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Effect of size non-uniformity on photoluminescence from ensembles of InAs quantum dots embedded in GaAs

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Abstract The photoluminescence (PL) spectrum from ensembles of InAs/GaAs quantum dots (QDs) is calculated. The effect of the dot size distribution and the variation of the associated confining potentials on the PL spectra are estimated. It is found that the intermixing of the interfaces causes an increase of the PL spectra energy. The size distribution determines the spectrum width and makes the PL line shape asymmetric with a high-energy tail. Moreover, the non-uniform size distribution also results in a redshift of the PL peak. The experimental PL spectrum is well explained by the size distribution and intermixing effect within the effective mass approximation.

In recent years, quantum dots (QDs) fabricated by molecular beam epitaxy (MBE) in the Stranski–Krastanow (S–K) modes have attracted much attention due to their excellent predicted properties for optoelectronic devices applications. However, a key issue is the random statistical size distribution frequently appearing in the S–K mode, which broadens photoluminescence (PL) peaks. In order to optimize device performance such as detector sensitivity at a particular wavelength and laser gain, the PL linewidth should be as narrow as possible. Production of ensembles of quantum dots with the highest possible uniformity is a major problem in this area. Many groups have developed different techniques to reduce the inhomogeneous distribution of dot size [1, 2]. However, the origin of the inhomogeneous broadening of the PL peak is still not clear and a few theoretical studies on the size distribution effect have been made.

The interface intermixing technology is a well-developed post growth process for energy band engineering in low-dimensional system. For these purposes, the shape of the barrier potential is modified by interface intermixing induced by the rapid thermal annealing and the ion-implantation process. Generally, the intermixing leads to a blueshift and a narrowing of optical emission from QDs [3, 4]. So that, both size distribution and interface intermixing will affect the PL spectrum. There is, hence, some needs to systematically study these effects of QD on PL characterization.

Recently, similar to those of Kane's mean field approach and Lifshitsz's probabilistic arguments [5–7], Chen et al. [8] have established a model for the PL spectrum of ensembles of Si nanoclusters influenced by the size distribution effect. In the present paper, we propose to use this model to investigate the effect of the size distribution and the interface intermixing on the PL spectrum of InAs/GaAs QDs. Using the effective mass approximation reported by Wojs [9], we get the energy states by solving the Schrödinger equation. In addition, the intermixing in epitaxially grown InAs/GaAs quantum dot systems involves much more complex three-dimensional processes. To better understand the observed effects of intermixing, the interdiffusion is modeled using a simple one-dimensional Fickian diffusion for the growth direction. Indeed, because of the small aspect ratio of QDs, the principle effects of the intermixing can be expected to come mainly from diffusion in the vertical direction [10–12].

All the QD samples investigated were grown by MBE [3]. To estimate the size of the QDs, an uncapped referenced sample was also grown under the same growth conditions. Then the referenced sample was characterized

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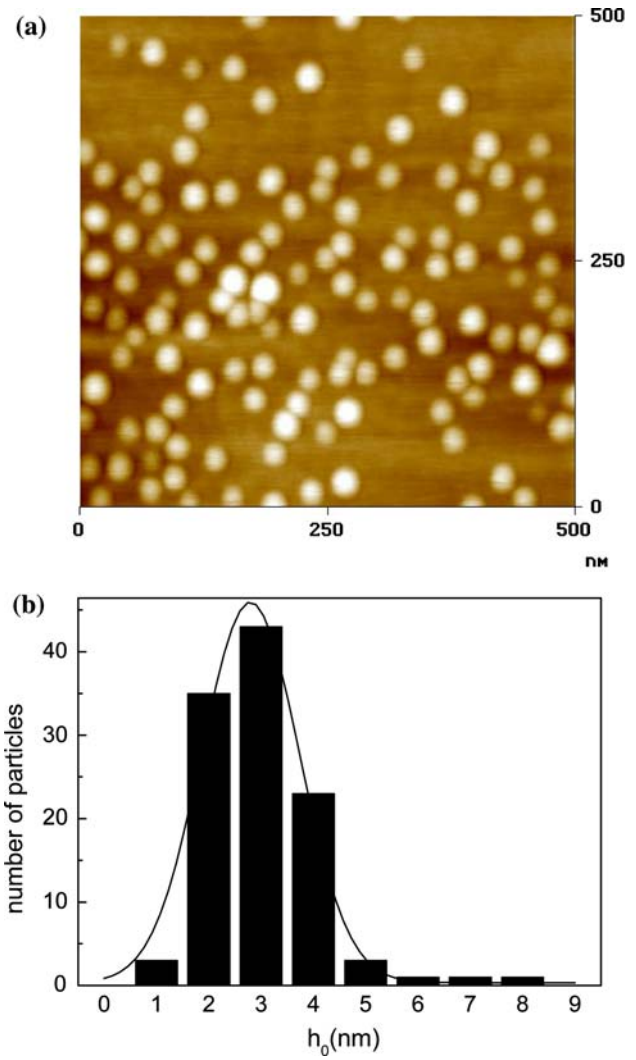

