Reprinted from

JAPANESE JOURNAL OF
APPLIED
PHYSICS

RAPID COMMUNICATION

Carrier Relaxation in InAs/InGaAs Dots-in-a-Well Structures

Rui Chen, Hui-Yun Liu, and Han-Dong Sun

Jpn. J. Appl. Phys. **49** (2010) 020203

Japanese Journal of Applied Physics **49** (2010) 020203 **RAPID COMMUNICATION**

Carrier Relaxation in InAs/InGaAs Dots-in-a-Well Structures

Rui Chen, Hui-Yun Liu[1], and Han-Dong Sun*

Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore
[1]*Department of Electronic and Electrical Engineering, University College London, Torrington Place, London, WC1E 7JE, United Kingdom*

Received October 29, 2009; accepted December 15, 2009; published online February 5, 2010

We report on the mechanism of electronic structure and different carrier relaxation processes in InAs/$In_xGa_{1-x}As$ dots-in-a-well (DWELL) structure investigated by comprehensive spectroscopic characterization. Selectively excited photoluminescence and photoluminescence excitation analyses reveal that when excited at different photon energies, carriers relax to the ground state of the quantum dots by distinct schemes. Our investigation clearly manifests the roles of longitudinal optical phonons and absorption continuum states played in the carrier relaxation process in DWELL structures.

DOI: 10.1143/JJAP.49.020203

Self-assembled InAs quantum dots (QDs) have been the object of intensive studies due to the potential laser device applications for optical fiber telecommunication. Long wavelength InAs QDs emission have been achieved by atomic layer epitaxy (ALE), covering InAs QDs with InGaAs and/or AlAs cap layer,[1)] or by strain compensation with GaNAs capping layers.[2,3)] Of the many techniques proposed and investigated, another approach is growing the dots within an InGaAs quantum well (QW), i.e., to form a so-called quantum dots-in-a-well (DWELL) structure.[4)] By embedding the QDs in a InGaAs QW, carrier capture into the QDs is greatly increased.[5)] After years of investigation and exploration, photodetectors and laser devices based on InAs/InGaAs DWELL structure have been demonstrated.[6–8)] However, electronic structure and carrier relaxation process in this structure has never been studied. Such investigation is crucial not only for high speed device performance but for understanding the fundamental physics of zero-dimensional system.

Processes of carrier relaxation have been controversially discussed over a decade for the conventional InAs/GaAs QDs.[9–16)] For relatively more complicated DWELL structure, however, there has been no report in this respect. In this letter, we investigate carrier relaxation processes in InAs/InGaAs DWELL structure by detailed selectively excited photoluminescence (SEPL) and photoluminescence excitation (PLE) measurements. We observed phenomena of carrier relaxation with and without multiple longitudinal (LO) phonon assistance. Different mechanisms of carrier excitation and relaxation in the InAs/InGaAs DWELL structure are analyzed and discussed in detail. Our investigation sheds light on the relevance between the electronic structure and carrier relaxation.

The sample used herein was grown in a VG Semicon V80H molecular beam epitaxy (MBE) system on Si-doped GaAs(100) substrates. QDs were formed by 2.9 monolayers (MLs) InAs grown upon a 2 nm $In_{0.12}Ga_{0.88}As$ strained buffer layer, and then covered with a 6 nm $In_{0.12}Ga_{0.88}As$ strain-reducing layer. After the DWELL structure growth at 505 °C, the nanostructure was capped with 15 nm GaAs follow by a 120 s long growth interruption and further GaAs cap growth at 615 °C. Dot density was estimated to be around 3.1×10^{10} cm^{-2} from atomic force microscopy (AFM) measurement. Detailed growth and characterization of this structure can be found in refs. 7, 17, and 18. The optical characterizations were performed at 10 K within a closed cycle helium cryostat. A cw He–Cd laser emitting at 442 nm was used for PL excitation source and the signal was detected by a Peltier cooled InGaAs photodiode using standard lock-in amplifier technique. The SEPL and PLE signal was detected using the same system, but the excitation source was replaced by a 250 W tungsten halogen lamp combined with a 0.27 m grating monochromator and suitable filters.[19,20)]

