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Negative Interband Photoconductivity in Ge/Si Heterostructures with Quantum Dots of the Second Type

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It was found that irradiating an array of Ge nanoclusters in *n*-Si with light that induced interband transitions gave rise to negative photoconductivity. This result was explained by localization of equilibrium electrons at the Si/Ge interface in the potential of the nonequilibrium holes trapped on deep states in Ge islands. © *2000 MAIK "Nauka/Interperiodica".*

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Interest in studying photoelectric properties of assemblies of nanosized atomic clusters (quantum dots, QDs) obtained during the growth of mismatched heteroepitaxial systems via a Stranski–Krastanov process is due to new possibilities of creating efficient light-emitting devices and photodetectors [1, 2]. Arrays of Ge islands grown on Si(001) are a typical example of such a system.

Strained Ge/Si(001) layers represent heterostructures of the second type. The characteristic properties of systems of this class are determined by the relative positions of the band edges of the source semiconductors at the interface. The conduction band of one of the materials (Si) here is close to the valence band of the other (Ge), which causes spatial separation of carriers. In this case, holes are localized in Ge quantum dots, whereas there is a potential barrier for electrons in Ge. However, the positive charge generated by holes and the nonuniform structural deformation of the silicon islands surrounding Ge because of the mismatch of the Ge and Si lattice constants may give rise to potential wells for electrons at the Si/Ge interface. Self-consistent calculations of the energy spectrum of electrons and holes were carried out in [3] within the effective mass approximation for pyramidal Ge nanoclusters with a base length of 15 nm and a height of 1.5 nm. The calculations showed that the localization energy of holes in QDs is $E_{lh} \approx 400$ meV and the localization energy of electrons in Si at QDs is $E_{le} \approx 40$–70 meV (depending on the number of holes in the dot). The contribution to E_{le} from the deformation of the silicon matrix comprises only a small part (7 meV [3]); therefore, electrons can be localized at a QD at sufficiently high temperatures only when the QD is charged by a hole. In our opinion, in structures with *n*-type conductivity, this may lead to such a rare phenomenon as negative photoconductivity (PC). In the majority of real cases, the conductivity increases upon illumination (positive PC), because band-to-band illumination increases the concentration of free charge carriers.

Consider *n*-type silicon in which undoped Ge nanoclusters have been introduced (Fig. 1a). The dark conductivity of the system is determined by the free electrons in the Si conduction band. Upon absorption of light that induces interband transitions and generates electron–hole pairs, holes will start to accumulate in Ge QDs, charging them positively. As a result, potential wells for electrons will arise at the Si/Ge interfaces in silicon. If $E_{le} > E_D$, where E_D is the ionization energy of donor impurities in Si, these potential wells will behave like photoinduced electron attachment centers (Fig. 1b). The localization of electrons on these centers will decrease the concentration of mobile carriers and reduce the conductivity of the system.

To verify the above considerations, we investigated the photoconductivity of the Ge/Si epitaxial structure grown on a Si(001) substrate with the phosphorus concentration $N_s \approx 5 \times 10^{15}$ cm^{-3}. The substrate thickness was $L_s = 300$ μm. The structure consisted of ten layers of Ge islands separated by Si interlayers 30 nm thick. The total thickness of the epitaxial layer was $L_{epi} \approx 0.3$ μm. The concentration of donors (Sb) in the epitaxial Si layer was $N_{epi} \approx 2.5 \times 10^{16}$ cm^{-3}. The Ge nanoclusters were of a pyramidal shape with a base length of 15 nm and a height of 1.5 nm [4]. The layer density of nanoclusters in each layer was $n_{QD} \approx 3 \times 10^{11}$ cm^{-2}. A GaAs light-emitting diode with an emission maximum at a wavelength of ~ 0.9 μm was used as a light source. The emission intensity of the light-emitting diode was modulated with a frequency of 2 kHz. The photoconductivity was measured at the modulation frequency.

