Temperature-dependent energy band gap variation in self-organized InAs quantum dots

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(Received 30 July 2011; accepted 21 September 2011; published online 12 October 2011)

We investigated the temperature-dependent variation of the photoluminescence emission energy of self-organized InAs/GaAs quantum dots (QDs) grown by conventional Stranski-Krastanov (SK) molecular beam epitaxy and migration-enhanced molecular beam epitaxy (MEMBE) and that of MEMBE InAs QDs in a symmetric and an asymmetric $In_{0.2}Ga_{0.8}As/GaAs$ well. The temperature-dependent energy variation of each QD is analyzed in low and high temperature regions, including a sigmoidal behavior of conventional SK quantum dots with the well-known Varshni and semi-empirical Fan models. © *2011 American Institute of Physics*. [doi:10.1063/1.3651492]

In(Ga)As/GaAs self-organized quantum dots (QDs) have been a mainstay as a promising zero-dimensional system for potential device applications and basic studies. A lot of research, therefore, has been conducted in a wide range of high-performance optoelectronic devices, such as high-temperature operating infrared photodetectors, low-threshold and temperature-stable laser diodes, wide bandwidth superluminescent diodes, and single photon sources for quantum cryptography.[1–5] However, the key characteristics and availability of self-organized QDs depend very much on the growth condition which strongly influence their size, shape, and the wetting layer (WL) structure. Accordingly, the self-organized QDs need to show a deterministic physical performance for improved optics and electronic devices. In this sense, the temperature-dependent optical study of QDs offers an effective guideline for in-depth research. A number of empirical and semi-empirical models have been proposed to analyze the seemingly quadratic decrease of the band gap at low temperature and linear decrease at high temperature.[6–16] In particular, some Stranski-Krastanov (SK) grown InAs QDs have shown a sigmoidal temperature-dependent variation of the energy gap with a temperature-driven photoluminescence (PL) FWHM change.[16–19] Thus, several efforts have been made to analyze the abnormal temperature behavior.[16]

In this work, we studied the temperature-dependence of the band gap energy in four different InAs QDs with the PL measurements. Considering the WL thickness and uniformity, applicable fitting models are employed to describe the temperature-dependent PL evolution in low and high temperatures. The empirical Varshni and semi-empirical Fan models are utilized for the temperature dependence of the PL energy and the physical meaning is discussed with the resulting Varshni and Fan parameters.

Four self-organized InAs QD samples were grown for temperature PL study. The QD samples were grown on (001) semi-insulating (SI) GaAs wafers in a V80 molecular beam epitaxy (MBE) system equipped with an ion getter pump and a tetramer arsenic source. Starting from the 70 nm GaAs buffer layer, a 50 nm $Al_{0.3}Ga_{0.7}As$ layer and a short period superlattice (SPS) consisting of 20 stacks of 2-nm $Al_{0.3}Ga_{0.7}As$ and 2-nm GaAs layers were deposited. Then on top of the 20-nm GaAs layer, 2.4 monolayers (MLs) of the InAs QDs were formed by conventional MBE for A0 and about 3 MLs by migration enhanced molecular beam epitaxy (MEMBE) for A1. The A0 and A1 that were prepared resulted in different thicknesses of WL, 4 nm and 2.1 nm, respectively.[19] The samples A2 and A3 consisting of 3 MLs MEMBE InAs QDs were inserted into two 5-nm undoped $In_{0.2}Ga_{0.8}As$ symmetric wells for A2 and into a 7.5-nm and a 1.25-nm undoped $In_{0.2}Ga_{0.8}As$ asymmetric well for A3 on top of a 20-nm GaAs layer. Each InAs QD group was then capped with a 20-nm GaAs layer, 20 SPS stacks of 2-nm $Al_{0.3}Ga_{0.7}As$ and 2-nm GaAs layers, a 10-nm $Al_{0.3}Ga_{0.7}As$ layer, and a 4-nm GaAs layer. The detailed sample preparation conditions can be found elsewhere.[19,20] The PL measurements were performed using an Ar-ion laser with excitation wavelength of 514.5 nm line in a He-cryostat system. The QD emission was detected by using a 105-cm monochromator equipped with a liquid nitrogen cooled InGaAs detector.

