Available online at www.sciencedirect.com
SCIENCE DIRECT®

ELSEVIER

Solid State Communications 133 (2005) 191–195

solid state communications

www.elsevier.com/locate/ssc

Transition behavior from uncoupled to coupled multiple stacked CdSe/ZnSe self-assembled quantum-dot arrays

T.W. Kim[a,*], K.H. Yoo[b], Gil-Ho Kim[c], S. Lee[d], J.K. Furdyna[e], M. Dobrowolska[e]

[a]*Division of Electrical and Computer Engineering, Advanced Semiconductor Research Center, Hanyang University, 17 Haengdang-dong, Seongdong-gu, Seoul 133-791, South Korea*
[b]*Department of Physics and Research Institute for Basic Sciences, Kyung Hee University, Seoul 130-701, South Korea*
[c]*Department of Electronic and Electrical Engineering, Sungkyunkwan University, Suwon 440-746, South Korea*
[d]*Department of Physics, Korea University, Seoul 136-701, South Korea*
[e]*Department of Physics, University of Notre Dame, Norte Dame, IN 46556, USA*

Received 15 September 2004; accepted 18 October 2004 by C.N.R. Rao

Available online 4 November 2004

Abstract

Transition behavior from uncoupled to coupled multiple stacked CdSe/ZnSe quantum-dot (QD) arrays grown by molecular beam epitaxy were investigated. Transmission electron microscopy showed that vertically stacked self-assembled CdSe QD arrays were embedded in the ZnSe barriers. The results for the photoluminescence (PL) data at 18 K demonstrated clearly that the transition behavior from uncoupled to coupled peaks depended on the ZnSe barrier thickness. The temperature-dependent PL measurements showed that the activation energy of the electrons confined in the CdSe QDs increased dramatically with decreasing ZnSe spacer layer thickness due to the strong coupling between CdSe/ZnSe QD arrays. The present observations can help improve understanding of the dependence of the coupling behavior and activation energy in CdSe/ZnSe QDs on the spacer layer thickness.

PACS: 68.55.Bd; 78.20.−e

Keywords: A. Nanostructures; B. Epitaxy; D. Optical properties

Semiconductor quantum dots (QDs) have become particularly attractive because of both scientific and technological points of view [1–7]. Recently, there has been increasing interest in II–VI wide-gap QDs for potential applications in optoelectronic devices operating in the blue-green region of the spectrum [8–10]. However, relatively little work has been performed on group II–VI/II–VI QDs in comparison with III–V/III–V QDs because of delicate problems encountered during growth. Among the many QD structures, closely stacked QDs, consisting of smaller band-gap QD arrays separated by a thin embedded barrier, have been currently receiving considerable attention for the investigations of fundamental physical properties [11,12] and for application devices in next-generation optoelectronic and quantum information processing devices [13,14]. Even though some works on the optical properties of CdSe/ZnSe QDs have been reported [15–17], the dependence of the coupling behavior and activation energy on the ZnSe spacer thickness in vertically stacked CdSe/ZnSe self-assembled QDs has not been reported yet.

This communication reports data for the transition behavior from uncoupled to coupled multiple stacked CdSe QD arrays embedded in ZnSe barriers grown by molecular beam epitaxy (MBE). Transmission electron

* Corresponding author. Fax: +82 2 2292 4135.
E-mail address: twk@hanyang.ac.kr (T.W. Kim).

0038-1098/$ - see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ssc.2004.10.029

microscopy (TEM) measurements were performed to characterize the microstructural properties of the CdSe QD arrays inserted in ZnSe barriers, and temperature-dependent photoluminescence (PL) measurements were carried out in order to investigate the interband transitions and to determine the dependence of the coupling behavior and activation energy on the ZnSe spacer layer thickness.

Three samples used in this work were grown on semi-insulating (100)-oriented GaAs substrates by MBE and consisted of the following structures: a 300 Å undoped ZnSe capping layer; a CdSe/ZnSe QD layer; and an 1000 Å undoped ZnSe buffer layer. The QD layer of sample (a) is one period of 2.5 monolayer (ML) CdSe/ZnSe QDs, and those of samples (b) and (c) are five periods of 2.5 ML CdSe/ZnSe QDs with spacer layer thicknesses of 60 and 300 Å, respectively. The thickness of each layer was determined by using cross-sectional TEM.

