

Spectral engineering of carrier dynamics in In(Ga)As self-assembled quantum dots

Thomas F. Boggess^{a)} and L. Zhang

Optical Science and Technology Center, Department of Physics and Astronomy, University of Iowa, Iowa City, Iowa 52242

D. G. Deppe, D. L. Huffaker, and C. Cao

Microelectronics Research Center, Department of Electrical and Computer Engineering, The University of Texas at Austin, Austin, Texas 78712-1084

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Time-resolved photoluminescence upconversion with 200 fs resolution is used to investigate the carrier capture, energy relaxation, and radiative recombination in two self-assembled quantum-dot ensembles with distinctly different sizes and energy spectra. When carriers are excited into the wetting layer at low density and low lattice temperature, the relaxation time to the ground state of the larger dots is ~ 1 ps, but the corresponding time for the smaller dots with larger energy spacings is ~ 7 ps. This, along with the observed temperature dependence, suggests phonon participation in the relaxation process. At low temperatures, the radiative recombination time in the smaller dots is approximately twice that of the larger dots. The reduced oscillator strength in the smaller dots may be due to a reduced electron-hole wave-function overlap in the smaller dots, in addition to a size-dependent super-radiance effect. © 2001 American Institute of Physics.

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Innovations in the controlled growth of InAs/GaAs and InGaAs/GaAs self-assembled quantum dots (SAQDs) have recently led to their implementation as the active region of high-performance lasers.¹⁻⁸ The gain and maximum modulation rate of these lasers are intimately coupled to the radiative recombination and carrier energy relaxation rates in the SAQDs. The capture of carriers from the wetting layer and their subsequent relaxation through the discrete states of the SAQDs are particularly interesting problems, as the reported time scales for such processes vary from approximately a picosecond to hundreds of picoseconds.⁹⁻²²

The observation of slower relaxation times has been pointed to as evidence of the phonon bottleneck.^{23,24} On the other hand, a substantial body of research exists, including SAQD laser studies, that indicates that relaxation within SAQDs proceeds at a rate greatly exceeding that originally thought possible in the presence of a phonon bottleneck. Several explanations have been posed for this fast relaxation, including multiphonon emission,²² Auger processes,²⁵ and electron-hole scattering.²⁶

The wide range of reported values for the energy relaxation time in SAQDs is likely a result of multiple factors. For instance, reported results vary with regard to experimental conditions, such as lattice temperature, excitation intensity, and excitation energy. In addition, varying growth conditions lead to SAQDs with differing size, shape, ground-state energy, and electron and hole potential depth. This results in wide variations in the number, degeneracy, and distribution of confined energy levels in the SAQDs. While this adds complexity to the problem of understanding the carrier dynamics in the SAQDs, it also points to the potential for engineering SAQD laser properties by manipulation of the en-

ergy spectrum through the control of dot size and composition. Here, we demonstrate dramatically differing carrier dynamics in SAQDs with distinctly different energy spectra.

SAQDs are grown in a thin (2000 Å) GaAs region bounded by AlAs diffusion barriers. One sample consists of two layers of large (350 Å diam by 110 Å height after covering) InGaAs/GaAs dots with a density of 10^{10} cm⁻². The alloy composition during growth is 50%, but the center of the dots is indium rich, resulting in nearly parabolic potential wells.²⁷ The two SAQD layers are separated by 1000 Å. The photoluminescence spectra, shown in Fig. 1(a), illustrate that the 300 K ground-state emission occurs at 1.32 μm and that multiple emission peaks are observed for high excitation. Indium grading results in transition energies that are nearly equally spaced with an average separation of ~ 65 meV. Any effect of indium grading on the carrier dynamics will likely be manifest in the emission spectrum. The second sample consists of a single layer of much smaller (250 Å diam by 30 Å height after covering) InAs/GaAs SAQDs with a density of 3×10^{10} cm⁻². As shown in Fig. 1(b), these dots exhibit ground-state emission at 1.23 μm and, for high excitation, multiple emission peaks with much larger separation relative to the larger SAQDs (~ 93 meV).