The dependences of the relative PC $\Delta G/G$ on the illumination power P for a Ge/Si structure with quan-

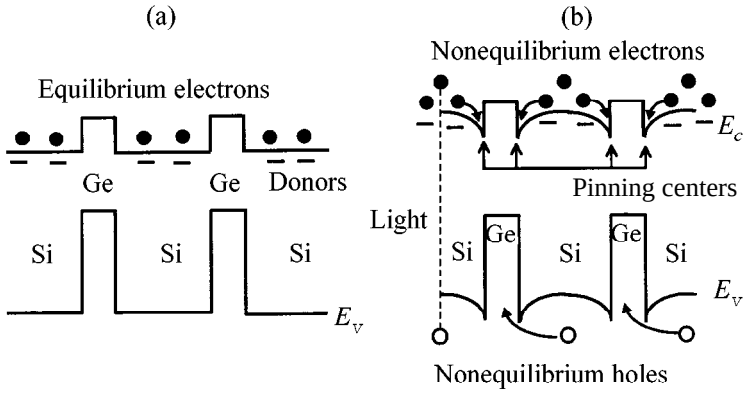

Fig. 1. Band structure of a multilayer *n*-type Ge/Si heterostructure with Ge quantum dots (a) in the absence of light and (b) under illumination. The figure illustrates the mechanism of negative interband photoconductivity (see text).

tum dots at various temperatures are shown in Fig. 2. The lux–ampere characteristic at $T = 77$ K for a sample containing no epitaxial layer with islands is also presented there (dashed line). The photoconductivity in the sample containing no quantum dots is positive and increases almost linearly with increasing P. The structure containing Ge nanoclusters exhibits negative PC in the range of illumination intensities $P = 0$–100 mW/cm^2. The effect of a decrease in photoconductivity upon illumination is suppressed with an increase in temperature, because the thermal energy kT at high T becomes of the order of the localization energy of electrons E_{le} and most of the electrons are no longer confined by the positive potential of the holes.

As the illumination intensity increases, a transition to positive PC is observed (at $P > 100$ mW/cm^2). We associate the change of the PC sign with the saturation of the number of electrons that can be localized in the potential of quantum dots charged by holes. The calculations given in [3] demonstrate that the third or the fourth electron is already displaced to the region of conduction-band delocalized states through the Coulomb interaction with the trapped electrons. Optical generation directly in Ge cannot markedly affect the filling of islands because of the small thickness of Ge. Note, however, that the diffusion length of nonequilibrium carriers in Si (100–1000 μm [5]) is much larger than the light penetration depth (~10 μm). Therefore, virtually all the photoexcited holes and electrons in Si can be trapped, diffusing from the bulk, on the bound states in the region of QDs. Then, taking into account that the interband recombination times of electrons and holes in Si and Ge nanoclusters are much the same [3], we obtain the average number of nonequilibrium holes and electrons per island $N_h = P\tau/h\nu n_{QD}N$, where τ is the interband recombination time, $h\nu$ is the photon energy, and N is the number of QD layers. For $\tau \sim 15$ μs, $h\nu = 1.4$ eV, $n_{QD} \approx 3 \times 10^{11}$ cm^{-2}, and $N = 10$, we actually obtain the degree of filling $N_h = 2.5$. This means that the potential wells in the silicon conduction band are already filled with electrons at an illumination intensity of ~ 100 mW/cm^2. Additional generation of nonequilibrium electrons will lead to growth of the concentration of free carriers. Therefore, the PC has a positive sign.

If the maximum number of bound electron states at each island equals two, a minimum of the photoconductivity is reached when each QD contains one nonequilibrium hole. In this case, both equilibrium and nonequilibrium electrons can be confined at a QD, thus making no contribution to the conductivity.