Fig. 1 (**a**) AFM image, (**b**) the size distribution of the as-grown uncapped reference QD sample

by atomic force microscopy (AFM), cf. Fig. 1a. The size distribution of the referenced InAs quantum dots is shown in Fig. 1b. InAs QDs are formed as the lens-shaped and the density is about 4×10^{10} cm^{-2}, while the mean radius and the height are about 15 and 2.8 nm, respectively. The ratio of radius to height is 5.4. As reported by Ji [13], the wetting layer (WL) thickness is estimated to be 0.5 nm. The distribution of the QDs' height can be described by a Gaussian function $P(h)$ [8, 14, 15] as follows:

$$P(h) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(\log h - \log h_0)^2}{2\log^2 \sigma}\right]. \quad (1)$$

where h is the height of QD, h_0 is the mean height, and σ is the standard deviation. However, the experimentally measured dot height distribution displayed in Fig. 1b suggests a more asymmetric distribution characterized by a higher fraction of larger dots, the dot height is best to be approximated by a Laplacian height distribution.

As shown in Fig. 1b, which was measured from the AFM experimental data displayed in Fig. 1a, we get the parameters h_0 and σ being 2.8 and 1.1 nm, respectively.

The samples have been implanted with protons at room temperature with the proton doses of 1×10^{15} ions/cm^2. Rapid thermal annealing (RTA) is carried out at the temperature of 600, 700, and 750 °C for sample B, C and D, respectively [3]. All samples are studied by the PL spectrum at 77 K.

Figure 2 presents the PL spectrum for sample A (dots). The main emission peak from optical recombination is at 1.220 eV and its full-width-half-maximum (FWHM) is 67 meV.

In Fig. 3, we show the calculation of the transition energy E between the first electron ground state and the heavy hole ground state as a function of the height h of the QD [9]. By adapting a first-order polynomial to this discrete data, we obtain the functional behaviour between E and h in our QDs as

$$E = Eg + \frac{c}{h} \quad (2)$$

where Eg is the band gap and c is an appropriately dimensioned constant. As we calculate, Eg = 0.77 eV, c = 1.37 eV/nm.

In order to simplify the calculation, we set the ratio of radius to height fixed. It is well known that the aspect ratio of the QDs decreases with increasing dot radius. In our calculation, the fixed ratio will result in the larger transition energy E, which will cause the calculated PL spectrum

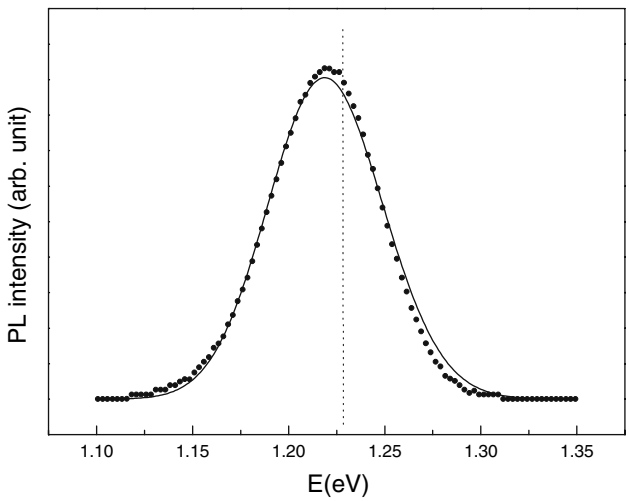

Fig. 2 Comparison of the experimental (*dots*) and theoretical (*solid line*) PL spectra at 77 K