Time-resolved photoluminescence upconversion measurements with ~ 200 fs resolution were conducted under similar conditions for each sample to examine both the relaxation time to the ground state and the radiative recombination. Carriers are excited below the GaAs band edge (i.e., into the wetting layer) and the SAQD ground-state emission is upconverted in a 0.5-mm-thick LiIO₃ crystal to provide the temporal resolution. The upconverted radiation is passed through a monochromator prior to detection to provide ~ 10 meV spectral resolution. The time dependence of the ground-

^{a)}Electronic mail: thomas-boggess@uiowa.edu

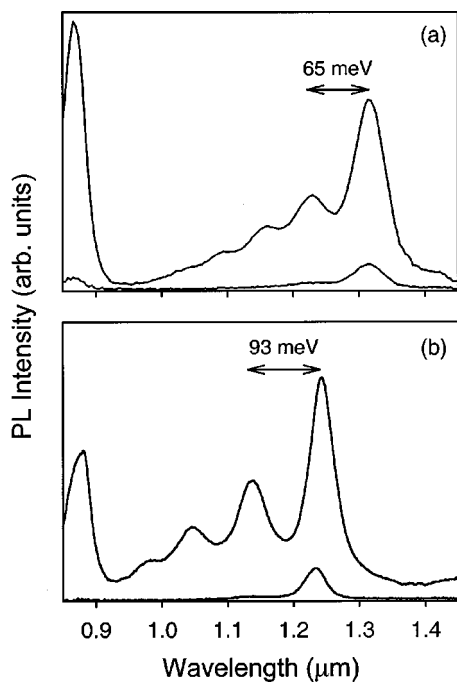


FIG. 1. 300 K photoluminescence for (a) large InGaAs SAQDs and (b) small InAs SAQDs under 800 nm continuous-wave excitation. In each case, spectra are shown at low excitation, where emission only from the ground state is observed, and significantly higher excitation, where excited-state emission is evident. The feature below 900 nm is associated with emission from GaAs.

state emission is measured as a function of lattice temperature and excitation level for each of the two samples.

The initial rise of the ground-state emission for each sample is well described by a simple exponentially increasing function, and fits to the data provide a characteristic rise time. The rise time as a function of temperature for each sample is shown in Fig. 2. Each data point represents the average of measurements conducted over the range of excitation fluences indicated in the insets. There are several important features evident in these data. First, as we have reported previously,²⁸ the rise time for the large SAQDs is extremely short, particularly at lower temperatures. Second, it is clear that at all temperatures the relaxation times for the smaller SAQDs are significantly longer. Third, the temperature dependencies of the rise times are distinctly different for the two structures.

A further significant feature is shown in the insets in Fig. 2, i.e., the observed rise times (for both structures) show little, if any, dependence on excitation fluence for the range of fluences investigated. This suggests that the rapid rise times are not associated with Auger-like processes involving carriers in the wetting layer. On the other hand, it is possible that intradot electron-hole scattering may account for the fast relaxation. It is evident, however, that in the presence of the smaller energy-level spacing in the larger SAQDs, the relaxation rate is enhanced. This behavior is suggestive of phonon participation. If we take the electron effective mass as $0.04 m_0$, the energy separation between the low-lying *electron* states is estimated to be 33 meV. Given the observed spacing between optical transitions, the separation between optically active *hole* states would be comparable. This energy separation, which again is nearly constant for all ob-

