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Research Paper

Analysis of In/Ga Inter-Diffusion Effect on the Thermodynamical Properties of InAs Quantum Dot

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Abstract Debye temperature is an important thermodynamical factor in quantum dots (QDs); it can be used to determine the degree of homogeneity of a QD structure as well as to study the interdiffusion mechanism during growth. Direct estimation of the Debye temperature can be obtained using the Varshni relation. The Varshni relation is an empirical formula that can interpret the change of emission energy with temperature as a result of phonon interaction. On the other hand, phonons energy can be calculated using the Fan Expression. The Fan expression and Varshni relation are considered equivalent at a temperature higher than Debye temperature for InAs quantum dot. We investigated InAs quantum dot optically, the photoluminescence spectra and peak position dependency on temperature has been discussed. We applied a mathematical treatment using Fan expression, and the Varshni relation to obtain the Debye temperature and the phonon energy for InAs quantum dots sample. Debye temperature increase about double compared to bulk crystal. We concluded that the In/Ga interdiffusion during growth played a major role in altering the quantum dot thermodynamical parameters.

Keywords: Photoluminescence, InAs Quantum dot, Debye temperature Varshni relation, Fan expression

I. Introduction

Self-assembled quantum dots (QDs) are of great interest because of the nature of their self-assembled growth [1-7]. InAs QDs have received special interest because of their wide usage in various applications in optoelectronics [8-10]. Debye temperature is known to be related to the distance that allows minimum phonon energy to exist. In other words, we can estimate that phonons can exist if a large enough amount of k_BT is supplied to the quantum dots, for which the minimum distance corresponds to the phonon frequency. For InAs QDs, the Debye temperature is related to the volume of the quantum dots. When the minimum phonon energy corresponds to the atomic separations, there is a finite number of phonon energy states because phonon propagation is bound by the medium. Various methods have been used to fit the observed emission energy temperature dependency. The most widely known expressions are the Varshni expression and the Fan relation [11,12]. The applicability of the empirical Varshni expression to the QD system and its interrelationship to Fan relation was introduced by I.A Vainshtein et al [13]. It is clear that the Fan expression explicitly satisfies the phonon statistics of the crystals.

In this work, we found a high value of Debye temperature for InAs QDs. Our fitting values were found to be higher than those reported by other authors. We attribute this to In/Ga interdiffusion during growth which changes the QD volume. The variation of the band gap width with temperature is attributed to the thermal expansion of the QDs, and is related to the quantum confinement of the electrons and the holes that form excitons in the QDs. The temperature dependence of the band gap energy is introduced; the phonon energy, as well as the Debye temperature, are obtained by fitting of the temperature dependence of the peak position with the Varshni relation and the Fan expression. Empirical evidence for the Varshni relation and the Fan expression is introduced to explain the temperature dependence, of the semiconductor band gap.

II. Materials and Methods

InAs QDs were prepared by Migration Enhanced Molecular Beam Epitaxy (MEMBE) [20]. The schematic diagram of the sample structure is shown in Figure 1. Under pressure of 1×10-9 Torr, the sample was heated at 400°C for 30 minutes to start cleaning process in the

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Figure 1. Schematic diagram of InAs quantum dot structure.

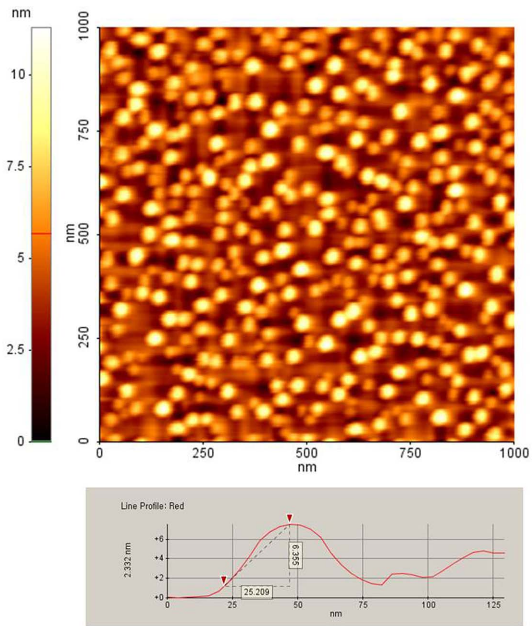

Figure 2. Atomic force microscope for InAs quantum dot sample and line profile of a typical InAs QD.

