Evidence for two-dimensional correlated hopping in arrays of Ge/Si quantum dots

A. I. Yakimov,* A. V. Dvurechenskii, A. V. Nenashev, and A. I. Nikiforov
Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia
(Received 27 July 2003; published 18 November 2003)

We report an analysis of a two-dimensional variable-range hopping conductance in doped and gated Ge/Si heterostructures with arrays of Ge quantum dots. We found that the conductivity σ versus temperature T follows the Efros-Shklovskii behavior $\sigma=\sigma_0\exp[-(T_0/T)^{1/2}]$ with the temperature-independent prefactor $\sigma_0 \sim e^2/h$. A strong reduction of the measured value of the characteristic temperature T_0 from that calculated for single-particle hopping was observed. The data provide evidence for interaction-driven many-electron excitations in dense arrays of quantum dots.

DOI: 10.1103/PhysRevB.68.205310 PACS number(s): 73.21.La, 73.40.Gk, 73.63.Kv

I. INTRODUCTION

Systems of randomly localized electronic states with long-range Coulomb interactions between the particles are often referred to as ''Coulomb glasses.'' A central issue of Coulomb glasses is the influence of electron-electron interactions on the transport properties. In general, the temperature dependence of the conductivity for variable-rangle hopping (VRH) is given by

$$\sigma(T)=\sigma_0(T)\exp[-(T_0/T)^x]. \tag{1}$$

VRH conductivity in the presence of long-range Hartree interactions between localized single-particle excitations obeys the Efros-Shklovskii (ES) law[1]

$$\sigma(T)=\sigma_0\exp[-(T_0/T)^{1/2}], \tag{2}$$

where

$$k_BT_0=Ce^2/(\kappa\xi) \tag{3}$$

is the characteristic interaction energy scale, C is a numerical coefficient that depends on dimensionality, k_B is the Boltzmann constant, κ is the relative permittivity of the host lattice, and ξ is the localization length of electrons. Within the mechanism of phonon-assisted VRH, the prefactor σ_0 takes the form $\sigma_0=\gamma/T^m$, where γ is a temperature-independent parameter and $m\sim1$.[2] The theoretical value of the constant C for single-particle hopping in two dimensions (2D) is $C \simeq 6$.[2,3]

Several authors have argued that under certain conditions dc VRH conduction can be dominated by many-particle Coulomb correlations between electronic transitions.[4,5] Sequential correlations appear when the hops of an electron facilitates the hopping probability of another electron due to rearrangement of the local potentials and/or site occupations in the vicinity of the initial and final states for tunneling process. There can be also interaction-driven simultaneous hopping of several electrons, resulting in a lowering of the energy configuration of the system. Because the formation of such many-particle dressed excitation provides screening of Coulomb interactions at large distances, the characteristic interaction energy in correlated hopping is reduced relative to its single-particle value.[6,7] Pérez-Garrido and co-workers[4] considered continuous paths through the space of many-charge configurations and showed that, in a regime of many-particle excitations, $\sigma(T)$ dependence has the ES form with numerical constant $C=0.6\pm0.2$; i.e., the parameter T_0 turns out to be about one order of magnitude smaller than Efros and Shklovskii's prediction for single-electron hopping. A reduction of hopping constants from the single-particle value in gated 2D GaAs/AlGaAs heterostructures has been observed in Ref. 8. Kozub, Baranovskii, and Shlimak,[5] assuming that interaction-assisted fluctuations of energies of hopping sites have spectral density $1/f$, demonstrated that sequential Coulomb correlations in a Coulomb glass can result in a phononless VRH with a temperature-independent universal prefactor $\sigma_0\approx e^2/h$ and a hopping exponent close to the ES value $x=1/2$.

