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Non-equilibrium transport in arrays of type-II Ge/Si quantum dots

N.P. Stepina[*], **A.I. Yakimov, A.V. Dvurechenskii, A.V. Nenashev,** and **A.I. Nikiforov**

Institute of Semiconductor Physics, prospekt Lavrenteva13, 630090 Novosibirsk, Russia

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We study the effect of interband light excitation on hopping conductivity in dense arrays of Ge/Si quantum dots. Both negative and positive photoeffects depending on dot occupations with holes were observed. Long-time conductivity dynamics (typically, $10^2 - 10^4$ sec at T=4.2 K) has been revealed as well as after switch on and switch off the illumination, displaying a sluggish temporal dependence. Our observation is explained by spatial separation of electrons and holes due to the presence of potential barriers created by positively charged Ge quantum dots. The time-dependent equalization of barrier heights as a result of hole trapping into the dots was proposed as an additional effect explaining persistent photoconductivity.

1 Introduction Ge/Si (001) quantum dots (QDs) structures exhibit a type-II band lineup. The large (~0.7 eV) valence-band offset characteristic of this heterojunction leads to an effective localization of holes in Ge regions. It has been shown [1, 2] that at low temperatures (<20K) charge transport through such a system is dominated by hole hopping between the dots. The Hartree potential of holes induces a quantum well for free electrons of Si at Ge/Si interface. Thus a fundamental feature of these dots is that trapping potential for electrons would be greatly enhanced with charging the dots with holes. The spatial separation of electron and hole in Ge/Si QDs causes many exciting phenomena, such as negative interband photoconductance [3], anomalous quantum-confined Stark effect [4], blue shift of the interband transition with formation of exciton complexes [5]. In this paper we describe experimental results that demonstrate anomalous dynamics of hopping photoconductivity (PC) created by exposure to interband illumination.

2 Experimental details The samples were grown on a (001) oriented Si substrate with a resistivity of 1000 Ω cm by molecular-beam epitaxy in the Stranskii-Krastanov growth mode. To supply holes on the dots, a boron δ-doping Si layer inserted 5 nm below the Ge QD layer was grown. Si cap and buffer layers were boron doped at a level of ~10^{16} cm^{-3}. Details of sample preparation have been described elsewhere [1]. To avoid influence of surface effects on photoconductivity measurements, Ohmic contacts were fabricated by Al evaporation into the preliminary etched pits followed by annealing at 400 °C in N_2 atmosphere. The etching depth was such as the electrical contacts to buried Ge QD layer are formed. To separate response from the dots, the test samples were grown under conditions similar to the dot samples, except that no Ge was deposited. Photoconductivity experiments have been carried out using a GaAs light-emitting diode (LED) with an emission maximum at a wavelength of 0.9 μm for excitation.

[*] Corresponding author: e-mail: stepina@isp.nsc.ru, Phone: +7 3832 33 26 24, Fax: +7 3832 33 26 24

3 Experimental results Hopping transport of holes along two-dimensional layers of Ge quantum dots in Si have been investigated in [1, 2]. From the temperature dependence of conductivity the mechanism of charge transfer was identified as being variable-range hopping in a density of state determined by long-range Coulomb interaction between the dots. Interband illumination of such a system results in a complicated transient behavior of the photoconductivity during reaching illuminated or dark steady state. Our experiments involve the following procedure. The samples are cooled to the measuring temperature with a zero drain voltage, and are allowed to equilibrate for several hours. The time dependence of conductance under the condition of LED illumination (on) and stopping illumination (off) was taken several times serially. Figure 1 a shows typical conductance transient traces for two structures with different δ-boron doping. In contrast to the test Si samples, in which the PC is always positive, both positive and negative photoeffects depending on dot filling factor are observed in dot samples. In both cases, kinetics of the recovery as well as of the decay are extremely slow. When the LED illuminates on the sample, the resistivity changes rapidly at the beginning and slowly at the end, while the residual photoconductance can persist for several hours after the light switch off. This phenomenon characterizes a typical effect of persistent photoconductivity (PPC). The PPC level is about 90% of the initial PC value after more than 5000 seconds of decay. Long-time relaxation process was not observed in test samples containing no dots (Fig. 1b).