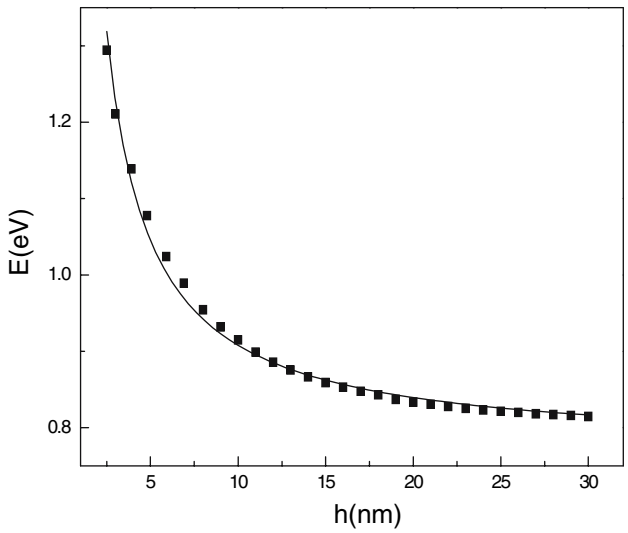

Fig. 3 Calculated (*dots*) transition energy E of self-assembled QDs as a function of the QD height h together with an adopted polynomial (*solid line*) according to Eq (2)

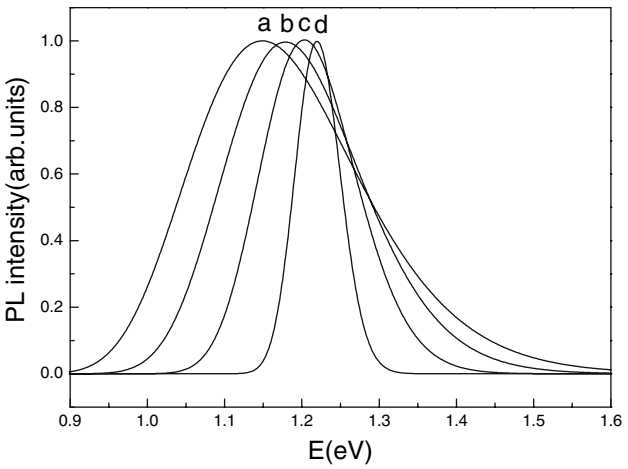

Fig. 4 PL spectra of ensembles of quantum dots characterized by different size distributions. The parameter sigma is 1.4 nm of (**a**), 1.3 nm of (**b**), 1.2 nm of (**c**), 1.1 nm of (**d**) assuming average island heights and width 15 and 2.8 nm

$P(E)$ has slightly certain blueshift. With the fixed ratio, the number of available carriers N_c in a quantum dot for radiative recombination is proportional to the cubic of h [8]:

$$N_c \propto h^3 \tag{3}$$

According to Eqs. (1–3), $P(E)$ can be represented by

$$P(E) = \frac{1}{\sqrt{2\pi}\log\sigma} b \int_0^\infty \delta(E - E_0) h^3 \exp\left(-\frac{(\log h - \log h_0)^2}{2\log^2\sigma}\right) \mathrm{d}h \tag{4}$$

where b is a suitable normalization constant.

We present the theoretical photoluminescence spectrum (solid line) calculated by Eq. (4) in Fig. 2. The theoretical spectrum is obtained with the mean height h_0 = 2.8 nm and the standard deviation σ = 1.1 nm. The theoretical curve is in good agreement with the experimental data (shown by dots). Supposing the ensemble of quantum dots being uniform (σ = 0 nm) and using the average height in calculation, the position of the PL peak (shown by the vertical dash line) is at 1.228 eV. It is 8 meV higher than the experimental one. This is caused by non-linear dependence of transition energy on the QD size, as shown in Fig. 3. This behaviour also indicates that our delicated model is most suitable for a right way to describe the QD property by PL measurement.

Figure 4 shows the PL spectra assuming different standard deviations σ of QD size distribution. The calculated photoluminescence spectra are obtained with the mean height h_0 of 2.8 nm and σ in the range 1.1–1.4 nm. As expected, the PL peak position has a redshift and the spectrum becomes broad at increasing standard deviation σ. With a small value of σ, the PL spectrum is approximately Gaussian shaped. With an increase of σ, the PL lineshape becomes asymmetric in accompanying with a short-wavelength (high-energy) band tail. This is in agreement with the experimental results reported by Cheng et al. [16].