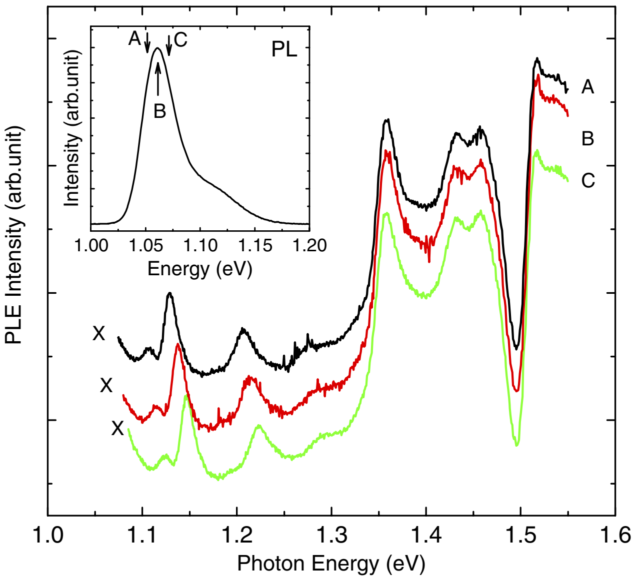

Fig. 1. (Color online) PLE spectra of the sample taken at 10 K and the crosses represent the detection energies. Inset is the PL excited by 442 nm, the arrows also denote different detection positions of PLE spectra.

Figure 1 shows the PLE and PL spectra (inset, excitation $\lambda = 442$ nm) of the investigated sample taken at 10 K, where the crosses and arrows in the inset indicate different detection energies for PLE spectra. Excitation power density was controlled as low as 0.07 W cm^{-2} for PL measurement to avoid excited state emission due to the saturation of the ground state (GS). Upon excitation above the GaAs band gap, the GS transition of the InAs QD leads to PL peaks at 1.06 eV and an asymmetrical PL line shape is observed due to the inhomogeneity of the QDs ensemble. For the PLE spectra, the "resonant-like" peaks in the low energy region (1.10–1.33 eV) are sensitive to the detection energy, which

---

*E-mail address: hdsun@ntu.edu.sg

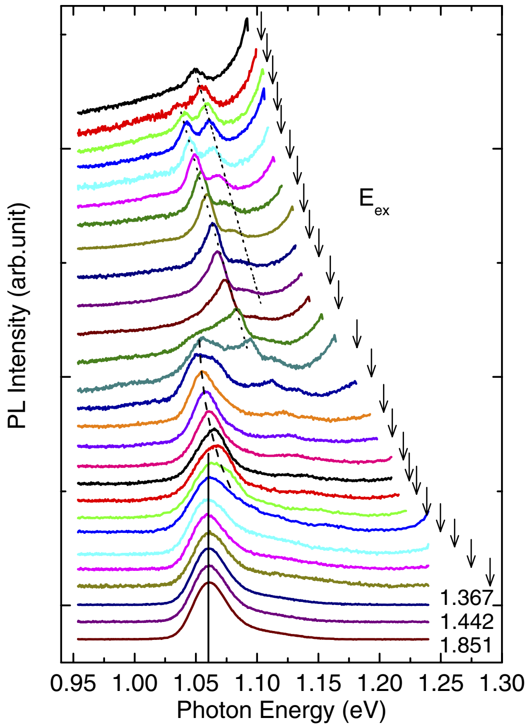

Fig. 2. (Color online) PL spectra of the sample under different excitation energy. The arrows on the right hand side indicate the positions of excitation energies for SEPL.