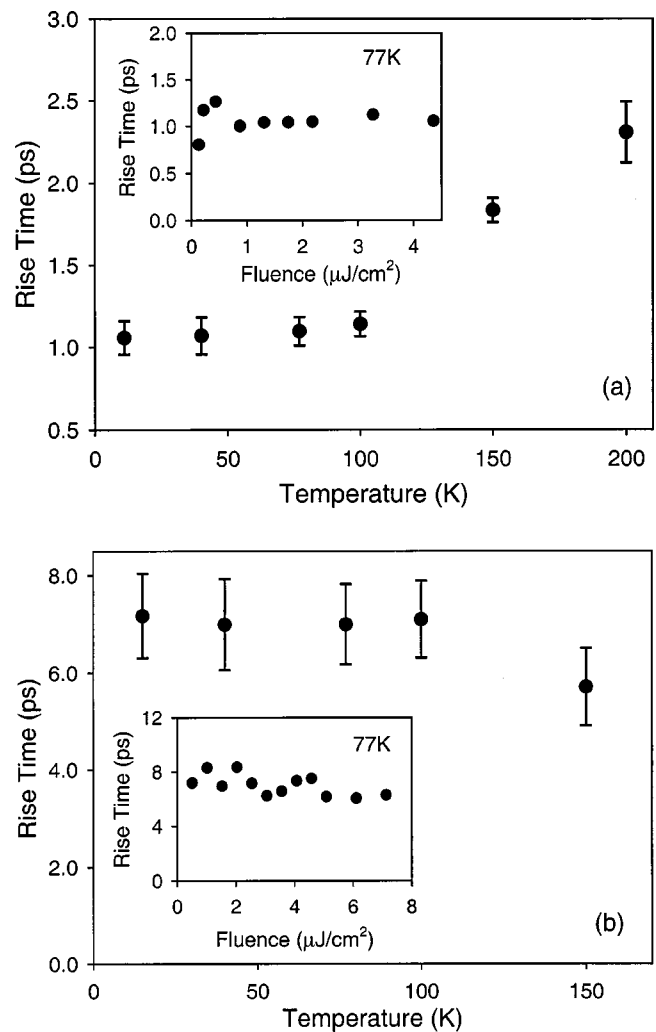


FIG. 2. Rise time of the ground-state emission as a function of temperature for (a) the large InGaAs SAQDs and (b) the small InAs SAQDs. Each data point represents an average photoluminescence rise time for the range of excitation fluences shown in the corresponding inset. The insets provide example fluence-dependent data at 77 K, illustrating that the rise time is essentially independent of carrier density.

served transitions, is nearly equal to the LO phonon energy measured in InGaAs SAQDs.²⁹ This indicates that relaxation through optical phonon emission is a possible explanation of the extraordinarily fast low-density relaxation times measured in the InGaAs SAQDs. Conversely, the relaxation process is clearly impeded in the structure with larger energy-level separations, which is suggestive of a phonon bottleneck, but one that is not nearly as severe as that originally predicted.^{23,24}

The temperature dependence of the rise time for the two structures is also strikingly different, with the larger dots exhibiting an increase in relaxation time with increasing temperature and the smaller dots showing the opposite behavior at the highest temperature. A detailed rate equation³⁰ analysis including the phonon bath shows that the temperature dependence of the relaxation dynamics depends critically on the degree of thermal coupling between the wetting layer and the high-energy SAQD states involved in the capture process. If the coupling is weak, the relaxation rate will increase with increasing temperature due to stimulated phonon emission. Conversely, with strong coupling the rate will decrease

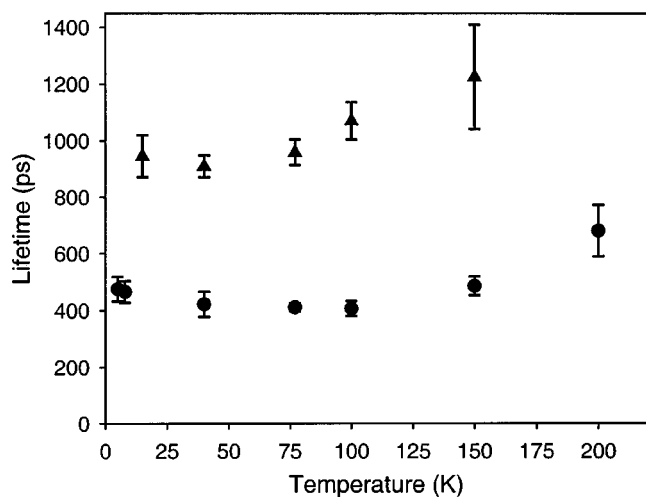


FIG. 3. Radiative lifetime as a function of temperature for the InGaAs (circles) and InAs (triangles) SAQDs illustrating the decreased radiative lifetime for the larger InGaAs SAQDs. Each data point represents an average lifetime measured over numerous excitation fluences. The larger error bars for the highest temperatures reflect a slight increase in the recombination time with density at these temperatures.