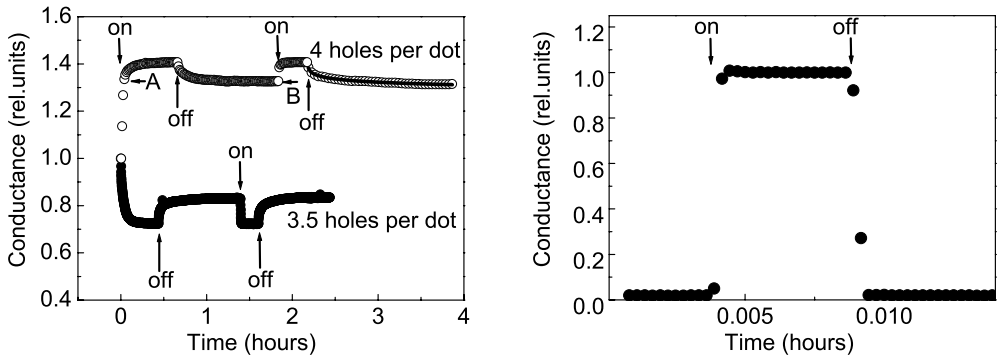

Fig. 1 a) Conductivity traces for the samples with different average numbers of holes per one Ge dot. The solid line is the fitting to the experimental data by a logarithmic law. b) Photoconductivity for the Si sample without Ge quantum dots.

Transient curves for the samples with positive photoconductance after the first light switch on are shown in Figure 2 a for different light intensities. All curves were normalized to conductivity values in saturation ($G_{\max}$). The negative photoconductivity behaviour is similar to sign that for the positive one. One can see that the rate of PC changing at the initial part of the curves increases with intensity of illumination. Figure 3 shows the conductance decay traces at different temperatures and demonstrates the quenching of the PPC effect at high temperatures. Experimental data have been normalized to unity at $t = 0$ (at a moment when the illumination is terminated) according to $G_{PPC}(t) = (G(t) - G_d)/(G(0) - G_d)$. Here, $G(0)$ is the conductance level immediately after the termination of the excitation source, G_d is the initial conductance in the dark.

4 Discussion To explain the experimental observations we propose the following model. Quantum dots occupied with holes induce a band bending which corresponds to a potential barrier for free holes and a potential well for electrons. When an electron-hole pair is photoexcited, electron is trapped by a dot while the hole cannot recombine with this electron because of the QD's repulsive potential. Recombination of electrons with equilibrium holes in Ge nanoclusters reduces the potential barrier for hole capture. Thus, under illumination, the hole trapping into QDs occurs in a condition of permanent decreasing of the potential barrier height. Changing of hole concentration in the dots can be described by

$$\frac{dp}{dt} = -J_e - C_{em} + C_{cap}^0 \exp\left(-\frac{V}{kT}\right), \tag{1}$$

where J_e is the electron flux into the dots, V is the barrier height, C_{em} and C_{cap} are the emission and capture rates of holes in the dots, correspondently. Since the equilibrium state, in which the hole emission and capture rates are equal, cannot be reached over the time-window of experiment, one may neglect the emission term in this equation. If concentration of non-equilibrium holes Δp in QDs is small

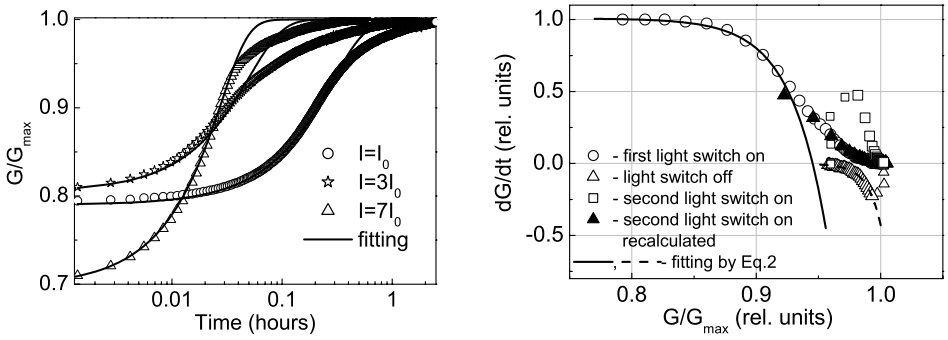

Fig. 2 a) Light intensity dependence of photoconductivity buildup. Solid lines are the fit to the experimental data by numerical solution of Eq. (2). I is the light intensity. b) Derivative of experimental relaxation curve as a function of conductivity.

