Resonant photoluminescence of InAs/GaAs quantum dots

© A.P. Gorshkov, V.D. Shvartsman¶, N.S. Volkova, A.A. Cvetkov, S.M. Plankina, P.B. Demina, A.S. Panfilov

Lobachevsky State University,
Nizhny Novgorod, Russia

¶ E-mail: shvartsman@unn.ru

Received March 31, 2026
Revised May 26, 2026
Accepted June 1, 2026

Resonant photoluminescence of self-assembled InAs/GaAs quantum dots grown by metal-organic vapor-phase epitaxy at atmospheric pressure has been studied. Under excitation by a Nd:YAG laser (1.165 eV), narrow peaks at 1.059 and 1.049 eV appear, which are absent under above-barrier excitation. Temperature dependences and numerical modeling of selective excitation in a size-dispersed quantum dot ensemble were analyzed. It is shown that excitation via the first excited state does not explain the temperature stability of the resonant peaks. The results indicate laser absorption accompanied by the emission of three and four LO phonons.

Keywords: quantum dots, indium arsenide, resonant photoluminescence, selective excitation, LO phonons.

DOI: 10.61011/SC.2026.02.63748.9419

1. Introduction

The paper is dedicated to the study of the possibility of resonant excitation of photoluminescence of self-organized quantum dots (QD) InAs/GaAs, grown by vapor phase epitaxy from metalloorganic compounds (VPE MOC) at atmospheric pressure of hydrogen. The study of QD optical properties is of great interest for understanding of fundamental processes of transfer and relaxation of charge carriers in zero-dimensional systems, including mechanisms of interlevel transitions and interaction of carriers with phonons.

Usually the experimental studies of photoluminescence (PL) use non-selective overbarrier excitation, under which the QD array radiation has a wide band provided for by the spread of dimensions and, accordingly, energies of transitions. Resonant photoluminescence arising under radiation with light having energy of photons lower than the width of the matrix bandgap, makes it possible to selectively excited only those QDs whose energy levels make it possible for the process of laser radiation absorption to occur in accordance with a certain mechanism. This enables the study of optical properties and processes of relaxation in a narrow, spectrally uniform subset of dots. It is especially relevant to conduct such studies to analyze the properties of QD arrays with highly widened peaks of optical transitions [1,2], and also with the pronounced bimodality of dimensions [3,4]. Such information may be received by the method of PL excitation spectroscopy [5], however, for this purpose a powerful tunable optical source is required. The attempts to apply this method to quantum dots grown by VPE MOC at atmospheric pressure prevented obtaining informative results due to insufficient power of the exciting radiation with the use of a monochromator and a halogen lamp, and limited sensitivity of standard detectors. Therefore, this paper implemented the approach of selective excitation of quantum dots at the fixed wavelength using an Nd:YAG-laser.

2. Experimental method

The studies were carried out using a structure with a InAs/GaAs QD grown by VPE-MOC method at atmospheric pressure of hydrogen. The substrate used was n^+-GaAs(100). The following were formed on the substrate surface sequentially: a n-GaAs buffer layer with the thickness of 0.6 μm grown at temperature of 600 °C, the layer of InAs quantum dots formed at temperature of 520 °C, and the undoped GaAs coating layer with thickness of 100 nm. The schematic view of the studied structure is presented in Figure 1. To measure the PL spectra under overbarrier (nonresonant) excitation, a laser was used with the wavelength of 462 nm and power of 1.5 W. For resonant excitation, an Nd:YAG-laser was used with the wavelength of 1064 nm ($h\nu = 1.165$ eV) and power of 0.5 W. Both lasers worked continuously. The spectra were recorded using a monochromator Acton SP500 with a 600-lines/mm diffraction grating and an InGaAs-photodiode. The signal was registered using the standard scheme of synchronous detection with an SR810 DSP lock-in amplifier. The measurements were done in the temperature range of 77–180 K.

The GaAs coating layer and the layer of quantum dots were etched in NH_4OH:H_2O_2:H_2O solution at the ratio of 1 : 1 : 5.

3. Results and discussion

Under overbarrier excitation with a laser having a wavelength of 462 nm, when electron-hole pairs are generated in a GaAs-matrix and grabbed by the entire QD array

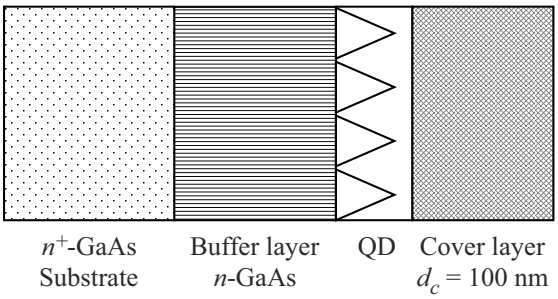

Figure 1. Scheme of studied sample with InAs/GaAs quantum dots.

