AIP Applied Physics Letters

Ground state splitting of vertically stacked indium arsenide self-assembled quantum dots

Shuwei Li and Kazuto Koike

Citation: Appl. Phys. Lett. **81**, 3594 (2002); doi: 10.1063/1.1515365
View online: http://dx.doi.org/10.1063/1.1515365
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v81/i19
Published by the American Institute of Physics.

Ground state splitting of vertically stacked indium arsenide self-assembled quantum dots

Shuwei Li[a)]
Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, No. 1 Yan-an Road 130021, Changchun, People's Republic of China

Kazuto Koike
New Materials Research Center, Osaka Institute of Technology, Asahi-ku Ohmiya, Osaka 535-8585, Japan

(Received 24 May 2002; accepted 27 August 2002)

An attractive feature of vertically stacked InAs/AlGaAs quantum dots (QDs), which were buried in AlGaAs high potential barrier and spacer epilayer and grown by molecular-beam epitaxy with size-controlled growth, exhibits an unknown macroscopic quantum phenomenon (i.e., phase-change splitting of the ground state). In the vertically aligned QDs, due to many-body effect and quantum-mechanical renormalization, the electron ground state splits into a series of peaks of which the intensity gradually, systematically decreases to redshift direction with a wavelength constant. By the way, energy levels of electrons and holes might really be "seen" by deep level transient spectroscopy to which the photoluminescence experiment is in an excellent agreement. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1515365]

In quantum dots (QDs) three-dimensional confinement of carriers leads to energy level discreteness to exhibit a rich spectrum of phenomena including quantum confinement, exchange splittings, Coulomb blockade, and multiexciton transitions.[1–4] Furthermore, artificial molecules can be realized by stacking layers of self-assembled QDs,[5,6] and in this letter, the vertically coupling effect can be controlled by changing the spacer thickness and QD size. The coupling effect and many-body effect of the strongly localized electrons, to which the artificial molecules of the stacked QDs are adjacent to Si-doped GaAs layer to be able to obtain the abundant tunneling resonant electrons, lead to a quantum dephase splitting of the ground state energy level and to exhibition in a recombination behavior of the photoexcited carriers. The vertically stacked QD systems not only get the abundance and readjustment spectrum within a large wavelength range but also are strongly driven by applications of semiconductor devices, which should be advantageous in many respects. In our case, the quantum confinement of the vertically stacked QDs is different from the previous studies to which the QDs are directly stacked without high potential barrier and tunneling resonant electrons. Our vertically stacked QDs are buried in AlGaAs spacer layers to exhibit a quantum-mechanical renormalization behavior in the stacked QD electron ground state.

The size-controlled growth of the vertically stacked self-organization might improve the uniformity of the self-assembled islands with a promising way. The five-period vertically stacked samples with the size-controlled growth were grown by molecular-beam epitaxy with solid sources of Al, Ga, In, and As in a noncracking K cell.[7] The structure of QD samples consists of a 500 nm Si-doped GaAs buffer on Si-doped GaAs substrates, a 500 nm GaAs layer, a 15 nm $Al_{0.5}Ga_{0.5}As$ barrier, five period vertically stacked InAs QDs after 2 ML GaAs layer, a 50 nm $Al_{0.5}Ga_{0.5}As$ barrier, and a 15 nm GaAs cap layer. From the morphology of the first and fifth sheets and histograms of width and height of the dots shown in Fig. 1, the atomic force microscopy (AFM) data do not show the difference between the first and the last QD layers. It is reasonably believed that the vertically stacked QDs are well distributed, which can make the vertically stacked InAs QDs exhibit the effects of the strong quantum confinement and coupling. Two kinds stacked QD samples (i.e., the normal size dots and the small size dots) were grown and exhibited the different quantum confinement and coupling effects. The distributions of the normal and small size QDs are about 2.5 nm in height and about 25 nm width in size and about 2.5×10^{10} cm^{-2} in density, and about <2.5 nm height and <20 nm width in size and $<1\times10^{10}$ cm^{-2} in density, respectively.