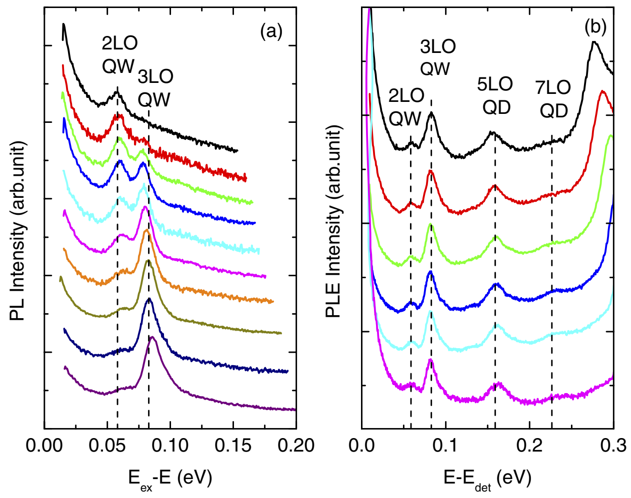

Fig. 3. (Color online) (a) SEPL spectrum of the sample plotted against the difference between the excitation energy and emergent photon energy ($E_{ex} - E$). (b) PLE spectrum as a function of energy difference between incident photon energy and the detection energy ($E - E_{det}$).

we will show below to result from selective carrier relaxation assisted by multiple LO-phonons. Comparatively, the shape of the PLE spectra in higher energy region is independent of the detection energy. The behavior in this "non-resonant" excitation region reflects the optical absorption of the surrounding InGaAs QW and GaAs, and is unrelated to the size of the QD in the sample.

Figure 2 depicts the detailed PL spectra of the sample under various excitation energies. The spectra are normalized and shifted vertically for better clarity, with respective excitation energies of SEPL indicated by the arrows. A closer inspection at the spectra reveals three different carrier relaxation schemes. Firstly, for excitation energy lower than 1.153 eV, the spectra consist of a series of sharp features (indicating by dotted lines), the energy of which depend upon excitation energy. However, for slightly higher excitation energy, those sharp features gradually disappear and are replaced by a broad peak, which also shift with the excitation energy. For excitation energy higher than 1.24 eV, the PL spectra maintains the same lineshape with peak energy at 1.06 eV marked by the solid line. By comparison with the inset of Fig. 1, the emissions located at 1.06 eV are exactly the same and can be attributed to the GS transition from the whole QDs ensemble.

The low energy SEPL (excitation energy lower than 1.153 eV) and PLE spectra are plotted in Fig. 3 as a function of energy difference between the excitation energy and emergent photon energy ($E_{ex} - E$), and incident photon energy and the detection energy ($E - E_{det}$). Clear resonant peaks are observed indicated by vertical dashed lines. It is well established that in zero-dimensional QD system, carrier relaxation can not simply achieved by interaction with acoustic phonons due to larger separation between discrete energy levels.[16] In SEPL spectra, only those dots having a energy level spacing equal to an integer multiple of the LO phonon energy are the only ones that can relax to the GS by LO phonon emission and contribute to the emission. The present resonant phonon peaks are 58 (2 × 29), 84 (3 × 28), 160 (5 × 32), and 227 (7 × 32.5) meV, respectively. Thus the four phonon resonant peaks can be ascribed to LO phonons in surrounding InGaAs QW or InAs QD, taking into account that the LO phonon energy is related to the strain and confinement.[21] From the calculation, the bulk InAs LO phonon energy is around 29.9 meV and has a reduction due to the strain. A LO phonon energy of 32.1 meV has been estimated from the strain distribution in InAs pyramids with {101} side facets,[21] which is in excellent agreement with the experimental value we obtained. It is also noticed that those obtained values are very close to that in ref. 13 regarding the conventional InAs/GaAs material system.

