08

Charge carrier localization in InAs self-organized quantum dots

© *A.N. Kosarev*[1,2], *V.V. Chaldyshev*[1,¶]

[1] Ioffe Institute, St. Petersburg, Russia
[2] Peter the Great Saint-Petersburg Polytechnic University, St. Petersburg, Russia
E-mail: kosarev@mail.ioffe.ru
¶ E-mail: chald.gvg@mail.ioffe.ru

Received August 30, 2021
Revised September 7, 2021
Accepted September 8, 2021

We considered the problem of localization of electrons and holes taking for instance the pyramidal InAs quantum dots in GaAs. The problem of quantum mechanics was solved for the localizing potential taking into account the geometry, chemical composition and built-in fields of the mechanical stress and strain. We found that the strongest localization of both types of charge carriers can be achieved if the ratio of the pyramid height to its base is about 0.2.

Keywords: quantum dots, elastic strain, charge carrier localization.

DOI: 10.21883/TPL.2022.15.55280.19006

The depth and nature of the localization of electrons and holes are the main parameters of quantum dots (QDs), which determine their electronic and optical properties and the possibility of their use in various electronic, optoelectronic and photonic devices [1,2]. Despite the significant difference in the band gaps of the material from which the QD is formed (self-organized) and the material of the surrounding barrier layer, the localization of charge carriers in the QD often turns out to be weak or even impossible. The reason for this is mechanical deformations, the presence of which is necessary for the process of self-organization according to the Stranski–Krastanov mechanism [3,4]. Glaring example of this phenomenon are InAs QDs, the technology of obtaining (self-organization) of which in the process of molecular beam epitaxy is well developed [5]. Since the band gap of InAs is 0.4 eV, and the band gap of the GaAs barrier is 1.5 eV, InAs QDs could potentially cover a very wide and practically important near-infrared optical range, including the transparency window of an optical fiber near $1.55\,\mu$m (0.8 eV), commonly used in optical communication lines. In reality, radiation from InAs QD takes place in the wavelength range $0.8-1.1\,\mu$m, increasing with the size of the QD [6].

In this work, the localization problem of electrons and holes on the example of pyramidal InAs QDs in gallium arsenide, is considered. We found that, for a given QD volume, the localization depth of both types of carriers has a nontrivial dependence on the ratio of the height of the pyramid to its base.

We performed calculations for typical InAs QDs obtained by molecular-beam epitaxy on (001) GaAs substrates in the Stranski–Krastanov mode with the growth of five InAs monolayers. Pyramidal InAs QDs formed on the growth surface were overgrown by a 30 nm thick GaAs barrier layer. The procedure for growing and forming the QD system is described in reference [7]. The results of detailed electron microscopic studies of such QDs are presented in reference [8]. These studies, in particular, showed that the typical height of InAs QDs overgrown by in GaAs is $H = 9.0 \pm 0.5$ nm, and the base of the pyramid $L = 28.0 \pm 0.5$ nm. In this case, the edges of the base of the pyramid were parallel [100] and [010]. The faces of the pyramid corresponded to the Miller indices {203}. Such dimensions and shape of QD were chosen by us as reference ones. It should be noted that overgrowth of QD leads to a decrease in the ratio of the height of the pyramid to its base. This effect is typical for QDs formed by self-organization on the surface [9,10]. The manifestation of this effect significantly depends on the overgrowth conditions. In addition to changing the size ratio, the overgrowth can also lead to In–Ga mixing. For the reference QD, we considered the indium concentration as homogeneous and equal to 0.92, which ensured the agreement between the energy level calculations and the observed energy position of the photoluminescence line from the QD.

The approach used in the calculations is described in our work [11]. To determine the localization depth of electrons and holes and then calculate the spectral position of the exciton radiation peak, the problem of quantum mechanics was solved numerically for both types of carriers in a single-zone model. In this case, the localizing potential was obtained as a result of solving the problem of the linear theory of elasticity for a given geometry and homogeneous chemical composition of QD. In process of calculation the ratio of the height to the base of the pyramid was $0.05 < H/L < 1.75$ on retention of its volume. The indium concentration corresponded to the reference QD and also remained unchanged. All calculations were carried out by the finite element method. The volume of the cell in which the calculation was performed was ~ 450 times greater than the volume of the QD. The parameters of the material for calculations were taken from references [4,12]. The determination of the parameters of the quasi-binary InAs–GaAs solid solution was carried out according to

the Vegard's law. The band gaps for GaAs and InAs were 1.51 and 0.4 eV, respectively, for the temperature of 77 K, at which the exciton radiation spectra from the reference QD were experimentally studied.