In Fig. 5, we compare our theoretical spectra (solid lines) with the experiment data (dots) for the intermixing of samples B, C and D. The line (A) is the PL spectrum of the

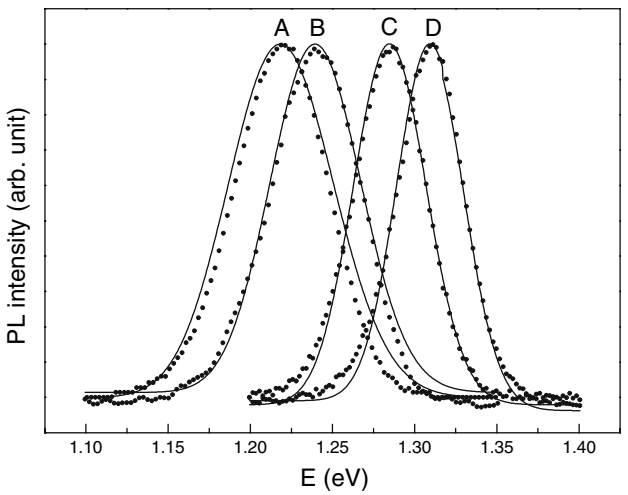

Fig. 5 Comparison of the experimental (*dots*) and theoretical (*solid line*) PL spectra for the as-grown sample A and for differently intermixed samples B, C, and D at 77 K

as-grown sample. Lines (B), (C), and (D) are the PL spectra of the samples B, C and D, respectively. The diffusion length induced by intermixing is set as an adjustable parameter in our calculation. The theoretical spectra are in good agreement with the experimental results. With the error function, based on the emission energy of the experimental PL spectra, the lengths of the interdiffusion of In–Ga atoms are deduced to be 0.23, 0.45 and 0.54 nm, respectively. The good agreement between the calculated and experimental PL curves indicates that the variations in the PL emission peak position and linewidths are mainly caused by the interdiffusion of In–Ga atoms at the interface between the QD and the GaAs barrier. An increased Ga concentration of the originally pure InAs QDs will increase the QD band gap. This will result in an increase of the transition energy. As observed in a previous report [17], for the same diffusion length, the higher height QD has stronger blueshift effect. This will lead to a narrowing of the FWHM of the PL spectra. It is clearly observed in Fig. 5 that with the increase of the interdiffusion length from 0.23 to 0.54 nm, the PL linewidth decreases from 65 to 46 meV. This linewidth narrowing phenomenon was also observed in a previous report [18].

In summary, we have studied the effect of the size distribution and the interface intermixing on the PL spectrum of self-assembled quantum dots. Based on the statistic distribution of the quantum dot sizes obtained from AFM images of uncapped dots, we have successfully modeled the associated PL spectra of caped InAs dots embedded in GaAs. The size distribution will not only broaden the PL line, but also shift the PL peak positions. Moreover, comparing the PL spectra between the as-grown sample and the intermixed samples, it is found that the intermixing induces a blueshift in the PL spectrum and a narrowing of the linewidth. This leads to the conclusion that both, the size distribution and the intermixing play important roles in the PL spectrum of QDs.

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References

1. Petroff PM, Schmidt KH, Ribeiro GM, Lorke A, Kotthaus J (1997) Jpn J Appl Phys 36:4068
2. Mukhametzhanov I, Wei Z, Heitz R, Madhukar A (1999) Appl Phys Lett 75:85
3. Ji Y, Lu W, Chen G, Chen X (2002) J Appl Phys 93:1028
4. Ji Y, Chen G, Tang N et al (2003) Appl Phys Lett 82:2802
5. Lifshitz IM (1964) Adv Phys 42:483
6. Kane CL, Lee PA, Ng TK et al (1990) Phys Rev B 41:2653
7. Swierkowski L, Szymanski J, Gortel ZW (1995) Phys Rev Lett 74:3245
8. Chen X, Zhao J, Wang G, Shen X (1996) Phys Lett A 212:285
9. Wojs A, Hawry P, Fafard S (1996) Phys Rev B 54:5604
10. Fafard S, Allen CN (1999) Appl Phys Lett 75:2374
11. Dubowski JJ, Allen CN, Fafard S (2000) Appl Phys Lett 77:3583
12. Perret N, Morris D, Franchomme-Fosse L, Cote R et al (2000) Phys Rev B 62:5092
13. Ji Y, Lu W, Chen G et al (2003) J Appl Phys 93:1208
14. Trwoga PF, Kenyon AJ, Pitt CW (1998) J Appl Phys 83:3789
15. Kohli S, Theil JA, Snyder RD et al (2003) J Vac Sci Technol B 21:719
16. Cheng W-Q, Xie XG, Zhong ZY (1998) Thin Solid Films 312:287
17. Dubowski JJ, Allen CN, Fafard S (2000) Appl Phys Lett 77:3583
18. Lobo C, Leon R (1998) Appl Phys Lett 72:2850