TEM observations were performed using a JEM 2000EX transmission electron microscope operating at 200 kV. The samples for TEM measurements were prepared by cutting and polishing with diamond paper to a thickness of approximately 30 μm and then argon-ion milling at liquid-nitrogen temperature to electron transparency. The PL measurements were carried out using a 75-cm monochromator equipped with an RCA 31034 photomultiplier tube. The excitation source was the 3250 Å line of a He-Cd laser, and the sample temperature was controlled between 18 and 300 K by using a He displex system.

A plane-view bright-field TEM image of sample (b) in the [001] zone shows CdSe QDs embedded into an undoped ZnSe matrix with spacer layer thickness of 60 Å. Strained CdSe QDs are clearly observed, as indicated by the characteristic dark and bright contrast related to an inhomogeneous lattice deformation in a three-dimensional structure [18]. The sizes of the QDs are approximately between 15 and 45 nm, and the surface density of the QDs is 2.1×10^9 cm^{-2}. These results are in reasonable agreement with those obtained from the atomic force microscope measurements. The TEM morphology can be explained by the thermodynamics of island growth [19]. Even though the formation of the QDs releases the strain energy, the white bands inside the CdSe with the black islands originate from strain effects due to the transition from two-dimensional to three-dimensional nucleation. A cross-sectional bright-field TEM image of the same sample depicts that five periods of CdSe QD arrays are embedded in an intrinsic ZnSe barrier with ZnSe spacer layer thickness of 60 Å. The typical lateral sizes of the CdSe QDs are between 15 and 40 nm, and their heights are between 3 and 4 nm. Coherently strained islands are clearly observed, which is indicated by characteristic dark and bright contrasts related to an inhomogeneous lattice deformation in the three-dimensional structure and to the absence of dislocations [20].

Fig. 1 shows the PL spectra measured at 18 K for (a) one period of the CdSe/ZnSe QDs and five periods of the CdSe/ZnSe QDs with spacer thicknesses of (b) 60 Å and of (c) 300 Å. The dominant PL peaks are related to interband transitions from the ground electron subband to the ground heavy-hole subband (E_1–HH_1) of the CdSe QDs. When the sample (a) is compared with the other samples, the (E_1–HH_1) peak position shifts to higher energy with increasing number of the stacking layers. When samples (b) and (c) are compared, the peak position corresponding to the (E_1–HH_1) transition shifts to lower energy with decreasing ZnSe spacer thickness. When the coupling of wave functions between QD layers occurs with decreasing ZnSe spacer thickness, the carriers flow into the largest size QDs resulting in the red shift. The luminescence intensity of one period sample (a) is smaller than those of five period samples (b) and (c), as expected. Between the two five period samples, the sample (b) with spacer thickness of 60 Å has the higher luminescence intensity. Furthermore, the luminescence intensity of this sample (b) does not disappear even at 251 K. Therefore, it may be concluded that the luminescence intensity of the CdSe/ZnSe QDs is enhanced due to closely stacked vertical layers, and this phenomenon can help improve the efficiency of optoelectronic devices.