Usually, gated doped semiconductors are exploited to look for many-particle correlations in 2D VRH. To drive the conductivity of the system, one is forced to change carrier concentration, thereby approaching inevitably the metal-insulator transition (MIT). However, since localization degrades the screening of electron-electron interactions, correlation effects should be particularly important far from the MIT on the insulator side. From this point of view, we believe that it is more reasonable to use dense arrays of quantum dots (QD's) for studying correlated hopping because, in such a case, it is possible to fix the QD density and vary only carrier wave functions by successive loading holes into different quantum states of the dots, being deep in the insulator phase. Furthermore, Ge/Si QD's are best suitable as the object of investigation, because (i) it is possible to obtain dense arrays with a QD density up to 10^{12} cm^{-2} (Ref. 9), in which the hopping transport of holes between QD's is a dominant mechanism of charge transfer at low temperatures; (ii) self-assembled QD's are all located at the same position in the growth direction—i.e., there is no disorder factor along the growth direction; (iii) as distinct from the majority of impurities in semiconductors, ensembles of QD's can serve as a system of multicharged localization centers, in which the role of Coulomb potential is especially important.

Previously, using an artificial screening provided by a metallic plane, parallel to a layer of Ge QD's in Si, we have proved that the VRH transport in arrays of self-assembled Ge/Si(001) QD's is strongly affected by long-range interactions.[10] In this paper, we examine in detail measure-

ments of 2D variable-range hopping conductance in Ge/Si heterostructures with a layer of Ge nanometer-scale QD's. The work spans a wide range of samples characterized by different QD densities, sizes, dot occupation with holes, and layer composition. The results obtained demonstrate the many-electron nature of dc transport in arrays of coupled QD's.

II. EXPERIMENT

We studied four series of Ge/Si heterostructures with self-assembled Ge quantum dots of pyramidal shape fabricated by molecular-beam epitaxy in the Stranskii-Krastanov growth mode.[11] All samples were grown on (001)-oriented Si substrates. Since the preparation details have been already published elsewhere,[10,12] we will make only a short description here.

Samples of series No. A were formed in Si substrates with a resistivity of 1000 Ω cm doped with boron up to a concentration of 10^{13} cm^{-3}. A Ge layer eight monolayers thick ($\sim$10 Å) was introduced into the middle of the 90-nm epitaxial Si layer. The average size of the dot base length is around 10 nm; the height is $\sim$1 nm. The areal density of the dots is $n_{QD} \approx 4\times10^{11}$ cm^{-2}. Their dimensions vary within a 20% range. In the samples of the next families Nos. B, C, and D, the thickness of the deposited Ge layer was ten monolayers, yielding Ge dots approximately 15 nm in diameter and 1.5 nm in height with $n_{QD} \approx 3\times10^{11}$ cm^{-2}.[13] The sequence of the bottom layers in series Nos. B and C was similar to that in samples No. A. In No. B, the thickness of the Si cap layer was 40 nm. In samples Nos. C, the array of Ge nanoislands was capped with a 10-nm Si layer and then by a top 25-nm-thick anodic SiO_2; the latter was fabricated to reduce permittivity of the media and thereby to change the long-range interactions.

To supply holes on the dots, a boron δ-doping Si layer inserted 5 nm below the Ge QD layer was grown. Because the ionization energy of boron impurities in Si is 45 meV and the energies of the first ten hole levels in Ge QD's of this size are 200–400 meV,[14] holes at low temperatures leave impurities and fill levels in QD's. After spatial transfer, the average number of holes per dot was varied from $N_h = 0.5$ to $N_h = 6.5$ by varying the doping.

Samples No. D were silicon metal-oxide-semiconductor field-effect transistors (MOSFET's) fabricated on a silicon-on-insulator wafer [p-type Si(001) substrate, 400 nm buried SiO_2 and 170 nm top Si] and containing about 3×10^7 Ge dots in a buried active channel. The average filling factor of the QD array was varied by the gate potential and can be determined from oscillations of the drain current as a function of gate voltage.[10]

Source and drain electrodes were made using Al evaporation and annealing at 450°C in a N_2 atmosphere. In all samples, the resistance along the QD layers was measured by a two-terminal method with a Keithley 6514 electrometer. The data were taken in the Ohmic regime and in the temperature range where condition $T \ll T_0$ is fulfilled.