Photoluminescence (PL) measurement is performed in a closed-cycle He cryostat (20–300 K) using a 514.5 nm line

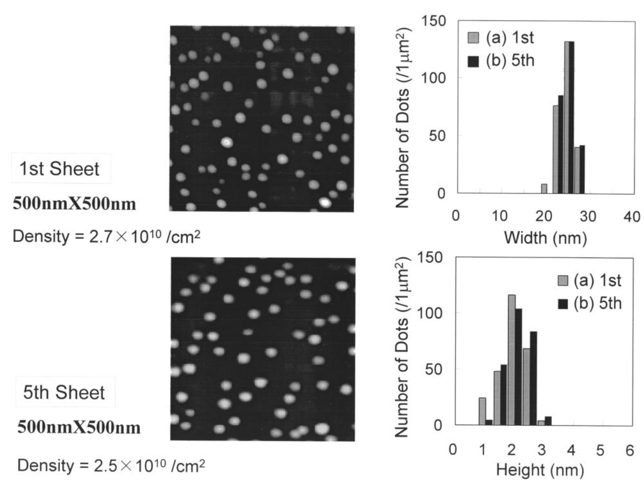

FIG. 1. Surface morphology of the first sheet and the fifth sheet quantum dots by AFM and histograms of width and height of QDs, to which the well distribution is shown.

[a)]Author to whom correspondence should be addressed; electronic mail: snmocvd@public.cc.jl.cn

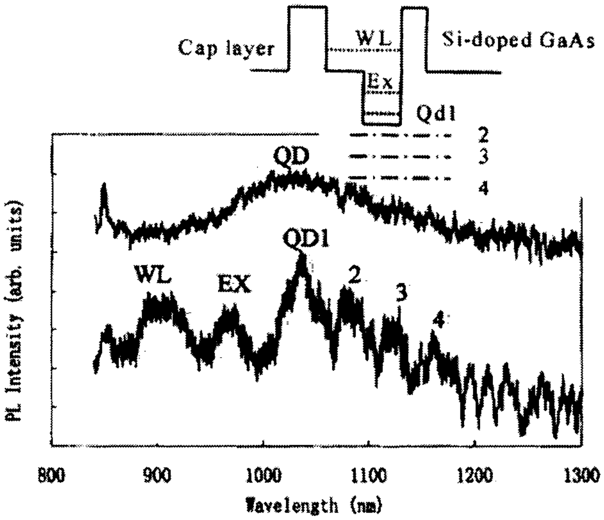

FIG. 2. Normal size PL spectra. The wetting layer (WL) ground state transition (E_{wl}-H_{wl}) is centered near 900 nm (1.38 eV), and the excited-state interband transition (E_{ex}–H_{ex}) is centered at 976 nm (1.27 eV) while the ground state transition (E_{qd}-H_{qd}) is centered near 1041 nm (1.19 eV), to which the forbidden transitions do not contribute to the PL spectrum. The temperatures of two curves are at 20 and at 60 K, the laser excitation power is 25 mW, and 20 K peak distance of the ground state splitting is about 43.7 nm. The inset is an energy diagram of the phase change splitting of the QD electron ground state.

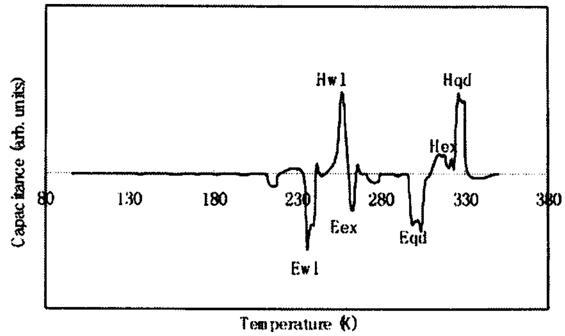

FIG. 3. Sample's DLTS spectrum. The negative peaks E_{wl}, E_{ex}, and E_{qd} represent the electron discrete energy levels of the ground states of WL, the first excited state, and the ground state of the QDs, respectively; the positive peaks H_{wl}, H_{ex}, and H_{qd} are the hole discrete energy levels of the ground state of the WL, the first excited state, and the ground state of the QDs, respectively.