It is clearly describes in Fig. 3(a) that with changing excitation energy, first the 2LO and then the 3LO resonant peak becomes dominant, which reflects the density of QD with matching transition energy. It was suggested that the order of LO phonon resonant features in SEPL spectra scales approximately with the degree of inhomogeneity resulting from dot size and shape fluctuations.[12] A larger size fluctuation will lead to a bigger energy region of excited state from a given GS and the SEPL can show higher order LO phonon resonate peaks. The linewidth of the investigated InAs/InGaAs DWELL structure is around 37 meV under nonresonant excitation at 10 K. The SEPL spectra only reveal 2LO and 3LO features which are consistent with previous reports regarding InAs/GaAs QDs.[13,16] For PLE spectra shown in Fig. 3(b), not only the well defined 2LO and 3LO phonon resonate peaks but also the higher orders can be clearly resolved. However, corresponding features at similar energies are not found in SEPL spectra under suitable excitation energy. Previous report have noticed similar phenomenon but without explanation.[13] The origin of these features will be discussed later.

For the conventional QD system, the crossed transitions between bound state and delocalized state[22] will lead to a quasi-continuous absorption background extending downward from the WL band edge.[23,24] In the InAs/InGaAs DWELL structure, the WL together with the surrounding InGaAs QW, formed a special bi-QW which also have a quasi-continuous density-of-state (DOS). However it is different from the normal QDs system. Evidence can be extracted from PLE spectra in Fig. 1 where we notice that when the photon energy is higher than 1.24 eV, a strong absorption tail link to the surrounding InGaAs QW emerges. Just as shown in Fig. 2, when gradually increase the excitation energy, the phonon resonates peaks will be replaced by a broad peak due to the selectively GS transition. That is because when the excitation energy is high enough to reach the tail of the continuous DOS, carrier can relax with the help of the continuum state in addition to the multi-LO phonon scattering. The QDs whose upper states resonate with the excitation energy thus yield this selectively GS transition.[16] This is the reason why higher orders phonon resonate peaks are absent in the SEPL spectra. Because the suitable excitation energy for higher order phonon resonate peaks is large enough to touch the continuum tail and will not satisfy the conditions of multi-phonon resonance.

While further increasing the excitation energy above 1.24 eV, a constant broad peak will dominate the SEPL related to the GS transition from the whole QDs ensemble. For such excitation energy with higher DOS than the discrete states of QDs, carriers can roll down with the interaction between the low frequency acoustic phonons and cascade by multi-LO phonon emissions. Although the excitation energy is lower than the InGaAs QW band edge, no selective excitation will occur because of the high quasi-continuous absorption. In this regime, all different sizes of QDs will be activated and the spectrum is basically the same as the inset of Fig. 1.

In conclusion, the carrier relaxation and recombination for the InAs/InGaAs DWELL structure is investigated by combining PL, SEPL and PLE spectra. At low excitation energy regime, we observed phonon resonant peaks in both SEPL and PLE spectra which are the strong evidence for phonon assisted carrier relaxation. At high excitation energy regime, the whole QDs ensemble is excited and contributed to emission with the help of quasi-continuous absorption band. While in the medium regime, QDs are selectively activated and the photon is generated from selectively GS transition. The analyses provide detailed carrier relaxation mechanism which is important for better understanding of this material structure. Through the optimal of interaction between quantum well and wetting layer, the internal quantum efficiency of this kind of material structure can be enhanced which is crucial for photonic device application.

