Temperature dependencies of radiative and nonradiative carrier lifetimes in InGaAs quantum well-dots

© A.M. Nadtochiy[1,2], I.A. Melnichenko[1], K.A. Ivanov[1], S.A. Mintairov[3], N.A. Kalyuzhnyy[3], M.V. Maximov[2], N.V. Kryzhanovskaya[1], A.E. Zhukov[1]

[1] National Research University Higher School of Economics,
190008 St. Petersburg, Russia
[2] Alferov University,
194021 St. Petersburg, Russia
[3] Ioffe Institute,
194021 St. Petersburg, Russia
E-mail: al.nadtochy@mail.ioffe.ru

Received September 20, 2022
Revised September 26, 2022
Accepted September 26, 2022

Heterostructure with InGaAs/GaAs quantum well-dots was investigated in temperature range 10−300 K using photoluminescence spectroscopy in CW mode as well with time resolution. Obtained decay times were splitted into radiative and nonradiative components of carrier lifetime. It is found that radiative lifetime demonstrates exponential growth with temperature rise, while temperature dependence of nonradiative one is much weaker.

Keywords: semiconductors, quantum well-dots, photoluminescence, time resolution, lifetime, temperature dependence.

DOI: 10.21883/SC.2022.10.54907.9963

1. Introduction

In recent decades, semiconductor devices based on quantum-dimensional heterostructures have been increasingly used in various fields of science and technology. Modification of the density of states in the active region of such devices makes it possible to improve their characteristics, for example, to reduce the threshold current density [1] and increase the temperature stability [2], and also opens up opportunities for creating fundamentally new devices, such as single photon sources [3]. In this regard, the study of the physical properties of quantum-dimensional heterostructures is extremely relevant from both practical and fundamental points of view. A deep understanding of the processes occurring in semiconductor structures can be provided by optical studies with high time resolution. Two-dimensional (quantum wells, QW [4]), one-dimensional (quantum wires [5]) and zero-dimensional (quantum dots, QD [6]) semiconductors were studied in sufficient detail using time-resolved photoluminescence (PL) methods. At the same time, the recently proposed active medium of transition dimension (0D/2D) (so-called quantum well-dots, QWDs) has been studied rather poorly by these methods [7].

QWDs represent the InGaAs/GaAs quantum well with inhomogeneities in composition and thickness, and can also be considered as a superdense array of QDs with a relatively low localization energy of electrons and holes [8]. At the same time, they demonstrate a number of advantages inherent in both 2D quantum wells and 0D quantum dots. In particular, QWDs have a large gain/absorption coefficient. A record high material gain was achieved above $10^4\,cm^{-1}$, exceeding the material gain of QW and QD, as well as the modal optical gain of about $75\,cm^{-1}$ for one layer of QWDs [8,9]. Due to their unique properties, [9] QWDs seem to be very promising as an active area of photovoltaic converters, edge-emitting lasers, waveguide photodetectors, microlazers and other devices.

In this article, we present the results of the study of QWDs by the method of time-resolved PL at different temperatures and comparing these results with PL under continuous wave excitation in order to study in detail the processes of radiative and non-radiative recombination in a new type of nanostructures — QWDs.

2. Experiment

Laser heterostructures were synthesized by metalorganic vapor-phase epitaxy on slightly misoriented GaAs substrates. The research sample consisted of barriers $Al_{0.4}Ga_{0.6}As$ with a thickness of 50 nm, bounding a GaAs matrix with a thickness of 600 nm with one layer of QWDs in the center. The QWDs were formed by deposition of $In_{0.4}Ga_{0.6}As$ with an effective thickness of 8 monolayers. Details of the QWDs growth can be found in the review [8].

Time-resolved photoluminescence (PL) studies were carried out using optical gating by the up-conversion method. The sample was pumped using a Coherent Mira 900 D titanium-sapphire laser in femtosecond mode with a laser pulse duration of 120 fs (repetition rate — 76 MHz) and a radiation wavelength of 780 nm. The time-resolved PL signal was recorded using a FOG spectrometer (CDP systems) using a nonlinear BBO crystal. The experimental setup made it possible to register PL with a time resolution

of ~ 0.2 ps in the range of 8 ns. We estimate the optical resolution in such an experiment as 10 nm. The excitation energy density was $\sim 10\,\mu$J/cm^2, which corresponds to $3 \cdot 10^{13}$ photon/cm^2.

The sample was pumped under continuous excitation using a YAG:Nd laser with a wavelength of 532 nm and an excitation power density of 0.3W/cm^2. The PL was measured using a cooled germanium diode coupled to an MDR-23 monochromator. Closed-cycle helium cryostats were used for PL studies at low temperatures: Montana S-50 — in case of time-allowed experiments and Janis — for CW excitation.