Figures 1(a) and 1(b) show the temperature-dependent PL evolutions of the InAs QDs of samples A0–A3 ranging from 15 to 300 K. Figure 1(a) shows the WL reduction effects from A0 to A1. As the WL is reduced with a factor of 2 and its uniformity is enhanced, the temperature-dependent PL peak position of A1 shows a monotonous trend, while anomalous high temperature variation is shown with A0. In fact, a similar sigmoidal temperature-dependence shown in A0 has been observed in some SK-grown QDs having WL.[16–18] Indeed, a WL enhances the coupling with the wave function of QDs and carrier redistribution at elevated temperature;[19] therefore, carriers in the entire ensemble QD can be redistributed with a common Fermi level with the aid of WL at high temperature.[19] Figure 1(b) shows the monotonous redshift of the PL energy in MEMBE InAs dot-in-a-

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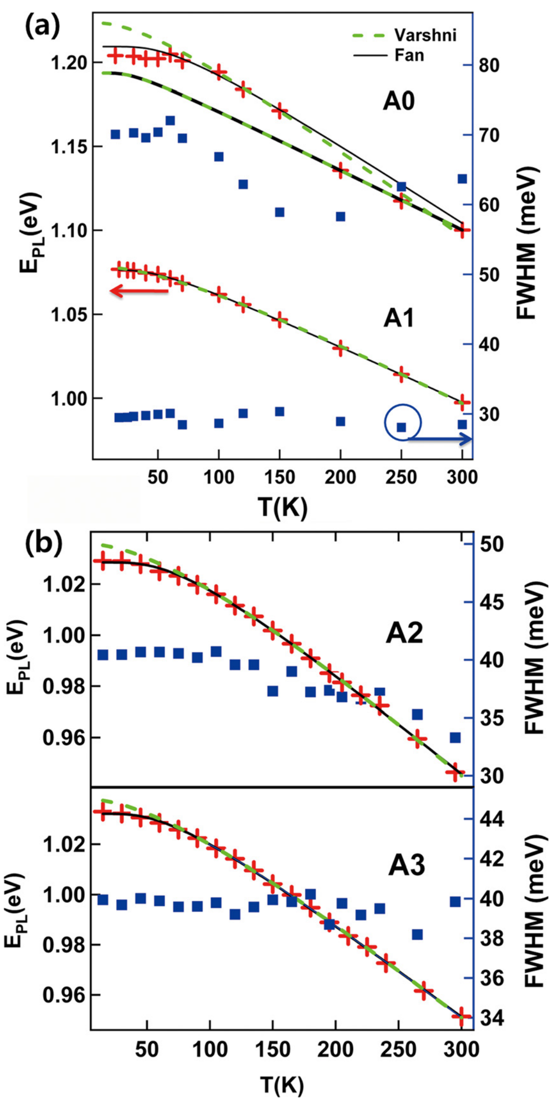

FIG. 1. (Color online) The temperature-dependent PL peak evolutions are presented at various temperatures (15–300 K) for (a) the InAs dots grown in conventional MBE (A0) and MEMBE (A1) and for (b) the InAs dots in undoped $In_{0.2}Ga_{0.8}As$ symmetric (A2) and asymmetric wells (A3). The red cross is the temperature dependence of the experimental PL peak energy (E_{PL}). The blue rectangle indicates the experimental PL linewidth (FWHM). The green dashed lines and the black solid lines represent the projected energy gap variations by using Varshni and Fan models, respectively.

well systems, yet significant change in the PL linewidth of A2 is accompanied by 20% reduction. The observed temperature sensitivity in PL linewidth of A2 could be suppressed in an asymmetric well (A3);[20] however, the change in linewidth of 7 meV for A2 is remarkably smaller than that of 11 meV for A0.

To further discuss these temperature-dependent optical properties, we employ two well-known models which originated from bulk semiconductors. The first one is the Varshni model[6,7] which is empirically expressed as

$$E(T) = E(0) - \frac{\alpha T^2}{\beta + T}, \tag{1}$$

where $E(0)$ is the band gap energy at 0 K and, α and β are the two Varshni coefficients to be determined. The Varshni equation was devised to fit the seemingly linear decrease in the high temperature region and the quadratic decrease in the low temperature region as observed in many semiconductor material systems.

The second model that we employed is the Fan equation which was developed for fitting the temperature-dependence of the band gap energy, based on the Bose-Einstein statistical distribution for phonons. The Fan equation[21,22] is written in the form of

$$E(T) = E(0) - A(\langle n \rangle + const), \tag{2}$$

where the Bose-Einstein statistics of phonons with the energy $\hbar w$ and the Fan parameter A are, respectively, given by $\langle n \rangle = 1/(e^{\hbar w/k_B T} - 1)$ and by $A = \frac{e^2}{\sqrt{2}\hbar}\sqrt{m_0 \hbar w}\frac{1}{4\pi\varepsilon}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)\left(\sqrt{\frac{m_e}{m_0}} + \sqrt{\frac{m_h}{m_0}}\right)$. Here, ε is the dielectric constant; ε_∞ and ε_0 are the static and high frequency dielectric constants, respectively; m_0 is the free electron mass; and m_e and m_h are the effective electron and hole masses, respectively. Note that Varshni coefficients can be obtained from Eq. (2) as the temperature coefficient of the bandgap width $\alpha = Ak_B/\hbar w$ and the phonon coefficient $\beta = \hbar w/2k_B$ in the limit of high temperature $k_B T \gg \hbar w$. The constant in Eq. (2) can either be taken as 0 (the energy level broadening), 1 (the energy level shift) or 1/2 (the exciton-phonon interaction).[9] Once the constant is fixed as 1/2 corresponding to the exciton-phonon interaction, the Fan equation is the same as the Vina model.[8] In this work, 0 is taken for the constant in Eq. (2) since the constant does not change the whole picture.