The results of calculating the elastic deformation energy for various ratios of the height and base of the pyramid H/L are shown in Fig. 1. In Fig. 1, *a*, the energy is integrated over the entire cell over which the calculation was performed, and Fig. 1, *b* shows part of this energy stored in the volume of the InAs QD. It can be seen that the decrease in the H/L form factor leads to the decrease in the total energy of the system, which creates thermodynamic driving force that causes the decrease in the H/L ratio during QD overgrowth. In this case, the part of the total elastic energy, which is localized inside the QD, increases.

The total elastic energy can be represented in terms of the components of the mechanical stress tensors σ and elastic deformation tensors ε:

$$W_{total} = \int \left[\frac{1}{6}\mathrm{tr}(\varepsilon)\mathrm{tr}(\sigma) + \left(\frac{1}{2}\varepsilon_{ii}\sigma_{ii} - \frac{1}{6}\mathrm{tr}(\varepsilon)\mathrm{tr}(\sigma) \right) + \frac{1}{2}\varepsilon_{ij}\sigma_{ij} \right] dV. \quad (1)$$

Here the first term in square brackets represents the hydrostatic energy. The second term, which we will call as the „deviatoric", corresponds to the biaxial distortion. The last term represents the energy of shear deformations described by the off-diagonal components of the tensors σ and ε. The variation of these contributions to the total elastic energy as a function of the H/L ratio is shown in Fig. 1. It can be seen from the Figure that the hydrostatic part of the elastic energy is mainly accumulated inside the QD, while the elastic energy outside the QD is mainly provided by the deviatoric and shear components of the field of mechanical stresses and strains. This result agrees well with the predictions of analytical inclusion models [13–15].

In contrast to the total elastic energy of the system, its part accumulated inside the QD increases with the decreasing in the H/L ratio. It is important that this increase is mainly provided by the deviatoric energy component. The explanation for this fact is that as the height decreases and the base increases, the distribution of mechanical stresses and deformations approaches to realized one in the thin film.

The influence of mechanical fields on the energies of electrons and holes is described in terms of deformation potentials and the piezoelectric effect. The change in the electron potential with respect to the undeformed material is proportional to the change in the volume of the lattice cell of the crystal lattice:

$$\delta E_e = a_e \mathrm{tr}(\varepsilon). \quad (2)$$

Here a_e is negative hydrostatic deformation potential of electron. The InGaAs valence band is degenerate at the center of the Brillouin zone, where its top is located. The

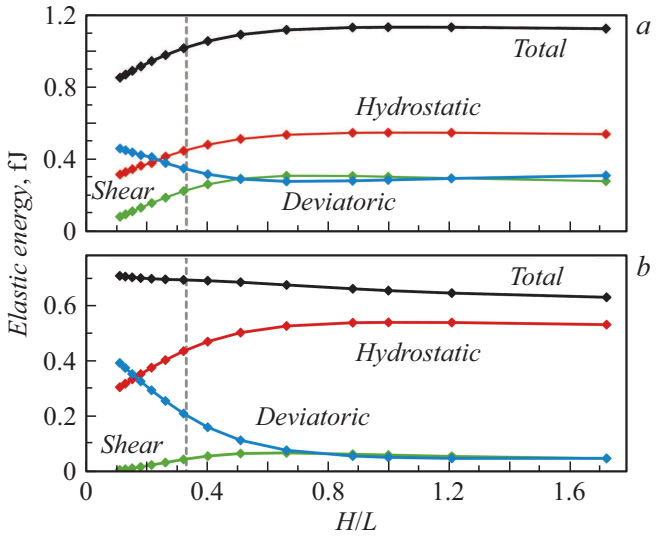

Figure 1. Dependence of the elastic deformation energy on the height-to-base ratio for the pyramidal InAs QD. Curves are presented for total energy (*Total*), hydrostatic component (*Hydrostatic*), deviatoric component (*Deviatoric*), shear component (*Shear*). *a* is the energy of the whole cell, *b* is the energy of the QD volume only.

deviatoric and shear components of deformations release this degeneracy of heavy and light holes in addition to the hydrostatic energy shift

$$\delta E_h = a_h \mathrm{tr}(\varepsilon) \pm \sqrt{\frac{b^2}{2}\left[(\varepsilon_{xx}-\varepsilon_{yy})^2+(\varepsilon_{xx}-\varepsilon_{zz})^2+(\varepsilon_{yy}-\varepsilon_{zz})^2\right]+d^2(\varepsilon_{xy}^2+\varepsilon_{xz}^2+\varepsilon_{yz}^2)}. \quad (3)$$

Here a_h is positive hydrostatic deformation potential for holes, b and d are deformation potentials for the deviatoric and shear components of the elastic deformation tensor.

The presence of elastic deformations causes the piezoelectric effect in InGaAs. Our calculations showed that the contribution of the piezoelectric effect to the QD localizing potential is relatively small and can be neglected in most cases. The authors of the work [12] came to the same conclusion earlier, when they carried out similar calculations for pyramidal InAs QDs with {101} faces.

