Nonradiative recombination in quantum dots via Coulomb interaction with carriers in the barrier region

Igor Yu. Solov'ev[a] and Georgy G. Zegrya
Ioffe Physico-Technical Institute, Politechnicheskaya St. 26, 194021, St. Petersburg, Russia

(Received 29 October 2002; accepted 26 February 2003)

A mechanism of nonradiative recombination of nonequilibrium carriers in semiconductor quantum dots (QDs) is suggested and discussed. Recombination of an electron-hole pair localized in a QD occurs via Coulomb (Auger) interaction with carriers in the barrier region. It is shown that the characteristic time of such an Auger process depends on QD parameters, temperature, and carrier density in the barrier region and, under certain conditions, is shorter than the characteristic time of radiative recombination. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1569424]

Semiconductor quantum dots (QDs) attract increasing attention in view of the possibility of their application in various electronic devices, from lasers to quantum computers.[1] Recently, the possibility has been demonstrated of using QDs as markers in biological investigations and in medicine.[2] In this connection, it is important to understand what physical processes affect the electrical and optical properties of QDs. By now, no detailed analysis has been made of processes governing the lifetime of carriers localized in QDs. The lifetime is determined by recombination of electron-hole pairs localized in QDs and by the escape of carriers from the dots to the barrier. The recombination channels can be divided into radiative and nonradiative.

The radiative recombination in QDs has been sufficiently well studied theoretically and experimentally.[3,4]

In typical semiconductor systems, the radiative lifetime in a QD is about 1 ns.[3,4] This value is in good agreement with the experimental time of radiative recombination.[3] The radiative lifetime can be altered by varying the transition frequency or the overlap integral between the electron and hole wave functions. However, the probability of radiative recombination cannot be changed significantly in this way.

On the other hand, the time of nonradiative recombination may vary with the QD parameters and temperature in wider limits than the time of radiative recombination does. Together with direct Auger recombination of carriers in a QD,[5] one more channel of nonradiative recombination is possible, which always exists in heterostructures. This is Auger recombination process in which the energy of an electron and a hole recombining in a QD is transferred, via Coulomb interaction, to a carrier in the barrier region (see Fig. 1). The probability of such an Auger recombination process is proportional to the density of electrons and holes in the semiconductor matrix. This process resembles the well-known mechanism of electron relaxation in QDs.[6] In QD laser structures, the density of injected electrons and holes in the semiconductor matrix is as high as 10^{18} cm^{-3} and more. As shown later, at these densities the probability of the earlier Auger process is comparable with, or exceeds the probability of radiative recombination. Consequently, taking this process into account is important in analyzing the threshold characteristics of QD lasers. In the present study, we are concerned with such a mechanism of nonradiative recombination [via Coulomb interaction of an electron-hole pair in a QD with carriers in the barrier region (Fig. 1)].

Auger recombination rate depends on the transition energy and overlap integral between the electron and hole wave functions. These values are determined by the dimensions and shape of a QD and depend on some characteristic sizes of QDs. For example, there are two sizes (axes) for lens-shaped dots, and only one (radius) for spherical dots. For simplicity, we consider noninteracting spherical QDs in a three-dimensional matrix. The application of a spherical approximation to QDs is undoubtedly a simplification, which gives no way of obtaining precise quantitative results. However, these results reflect the real situation qualitatively. More precise data can be obtained by considering pyramidal QDs with account of the piezoelectric effect, since in this case, wave functions of electrons and holes are expanded in the different directions and less overlapped with each other.[7] The calculations entail considerable computational costs and,