with increasing temperature because of thermal reexcitation from high-energy confined states into the wetting layer.³⁰

As illustrated in Fig. 3, we also find that the radiative recombination rate in the SAQDs is size dependent. Again, these data were taken in the low-excitation regime. At low temperatures, the radiative lifetime in the smaller dots is roughly twice that of the larger dots. This indicates a smaller oscillator strength for the smaller dots. A reduced overlap of the electron-hole wave function in the smaller SAQDs may be partly responsible for the lifetime change, but it has also been suggested that the radiative lifetime in SAQDs is size dependent due to super-radiance.³¹ The temperature dependence to the recombination is similar for the two samples; both exhibit a lifetime that increases with temperature. This has been reported previously^{16,28,32,33} and is attributed to the increasing occupation of excitonic dark states with increasing temperature.³⁴ At the highest temperatures, we observe a modest increase in lifetime with density, which is reflected in the larger error bars for the highest temperatures. This is also consistent with greater occupation of dark states under these conditions.

In summary, we have shown that when grown and measured under similar conditions two SAQDs with dramatically different sizes and energy spectra display significantly different relaxation and recombination dynamics. The results suggest an important role for phonons in the energy relaxation within these SAQDs. The shorter radiative lifetime for the larger SAQDs indicates a reduced oscillator strength arising from greater electron wave-function barrier penetration in the growth direction for the smaller dots, and is consistent with predictions of super-radiance. These studies indicate the potential for optimization of 1.3 μm SAQD lasers through size- and composition-controlled growth.