The fitted dependences are shown in Figure 1. The green dashed lines and the black solid lines correspond to the projected energy gap variations by the Varshni and Fan models, respectively. The associated fitting parameters obtained from the Varshni and Fan models are listed in Table I along with the parameters from the literature. Here, the Fan parameters are calculated based on the referred Varshni coefficients (*) (Refs. 23–25) for bulk InAs and InAs QDs from the literature. As reported in Figures 1(a) and 1(b), three different regions can be categorized for a fitting where low temperature is below 70 K, in which the Fan equation tracks more accurately than that of the Varshni (A0 and A2); a transition in which the PL energy decreases rapidly (A0: 150–200 K). For the high temperature region where PL linewidth changes considerably (A0 and A2), there have been discussions to understand and analyze the temperature-dependent energy

TABLE I. Parameters extracted from the projection of temperature-dependent PL emission to Varshni and Fan models. The Varshni coefficients from the literature are marked with the symbol*.

Sample	α(10^{-4} eV/K)	B (K)	A (meV)	$\hbar w$(meV)
A0 for low T	5.4	79	85	16
A0 for high T	3.6	40	30	7
A1	3.8	121	49	12
A2	4.6	153	91	19
A3	4.1	128	82	18
Bulk InAs[23]	3.2*	93*	29	10
Bulk InAs[24]	2.7*	83*	29	9.3
InAs QD[25]	4.2*	199*	60	15

gap variation.[9,11,16–18] Unfortunately, a recent proposal[16] for the sigmoidal behavior, $E = E(0) + a/(T + b)$, diverges significantly in our case.

In the low temperature region, the Fan model is found to be more accurate for A0 and A2, in particular, while the Varshni model overestimates the energy slope. The better accuracy with the Fan model reflects that the energy gap shrinkage of QD could be affected by the electron and phonon interaction than by thermal expansion[26] below 70 K. The calculated phonon energy of 16 meV from Fan's model for SK QDs (A0) is fairly comparable to the reported value of 15 meV.[25]

At high temperature, the Varshni and Fan models can both be used to describe the PL energy shift. In particular, at temperatures higher than 200 K for A0, the PL peak energies follow both models with the parameters ranging to those of the typical bulk InAs.[23,24] The calculation of $\hbar w \sim 7$meV from Fan's model at high temperature is well matched with the calculated phonon energy (2β) satisfying the relation $A = 2\alpha\beta$ in the high temperature limit. It is, however, 44% of the phonon energy of 16 meV at low temperature. Notice that the high temperature phonon energy of 7 meV is located close to bulk GaAs transverse acoustic (TA) phonon $\hbar w_{\mathrm{TA}}(L) = 7.7\,\mathrm{meV}$ and $\hbar w_{\mathrm{TA}}(X) = 9.8\,\mathrm{meV}$.[11,27] The extracted phonon energy of $\hbar w \sim 7$ meV then could indicate a dominant interaction with phonons of the GaAs host material.[11] However, the calculated $\hbar w$ from Fan's model of about 9.3–10 meV for bulk InAs do not match with 60% of the calculated[28] and experimentally observed values.[29]

In the case of quantum well structures, Rojas-Ramirez[30] observed the increase in all the parameters (α, β, A, and $\hbar w$) with the quantum confinement increasing via $In_{0.2}Ga_{0.8}As$ well-width reduction. In other words, the phonon energy is larger when the quantum confinement is enhanced. Concerning a quasi-equilibrium state of the entire QD ensemble at higher temperature,[19] the deconfinement via InAs WL would reduce the phonon energy. One can notice that all parameters for A0 decrease at high temperature. With other MEMBE QD samples A1–A3, the Varshni and Fan models give satisfactory results with each parameter ranged to those of InAs QDs (Ref. 25) tabulated in Table I. The extracted phonon energies of A2 and A3 from the Fan model are about 40% smaller than the experimentally obtained phonon energy[20] of 30 meV.

To conclude, we have demonstrated that the empirical Varshni and semi-empirical Fan models are valid for the temperature-dependent PL study of the MEMBE InAs QDs. The sigmoidal behavior of SK-grown QD is well analyzed by the Varshni and Fan models in the two different low and high temperature regions, respectively. The estimation with the Fan model has better accuracy especially at low temperature. All parameters are related to the phonon energy which increases with enhanced quantum confinement.

The authors would like to thank N.K. Cho and S.P. Ryu for their help with growth and measurements. This work was supported by MEST through the GRL program and was performed within the framework of the Center for Photonics and Nanostructures (CPN), KIST-CNRS LIA.

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