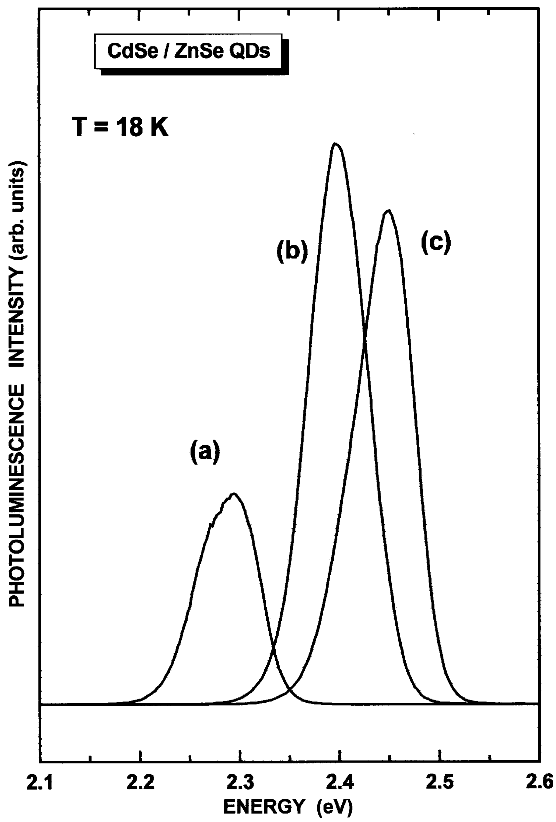

Fig. 1. Photoluminescence spectra at 18 K for the CdSe quantum-dot arrays embedded in ZnSe barriers: (a) one period of the CdSe/ZnSe quantum-dot array and five periods of CdSe/ZnSe quantum-dot arrays with ZnSe spacer thickness of (b) 60 Å or (c) 300 Å.

In order to determine and compare the dependence of the activation energies of the electrons confined in the CdSe/ZnSe QDs on the spacer thickness, temperature-dependent PL measurements were carried out. Fig. 2 shows the results of the PL spectra measured at several temperatures for the CdSe/ZnSe QDs with a ZnSe spacer thickness of 60 Å. Since the energy gaps of the CdSe QDs decrease with increasing temperature, the PL peaks corresponding to the (E_1–HH_1) transition shift to lower energy with increasing temperature. When the participating carriers move out through activation over an energy barrier, the PL intensity as a function of temperature is given by [21, 22]

$$I = I_0/[1 + C\exp(-\Delta E_A/k_B T)], \quad (1)$$

where I_0 is the PL intensity at 0 K, C is the ratio of the non-radiative transition probability to radiation transition probability, ΔE_A is the activation energy, and k_B is the Boltzmann constant. Integrated PL intensities as a function of the reciprocal temperature for three samples are shown in Fig. 3. The activation energies determined in Fig. 3 are approximately 110 ± 7, 150 ± 10, and 100 ± 5 meV for samples (a), (b) and (c), respectively. The activation energy of five periods of CdSe/ZnSe QD arrays with a ZnSe spacer thickness of 60 Å is much larger than that of the CdSe/ZnSe QD arrays with a ZnSe spacer thickness of 300 Å, while the latter is almost same as that of one period of the CdSe/ZnSe array. When the ZnSe spacer thickness is 60 Å, the electronic wave functions of the ground subbands in the CdSe QDs arrays are strongly coupled with each other. However, when the spacer layer thickness becomes 300 Å, the electronic states in the CdSe/ZnSe QD arrays are no