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- ¹N. Kirkstædter, N. Ledentsov, M. Grundmann, D. Bimberg, V. Ustinov, S. Ruvimov, M. Maximov, P. Kop'ev, and Zh. Alferov, *Electron. Lett.* **30**, 1416 (1994).
- ²H. Shoji, K. Mukai, N. Ohtsuka, M. Sugawara, T. Uchida, and H. Ishikawa, *IEEE Photonics Technol. Lett.* **7**, 1385 (1995).
- ³Q. Xie, A. Kalburge, P. Chen, and A. Madhukar, *IEEE Photonics Technol. Lett.* **8**, 965 (1996).
- ⁴K. Kamath, P. Bhattacharya, T. Sosnowski, T. Norris, and J. Phillips, *Electron. Lett.* **32**, 1374 (1996).
- ⁵D. L. Huffaker, G. Park, Z. Zou, O. B. Shchekin, and D. G. Deppe, *Appl. Phys. Lett.* **73**, 2564 (1998).
- ⁶L. F. Lester, A. Stintz, H. Li, T. C. Newell, E. A. Pease, B. A. Fuchs, and K. J. Malloy, *IEEE Photonics Technol. Lett.* **11**, 931 (1999).
- ⁷K. Mukai, N. Ohtsuka, H. Shoji, M. Sugawara, N. Yokoyama, and H. Ishikawa, *IEEE Photonics Technol. Lett.* **11**, 1205 (1999).
- ⁸G. Park, O. B. Shchekin, S. Csutak, D. L. Huffaker, and D. G. Deppe, *Appl. Phys. Lett.* **75**, 3267 (1999).
- ⁹S. Fafard, R. Leon, D. Leonard, J. L. Merz, and P. M. Petroff, *Phys. Rev. B* **52**, 5752 (1995).
- ¹⁰F. Adler, M. Geiger, A. Bauknecht, F. Scholz, H. Schweizer, M. H. Pikuhn, B. Ohnesorge, and A. Forchel, *J. Appl. Phys.* **80**, 4019 (1996).
- ¹¹R. Heitz, I. Mukhametzanov, H. Born, M. Grundmann, A. Hoffman, A. Madhukar, and D. Bimberg, *Physica B* **272**, 8 (1999).
- ¹²B. Ohnesorge, M. Albrecht, J. Oshinowo, A. Forchel, and Y. Arakawa, *Phys. Rev. B* **54**, 11532 (1996).
- ¹³K. Mukai, N. Ohtsuka, H. Shoji, and M. Sugawara, *Appl. Phys. Lett.* **68**, 3013 (1996).
- ¹⁴S. Raymond, S. Fafard, P. J. Poole, A. Wojs, P. Hawrylak, S. Charbonneau, D. Leonard, R. Leon, P. M. Petroff, and J. L. Merz, *Phys. Rev. B* **54**, 11548 (1996).
- ¹⁵Z. L. Yuan, E. R. A. D. Foo, J. F. Ryan, D. J. Mowbray, M. S. Skolnick, and M. Hopkinson, *Physica B* **272**, 12 (1999).
- ¹⁶H. Yu, S. Lycett, C. Roberts, and R. Murray, *Appl. Phys. Lett.* **69**, 4087 (1996).
- ¹⁷W. Yang, R. R. Lowe-Webb, H. Lee, and P. C. Sercel, *Phys. Rev. B* **56**, 13314 (1997).
- ¹⁸F. Adler, M. Geiger, A. Bauknecht, D. Haase, P. Ernst, A. Dornen, F. Scholz, and H. Schweizer, *J. Appl. Phys.* **83**, 1631 (1998).
- ¹⁹S. Marcinkevicius and R. Leon, *Phys. Rev. B* **59**, 4630 (1999).
- ²⁰R. Heitz, M. Grundman, N. N. Ledentsov, L. Ecke, M. Veit, D. Bimberg, V. M. Ustinov, A. Y. Egorov, A. E. Zhukov, P. S. Kop'ev, and Z. I. Alferov, *Appl. Phys. Lett.* **68**, 361 (1996).
- ²¹T. S. Sosnowski, T. B. Norris, H. Jiang, J. Singh, K. Kamath, and P. Bhattacharya, *Phys. Rev. B* **57**, R9423 (1998).
- ²²R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, and Zh. I. Alferov, *Phys. Rev. B* **56**, 10435 (1997).
- ²³U. Bockelmann and G. Bastard, *Phys. Rev. B* **42**, 8947 (1990).
- ²⁴H. Benisty, C. M. Sotomayor-Torres, and C. Weisbuch, *Phys. Rev. B* **44**, 10945 (1991); H. Benisty, *ibid.* **51**, 13281 (1995).
- ²⁵U. Bockelmann and T. Egeler, *Phys. Rev. B* **46**, 15574 (1992).
- ²⁶R. Ferreira and G. Bastard, *Appl. Phys. Lett.* **74**, 2818 (1999).
- ²⁷S. Krishna, D. Zhu, J. Xu, K. K. Linder, O. Qasimeh, P. Bhattacharya, and D. L. Huffaker, *J. Appl. Phys.* **86**, 6135 (1999).
- ²⁸L. Zhang, T. F. Boggess, D. G. Deppe, D. L. Huffaker, O. B. Shchekin, and C. Cao, *Appl. Phys. Lett.* **76**, 1222 (2000).
- ²⁹Y. Toda, O. Moriwaki, M. Nishioka, and Y. Arakawa, *Phys. Rev. Lett.* **82**, 4114 (1999).
- ³⁰D. G. Deppe, D. L. Huffaker, C. Cao, T. F. Boggess, and L. Zhang (unpublished).
- ³¹E. Sugawara, *Phys. Rev. B* **51**, 10743 (1995).
- ³²G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz, and P. M. Petroff, *Appl. Phys. Lett.* **64**, 2815 (1994).
- ³³M. Paillard, X. Marie, E. Vanelle, T. Amand, V. K. Kalevich, A. R. Kovsh, A. E. Zhukov, and V. M. Ustinov, *Appl. Phys. Lett.* **76**, 76 (2000).
- ³⁴H. Gotoh, H. Ando, and T. Takagahara, *J. Appl. Phys.* **81**, 1785 (1997).