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Time-resolved optical characterization of InAs/InGaAs quantum dots emitting at 1.3 μm

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We present the time-resolved optical characterization of InAs/InGaAs self-assembled quantum dots emitting at 1.3 μm at room temperature. The photoluminescence decay time varies from 1.2 (5 K) to 1.8 ns (293 K). Evidence of thermalization among dots is seen in both continuous-wave and time-resolved spectra around 150 K. A short rise time of 10 ± 2 ps is measured, indicating a fast capture and relaxation of carriers inside the dots. © *2000 American Institute of Physics.* [S0003-6951(00)05123-8]

There is an increasing interest for the development of GaAs-based active materials emitting in the telecom wavelength regions around 1.3 and 1.55 μm. These would allow the realization of telecom sources cheaper and less temperature sensitive than commercial InP-based devices. It has been shown that self-assembled InAs/GaAs quantum dots (QDs) on GaAs substrates can provide optical gain at 1.3 μm.[1–4] A recently proposed method[5,6] to achieve the 1.3 μm wavelength is to embed the QDs in an InGaAs quantum well (QW). This results in a lower transition energy as compared to GaAs-embedded QDs, presumably due to reduced strain in the QDs, suppression of In segregation from the QDs when covering with InGaAs, and lower barrier potential. Low-threshold lasers in the 1.3 μm region were demonstrated using this approach.[7,8] In order to assess this new gain material for application in high-speed laser sources, parameters such as carrier lifetime, capture, and relaxation times must be measured. In this letter, we investigate these parameters by time-resolved photoluminescence (PL) in InAs QDs embedded in a InGaAs QW and emitting at 1.3 μm at room temperature. We measure the carrier lifetime and the PL rise time as a function of temperature from 5 K to room temperature. This data demonstrates a very fast carrier relaxation from the QW to the QD ground state, indicating that this active material is indeed suitable for high-speed modulation.

The sample was grown by molecular beam epitaxy on a (001) GaAs substrate. The epitaxial structure has the following sequence: GaAs buffer layer, 20 nm AlAs, 100 nm GaAs, 20 nm $In_{0.15}Ga_{0.85}As$ including a single layer of QDs in the center, 100 nm GaAs, 20 nm AlAs, and a top 20 nm GaAs cap layer. The QDs were grown by depositing 2.6 monolayers of InAs. Figure 1 shows the continuous-wave (cw) PL spectra taken at different temperatures with a pump power density $I_{pump}=9$ W/cm². At this low excitation power

only the ground state contributes to the PL spectra at low temperatures. The first excited state is observed at higher pump levels (not shown) ≈50 meV above the ground state peak. The ground state PL peak energy varies from 1.02 eV (1210 nm) at 50 K to 0.946 eV (1311 nm) at room temperature (RT). At low temperature, the large inhomogeneous linewidth [full width at half maximum (FWHM)=82 meV at 50 K] is due to size and composition dispersion among different dots. Particularly noticeable is the PL tail on the high-energy side. This is only observed when dots are grown *inside* the InGaAs QW, whereas dots grown on GaAs and covered with InGaAs present a more symmetric PL lineshape. Little change is seen in the spectra in the temperature range 5 K<T<100 K. At temperatures above 100 K, carriers start to thermalize among different dots, which produces a line narrowing of the PL.[9,10] Above 200 K, the first excited state starts to be thermally populated, while the overall PL efficiency decreases with temperature.

For the time-resolved PL measurements, *p*-polarized pump pulses from a mode-locked Ti:Sa (130 fs, 76 MHz) were focused onto the sample at the Brewster angle. The PL emission was collected normal to the sample surface, spectrally resolved by a monochromator and detected by a streak camera with an infrared-enhanced photocathode. The spectral and temporal resolution are 4 nm and 3.5 ps, respectively, in the fastest time range. Figure 2 shows the measured PL decay at different temperatures, at a detection energy corresponding to the ground state PL peak for each temperature. The pump energy is 1.55 eV (i.e., above the GaAs band gap), and the average pump intensity incident on the sample is 9.4 W/cm², which corresponds to an estimated 10^{11} cm^{-2} excited electron/hole pairs per pulse. At this pump level, excited states only contribute as a high-energy tail in the PL spectra. The PL decay can be well fitted by a monoexponential for all temperatures ≤120 K and ≥250 K. In the intermediate temperature range 120 K<T<250 K (see for instance the 150 K decay in Fig. 2), the ground state PL is
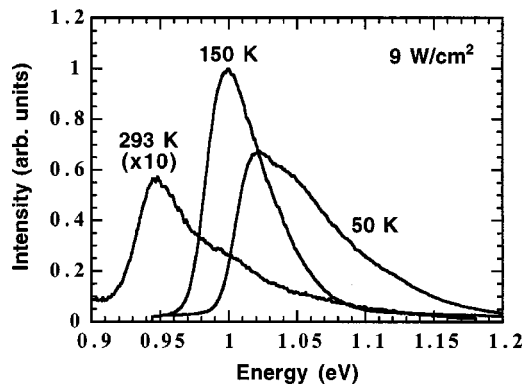