preparation chamber and then loaded into the growth chamber. The substrate temperature was raised up to 620°C for 20 minutes for deoxidation under arsenic tetramer introduction and then decreased to 590°C to allow growth of the GaAs buffer layer, which had a growth rate of 0.5 monolayer/s. This process was monitored by RHEED measurement. After the growth of the 0.5 µm-thick GaAs buffers had finished and there were 10 stacks of 2 nm thick $Al_{0.3}Ga_{0.7}As$ and 2 nm thick GaAs, the deposition process was interrupted under arsenic dimer overpressure and substrate temperature decreased to 480°C. All of the following structures were grown at this temperature. The GaAs layer containing the InAs QD layer had 10 stacks of 2 nm thick $Al_{0.3}Ga_{0.7}As$ and 2 nm thick GaAs over the buffer layer. The upper InAs QDs were grown for inspection via AFM measurement. Using the atomic force microscope (AFM) data, the QD height was found to be around 6-8 nm; the diameter was 38-50 nm. The density of the QDs in this sample was approximately 5×10^{10} cm^{-2}, as shown in Figure 2. The growth rate of InAs is equivalent to 0.07 monolayer/s. The set-up for the optical measurement was exactly described in the reference [21].

III. Results and Discussion

Figure 3 shows the temperature dependence of the PL spectra in the temperature region of 13 to 225 K; it can be seen that the PL intensity decreases with increase of the temperature due to thermal quenching. It should also be noted that the peak emission energy shifted to a lower energy as the temperature increased, which is clarified in Figure 2. The Varshni [14] relation is used to describe the temperature dependence of the band gap E_g on the temperature, it has the form of:

$$E_g(T) = E_g(0) - \frac{\alpha_1 T^2}{\alpha_2 + T} \tag{1}$$

where $E_g(0)$ is the width of the band gap at zero temperature, and a_1 and a_2 are empirical parameters, the constant α_2 has Kelvin units, and its value is close to the Debye temperature [11-13].

Another popular expression that describes the temperature dependence of the band gap is the Fan expression[17] it has the form

$$E_g(T) = E_g(0) - A(\langle n\rangle + \text{constant}) \tag{2}$$

Because phonon follow Bose-Einstein statistics, Equation 2 describes the temperature dependence of the phonon energy and its relation with the temperature dependence of the band gap $\langle n\rangle$ has the form:

$$\langle n\rangle = \frac{1}{\exp(\hbar\omega / k_B T) - 1} \tag{3}$$

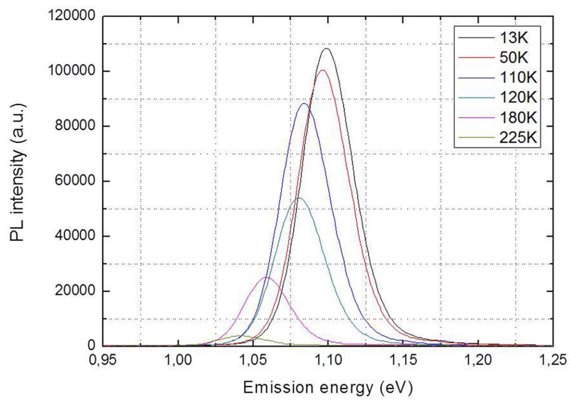

Figure 3. Photoluminescence spectra of the InAs quantum dot in the temperature range between 13 and 225 K.

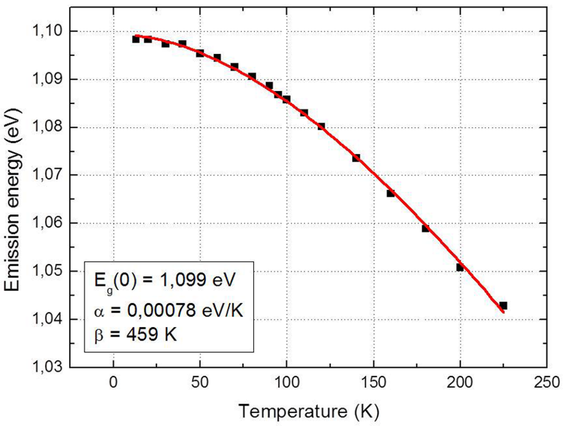

Figure 4. Fitting using the Varshni relation. Black squares are assigned to experimental data and the red solid line indicates the InAs band gap temperature dependence obtained by Varshni. In the inset-fitted values are shown.