3. Results and discussion

Fig. 1 presents the results of kinetic studies of the ground state of the QWDs at various temperatures (examples of spectra with pulsed and CW excitation are shown in the insets to Fig. 2). On all curves, it is possible to mark a section where the intensity of the PL practically does not change with time. This can be explained by the fact that the excessive concentration of carriers created at the initial moment by an optical pulse in the GaAs matrix requires a certain time for capture and recombination in the QWDs. During this period of time, the rate of capture of carriers from GaAs to the ground state of the QWDs is equal to the rate of recombination. However, the GaAs matrix is emptied faster than the levels in the QWDs and at long delay times, the recharge by the carriers of the ground state of the QWDs stops, after which the PL(t) signal intensity demonstrates a close to exponential decay.

In the present work, we have limited ourselves to the study of this part of the PL decay at different temperatures, since it characterizes the recombination processes occurring in the QWDs. By approximation using the expression $\mathrm{PL}(t) \propto \exp(-t/\tau_{\mathrm{PL}})$ the decay time of the PL signal from the QWDs was obtained, τ_{PL}, which was found to decrease with decreasing temperature (Fig. 2, *a*).

As shown, for example, in [4], the PL decay time can be divided into radiative τ_{R} and non-radiative τ_{NR} lifetime of charge carriers:

$$\frac{1}{\tau_{\mathrm{PL}}} = \frac{1}{\tau_{\mathrm{R}}} + \frac{1}{\tau_{\mathrm{NR}}}, \qquad (1)$$

the relationship between which can be characterized by the quantum efficiency of radiative recombination η:

$$\eta = \frac{\frac{1}{\tau_{\mathrm{R}}}}{\frac{1}{\tau_{\mathrm{R}}} + \frac{1}{\tau_{\mathrm{NR}}}} = \frac{\tau_{\mathrm{PL}}}{\tau_{\mathrm{R}}}. \qquad (2)$$

The expressions (1) and (2) allow us to express τ_{R} and τ_{NR} as follows:

$$\tau_{\mathrm{R}}(T) = \frac{\tau_{\mathrm{PL}}}{\eta(T)}, \quad \tau_{\mathrm{NR}}(T) = \frac{\tau_{\mathrm{PL}(T)}}{1-\eta(T)}. \qquad (3)$$

The temperature dependence of the quantum efficiency of radiative recombination can be estimated using the

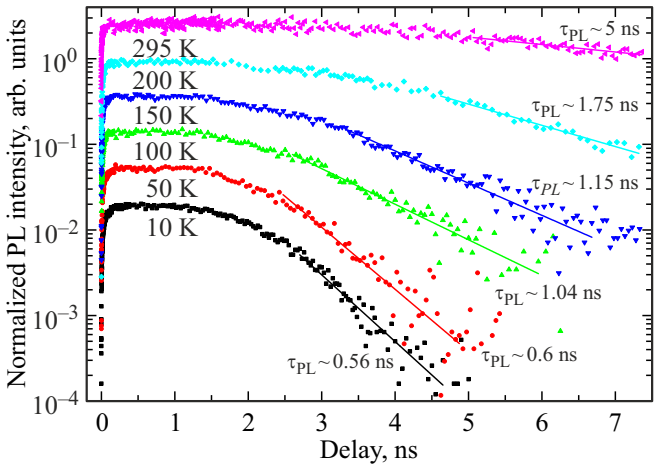

Figure 1. The kinetics of PL of QWDs at different temperatures. The curves are shifted along the vertical axis for clarity. An area of an exponential approximation of the PL decay is also shown.

temperature dependence of the integral intensity of the PL at low excitation densities, when the population of the GaAs matrix carriers and the contribution of the processes occurring there can be neglected. At the same time, it can be assumed that at low temperatures the quantum efficiency is close to 100%, since the emission of carriers into the matrix and nonradiative recombination are suppressed. Fig. 2, *b* shows the temperature dependence of the efficiency of radiative recombination of the QWDs, obtained by the described method from the integral intensity of the PL in CW mode, measured at an excitation density of 0.3W/cm^2, at which the contribution of GaAs to radiative recombination is negligible (see the spectra in the inset). We assume that $\eta = 100\%$ at 10 K.

Using the dependence η and the formula (3), we obtained the values τ_{R} and τ_{NR} at different temperatures (Fig. 3). We note that varying the low-temperature value of the efficiency of radiative recombination $\eta(10\,\mathrm{K})$ does not change the type of dependence $\tau_{\mathrm{R}}(T)$ and leads only to its displacement along the vertical axis, and the dependence $\tau_{\mathrm{NR}}(T)$ changes slightly in the low temperature range [4]. Thus, despite the impossibility to accurately estimate η, we can draw conclusions about the nature of temperature dependences of lifetimes in the QWDs.