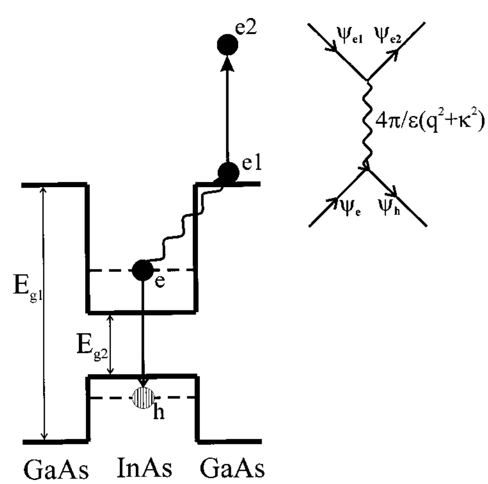

FIG. 1. Nonradiative recombination of an electron-hole pair in a QD (InAs/GaAs) via Coulomb interaction with a carrier from the barrier region. $E_{g1}=1.42$ eV, $E_{g2}=0.35$ eV. Inset: diagram describing the process. Ψ_e, Ψ_h are the wave functions of electron and hole localized in the QD; Ψ_{e1}, Ψ_{e2} are the initial and final wave functions of a carrier (electron or hole) from the barrier region.

[a]Electronic mail: igor@theory.ioffe.rssi.ru

even though refining the numerical values, do not change the results fundamentally. Consideration of cylindrical QDs, frequently used in calculations of this kind, does not refine the model any significantly, since in this case, as also in the case of spherical QDs, electrons and holes are localized in the same region and have similar symmetries. We choose as an example the well-understood InAs/GaAs system, which allows comparison of the results obtained with experimental data. A three-dimensional description of the barrier region largely corresponds to the experimental situation, since the depth of the wetting layer is small and carriers are, for the most part, outside this layer. The inverse time of Auger recombination of an electron-hole pair localized in a QD with carriers in the barrier region is calculated using the Fermi golden rule within the first-order perturbation theory in the electron–electron interaction

$$G=\frac{2\pi}{\hbar}\sum_{e,h,p1,p2}|W|^2 f_e f_h f_{p1}(1-f_{p2}) \times\delta(E_e+E_{p1}-E_{p2}-E_h). \quad (1)$$

Here $f_{p1,p2}$ are the Fermi distribution functions of particles (electrons and holes) from the barrier region, involved in the process; $f_{e,h}$ are the occupancies of electron and hole levels in the QD; E_e and E_{p1} are the energies of the initial states of an electron in the QD and the corresponding carrier in the barrier region; and E_h and E_{p2} are the energies of a hole and the final state of a carrier in the barrier region. W is the Coulomb matrix element between electrons localized in the QD and electrons in the matrix, having the form

$$W=\left\langle \Psi_f(\mathbf{r}_1,\mathbf{r}_2)\left|\frac{\exp(-\kappa|\mathbf{r}_1-\mathbf{r}_2|)}{\epsilon|\mathbf{r}_1-\mathbf{r}_2|}\right|\Psi_i(\mathbf{r}_1,\mathbf{r}_2)\right\rangle, \quad (2)$$

where e is the dielectric constant and

$$\kappa=\sqrt{\frac{4\pi n_0}{\epsilon k_B T}} \quad (3)$$

is the Debye wave vector and n_0 is the particle density in the matrix.

$\Psi_{i,f}(\mathbf{r}_1,\mathbf{r}_2)$ are the wave functions of the initial and final states of the system. In a single-particle approximation, they are products of the wave functions of electron and hole localized in a QD by wave functions of carriers in the barrier region. A diagram corresponding to this process is presented in the inset of Fig. 1.

In what follows we use the atomic system of units. We are interested in the ground dipole-active state of the electron-hole pair, since it is from this state that the major part of emission from a QD occurs. The wave functions of particles localized in the QD, used to calculate the matrix element of the Coulomb interaction, are obtained in terms of the multiband Kane model for a spherical QD of finite depth.[8]

The Coulomb interaction between the electron and hole is disregarded since we are mainly interested in small QDs.