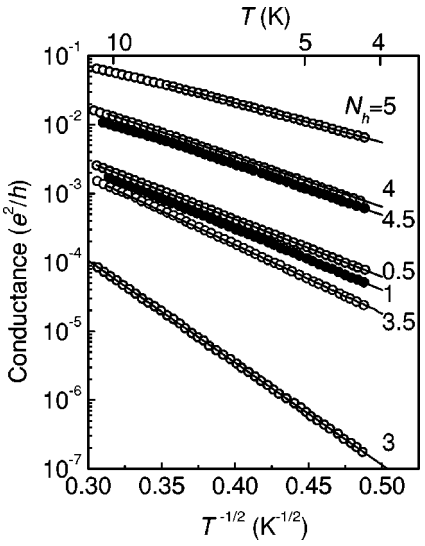

FIG. 1. The conductance $\sigma(T)$ vs $T^{-1/2}$ of samples No. A for different dot occupation. Symbols correspond to experimental points; solid lines are approximations of the experimental data using equation $\sigma(T) = \gamma/T^m \exp[-(T_0/T)^{1/2}]$. T_0, γ, and m are variable parameters.

III. RESULTS AND DISCUSSION

An example of experimental temperature dependences $\sigma(T)$ for samples Nos. A, C, and D is shown in Figs. 1 and 2. Conductance is given in units of e^2/h. In order to obtain detailed information on the functional form $\sigma(T)$, we used the differential method for an analysis of the temperature dependence of the reduced activation energy,[15] $w(T) = \partial \ln \sigma(T)/\partial \ln T = m + x(T_0/T)^x$. In this approach, if $m \ll x(T_0/T)^x$, then $\ln w(T) = A - x \ln T$, and $A = x \ln T_0 + \ln x$. Plotting $\ln w$ as a function of $\ln T$, one can find the hopping exponent x from the slope of the straight line. The parameter A can be found by the intersection point of the straight line with the ordinate axis, which gives the characteristic tem-

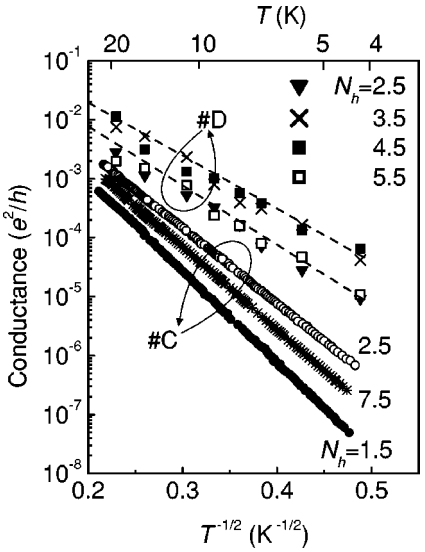

FIG. 2. Temperature dependence of VRH conductance of families Nos. C and D.

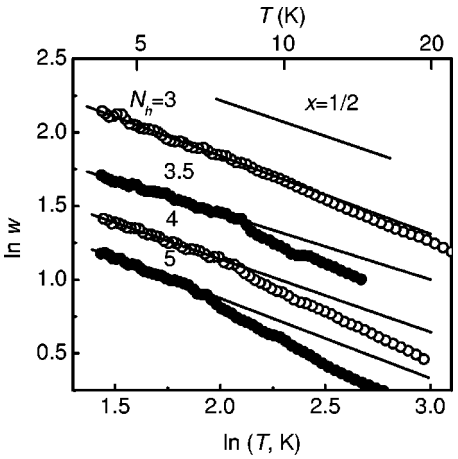

FIG. 3. Typical temperature dependence of the logarithmic derivative $w(T)=\partial \ln \sigma(T)/\partial \ln T$ (shown for samples No. A only) with different QD filling factor. The solid lines are least-squares fits to the linear dependence.