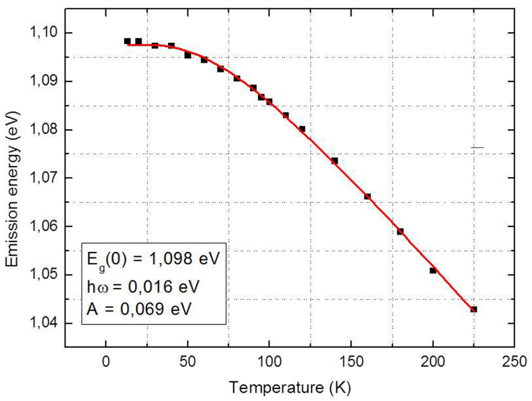

Figure 5. Fitting using the Fan equation. Black squares are assigned to experimental data and the red solid line indicates InAs band gap temperature dependence obtained by Fan. In the inset-fitted values are shown inset.

where is the phonon energy and A is the Fan parameter, the Fan parameter depends on microscopic properties of the material [13].

It should be noted that, due to the thermal lattice expansion, the Bose-Einstein statistical distribution of phonon will change, and so cause a shift in energy levels. The shift in energy level is proportional to the factor <n> [13,15].

This will lead us to the idea that increasing the temperature causes the occupation number of phonons in the phonon states to increase and so shift the emission energy to lower wavelength. It was shown by I.A Vainshtein et al. [13] that the parameter A has a dimension of energy that coincides in magnitude with the band gap at the temperature point, where the mean number of phonons responsible for the shift in the energy levels is equal to unity [13]. This temperature point is known as the Debye temperature. Therefore there must be a sufficient number of phonons to cause the shift. In Figures 4 and 5 fittings of the temperature dependent emission energy with the Varshni relation and the Fan expression respectively are shown. The fitting process yields the following values:

Table 1. The Varshni and the Fan coefficients for the PL peak position temperature dependence.

Peak name	Varshni coefficients		Fan coefficients	
	α_1, 10^{-4} eV/K	α_2, K	A, meV	ħω, meV
		InAs		
Ref. 11	3.08	70	40*	11.5*
Ref. 12	3.7	120	57.5*	14.7*
Ref. 12	2.5	75	26.2*	9.62*
Ref. 13	4.19	271	76.6*	20.8*
Ref. 14	5.405	204	96.2*	18.66*
Ref. 15	4.58	243	83.2*	20*
Ref. 15	4.54	210	81.2*	18.9*
InAs QD (this work)	7.8	459	70	16

phonon energy=16 meV, $E_g(0)$=1.099 eV, Fan parameter= 70 meV, and Debye temperature=459 K, this last of which is very high with respect to values reported by other authors. Comparison of the fitting parameters from this work with those of other authors's work is shown in Table 1. It is clear that the Varshini coefficients and the Fan coefficients have a linear relationship, however our results shows relatively large Debye temperature.

Since phonons are lattice vibrations, as the phonon energy increase the atom displacement due to oscillation increase. The constant D is defined as the deformation potential due to lattice oscillation; this constant is related to the Fan Parameter through Equation [13].

$$A = \frac{\hbar}{\omega M} D \tag{4}$$

where M is the mass of the oscillator; then, it is clear that the Debye temperature, as well as parameter A, defines the lattice separation and deformation potential due to oscillation. Large values of the fitting parameters, are attributed to In/Ga interdiffusion during growth [16]; the interdiffusion process is responsible for changing the lattice separation. Full understanding of the interdiffusion mechanism and its effect on the thermodynamical parameters are essential for device fabrication based on the InAs QD structure [17-20].

IV. Conclusions

The PL of a MBE-grown InAs QD sample is investigated. The temperature dependence of the peak position is studied on the basis of the Varshni relation and the Fan expression. The obtained thermodynamical parameters are found to be higher than values reported by other authors. This discrepancy is attributed to the occupation number of phonons in phonon energy states which is directly attributed to the In/Ga interdiffusion that took place during QD growth.

Acknowledgements

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