The results of the quantum-mechanical calculation of the ground state energies of electrons E_e and holes E_{hh} in InAs QD surrounded by a GaAs barrier, depending on the ratio H/L are shown in Fig. 2, *b* (filled circles and squares). Filled circles in Fig. 2, *a* show the corresponding dependence for the ground state energy of the exciton in QD, calculated under the assumption that the exciton binding energy is independent of the H/L ratio. When the form factor was changed, the QD volume remained unchanged and equal to the volume of the reference QD. The form factor of the reference QD $H/L = 0.32$ is marked in Figs 1 and 2 by vertical dashed lines. Assuming the binding energy of exciton in the QD of the given volume equal

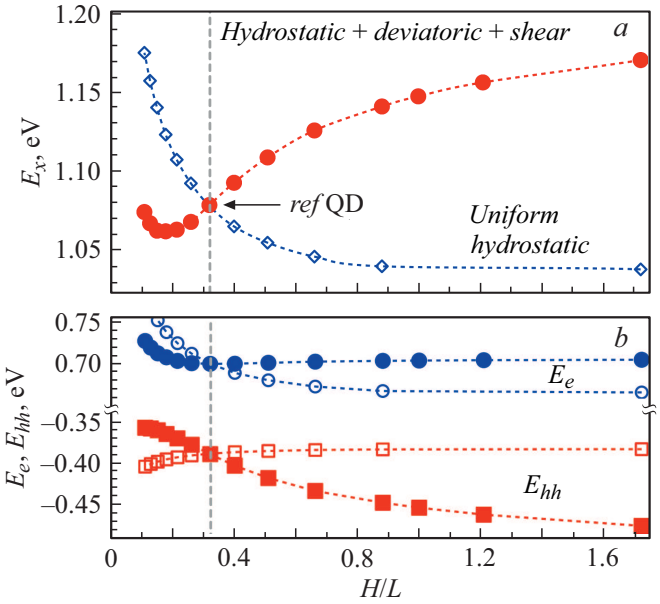

Figure 2. Dependences of the exciton energy (a), electron and heavy hole energies (b) on the height-to-base ratio for the pyramidal InAs QD. Filled symbols show the result of calculation taking into account all components of the deformation tensor, light symbols show the result of calculation taking into account only the hydrostatic part of deformations. The reference QD form factor $H/L = 0.32$ is marked with vertical dashed lines and corresponds to the intersection points of the curves.

to 10 meV [12], we obtained the quantitative agreement between the calculation results and the photoluminescence data obtained at 77 K. The corresponding point is marked in Fig. 2, a by an arrow.

It can be seen from Fig. 2, b that the energy of the ground state of electron has the minimum in the vicinity of $H/L = 0.2$. This energy increases rapidly at small H/L due to the enhancement of quantum confinement along the pyramid axis. The relatively slow increase in the electron energy with the increase in H/L is associated with the increase in the hydrostatic part of the deformations, as can be seen from Fig. 1.

The influence of size quantization on the energy of the ground state of hole in the QD is less pronounced due to the larger effective mass than in the case of the electron. In this case, E_{hh} shifts upward on the energy scale due to the transformation of hydrostatic deformations into deviatoric ones with the decrease in the H/L ratio.

As a result of the non-monotonic change in both the electron and hole energies, the energy of excitons localized in QDs has the well-defined minimum corresponding to the ratio of the height and base of the pyramid $H/L = 0.2$. Compared to pyramidal QDs with nonoptimal form factor, the benefit for the exciton energy can exceed 0.1 eV at the constant QD volume.

To understand the nature of the discovered phenomenon, we carried out additional calculations, in which only the hydrostatic component of the deformation tensor was taken into account. The results of such calculations are shown in Fig. 2 by light symbols. It can be seen that in this approximation, neither the energy of electrons, nor the energy of holes, nor, respectively, the energy of excitons have the minimum, i.e., the presence of the optimal form factor of the pyramidal QD is caused by the complex structure of the stressed state.

Obvious and natural approach to changing the localization of charge carriers in a QD is to change its dimensions. In this case, the enhancement of localization is achieved solely by reducing the quantum confinement energy. This approach, however, has two significant limitations. First, the given and, possibly, nonoptimal structure of the elastic deformation field does not change significantly with the change in the QD volume. Secondly, an increase in the size of QD above a certain limit inevitably leads to the formation of misfit dislocations [16,17], which is usually unacceptable for the use of QD in electronic and optoelectronic devices. Important advantage of strengthening the localization of charge carriers in QDs due to the shape effect is that this process should not lead to the formation of misfit dislocations, since the total mechanical energy of the system decreases in this process.