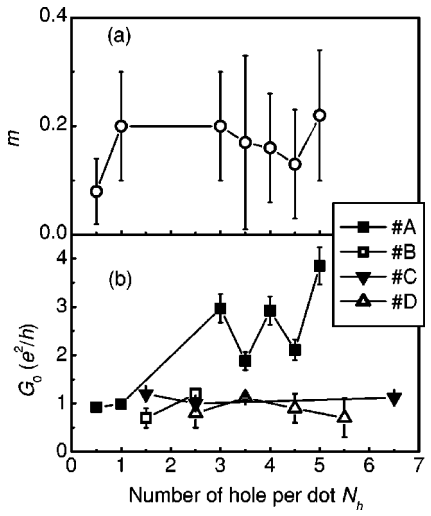

FIG. 4. Dependence of (a) the exponent m characterizing the temperature dependence of the conductivity prefactor σ_0 (shown for samples No. A only) and (b) the prefactor σ_0 (shown for all sample families) on the dot occupation with holes N_h.

perature $T_0=(10^A/x)^{1/x}$. Typical plots of $\ln w(T)$ versus $\ln T$ for several samples in series No. A are given in Fig. 3. At $T<10$ K, a linear relationship is observed between $\ln w(T)$ and $\ln T$, implying that $m \ll x(T_0/T)^x$ at these temperatures. The best fit of all the data gave values of the hopping exponent $x=0.51\pm0.05$. Similar behavior was found in samples Nos. B, C, and D.

Because it was already established that $x \simeq 0.5$, the method of nonlinear regression can be used for further determining the exponent m in the region of low temperatures. With this aim, the experimental data $\sigma(T)$ at $T<10$ K were approximated by the equation $\sigma(T)=\gamma/T^m \exp[-(T_0/T)^{1/2}]$, and the parameters γ, m, and T_0 were varied to obtain the best fit. In Fig. 1, the symbols are the experimental points and the solid lines are the least-squares fits to the ES equation. We found that m lies in the region 0.16 ± 0.09 [Fig. 4(a)]. Parameter m appears to be much less than the value predicted for phonon-assisted VRH ($m=1$). This means that the conductivity prefactor σ_0 depends weakly on temperature at low T and signals against the conventional phonon-assisted hopping mechanism.

The σ_0 data are presented in Fig. 4(b). An impressive feature is that the prefactor has a value close to e^2/h, the conductance quantum. Following the ideas of Ref. 5, we consider the universality of the prefactor as a manifestation of the 2D VRH conduction stimulated by the sequential Coulomb correlations.

To obtain further evidence for correlated VRH in arrays of Ge/Si QD's, it is necessary to measure the ES characteristic temperature T_0 and compare it with the theoretical predictions. In fact, since T_0 depends on the localization length ξ and hence on the electronic configuration of the occupied hole state in the dots, it is more convenient to find the universal constant C whose value is inversely proportional to intensity of the many-particle effects. It may be done taking into account the values of T_0 and ξ.

Asymptotic values of the hole localization length along 2D arrays of coupled Ge/Si quantum dots were obtained by computer modeling. The simulation was performed on a square lattice of 15×15 sites with the lattice constant $n_{\mathrm{QD}}^{-1/2}+\delta r$, where δr is a random value with a normal distribution. Only overlapping between nearest neighbors was included. We use the Hamiltonian

$$\hat{H}=\sum_{i,\alpha} E_{i,\alpha}\hat{a}^{\dagger}_{i,\alpha}\hat{a}_{i,\alpha}+\sum_{i,j,\alpha,\beta} J_{i,j,\alpha,\beta}\hat{a}^{\dagger}_{j,\beta}\hat{a}_{i,\alpha}, \qquad (4)$$

where index i counts the dots, index α denotes the hole bound-state number in QD's (we consider only nine bound states in each dot); $\hat{a}^{\dagger}_{i,\alpha}c$ ($\hat{a}_{i,\alpha}$) the creation (annihilation) operator for a hole in state α of the ith QD, $E_{i,\alpha}$ is the hole energy in this state, and $J_{i,j,\alpha,\beta}$ is the integral of overlapping between the αth state in the ith QD and βth state in the jth QD (elastic tunneling energy).