Ar^+ laser as an excitation source. In Fig. 2, PL efficiency of the QDs is influenced by severe restrictions of phonon bottleneck and long relaxation lifetime of energy levels. At the low temperature the thermal population of electrons in the discrete energy states can be neglected.[8] Due to the coupling effect of the vertically aligned QD InAs islands, the strong many-body interaction gives rise to the renormalization of the dot energy levels and domination by the lowest-kinetic-energy configuration. Further, the quantum mechanically coherent wave coupled from the special electrons, which strongly is localized on the individual dots, is delocalized over the stacked QD molecule. The many-body interaction which added the abundant resonant tunneling carriers can lead to quantum dephase resulting in a splitting breakdown of the electron conduction ground states. In our theoretical calculation, the captured number of every vertically stacked QD molecular in HFET memory device is about 3 or 4,[6] which means every dot of the vertically stacked QDs can capture only 3/5 or 4/5 electron. If the abundant resonant tunneling electrons are not provided, the electrons of the many-body interaction are too small to lead the phase-change splitting. In Fig. 2, the ground state transition is observed in four chief peaks with about 43.7 nm equal wavelength distance to which the intensity gradually, systematically decreases to the redshift direction with a constant wavelength value. In our other experiments (i.e., the five-period vertically stacked QDs with undoped GaAs layer instead of the Si-doped GaAs layer), the phase-change splitting of the QD ground state was not observed. The hole valence ground state has not split due to hole coupling weakness and heavier masses.[9] Of course, the observation of the quantum dephase splitting of the electron ground state serves as a macroscopic confirmation of some fundamental postulates of quantum mechanics. This question to which the phase-change splitting produces a lowest energy distribution is very complicated. The interaction between the located electrons in QDs and the delocated tunneling electrons is in a strong confinement situation, and the calculation of the wave-function overlap to produce the many-body interaction is too complicated.

Deep level transient spectroscopy (DLTS) measurement has been used to study the properties of quantum dots,[10,11] and the thermal emission from QDs can mask that from the deep levels in the DLTS data with an exponential dependence on emission energy.[12] When the vertically stacked InAs QDs are embedded in $Al_{0.5}Ga_{0.5}As$ barrier, the capture of carriers is caused by the tunneling effect through the high barrier potential instead of that of direct injection.[11,12] The activation energies of electrons and holes in QDs are so large that the DLTS peaks might be observed near room temperature,[13] and the hole thermal emission in dot can match with the electron thermal emission. As a result, in the DLTS experiment, the peaks of electron and hole become narrow to produce a possibility of the separation of electron and hole peaks (i.e., the positive signals corresponding to hole energy levels and the negative signals corresponding to electron energy levels, respectively). Due to the huge capture center of QDs, the emission of El_2 antisite donor deep level and DX center are ‘‘eaten’’ by electron and hole emission peaks.

The DLTS measurement is made using a 1 MHz capacitance meter, a pulse generator, a temperature controller, and a computer. The mean voltage is -2 V, the pulse voltage is 1 V, the pulse hold time is 1 ms, and the rate windows t_1-t_2 is 0.2–0.6 ms for the sample with the normal size dots. The temperature of the samples is cooled down below 90 K using liquid nitrogen heated until 400 K. Due to the lack of phonons, the relaxation in QDs is significantly slowed so that the discrete levels might be directly observed in Fig. 3 of the DLTS measurement. This is a consequence of the increased thermal escape of the carriers from the dots, and electrons and holes would be emitted from the wetting layer (WL) ground state, the QD excited state, and the QD ground state to the barrier of the conduction and valence bands, respectively. The transition of PL peaks has a natural link to the DLTS peaks. It is surprising that the energy distance ratio (i.e., between from the WL ground state to the excited state and from the excited state to the QD ground state) in PL

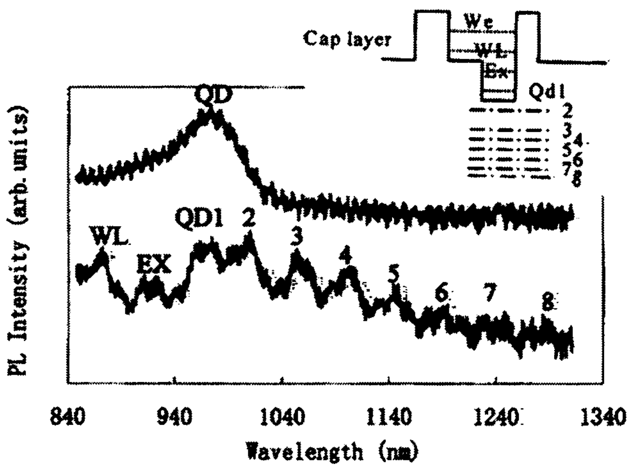

FIG. 4. The other PL spectra with the small size. Three peaks are the recombination of WL, QD excited state, and QD ground state. Temperatures of two curves are at 21 and at 60 K, the laser excitation power is 26 mW, and the peak distance of the ground state splitting is about 46.9 nm. The inset is the energy diagram.

