIB model

One of the suggestions made by ES in Ref. 3 is that since the largest energy scale in the CIB model is the Coulomb interaction on the mean distance between two sites (1 in our units), it will define the ac conductance behavior and for small ω one should expect the super-linear behavior described by (11) without the ω term in the brackets. For larger ω, one should see a rapidly decreasing conductivity with ω without transition to sub-quadratic form. But, as we have seen in case of the AS model, the sub-quadratic region could be formed by sites from within the Coulomb gap region, thus it is interesting to see whether such a kind of transition takes place in the CIB model.

In Fig. 2, results for the ac conductivity for the CIB model with $\xi=0.8$, $I_0=1/\xi=1.25$, and $L=80$ are presented. Solid lines are fits for low- ($\gamma=1.6$) and high- ($\gamma=2.4$) frequencies. The larger slopes for this model could be explained by its more peaked DOS (see inset in Fig. 2). Again, as for the AS model, the transition between the two regions with different ω-dependence occurs long before sites outside the Coulomb gap begin to participate in the ac conductance.

Based on the results of our simulations we argue that, if one performs experiments analogous to Refs. 5 and 6, but on compensated samples, the same behavior of the ac conductivity should be observed, namely, a sharp increase of the exponent over a small frequency range with increasing frequency before it rapidly drops.

IV. CONCLUSION

In conclusion, we have proposed a method for calculating the phononless hopping ac conductance of a Coulomb glass by means of a fast ground-state relaxation algorithm. We have calculated the ac conductance for the AS model and our calculations show larger exponents and a more abrupt transition than predicted of the ac conductance from a power law of ES type at low frequencies to Mott type at higher ones. We performed calculation for the CIB model as well and showed that in the ac conductance there is a region of super-quadratic behavior in contradiction to ES arguments about its absence.[3] The calculation of the relevant number of sites outside the Coulomb gap that take part in the ac conductance shows that the transition is driven by the Coulomb energy of the sites forming the resonant pairs rather than by the width of the Coulomb gap.

ACKNOWLEDGMENTS

S.B. is thankful to Professor A. A. Ovchinnikov and Professor L. A. Maksimov for helpful discussions. This research was supported by The Swedish Royal Academy of Sciences and by The Swedish Research Council. V.O. and S.B. were partially supported by the Russian Foundation for Fundamental Research through the Grant No. RFFR 00 15 97334 and by ISTC through the Grant No. 2207.

---

*Electronic address: sergey@condmat.theophys.kth.se

†Deceased.

[1] D. Belitz and T. R. Kirkpatrick, Rev. Mod. Phys. **66**, 261 (1999).

[2] B. I. Shklovskii and A. L. Efros, Zh. Eksp. Teor. Fiz. **81**, 406 (1981) [Sov. Phys. JETP **54**, 218 (1981)].

[3] B. I. Shklovskii and A. L. Efros, in *Electron-Electron Interactions in Disordered Systems*, edited by A. L. Efros and M. Pollak (Elsevier, New York, 1985).

[4] N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials*, 2nd ed. (Oxford University Press, Oxford, 1979).

[5] Mark Lee and M. L. Stutzmann, Phys. Rev. Lett. **87**, 056402 (2001).

[6] E. Helgren, N. P. Armitage, and G. Grüner, Phys. Rev. Lett. **89**, 246601 (2002).

[7] A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).

[8] S. D. Baranovskii, A. L. Efros, B. L. Gelmont, and B. I. Shklovskii, J. Phys. C **12**, 1023 (1979).

[9] J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. B **29**, 4260 (1984).

[10] A. Möbius, M. Richter, and B. Drittler, Phys. Rev. B **45**, 11568 (1992).

[11] S. A. Basylko, P. J. Kundrotas, V. A. Onischouk, E. E. Tornau, and A. Rosengren, Phys. Rev. B **63** 024201 (2000).

[12] Q. Li and P. Phillips, Phys. Rev. B **49**, 10269 (1994).

[13] A. Pérez-Garrido, M. Ortuño, E. Cuevas, J. Ruiz, and M. Pollak, Phys. Rev. B **55**, R8630 (1997).

[14] D. N. Tsigankov and A. L. Efros, Phys. Rev. Lett. **88**, 176602 (2002).

[15] A. Díaz-Sánchez, M. Ortuño, M. Pollak, A. Pérez-Garrido, and A. Möbius, Phys. Rev. B **59**, 910 (1999).

[16] R. N. Bhatt and T. M. Rice, Phys. Rev. B **23**, 1920 (1981).

[17] Mark Lee, J. G. Massey, V. L. Nguyen, and B. I. Shklovskii, Phys. Rev. B **60**, 1582 (1999).

[18] R. J. Deri and T. G. Castner, Phys. Rev. Lett. **57**, 134 (1986).

[19] M. Mochena and M. Pollak, Phys. Rev. Lett. **67**, 109 (1991); A. Díaz-Sánchez, A. Möbius, M. Ortuño, A. Neklioudov, and M. Schreiber, Phys. Rev. B **62**, 8030 (2000) and references therein.

[20] T. G. Castner, in *Hopping Transport in Solids*, edited by M. Pollak and B. Shklovskii (Elsevier, New York, 1991).

[21] E. Helgren, N. P. Armitage, and G. Grüner, Phys. Rev. B **69**, 014201 (2004).

[22] The repulsion energy of electrons on the same site is supposed to be much larger than the Coulomb energy at the mean distance between sites, which forbids double site occupation. Occupation numbers satisfy the electroneutrality condition $\Sigma_i n_i = KN$, where $K \leqslant 1$ is the compensation.

[23] On the entrance of M22, we have the condition $\varepsilon_i \leqslant \varepsilon_j$ satisfied for all $\{i, j : n_i = 0, n_j = 1\}$ and the condition (2) satisfied for all the pairs with $r_{ij} < r^*$ (or $1/r_{ij} > 1/r^*$). So the condition for a transition, $\delta H < 0$, takes the form $\varepsilon_i - \varepsilon_j - 1/r_{ij} < 0$ or $(\varepsilon_i - \mu) + (\mu - \varepsilon_j) < 1/r_{ij}$, where μ is defined in (3). Note, that by the definition of μ, $\varepsilon_i - \mu \geqslant 0$ and $\mu - \varepsilon_j \geqslant 0$, so to have $\delta H < 0$, we must demand $|\varepsilon_i - \mu| < 1/r_{ij}$ and $|\varepsilon_j - \mu| < 1/r_{ij}$. Since in subroutine M21 we have tested all the pairs with $r_{ij} < r^*$, we must search for a transition among the pairs with $r_{ij} \geqslant r^*$ (or $1/r_{ij} \leqslant 1/r^*$). So we can write $|\varepsilon_i - \mu| < 1/r_{ij} 1/r^*$ and $|\varepsilon_j - \mu| < 1/r_{ij} \leqslant 1/r^*$, which gives us the condition for the energies on the sites. On the exit of subroutine M22 we have a state in which condition (2) is satisfied for all pairs $\{i, j : n_i = 0, n_j = 1\}$. We have tested this directly for samples with the number of sites up to $N = 27000$.

[24] P. J. Kundrotas, V. A. Onischouk, and A. Rosengren, Phys. Lett. A **287**, 278 (2001).

[25] W. L. McMillan, Phys. Rev. B **24**, 2739 (1981).

[26] S. A. Basylko, V. A. Onischouk, and A. Rosengren, Phys. Lett. A **298**, 416 (2002).