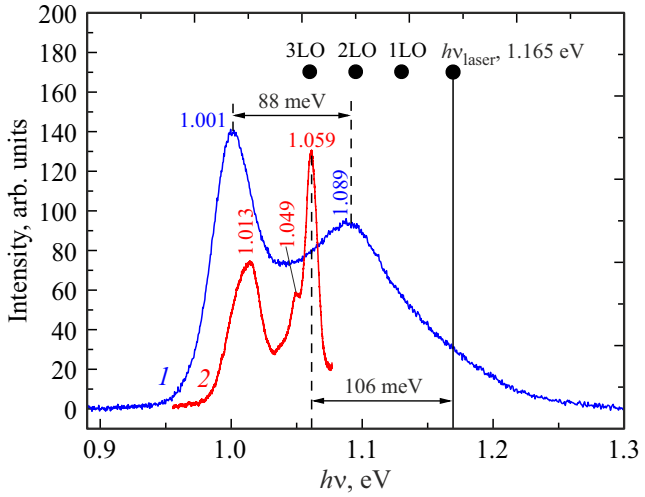

Figure 2. Photoluminescence spectra of the structure with the InAs/GaAs QD at $T = 77$ K: *1* — overbarrier excitation; *2* — resonant excitation. The vertical solid line marks the energy of the resonant excitation. The points specify the position of the expected phonon replicas at $\hbar\omega_{LO} = 36$ meV.

(Figure 2, curve *1*), the photoluminescence spectrum shows two wide peaks corresponding to the ground-state (E_0) and first excited (E_1) optical transitions with energies of 1.001 and 1.089 eV accordingly. These values reflect the median parameters of the entire QD array.

Under excitation with the Nd:YAG-laser (1.165 eV) at temperature of 77 K the PL spectrum acquires a significantly more complicated nature. A narrow peak for 1.059 eV with an additional arm at 1.049 eV appears in it, apart from a wide band at 1.013 eV, close to the ground-state transition under overbarrier excitation. Such spectrum structure was observed previously in other papers, too, for example, in [6].

The QD layer was etched to test the nature of these peaks. Then, under the same conditions of the resonant excitation, the optical signal completely vanished in the entire spectral region, which confirms their connection to the optical transitions between quantum-dimensional stages in QD, and not to the impurity levels or defects in the epitaxial layers.

The cause for occurrence of narrow photoluminescence peaks under excitation with photons with energy below the width of the matrix bandgap consists in the fact that under such conditions not the entire array of the quantum dots is excited, but only a part of it. The literature considers two possible mechanisms of such selective excitation. The first suggests absorption of a photon with formation of an electron-hole pair on the excited level of QDs [6] provided that the following condition is met (1):

$$h\upsilon_{\text{laser}} \approx E_e^1 - E_{hh}^1, \qquad (1)$$

where $h\upsilon_{\text{laser}}$ — energy of laser radiation, E_e^1 — energy of the first excited electron level, E_{hh}^1 — energy of the first excited hole level.

The second mechanism is related to the excitation of carriers to the ground states of the QDs with simultaneous emission of one or several optical phonons [7–9] with the energy that meets the condition (2):

$$h\upsilon_{\text{laser}} \approx E_e^0 - E_{hh}^0 + m \cdot \hbar\omega_{\text{LO}}, \qquad (2)$$

where E_e^0 — energy of the ground electron state, E_{hh}^0 — energy of the ground hole state, $\hbar\omega_{\text{LO}}$ — phonon energy, m — number of phonons participating in the process. These processes in GaAs may have a noticeable probability even with participation of several phonons, which is related to the value of the Huang–Rhys parameter, characterizing the force of the electron-phonon interaction. This parameter is defined as the ratio of the intensity of the first phonon replica to the intensity of phonon-free transition. For polar semiconductors, and especially for quantum-dimensional structures on their basis, its value may reach 0.01–0.5, which indicates a significant role of phonon processes in the optical transitions [10–13].