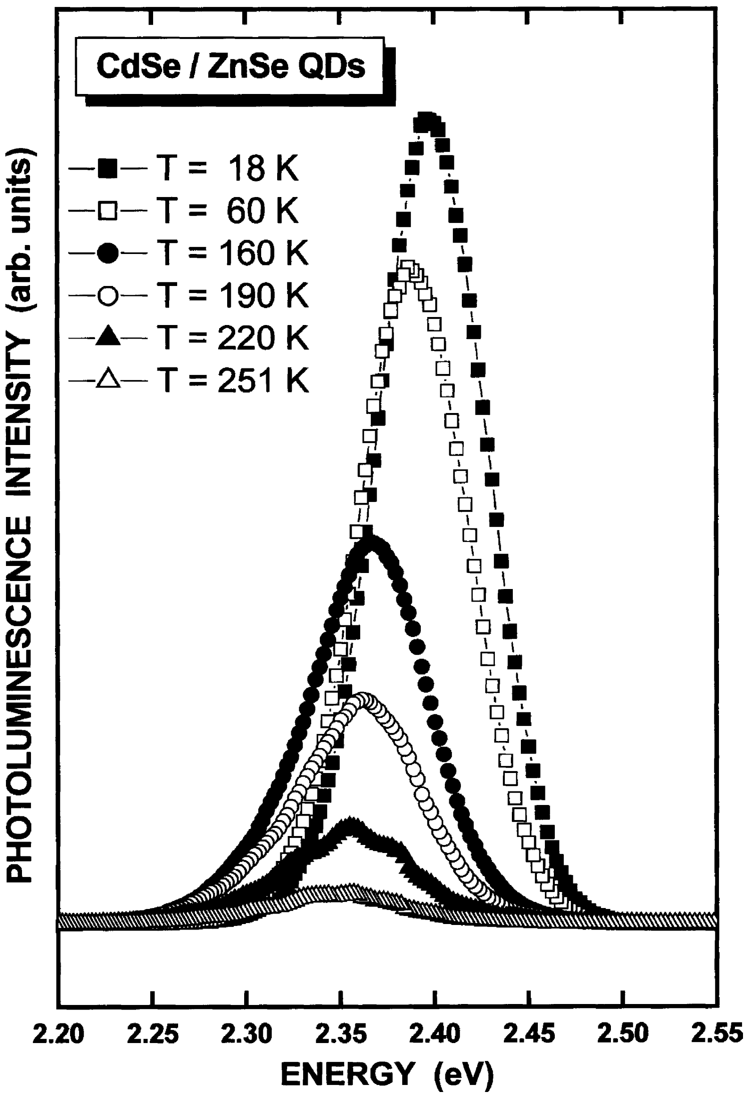

Fig. 2. Photoluminescence spectra measured at several temperatures for CdSe/ZnSe quantum-dot arrays with ZnSe spacer thickness of 60 Å.

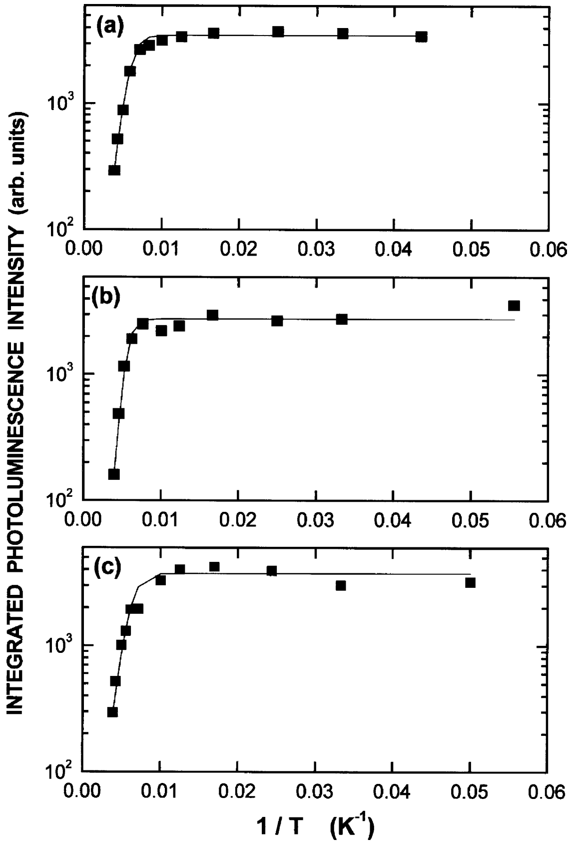

Fig. 3. Integrated photoluminescence intensities as a function of the reciprocal temperature for (a) one period of the CdSe/ZnSe quantum-dot array and five periods of CdSe/ZnSe quantum dots with ZnSe spacer thickness of (b) 60 Å or (c) 300 Å.

longer coupled with each other. Therefore, coupling between the zero-dimensional states of the QDs increases the activation energy of the electrons confined in the CdSe QDs. These results indicate that the activation energy of multiple vertically stacked CdSe/ZnSe QD arrays is strongly affected by the coupling and uncoupling behaviors of the multiple stacked CdSe/ZnSe QD arrays dependent on the CdSe spacer thickness.

In summary, the results of TEM measurements showed that high-quality CdSe QD arrays embedded in ZnSe barriers could be grown on GaAs substrates. The results of the temperature-dependent PL measurements showed that the activation energy of the electrons confined in the CdSe QDs increased with decreasing ZnSe spacer thickness due to the strong coupling between CdSe/ZnSe QD arrays and that the transition behavior from coupled to uncoupled (E_1–HH_1) peaks depended on the ZnSe barrier thickness. These present observations can help improve understanding of the dependence of the coupling behavior and activation energy on the ZnSe spacer thickness in CdSe/ZnSe QD structures. Furthermore, these results suggest that closely stacked CdSe/ZnSe QD arrays hold promise for high-efficiency optoelectronic devices utilizing strong excitonic transitions operating at room temperature.