The random hole energies $E_{i,\alpha}$ were taken as the size-quantization energies in quantum dots whose dimensions are characterized by a Gaussian distribution with a mean-square deviation of 20%. The dependence of the energy levels on dot size has been calculated previously using the sp^3 tight-binding model with inclusion of the spin-orbit interaction and deformation effects.[14]

The following procedure was used to determine the overlap integrals. We calculated the energies of hole states in a model structure containing Ge quantum dots inside a Si parallelepiped. Periodic boundary conditions

$$\psi(-d/2,y,z)=\psi(d/2,y,z) \qquad (5a)$$

or

$$\psi(-d/2,y,z)=-\psi(d/2,y,z), \qquad (5b)$$

where d is the parallelepiped size in the x direction and $\psi(x,y,z)$ the hole wave function, were considered. The same boundary conditions were used for the y and z directions.

TABLE I. Asymptotic values of the localization length (in nanometer units) of holes confined in array of Ge/Si QD's in the ground, first, and second excited states.

Sample family	Ground state	First excited state	Second excited state
A	2.4	2.8	–
B, C, and D	2.1	2.2	2.3

The overlap integral in the plane of the QD array in the x direction was defined as $J(d)=|E_+-E_-|/4$, where E_+ and E_- are the hole energies corresponding to the boundary conditions (5). The obtained dependence $J(d)$ can be rewritten in the form

$$J(d)=A_\alpha \exp(-B_\alpha d), \tag{6}$$

where coefficients A_α and B_α depend on the energy level number α, and B_α equals to the inverse localization length of a hole in α state of isolated QD's. Integrals $J_{i,j,\alpha,\beta}$ were determined as the geometrical mean of overlap integrals between α and β states:

$$J_{i,j,\alpha,\beta}=\sqrt{A_\alpha A_\beta}\exp\left(-\frac{B_\alpha+B_\beta}{2}d_{ij}\right), \tag{7}$$

where $d_{i,j}$ is the distance between QD's. The simulation was carried out using 5000 random realizations of the QD array with filling factor 1/2 for the ground state or for the first two excited states in the dots. For each realization, we calculated probability p_i of holes to occupy the corresponding state in each dot. The probability value was then approximated by the equation $p_i=a\exp(-2d_i/\xi_r)$, where d_i is the distance between the ith QD and dot with maximum local hole wave function amplitude. The localization length ξ was obtained by averaging ξ_r through all array realizations and is listed in Table I.

It now remains to determine the magnitude of C using the equation $C=k_BT_0/(e^2/\kappa\xi)$ and experimental values of T_0. These results are presented in Fig. 5. The value of the hopping constant turns out to be $C=0.9\pm0.4$. This implies that the characteristic interaction temperature T_0 is considerably smaller than the value from simulations of single-electron transport, which provides an additional argument in favor of correlated hopping in 2D arrays of Ge/Si quantum dots.

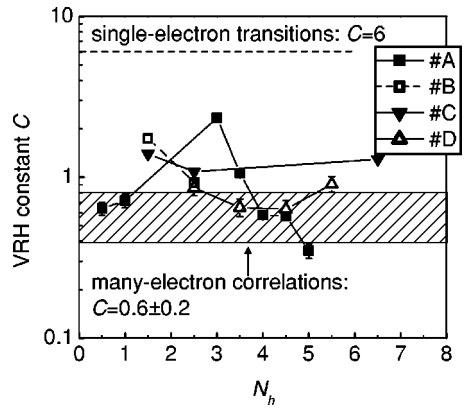

FIG. 5. Dependence of the numerical parameter C associated with variable-range hopping in arrays of Ge/Si quantum dots on the average number of holes in QD's. A crosshatched region corresponds to the prediction of the many-electron model (Ref. 4). The dashed line gives the value of C for single-particle hopping (Ref. 3).