The results of calculating the elastic deformation energy for various height ratios of typical InAs pyramidal QDs showed that, at the constant QD volume, deeper localization of carriers can be achieved by implementing the optimal ratio of the pyramid height to its base $H/L = 0.2$. For such QDs, the gain in energy can reach 0.1 eV. The shape effect found is not specific to the material system chosen. It can be stated with confidence that similar effects should be observed in QDs based on various materials, including traditional III–V, III-N, II–VI, Si–Ge.

Acknowledgments

The authors are grateful to N.A. Cherkashin and A.V. Konovalov for useful discussions. The authors are also grateful to N.A. Bert, V.N. Nevedomskiy, A.L. Kolesnikova, A.E. Romanov, V.V. Preobrazhenskiy, B.R. Semyagin, and M.A. Putyato for many years of collaboration in the study of quantum dots.

Funding

The work was supported by the Russian Foundation for Basic Research (grant 19-32-90116).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] Zh.I. Alferov, FTP, **32** (1), 3 (1998). [Zh.I. Alferov, Semiconductors, **32** (1), 1 (1998). DOI: 10.1134/1.1187350].

[2] M. Bayer, Ann. Phys. (Berlin), **531**, 1900039 (2019). DOI: 10.1002/andp.201900039
[3] I.N. Stranski, L. Krastanow, Abhandlungen der Mathematisch-Naturwissenschaftlichen Klasse IIb. Akad. Wiss. (Wien), **146**, 797 (1938).
[4] S. Adachi, *Physical properties of III–V semiconductor compounds: InP, InAs, GaAs, GaP, InGaAs, and InGaAsP* (John Wiley & Sons, N.Y., 1992).
[5] K.E. Sautter, K.D. Vallejo, P.J. Simmonds, J. Appl. Phys., **128**, 031101 (2020). DOI: 10.1063/5.0012066
[6] S. Ruvimov, P. Werner, K. Scheerschmidt, U. Gosele, J. Heydenreich, U. Richter, N.N. Ledentsov, M. Grundmann, D. Bimberg, V.M. Ustinov, A.Yu. Egorov, P.S. Kop'ev, Zh.I. Alferov, Phys. Rev. B, **51**, 14766 (1995). DOI: 10.1103/PhysRevB.51.14766
[7] V.N. Nevedomskii, N.A. Bert, V.V. Chaldyshev, V.V. Preobrazhenskii, M.A. Putyato, B.R. Semyagin, Semiconductors, **43** (12), 1617 (2009). DOI: 10.1134/S1063782609120082.
[8] N. Cherkashin, S. Reboh, M.J. Hÿtch, A. Claverie, V.V. Preobrazhenskii, M.A. Putyato, B.R. Semyagin, V.V. Chaldyshev, Appl. Phys. Lett., **102**, 173115 (2013). DOI: 10.1063/1.4804380
[9] I. Daruka, J. Tersoff, A.-L. Barabási, Phys. Rev. Lett., **82**, 2753 (1999). DOI: 10.1103/PhysRevLett.82.2753
[10] P. Kratzer, Q.K.K. Liu, P. Acosta-Diaz, C. Manzano, G. Costantini, R. Songmuang, A. Rastelli, O.G. Schmidt, K. Kern, Phys. Rev. B, **73**, 205347 (2006). DOI: 10.1103/PhysRevB.73.205347
[11] A. Kosarev, V.V. Chaldyshev, Appl. Phys. Lett., **117**, 202103 (2020). DOI: 10.1063/5.0032110
[12] O. Stier, M. Grundmann, D. Bimberg, Phys. Rev. B, **59**, 5688 (1999). DOI: 10.1103/PhysRevB.59.5688
[13] J.D. Eshelby, Proc. R. Soc. Lond. A, **241**, 376 (1957). DOI: 10.1098/rspa.1957.0133
[14] J.D. Eshelby, Proc. R. Soc. Lond. A, **252**, 561 (1959). DOI: 10.1098/rspa.1959.0173
[15] N.A. Bert, A.L. Kolesnikova, A.E. Romanov, V.V. Chaldyshev, Phys. Solid State, **44** (12), 2240 (2002). DOI: 10.1134/1.1529918.
[16] N.A. Bert, A.L. Kolesnikova, V.N. Nevedomsky, V.V. Preobrazhenskii, M.A. Putyato, A.E. Romanov, V.M. Seleznev, B.R. Semyagin, V.V. Chaldyshev, Semiconductors, **43** (10), 1387 (2009). DOI: 10.1134/S1063782609100236.
[17] V.V. Chaldyshev, N.A. Bert, A.L. Kolesnikova, A.E. Romanov, Phys. Rev. B, **79**, 233304 (2009). DOI: 10.1103/PhysRevB.79.233304