The radiative lifetime of τ_{R} with an increase in temperature > 50 K demonstrates a rapid exponential growth of $\tau_{\mathrm{R}}(T) \sim \exp(T/T_0)$, described by the characteristic temperature $T_0 \sim 55$ K. The nonradiative lifetime of carriers τ_{NR} demonstrates a much weaker nonmonotonic variation within the order of magnitude in the range of 50–300 K. This fact indicates either that the widespread approach of describing luminescence quenching with an increase in temperature due to temperature activation of non-radiative recombination channels is inapplicable in case of QWDs (see, for example, [10]), or that the activation energy of these channels is high enough, and this process manifests

itself at temperatures noticeably higher than 300 K. Nevertheless, if at temperatures < 150 K radiative recombination dominates, which corresponds to a high quantum efficiency of luminescence, then at higher temperatures τ_{NR} becomes smaller than τ_R (Fig. 3). Due to the exponential growth of the radiative lifetime with temperature, in order to realize a situation, where $\tau_{NR} > \tau_R$ at 300 K, it would be necessary to increase the τ_{NR} by an order of magnitude, which implies a radical improvement in the structural and optical quality of the material. Taking into account that the studied QWDs were used in the active region of high-efficiency lasers, it can be concluded that at room and elevated temperatures, the lifetime of carriers in the QWDs, even in highly perfect samples, will be significantly determined by non-radiative recombination.

In early works on the study of carrier lifetime in QWs, an exciton recombination theory was developed that predicts a linear increase in the radiative recombination time with temperature, which was experimentally confirmed by [11,12]. However, later there were reports about the observation both in the QWs [4,13] and in the quantum wires [5] and in the QDs [6] of an exponential temperature increase in the radiative lifetime of carriers with a comparable T_0 (τ_R increases approximately by ~ 10 times per 100 K). The proposed more complex theory, considering the contribution of free carriers to the recombination [14], also failed to satisfactorily describe the experiment [4,15]. Thus, at the moment there are no theoretical models explaining the exponential growth of the radiative lifetime of charge carriers in quantum-dimensional structures. It is also worth noting that the authors in the studies [15,16] note a strong influence of the inhomogeneity and thickness of the QW or wire on the temperature dependence of τ_R as a consequence of the change in the degree of localization of excitons.

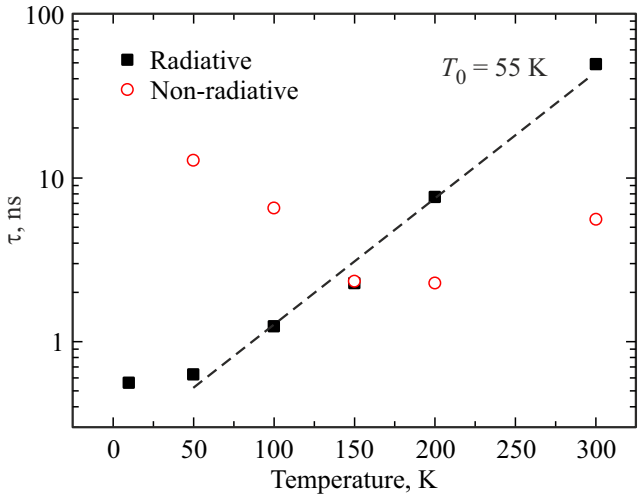

Figure 3. Calculated values of the characteristic times of radiative and non-radiative recombination (τ_R and τ_{NR}) at different temperatures, as well as approximation of the dependence of the radiative time by an exponential function with $T_0 = 55$ K.