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FIG. 1. cw photoluminescence spectra of a single InAs QD layer embedded in a 20 nm $In_{0.15}Ga_{0.85}As$ QW. The pump intensity and energy are $I_{pump}=9$ W/cm^2 and $E_{pump}=1.59$ eV.

FIG. 3. PL decay time (filled dots, left axis) and integrated intensity (empty squares, right axis) as a function of temperature. The decay time is obtained from a monoexponential fit to the ground state PL ($I_{pump}=9.4$ W/cm^2, $E_{pump}=1.55$ eV), whereas the integrated intensity is measured in a cw experiment with $I_{pump}=9$ W/cm^2 and $E_{pump}=1.59$ eV.

initially constant over the first 1–2 ns. This behavior may be due to thermalization among different QDs, the low-energy QDs being refilled by carriers escaped from high-energy QDs. In support of this interpretation, we notice that in this temperature range thermalization among different QDs is observed also in cw PL experiments (see Fig. 1), implying that the carrier escape time from the QDs is becoming comparable to the interband recombination lifetime. Thermalization between dots was also observed in short-wavelength InAs/GaAs QDs[10–12] and is responsible for reduced lasing linewidth and increased efficiency at high temperatures in QD lasers.[13,14]

Figure 3 shows the measured lifetime at the ground state energy as a function of temperature (filled dots, left axis). The decay times in the intermediate temperature range 120 K$<T<$250 K are obtained by fitting on the last portion of the decay curve, with a correspondingly larger fit uncertainty. The lifetime is 1.2 ns independent of temperature for $T\leq50$ K, increases with temperature up to ≈2 ns at 150 K, then decreases to 1.8 ns at RT. The constant lifetime value at low temperatures and the subsequent linear increase with temperature has been predicted theoretically[15] and observed before in self-assembled QDs.[16,17] The lateral confinement in the QDs produces nonzero (i.e., nonradiative) in-plane wave vector $k_\parallel$ components in the exciton wave function, thus reducing the radiative emission rate as compared to QWs.[15] As the temperature increases, excited states having larger values of $k_\parallel$ become populated, and the radiative rate decreases further. The measured low-temperature lifetime of 1.2 ns is
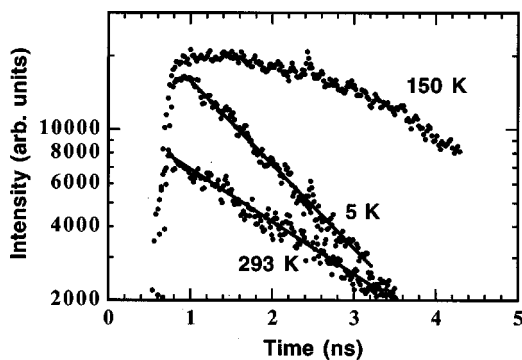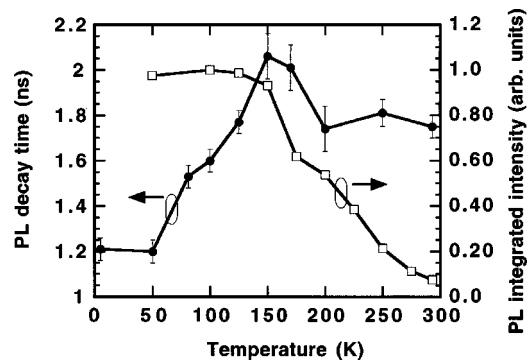
much larger than the value calculated by using Citrin's formula,[15] $\tau_{QD}(T=0$ K$)\approx370$ ps. This discrepancy may be related to the effect of the specific shape of InAs/InGaAs QDs and of piezoelectric fields on the ground state wave function.[18] The slight lifetime decrease at temperatures ≥200 K can be attributed to nonradiative recombination. Nevertheless, the lifetime remains rather large up to RT, demonstrating the high structural quality of these embedded QDs. This should be contrasted with recent measurements of ≈100 ps nonradiative lifetime at room temperature in short-wavelength InGaAs/InAs QDs[19] and ≈600 ps in long-wavelength QDs obtained by cycled submonolayer deposition.[20] On the right axis of Fig. 3 the temperature dependence of the integrated cw PL intensity is plotted (empty squares), for a pump intensity $I_{pump}=9$ W/cm^2. In order to explain the factor of 13 decrease in radiative efficiency at RT along with the relatively large decay time, we must assume either a long RT radiative lifetime of ≈18 ns, or the existence of nonradiative traps in the (In)GaAs matrix around the QDs, which would capture the carriers before they relax into the QDs.