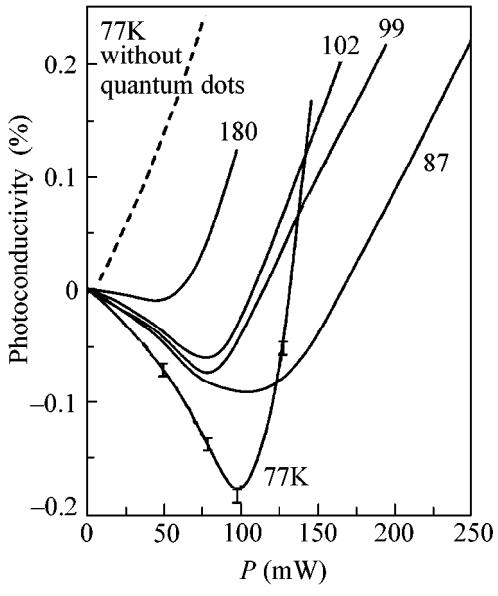

Fig. 2. Dependence of the relative photoconductivity on the interband illumination intensity in *n*-type Si with Ge quantum dots and in a sample without Ge nanoclusters at various temperatures. The light source is a GaAs light-emitting diode whose emission maximum corresponds to a photon wavelength of ~0.9 μm.

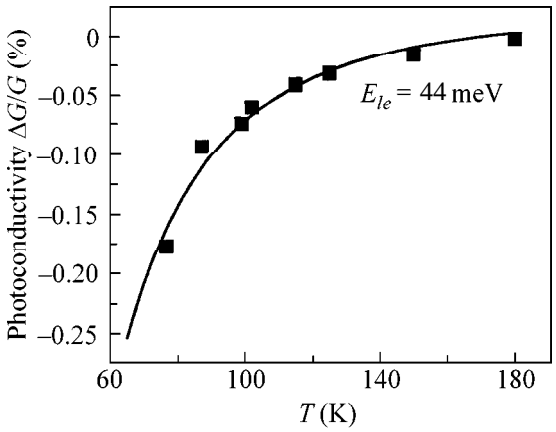

Fig. 3. Temperature dependence of the photoconductivity minimum. The solid line shows the result of approximating the experimental data (points) by Eq. (4) for the localization energy of electrons on the photoinduced attachment center E_{le} = 44 meV.

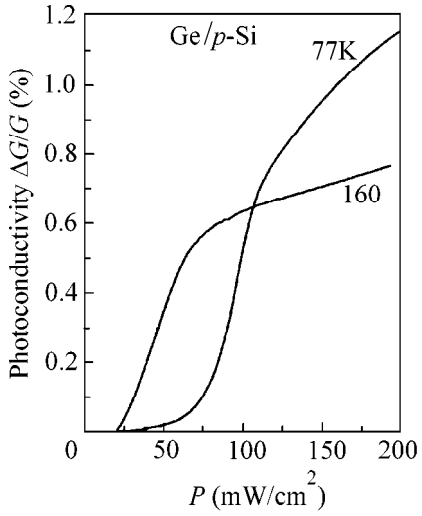

Fig. 4. Dependence of the relative photoconductivity on the interband illumination intensity in *p*-type Si with Ge quantum dots.

The expected value of negative PC can be estimated within a simple model. When photoinduced attachment centers appear at the interface of positively charged QDs, free electrons, on leaving Si, will start to quit the conduction process. Because the relationship $Nn_{\mathrm{QD}} > N_{epi}L_{epi}$ is fulfilled at the given parameters of the epitaxial Si layer (impurity concentration and thickness), part of the electrons will go out of the substrate to the layer with islands, leaving there a region of space charge of ionized donors with the width *w*. The value of *w* can be found from the electroneutrality condition

$$Nn_{\mathrm{QD}} = N_{epi}L_{epi} + N_s w. \tag{1}$$

The layer conductivity in the absence of illumination is determined by the equation

$$\begin{aligned} G &= e\mu_s N_s L_s \exp(-E_{\mathrm{P}}/kT) \\ &+ e\mu_{epi} N_{epi} L_{epi} \exp(-E_{\mathrm{Sb}}/kT), \end{aligned} \tag{2}$$

where μ_s is the electron mobility in the substrate, μ_{epi} is the electron mobility in the epitaxial Si layer, E_{P} is the ionization energy of the phosphorus level in the substrate, and E_{Sb} is the ionization energy of the antimony level in the epitaxial layer. The layer conductivity under illumination takes the form