In our case, it suffices to take planar waves as wave functions of carriers in the barrier region. Substituting these wave functions in the matrix element of Coulomb interaction, we pass to a Fourier transform and, making some rearrangements, represent it as

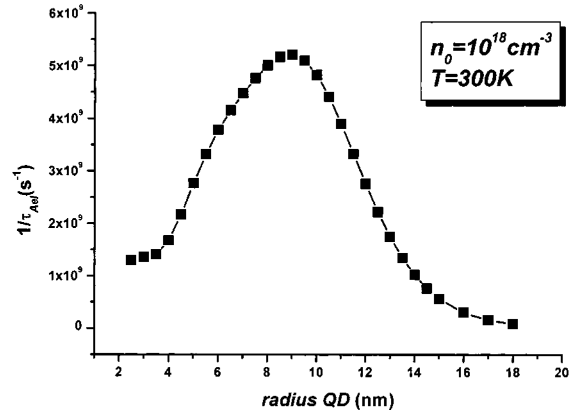

FIG. 2. Inverse time of nonradiative recombination of an electron-hole pair in a QD vs QD radius. $T=300$ K, $n_0=10^{18}$ cm^{-3}.

$$W(\mathbf{q})=\frac{4\pi}{\epsilon(q^2+\kappa^2)}\int d\mathbf{r}^3 e^{-i\mathbf{qr}}\Psi_e(\mathbf{r})\Psi_h(\mathbf{r}), \quad (4)$$

where $\Psi_e(\mathbf{r})$, $\Psi_h(\mathbf{r})$ are the wave functions of electron and hole in the QD, and $\mathbf{q}=\mathbf{k}_2-\mathbf{k}_1$ is the momentum transferred in the Coulomb interaction. The matrix element can be conveniently calculated with the exponent expanded in multipoles.

As already mentioned, the transition probability is calculated using the Fermi golden rule. As usual, it is necessary to make summation over all initial and final states of the system. We consider the case when electron and hole are at the ground quantum-well level; other levels can be taken into account without any difficulty, but this does not change significantly the results obtained.

To simplify the calculations, we assume that carriers in the barrier region are distributed in accordance with the Boltzmann distribution. This is true for holes in virtually any case, and for electrons when their density does not exceed at least 10^{18} cm^{-3}. Use of the Fermi distribution does not lead to any significant changes.

Figure 2 presents the dependence of the inverse time of Auger recombination, $1/\tau_A$, on the QD radius at room temperature ($T=300$ K) and constant electron density in the barrier region ($n=10^{18}$ cm^{-3}) for an Auger process involving a free electron. This process dominates over that involving a free hole. This is due to the fact that, in energy transfer to the hole, the momentum $\mathbf{q}$ transferred exceeds that in the case of energy transfer to an electron by approximately a factor $\sqrt{m_h/m_e}$.

The nonmonotonic dependence of $1/\tau_A$ on R is due to the dependence of the overlap integral of the electron-hole pair on the QD radius. The hole effective mass exceeds that of the electron, and, consequently, the hole wave function is virtually entirely localized within the QD. At small R, the electron wave function partially penetrates into the barrier region. With increasing R, the overlap integral grows and, at a certain radius, reaches its maximum value. Further, the overlap integral starts to decrease because of the delocalization of the electron and hole wave functions. Figure 3 presents the maximum $1/\tau_A$ value as a function of temperature. As expected, $1/\tau_A$ grows with increasing T, which is due to an increase in the phase volume of carriers in the barrier region.

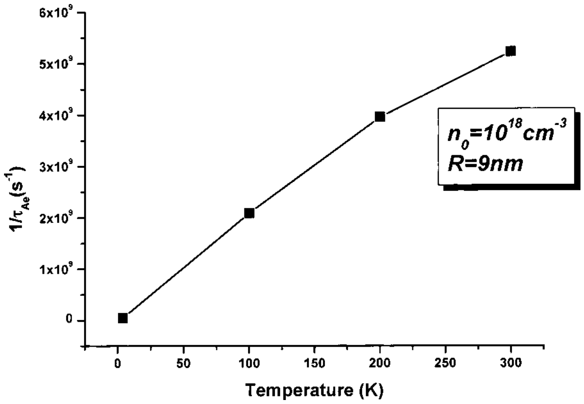

FIG. 3. Probability of nonradiative recombination of an electron-hole pair in a QD vs temperature T. $R=9$ nm, $n_0=10^{18}$ cm^{-3}.