enough, then the change in the barrier height and conductivity would be proportional to Δp. In this case, expression for conductivity takes the form

$$\frac{dG}{dt} = AI + B\exp(-\gamma G), \tag{2}$$

where *I* is the light intensity, *A*, *B* and γ are the constants. Oscillating behaviour of conductivity with dot occupancies was found in [1]. Then the different sign of photoconductivity observed in our experiment can be due to the change of hole number in dots under interband illumination. Solid lines in Fig. 2 a depict the fit to the experimental data by a numerical solution of Eq. (2). The initial part of the curves is described satisfactorily by Eq. (2), while the further conductivity growth becomes slower than the calculated one. In Fig. 2 b, we show derivative of the experimental relaxation curve *dG/dt* as a function of *G*. The exponential behavior characterized by Eq. (2) (solid line) is really violated when the conductivity reaches the value $G \approx 0.93 G_{\max}$. Discrepancy between the predicted and experimental data was proposed to be due to the alignment of barrier heights which, in turn, is a result of photohole capture into the dots under illumination. Nonuniformity of dot sizes yields the dispersion of the barrier heights around them; the effective barrier height (EBH) corresponds to a barrier with minimum value. During the capture process, holes enter the dots with predominately smallest positive charge, being resulted in equalization of the dot occupation. Obviously, for the uniform hole distribution through the dots, the EBH is in excess of that for the equilibrium state. Increasing of the effective barrier height causes the conductivity to rise slowly.

After the light switch off, holes continue to be captured by QDs and the barrier height steadily rises. Then the conductivity obeys the following equation:

$$\frac{dG}{dt} = B\exp(-\gamma G). \tag{3}$$

It has an analytical solution $G(t) = G_0 + C\ln(t - t_0)$, where G_0, C and t_0 are the constants. One can see in Fig. 1 a that the experimental data are well described by logarithmic law (solid line in Fig. 1 a) that corresponds to exponential dependence of *dG/dt* on *G* depicted by open triangles in Fig. 2 b. Dash line in Fig. 2 b indicates fitting to the experimental data by Eq. (3).

New conductive state resulting from the relaxation of the system after light termination differs from the initial one, because the PPC is realized at low temperature when the hole trapping into the dots is strongly limited. Moreover, this state as well as the stationary state under illumination is characterized by uniform distribution of holes through the dots. The second switching on of light takes place when the barrier heights are equal with their absolute value being lower then after the first illumination. Thus the point A in Fig. 1 corresponds to the barrier height larger than that at point B. If it is the case, we should get a coincidence of derivative curves depicted in Fig. 2 b for the first and second light switching on by extending the latter curve toward the values characterized by lower barrier height. It can be seen that after such a transformation (filled triangles in Fig. 2 b) these curves are really superposed.

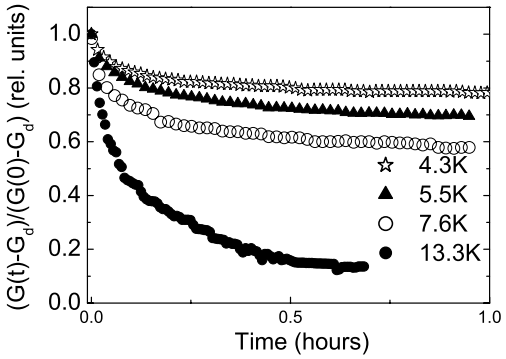

Fig. 3 PC decay at different temperatures.

Increase of the temperature accelerates the hole capture by dots results in the quenching of PPC effect (Fig. 3), that give further evidence to support the proposed model.

5 Summary Non-exponential slow kinetics of photoconductivity excitation and effect of persistent photoconductivity have been observed in *p*-type Ge/Si heterostructures containing Ge quantum dots. The essential points for the explanation of the experimental data are proposed to be (i) dependence of the hole trapping into the quantum dots on QD charge state, and (ii) equalization of the number of holes per each dot in a QD ensemble during the photoconductivity excitation and relaxation.

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