measurement completely corresponds to the distance ratio [i.e., between the central position (247 K of the E_{wl}–H_{wl} transition) to the central position (286 K of the E_{ex}–H_{ex} transition) and from the central position (286 K of the E_{ex}–H_{ex} transition) to the central position (313 K of the E_{wl}–H_{wl} transition)] in the DLTS spectrum. This result is unanimous for a simple theoretical calculation, and we have to recognize that the discrete levels really are ''seen'' by the DLTS spectrum. However, in the DLTS experiment, the dephase splitting of the ground state is not observed to reflect the different mechanism compared to PL recombination.

Figure 4 shows the PL spectra of the other sample with the small size QDs. The stronger quantum confinement in small size sample exploits intrinsic quantum mechanical correlations. Due to stronger quantum confinement, the ground state transition behaves a blueshift and becomes so strong that it splits into eight peaks with about 46.9 nm wavelength value by the dephase splitting process. The fundamental requirement for the experimental realization of such proposals is the successful generation. The many-body interaction and the abundant resonant tunneling electrons take place in the stronger confinement situation to enhance the phase-change splitting of such quantum-based phenomena.

The energy level structure and the relaxation mechanisms significantly influence the carrier capture and emission processes. In certain circumstances, the strongly localized electrons in QDs are affected by many-body interaction to create a fascinating phase and make the ground state dephase splitting. In order to address these questions, PL spectroscopy and DLTS are applied to the samples of InAs QDs which are vertically stacked in $Al_{0.5}Ga_{0.5}As$ high potential barrier. Due to their rich spectrum and discrete density of energy levels, vertically stacked QDs demonstrate a number of unique material and optical properties that might find their use in novel optoelectronic device applications.

The authors would like to thank Professors M. Yano, M. Inoue, and S. Sasa of the Osaka Institute of Technology for their support. Dr. Shuwei Li was supported by the Monbusho Grant-in-Aid for JSPS Fellows relating to the JSPS Postdoctoral Fellowship for Foreign Researchers.

[1] L. Wang, A. Williamson, A. Zunger, H. Jiang, and J. Singh, Appl. Phys. Lett. **76**, 339 (2000).
[2] J. Phillips, K. Kamath, X. Zhou, N. Chervela, and P. Bhattacharya, J. Vac. Sci. Technol. B **16**, 1343 (1998).
[3] M. Bayer, O. Stern, P. Hawrylak, S. Fafard, and A. Forchel, Nature (London) **405**, 923 (2000).
[4] L. Quiroga and N. Johnson, Phys. Rev. Lett. **83**, 2270 (1999).
[5] B. Partoens and F. Peeters, Phys. Rev. Lett. **84**, 4433 (2000).
[6] K. Koike, K. Saitoh, S. Li, S. Sasa, M. Inoue, and M. Yano, Appl. Phys. Lett. **76**, 1464 (2000).
[7] K. Koike, S. Li, and M. Yano, Jpn. J. Appl. Phys., Part 1 **39**, 1622 (2000).
[8] W. Chang, T. Hsu, K. Tsai, T. Nee, J. Chyi, and N. Yeh, Jpn. J. Appl. Phys., Part 1 **38**, 554 (1999).
[9] S. Fafard, M. Spanner, J. McCaffrey, and Z. Wasilewski, Appl. Phys. Lett. **76**, 2268 (2000).
[10] D. Lang, J. Appl. Phys. **45**, 3023 (1974).
[11] S. Anand, N. Carlsson, M. Pistol, L. Samuelson, and W. Seifery, Appl. Phys. Lett. **67**, 3016 (1995).
[12] M. Sobolev, A. Kovsh, V. Ustinov, A. Egorov, and A. Zhukov, J. Electron. Mater. **28**, 491 (1999).
[13] S. Li, K. Koike, S. Sasa, M. Inoue, and M. Yano Solid State Commun. (accepted for publication).