Figure 4 shows the the rise dynamics of the QD PL at 5 K, for an average pump intensity of 300 W/cm^2, corresponding to 3.1×10^{12} cm^{-2} excited electron–hole pairs per pulse. At this excitation level, the first 3–4 states in the dots are filled by the pump pulse. The continuous, dashed, and dotted

FIG. 2. Time-resolved PL decays as detected at the QD ground state for each temperature (average $I_{pump}=9.4$ W/cm^2, $E_{pump}=1.55$ eV, detection window=10 meV). The continuous lines show monoexponential fits.
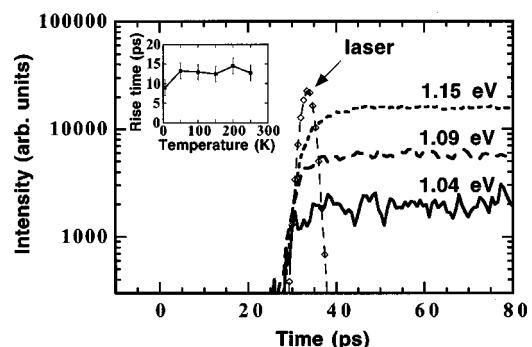

FIG. 4. PL rise dynamics at detection energies corresponding to the three lowest energy states in the QDs at 5 K and under strong excitation ($I_{pump}=300$ W/cm^2, $E_{pump}=1.55$ eV, detection energy window=40 meV). The dashed line represents the scattered pump laser pulse. Inset: Ground state rise time vs temperature ($I_{pump}=300$ W/cm^2).

lines represent the PL detected at energies corresponding to the ground, first, and second excited states, respectively, integrated over a 40 meV energy range. The PL signals first rise with a time constant close to the system resolution (FWHM of laser pulse=3.5 ps), then approach the final value with a smaller slope. The rise time, defined as the time from 10% to 90% of the maximum signal, is the same for the three energies within the system resolution, $\tau_{rise}=(10\pm2)$ ps. This value is comparable to the rise times reported for short-wavelength InAs/GaAs QDs at similar excitation levels.[19,21,22] The simultaneous rise of the PL signal from different states clearly proves that the relaxation time τ_0 between states in the dots is not the limiting factor in the PL rise, i.e., $\tau_0 \ll \tau_{rise}$. Since τ_{rise} is also much shorter than the measured carrier lifetime, we conclude that there is no relaxation bottleneck in these QDs, i.e., all carriers relax to the ground state before recombining. We measured a similar rise dynamics at excitation levels down to 10^{11} cm^{-2} photocreated electron–hole pairs per pulse. Lower excitation levels were not achievable due to the detector sensitivity. We suggest that the fast intraband relaxation time τ_0 is due to Auger relaxation. In fact, Auger relaxation was shown to be more effective than multiphonon relaxation for excitation densities corresponding to ≥ 1 electron–hole pair per dot per pulse in InGaAs/GaAs self-assembled QDs.[21] This also agrees with the theoretical prediction of Ferreira and Bastard,[23] who have calculated relaxation times $\tau_0 \approx 0.1-10$ ps for an Auger process involving only two particles in a QD. The inset in Fig. 4 shows the ground state rise time as a function of temperature, for $I_{pump}=300$ W/cm^2. The rise dynamics is roughly independent of temperature up to 250 K, the maximum measurement temperature allowed by the system sensitivity. We also obtained the same rise time when pumping with a photon energy below the GaAs band gap. This indicates that the measured rise time is not related to carrier diffusion inside the GaAs matrix, but more likely to carrier capture from the wetting layer into the QDs. We stress that this fast carrier relaxation makes embedded InAs/InGaAs QDs suitable for the realization of lasers with large modulation band widths.

In conclusion, we have measured the rise and decay dynamics of long-wavelength QDs obtained by embedding InAs QDs within an InGaAs QW. The PL spectra show evidence of thermalization among the different dots at temperatures ≥ 150 K. The decay time increases from 1.2 ns at low temperature to 1.8 ns at 293 K. This long carrier lifetime proves that nonradiative recombination inside the QDs and carrier evaporation to the wetting layer are not critical to the operation of long-wavelength QD-based devices even at room temperature. The short PL rise time $\tau_{rise}=(10\pm2)$ ps, almost independent of temperature, shows a very rapid, possibly Auger-mediated intraband relaxation and the potential for high modulation speeds in 1.3 μm QD lasers.