$$\begin{aligned} G_{ph} &= e\mu_s N_s (L_s - w) \exp(-E_{\mathrm{P}}/kT) \\ &+ e\mu_{\mathrm{QD}} N n_{\mathrm{QD}} \exp(-E_{le}/kT), \end{aligned} \tag{3}$$

where μ_{QD} is the electron mobility at the Si/Ge interface. Assume for the estimations that all the mobilities μ_s, μ_{epi}, and μ_{QD} are equal to each other. Then the relative value of PC can be obtained from Eqs. (1), (2), and (3):

$$\begin{aligned} \frac{\Delta G}{G} &= -\left\{1 + \frac{Nn_{\mathrm{QD}}}{N_{epi}L_{epi}}\left[\exp\left(-\frac{E_{\mathrm{P}} - E_{\mathrm{Sb}}}{kT}\right)\right.\right. \\ &\left.\left. - \exp\left(-\frac{E_{le} - E_{\mathrm{Sb}}}{kT}\right)\right] - \exp\left(-\frac{E_{\mathrm{P}} - E_{\mathrm{Sb}}}{kT}\right)\right\} \\ &\times \left\{1 + \frac{N_s L_s}{N_{epi}L_{epi}} \exp\left(-\frac{E_{\mathrm{P}} - E_{\mathrm{Sb}}}{kT}\right)\right\}^{-1}. \end{aligned} \tag{4}$$

The points in Fig. 3 show the experimental temperature dependence of the minimum of the relative photoconductivity. The calculated curve obtained using Eq. (4) is also drawn there as a solid line. The electron energy on the attachment centers E_{le} was the only fitting parameter. For E_{P} = 45.2 meV and E_{Sb} = 42.5 meV [6], the least deviation of the calculated curve from the experimental data is reached at E_{le} = 44 meV. The self-consistent calculations [3] give the following values of E_{le} for various variants of QD filling: 38 meV for one electron in the potential of one hole, 70 meV for one electron in the potential of two holes, and 50 meV for the first of two electrons in the field of two holes. The experimental energy E_{le} falls within the same range and has quite a reasonable value. It is evident in Fig. 3 that the simple model reproduces well both the value and the temperature dependence of negative photoconductivity. However, the temperature and concentration dependence of the mobilities must be correctly taken into account for obtaining results that are more accurate.

As noted at the beginning, the appearance of an electron-localizing potential when holes are trapped in QDs is the crucial factor in the occurrence of negative PC. It is clear that illumination should not lead to a decrease in the concentration of equilibrium carriers in

the *p*-type structures. This is because the occurrence of a potential well in the Ge valence band is mainly determined by the discontinuity between the Ge and Si valence bands. However, the Coulomb interaction energy of an electron and a hole bound into a spatially indirect exciton (≈25 meV [3]) is much lower than the localization energy of a hole (~400 meV in the ground state).

The lux–ampere characteristics of a Ge/*p*-Si structure are presented in Fig. 4. The sample was grown under conditions similar to those for the synthesis of the Ge/*n*-Si sample, except for the conductivity type of the substrate and the upper epitaxial Si layer with Ge islands (the concentration of the acceptor boron impurity was $\approx 10^{15}$ cm^{-3} in the substrate and $\approx 10^{16}$ cm^{-3} in the epitaxial Si layer). It is clear that the region with negative PC is absent in the *p*-type structure. Instead, an extended region with low positive PC is observed, after which a step increase in the photocurrent occurs. This behavior at low illumination intensities is due to trapping of nonequilibrium carriers of both types on the bound states in the vicinity of Ge islands (holes on the states in Ge islands and electrons on the states at the Si/Ge interface). The growth of PC at high intensities is explained by filling of the electron levels and the appearance of free nonequilibrium electrons.

We emphasize in conclusion that the mechanism of negative photoconductivity proposed in this work is accomplished only in quantum dots of the second type, because both electrons and holes in heterostructures of the first type are localized in the same semiconductor independently of the charge state of QDs.

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