IV. SUMMARY

In summary, we have analyzed the variable-range hopping transport of holes in Ge/Si self-assembled quantum dots. The conductivity prefactor has the temperature-independent value of around the conductance quantum e^2/h. The hopping constant is found to be 0.9 ± 0.4. These features are inconsistent with a model of single-particle hopping and provide experimental evidence for many-electron correlations in the dc transport of two-dimensional arrays of coupled quantum dots.

ACKNOWLEDGMENTS

The authors are much obliged to A. A. Bloshkin for experimental assistance and M. N. Timonova for the development of a simulation procedure. The work was supported in part by the Russian Foundation of Basic Research (Grant No. 03-02-16526). A.Y. acknowledges financial support from the State program for support of young Russian doctors of sciences (Grant No. MD-28.2003.02). A.V.N. is also grateful to Lavrentev Foundation (Grant No. 27).

*Electronic address: yakimov@isp.nsc.ru

[1] A.L. Efros and B.I. Shklovskii, J. Phys. C **8**, L49 (1975).
[2] D.N. Tsigankov and A.L. Efros, Phys. Rev. Lett. **88**, 176602 (2002).
[3] V.L. Nguen, Fiz. Tekn. Poluprovodn. **18**, 335 (1984) [Sov. Phys. Semicond. **18**, 207 (1984)].
[4] A. Pérez-Garrido, M. Ortuño, E. Cuevas, and J. Ruiz, Phys. Rev. B **55**, R8630 (1997).
[5] V.I. Kozub, S.D. Baranovskii, and I.S. Shlimak, Solid State Commun. **113**, 587 (2000).
[6] A.G. Zabrodskii and A.G. Andreev, Pis'ma Zh. Éksp. Teor. Fiz. **58**, 809 (1993) [JETP Lett. **58**, 756 (1993)].
[7] M. Lee and J.G. Massey, Phys. Status Solidi B **205**, 25 (1998).
[8] F.W. Van Keuls and A.J. Dahm, Phys. Status Solidi B **205**, 21 (1998).
[9] A.I. Yakimov, A.V. Dvurechenskii, A.I. Nikiforov, V.V. Ulyanov, A.G. Milekhin, A.O. Govorov, S. Schulze, and D.R.T. Zahn, Phys. Rev. B **67**, 125318 (2003).
[10] A.I. Yakimov, A.V. Dvurechenskii, V.V. Kirienko, Yu.I. Yakovlev, A.I. Nikiforov, and C.J. Adkins, Phys. Rev. B **61**, 10 868 (2000).
[11] O.P. Pchelyakov, Yu.B. Bolhovityanov, A.V. Dvurechenskii, A.I.

Nikiforov, A.I. Yakimov, and B. Voigtländer, Thin Solid Films **367**, 75 (2000).

[12] A.I. Yakimov, A.V. Dvurechenskii, A.I. Nikiforov, and A.A. Bloshkin, Pis'ma Zh. Éksp. Teor. Fiz. **77**, 445 (2003) [JETP Lett. **77**, 376 (2003)].

[13] A.I. Yakimov, A.V. Dvurechenskii, Yu. Proskuryakov, A.I. Nikiforov, O.P. Pchelyakov, S.A. Teys, and A.K. Gutakovskii, Appl. Phys. Lett. **75**, 1413 (1999).

[14] A.V. Dvurechenskii, A.V. Nenashev, and A.I. Yakimov, Nanotechnology **13**, 75 (2002).

[15] A.G. Zabrodskii and K.N. Zinoveva, Zh. Éksp. Teor. Fiz. **86**, 727 (1984) [Sov. Phys. JETP **59**, 425 (1984)].