Let us consider both hypotheses. Since the peak of the excited transition is rather wide (Figure 2, curve *1*), it is evident that the array contains small QDs with $E_1 = 1.165$ eV, capable of resonant absorption of the Nd:YAG-laser energy. Electron-hole pairs that arose in such QDs may either relax to the ground state, which results in a narrow peak at 1.059 eV (Figure 2, curve *2*), or enter the matrix with subsequent capture to other QDs forming a wide nonresonant component 1.013 eV (Figure 3, *a*). Non-compliance of the energy separation and energy of excitation to the resonant peak (106 meV) and difference of peak centers obtained under excitation with laser 462 nm (88 meV) may be explained by the fact that small QDs (responsible for the resonant PL) and medium QDs (responsible for the centers of non-resonant peaks on curve *1*) differ in the value of the difference $E_1 - E_0$. The final spectral width of the resonant peak is caused by random spread of the value of energy separation $E_1 - E_0$ between the ground and excited states in different small QDs absorpbing Nd:YAG-laser energy.

To test this hypothesis, numerical modeling of the spectrum of the resonant PL arising in the QD array with the spread of dimensions was conducted. The simplest

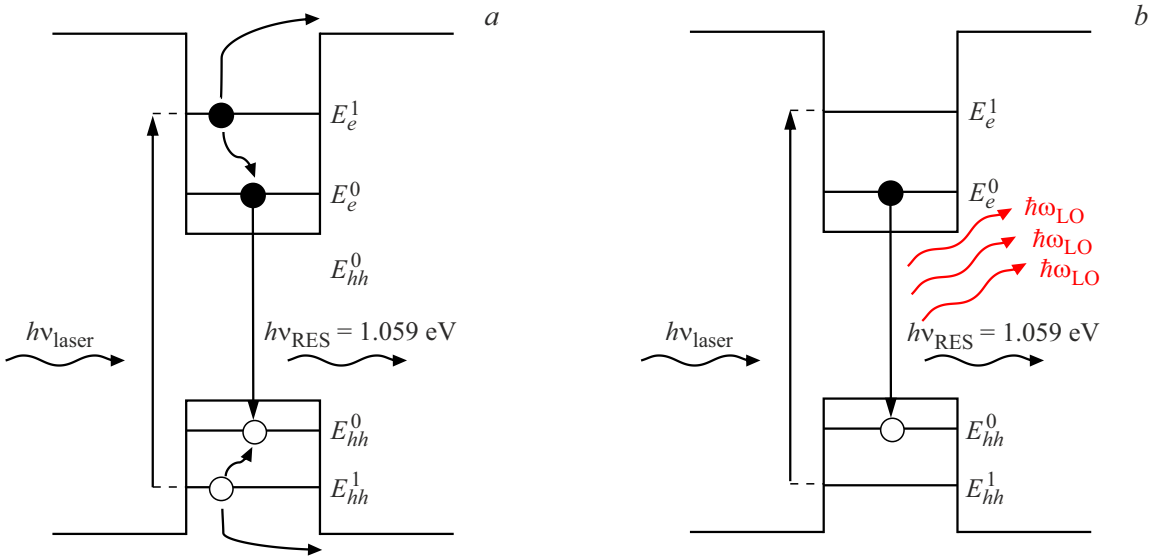

Figure 3. The possible mechanisms for resonant peak generation: *a* — generation of the electron-hole pair on the first excited level (E_1) with subsequent relaxation to the ground state (E_0); *b* — photon absorption with simultaneous emission of three LO phonons and generation of carriers in the ground state.

model of a three-dimensional infinite potential well, within which the energy levels E_n are defined by expression (3):

$$E_n = \frac{\hbar^2\pi^2}{2m_e}\left(\frac{n_1^2}{L_x^2} + \frac{n_2^2}{L_x^2} + \frac{n_3^2}{L_z^2}\right), \qquad (3)$$

where m_e — effective mass of the charger carrier, L_x — lateral dimension of the quantum dot, L_z — height of the quantum dot. The average dimensions, the dispersion value and also the width of the QD material bandgap were selected so that the model spectrum of optical transitions of the entire QD array would be best described by the PL spectrum measured at non-resonant excitation. Then those QDs were selected from the array, in which the energy of the excited transition E_1 was in the range of $h\upsilon_{\text{laser}} \pm 1$ meV, and the energy of the ground-state transition E_0 of these selected QDs was analyzed. Indeed, the energy separation between the excitation line $h\upsilon_{\text{laser}}$ and the resonant peak E_{res} was not constant, but varied with the laser energy variation. At $h\upsilon_{\text{laser}} = 1.165$ eV the model peak of the ground-state transition had the energy of 1.057 eV close to the one observed experimentally. However, the estimates predict another temperature dependence of the resonant peak position compared to the one observed experimentally. In the experiment the resonant peak at 1.059 eV is practically not displaced at temperature change in the range of 80–180 K (Figure 4), whereas the model peak under these conditions is displaced by 19 meV to the red region of the spectrum. Such displacement, if the resonant peak occurs in accordance with the considered mechanism, is quite expected and is related to the fact that temperature variation leads to the change in the composition of the subarray of the quantum dots, for which the excited optical transition turns out to be in resonance with the laser