It was shown that intradot Auger processes show very strong dependence on QD radius;[5,8] at the same time, the Auger process analyzed in this article exhibits an almost linear dependence on the carrier densities in the barrier region. Therefore, it is possible that either the former process dominates over the later, or vice versa, depending on the QD dimensions and carrier density in the barrier region.

Thus, our estimates demonstrate that the time of Auger recombination of an electron-hole pair in a QD with excitation of an electron (hole) in the barrier region may be shorter than the time of radiative recombination: at $T=300$ K, $n_0=3\times10^{18}$ cm^{-3}, and $R=9$ nm, the time of nonradiative recombination $\tau_A\simeq2\times10^{-10}$ s. This means that the Auger recombination process we consider strongly affects the lifetime of nonequilibrium carriers in the QD. In the case of QD lasers, such an Auger recombination process governs the internal quantum efficiency of emission and strongly affects the threshold current density. The quantum efficiency of a laser is a nonmonotonic function of the injected carrier density: it first grows with increasing density, while $\tau_A>\tau_R$, and then decreases with increasing density of injected carriers, provided that $\tau_A<\tau_R$. With increasing temperature and injected carrier density, the threshold current density of a laser grows owing to the appearance of an additional component, Auger current.

To conclude, we have shown that, in a QD heterostructure there exists, together with others, one more process of nonradiative recombination, which may compete with the well-know recombination processes and can strongly affect the threshold characteristics of QD lasers and govern the lifetime of nonequilibrium carriers.

Such an Auger recombination process affects the maximum power of QD lasers. With increasing current (i.e., growing carrier density in the barrier region) the probability of nonradiative Auger recombination of carriers in a QD becomes higher, which reduces the quantum efficiency of the laser and puts limitations upon its output power.

This work was partially supported by the Russian Foundation for Basic Research (Grants Nos. 01-07-90299 and 02-02-17610) and INTAS (Grant No. 0175wp).

[1] G. Yusa and H. Sakaki, Appl. Phys. Lett. **70**, 345 (1997); P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, L. Zhang, and A. Imamoglu, Science **290**, 2282 (2000); E. Dekel, D. Gershoni, and E. Ehrenfreund, Phys. Rev. B **61**, 11009 (2000).
[2] M. Bruchez, Jr., M. Moronne, P. Gin, S. Weiss, and A. P. Alivisatos, Science **281**, 2013 (1998); W. C. W. Chan and S. Nie, *ibid.* **281**, 2016 (1998); M. Han, X. Gao, J. Su, and S. Nie, Nature Biotechnol. **19**, 631 (2001).
[3] A. D. Yoffe, Adv. Phys. **50**, 1 (2001); D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, Chichester, 1999).
[4] L. V. Asryan and R. A. Suris, Semicond. Sci. Technol. **11**, 554 (1996).
[5] V. A. Kharchenko and M. Rosen, J. Lumin. **70**, 158 (1996); E. B. Dogonkine, V. N. Golovatch, A. S. Polkovnikov, A. V. Pozdnyakov, and G. G. Zegrya, 8th Int. Symp. Nanostructures: Physics and Technology, St. Petersburg, Russia, 19–23 June 2000, pp. 399–401.
[6] U. Bockelmann and T. Egeler, Phys. Rev. B **46**, 15574 (1992); A. V. Uskov, F. Adler, H. Schweizer, and M. H. Pilkuhn, J. Appl. Phys. **81**, 7895 (1997).
[7] O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999).
[8] D. I. Chepic, Al. L. Efros, A. I. Ekimov, M. G. Ivanov, V. A. Kharchenko, I. A. Kudriavtsev, and T. V. Vazeva, J. Lumin. **47**, 113 (1990).