Acknowledgements

The work at Hanyang University was supported by grant No. R02-2003-000-10030-0 from the Basic Research Program of the Korea Science and Engineering Foundation and also supported by the Korea Science and Engineering Foundation through Quantum-functional Semiconductor Research Center at Dongguk University. The work at University of Notre Dame supported by the National Science Foundation Grant DMR 0072897.

References

[1] D. Leonard, M. Krishnamurthy, C.M. Reaves, S.P. Denbaars, P.M. Petroff, Appl. Phys. Lett. 63 (1993) 3203.
[2] A. Ponchet, A. Le Corre, H. L'Haridon, B. Lambert, S. Lalaun, Appl. Phys. Lett. 67 (1995) 1850.
[3] H. Merchand, P. Desuardins, S. Guillon, H. Paultre, R. Mascut, Appl. Phys. Lett. 71 (1997) 527.
[4] V. Ustinov, E.R. Weber, Appl. Phys. Lett. 72 (1998) 362.
[5] H. Li, J. Wu, B. Xu, J. Liang, Z. Wang, Appl. Phys. Lett. 72 (1998) 2123.
[6] L. Chu, M. Arzberger, A. Zrenner, G. Böhm, G. Abstreiter, Appl. Phys. Lett. 75 (1999) 2247.
[7] Y. Toda, O. Moriwaki, M. Nichioka, Y. Arawaka, Phys. Rev. Lett. 82 (1999) 4114.
[8] G. Karczewski, S. Mackowski, M. Kutrowski, T. Wojtwicz, J. Kossut, Appl. Phys. Lett. 74 (1999) 3011.
[9] G. Bacher, T. Kümmel, D. Eisert, A. Forcel, B. König, W. Ossau, C.R. Becker, G. Landwehr, Appl. Phys. Lett. 75 (1999) 956.
[10] Y. Terai, S. Kuroda, K. Takita, T. Takamasu, G. Kido, J. Lumin. 87 (2000) 396.
[11] L.E.F. Foa Torres, C.H. Lewenkopf, H.M. Pastawski, Phys. Rev. Lett. 91 (2003) 116801.
[12] H. Heidemeyer, S. Kiravittaya, C. Müller, N.Y. Jin-Phillipp, O.G. Schmidt, Appl. Phys. Lett. 80 (2002) 1544.
[13] H. Jeong, A.M. Chang, M.R. Mellock, Science 293 (2001) 2221.
[14] N.J. Craig, J.M. Taylor, E.A. Lester, C.M. Marcus, M.P. Hanson, A.C. Gossard, Science 304 (2004) 565.
[15] S.H. Xin, P.D. Wang, A. Yin, C. Kim, M. Dobrowolska, J.L. Merz, J.K. Furdyna, Appl. Phys. Lett. 69 (1996) 3884.
[16] F. Flack, N. Samarth, V. Nikitin, P.A. Crowell, J. Shi, J. Levy, D.D. Awschalom, Phys. Rev. B 54 (1996) R17312.
[17] J. Seufert, M. Obert, M. Scheibner, N.A. Gippius, G. Bacher, A. Forchel, T. Passow, K. Leonardi, D. Hommel, Appl. Phys. Lett. 79 (2001) 1033.
[18] D.L. Smith, C. Mailhiet, J. Appl. Phys. 63 (1998) 2717.

[19] A. Barabasi, Appl. Phys. Lett. 70 (1997) 2565.
[20] X.Z. Liao, J. Zou, X.F. Duan, D.J.H. Cockayne, R. Lean, C. Lobo, Phys. Rev. B 58 (1998) R4235.
[21] See, for example, E.W. Williams, H.B. Bebb, in: R.K. Willardson, A.C. Beer (Eds.), Semiconductors and Semimetals vol. 8, Academic Press, New York, 1972.
[22] P.O. Holtz, B. Monemar, H.J. Lozykowski, Phys. Rev. B 32 (1985) 986.