[1] K. Mukai, N. Ohtsuka, M. Suguwara, and S. Yamazaki, Jpn. J. Appl. Phys., Part 2 **33**, L1710 (1994).

[2] R. P. Mirin, J. P. Ibbetson, K. Nishi, A. C. Gossard, and J. E. Bowers, Appl. Phys. Lett. **67**, 3795 (1995).

[3] D. L. Huffaker, G. Park, Z. Zou, O. B. Shchekin, and D. G. Deppe, Appl. Phys. Lett. **73**, 2564 (1998).

[4] J. M. Gérard, A. Lemaître, B. Legrand, A. Ponchet, B. Gayral, and V. Thierry-Mieg, J. Cryst. Growth **201**, 1109 (1999).

[5] K. Nishi, H. Saito, S. Sugou, and J.-S. Lee, Appl. Phys. Lett. **74**, 1111 (1999).

[6] V. M. Ustinov, N. A. Maleev, A. E. Zhukov, A. R. Kovsh, A. Y. Egorov, A. V. Lunev, B. V. Volovik, I. L. Krestnikov, Y. G. Musikhin, N. A. Bert, P. S. Kop'ev, Z. I. Alferov, N. N. Ledentsov, and D. Bimberg, Appl. Phys. Lett. **74**, 2815 (1999).

[7] Y. M. Shernyakov, D. A. Bedarev, E. Y. Kondrat'eva, P. S. Kop'ev, A. R. Kovsh, N. A. Maleev, M. V. Maximov, S. S. Mikhrin, A. F. Tsatsul'nikov, V. M. Ustinov, B. V. Volovik, A. E. Zhukov, Z. I. Alferov, N. N. Ledentsov, and D. Bimberg, Electron. Lett. **35**, (1999).

[8] K. Mukai, Y. Nakata, K. Otsubo, M. Sugawara, N. Yokoyama, and H. Ishikawa, IEEE Photonics Technol. Lett. **11**, 1205 (1999).

[9] S. Fafard, S. Raymond, G. Wang, R. Leon, D. Leonard, S. Charbonneau, J. L. Merz, P. M. Petroff, and J. E. Bowers, Surf. Sci. **362**, 778 (1996).

[10] A. Patanè, M. G. Alessi, F. Intonti, A. Polimeni, M. Capizzi, F. Martelli, M. Geddo, A. Bosacchi, and S. Franchi, Phys. Status Solidi A **164**, 493 (1997).

[11] L. Brusaferri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri, and S. Franchi, Appl. Phys. Lett. **69**, 3354 (1996).

[12] H. Htoon, H. Yu, D. Kulik, J. W. Keto, O. Baklenov, J. A. L. Holmes, B. G. Streetman, and C. K. Shih, Phys. Rev. B **60**, 11 026 (1999).

[13] H. Shoji, Y. Nakata, K. Mukai, Y. Sugiyama, M. Sugawara, N. Yokoyama, and H. Ishikawa, IEEE J. Sel. Top. Quantum Electron. **3**, 188 (1997).

[14] A. Patanè, A. Polimeni, M. Henini, L. Eaves, P. C. Main, and G. Hill, J. Appl. Phys. **85**, 625 (1999).

[15] D. S. Citrin, Superlattices Microstruct. **13**, 303 (1993).

[16] G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz, and P. M. Petroff, Appl. Phys. Lett. **64**, 2815 (1994).

[17] H. Yu, S. Lycett, C. Roberts, and R. Murray, Appl. Phys. Lett. **69**, 4087 (1996).

[18] O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999).

[19] S. Marcinkevicius and R. Leon, Phys. Rev. B **59**, 4630 (1999).

[20] L. Zhang, T. F. Boggess, D. G. Deppe, D. L. Huffaker, O. B. Shchekin, and C. Cao, Appl. Phys. Lett. **76**, 1222 (2000).

[21] B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, Phys. Rev. B **54**, 11 532 (1996).

[22] C. Lobo, R. Leon, S. Marcinkevicius, W. Yang, P. C. Sercel, X. Z. Liao, J. Zou, and D. J. H. Cockayne, Phys. Rev. B **60**, 16647 (1999).

[23] R. Ferreira and G. Bastard, Appl. Phys. Lett. **74**, 2818 (1999).