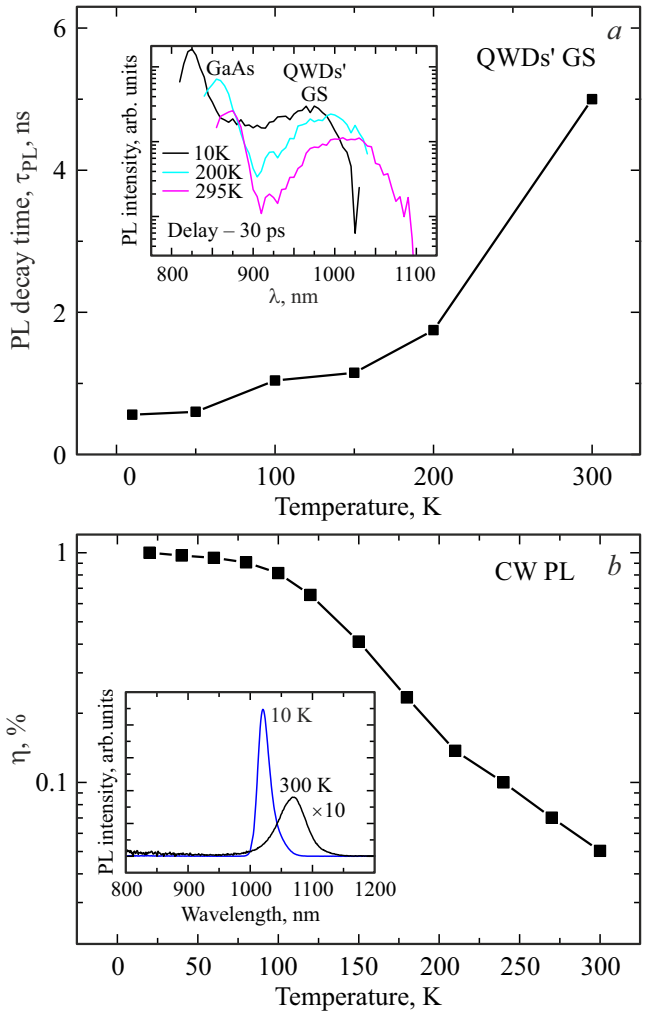

Figure 2. *a* — temperature dependence of the measured PL decay time, τ_{PL} (the inset shows examples of PL spectra with pulsed excitation at delay of 30 ps); *b* — temperature dependence of quantum efficiency η (the inset shows examples of PL spectra under CW excitation).

4. Conclusion

The radiative and non-radiative components of the lifetime of charge carriers in the InGaAs/GaAs QWD structure have been studied in a wide temperature range. It is shown that a decrease in luminescence intensity with an increase in temperature is associated with a significant increase in the time of radiative recombination, and not with the activation of non-radiative recombination channels.

Funding

This work was supported by the Russian Science Foundation (grant No. 22-72-10002). Time-permitted studies were carried out with the support of the HSE Fundamental Research Program. M.V. Maximov and A.M. Nadtochiy express their gratitude to the Ministry of Science and Higher

Education of the Russian Federation (project 0791-2020-0002).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] Z. Alferov. IEEE J. Select. Top. Quant. Electron., **6** (6), 832 (2000).
[2] O.B. Shchekin, J. Ahn, D.G. Deppe. Electron. Lett., **38** (14), 712 (2002).
[3] P. Senellart, G. Solomon, A. White. Nature Nanotechnol., **12** (11), (2017).
[4] M. Gurioli, A. Vinattieri, M. Colocci, C. Deparis, J. Massies, G. Neu, A. Bosacchi, S. Franchi. Phys. Rev. B, **44** (7), 3115 (1991).
[5] D. Y. Oberli, F. Vouilloz, E. Kapon. Phys. Status Solidi A, **164** (1), 353 (1997).
[6] O. Nasr, N. Chauvin, M. H. H. Alouane, H. Maaref, C. Bru-Chevallier, L. Sfaxi, B. Ilahi. J. Opt., **19** (2), 025401 (2017).
[7] A.M. Nadtochiy, S.A. Mintairov, N.A. Kalyuzhnyy, M.V. Maximov, D.A. Sannikov, T.F. Yagafarov, A.E. Zhukov. Semiconductors, **53** (11), 1489 (2019).
[8] M.V. Maximov, A.M. Nadtochiy, S.A. Mintairov, N.A. Kalyuzhnyy, N.V. Kryzhanovskaya, E.I. Moiseev, N.Y. Gordeev, Y.M. Shernyakov, A.S. Payusov, F.I. Zubov, V.N. Nevedomskiy, S.S. Rouvimov, A.E. Zhukov. Appl. Sci., **10** (3), 1038 (2020).
[9] N.Y. Gordeev, M.V. Maximov, A.S. Payusov, A.A. Serin, Y.M. Shernyakov, S.A. Mintairov, N.A. Kalyuzhnyy, A.M. Nadtochiy, A.E. Zhukov. Semicond. Sci. Technol., **36** (1), 015008 (2021).
[10] E.C. Le Ru, J. Fack, R. Murray. Phys. Rev. B Condens. Matter Mater. Phys., **67** (24), 1 (2003).
[11] G.W.'t Hooft, M.R. Leys, H.J. Talen-v.d. Mheen. Superlat. Microstr., **1** (4), 307 (1985).
DOI: 10.1016/0749-6036(85)90092-8
[12] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, R. J. Elliott. Phys. Rev. Lett., **59** (20), 2337 (1987).
[13] G. Bacher, C. Hartmann, H. Schweizer, T. Held, G. Mahler, H. Nickel. Phys. Rev. B, **47** (15), 9545 (1993).
[14] B.K. Ridley. Phys. Rev. B, **41** (17), 12190 (1990).
[15] D.Y. Oberli, M.-A. Dupertuis, F. Reinhardt, E. Kapon. Phys. Rev. B, **59** (4), 2910 (1999).
[16] S. Yamauchi, A. Shikanai, I. Morohashi, S. Furue, K. Komori, T. Sugaya, T. Takagahara. J. Appl. Phys., **102** (9), 094303 (2007).