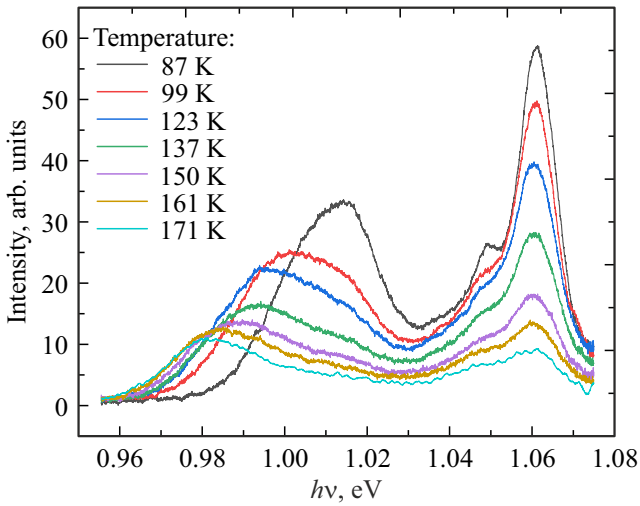

Figure 4. Temperature evolution of the photoluminescence spectra of the structure with the InAs/GaAs QDs at resonant excitation with an Nd:YAG-laser.

energy. As temperature rises, smaller QDs become involved in the resonance, which are characterized by both higher energies of optical transitions as such and higher value of the energy separation $E_1 - E_0$, which defines the position of the resonant peak relative to the fixed energy of excitation.

The second explanation of the observed effect is based on the absorption of the exciting radiation photon with simultaneous emission of one or several optical phonons. Such mechanism is free from the above disadvantage and makes it possible to explain the absence of the temperature shift of the resonant peak, since the energy of the LO phonon in GaAs (~ 36 meV) [14] practically does not

depend on temperature. Similar processes of absorption with the participation of phonons were discussed previously for InAs/GaAs-quantum dots [8,15–17]. Within this approach the narrow peak at 1.059 eV may be explained by excitation in very small QDs with energy of the ground-state transition $E_0 = 1.059$ eV provided that the condition (2) is met for $m = 3$, which corresponds to the emission of three LO phonons (Figure 3, *b*). With the substitution of $\hbar\omega_{LO} = 36$ meV the difference between the estimated and experimental value of the peak energy is ~ 2 meV, which is within the limits of its detection error. This hypothesis makes it possible to also explain the origin of the additional peak at 1.049 eV, which, as the ground-state resonant peak, does not demonstrate the temperature shift. For this peak Eq. (2) is satisfied for four LO phonons with energy of 29 meV, which corresponds to bulk InAs [14].

Therefore, the combination of experimental data and results of modeling indicate that the resonant peaks at energies of 1.059 and 1.049 eV are formed as a result of multi-phonon absorption of the exciting laser radiation.

4. Conclusion

The paper studied the resonant photoluminescence of self-organized InAs/GaAs quantum dots grown by the VPE MOC method at atmospheric pressure of hydrogen. Upon excitation with an Nd:YAG-laser with energy of 1.165 eV the complex structure of narrow peaks was found at 1.059 and 1.049 eV, absent under overbarrier pumping. It is shown that their formation is related to the selective excitation of the subarray of quantum dots. Numerical modeling of the excitation mechanism through the first excited state reproduces the position of the peak at 77 K, however, predicts the temperature shift, which is not observed experimentally. The absence of the temperature dependence of the resonant peak position and energy separations close to integer multiples of the LO-phonon energy, indicate the mechanism of multi-phonon absorption of exciting radiation with the participation of three and four LO phonons.

Funding

The study was conducted with the support of the Ministry of Science and Higher Education of the Russian Federation (project FSWR-2025-0008).

Acknowledgments

The authors would like to express their gratitude to B.N. Zvonkov for growing the structures.

Conflict of interest

Authors declare no conflict of interest.

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Translated by M.Verenikina