---

1) H. Y. Liu, B. Xu, Y. Q. Wei, D. Ding, J. J. Qian, Q. Han, J. B. Liang, and Z. G. Wang: Appl. Phys. Lett. **79** (2001) 2868.
2) X. Q. Zhang, S. Ganapathy, I. Suemune, H. Kumano, K. Uesugi, Y. Nabetani, and T. Matsumoto: Appl. Phys. Lett. **83** (2003) 4524.
3) S. Ganapathy, X. Q. Zhang, I. Suemune, K. Uesugi, H. Kumano, B. J. Kim, and T. Y. Seong: Jpn. J. Appl. Phys. **42** (2003) 5598.
4) 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** (1999) 2815.
5) H. Y. Liu, M. Hopkinson, C. N. Harrison, M. J. Steer, R. Frith, I. R. Sellers, D. J. Mowbray, and M. S. Skolnick: J. Appl. Phys. **93** (2003) 2931.
6) A. Amtout, S. Raghavan, P. Rotella, G. von Winckel, A. Stintz, and S. Krishna: J. Appl. Phys. **96** (2004) 3782.
7) H. Y. Liu, I. R. Sellers, T. J. Badcock, D. J. Mowbray, M. S. Skolnick, K. M. Groom, M. Gutierrez, M. Hopkinson, J. S. Ng, J. P. R. David, and R. Beanland: Appl. Phys. Lett. **85** (2004) 704.
8) S. Krishna, D. Forman, S. Annamalai, P. Dowd, P. Varangis, T. Tumolillo, Jr., A. Gray, J. Zilko, K. Sun, M. Liu, J. Campbell, and D. Carothers: Appl. Phys. Lett. **86** (2005) 193501.
9) J. L. Pan: Phys. Rev. B **49** (1994) 2536.
10) U. Bockelmann and T. Egeler: Phys. Rev. B **46** (1992) 15574.
11) T. H. Gfroerer, M. D. Sturge, K. Kash, J. A. Yater, A. S. Plaut, P. S. D. Lin, L. T. Florez, J. P. Harbison, S. R. Das, and L. Lebrun: Phys. Rev. B **53** (1996) 16474.
12) R. Heitz, M. Grundmann, N. N. Ledentsov, L. Eckey, 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** (1996) 361.
13) M. J. Steer, D. J. Mowbray, W. R. Tribe, M. S. Skolnick, M. D. Sturge, M. Hopkinson, A. G. Cullis, C. R. Whitehouse, and R. Murray: Phys. Rev. B **54** (1996) 17738.
14) R. Heitz, M. Veit, N. N. Ledentsov, A. Hoffmann, D. Bimberg, V. M. Ustinov, P. S. Kopev, and Z. I. Alferov: Phys. Rev. B **56** (1997) 10435.
15) M. Bissiri, G. Baldassarri Höger von Högersthal, M. Capizzi, P. Frigeri, and S. Franchi: Phys. Rev. B **64** (2001) 245337.
16) H. S. Ling, C. P. Lee, and M. C. Lo: J. Appl. Phys. **103** (2008) 124311.
17) H. Y. Liu, C. M. Tey, I. R. Sellers, T. J. Badcock, D. J. Mowbray, M. S. Skolnick, R. Beanland, M. Hopkinson, and A. G. Cullis: J. Appl. Phys. **98** (2005) 083516.
18) A. Lenz, H. Eisele, R. Timm, L. Ivanova, H. Y. Liu, M. Hopkinson, U. W. Pohl, and M. Dähne: Physica E **40** (2008) 1988.
19) H. D. Sun, A. H. Clark, S. Calvez, M. D. Dawson, P. Gilet, L. Grenouillet, and A. Million: J. Appl. Phys. **97** (2005) 033517.
20) H. D. Sun, A. H. Clark, H. Y. Liu, M. Hopkinson, S. Calvez, M. D. Dawson, Y. N. Qiu, and J. M. Rorison: Appl. Phys. Lett. **85** (2004) 4013.
21) M. Grundmann, O. Stier, and D. Bimberg: Phys. Rev. B **52** (1995) 11969.
22) A. Vasanelli, R. Ferreira, and G. Bastard: Phys. Rev. Lett. **89** (2002) 216804.
23) Y. Toda, O. Moriwaki, M. Nishioka, and Y. Arakawa: Phys. Rev. Lett. **82** (1999) 4114.
24) V. Popescu, G. Bester, and A. Zunger: Appl. Phys. Lett. **95** (2009) 023108.