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Detection of optical emissions from deep localized states in a self-assembled InAs/GaAs QD structure

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ABSTRACT

This work reports on the optical properties of self-assembled InAs/GaAs quantum dots (QDs) grown by molecular beam epitaxy. A sigmoidal temperature-dependent variation of the bandgap energy of the dots is detected from the photoluminescence (PL) investigations of the studied sample. This sigmoidal dependence of the bandgap energy was accompanied by an anomalous increase in the integrated PL intensity (IPLI) over the temperature range of 10 K to 110 K. Such optical observations are related to the existence of deep localized states (LS) within the QD structure. To illustrate the presence of the LS, pump power-dependent PL measurements at different temperatures were performed. It is found that the sigmoidal dependence of the PL peak energy (PLPE) gradually converts into a monotonous variation with the gradual increase in the excitation density. These observations show a saturation behavior of a limited number of states within the bandgap of the QDs. Such optical results provide clear evidence for the existence of deep levels within the InAs dots.

1. Introduction

Semiconductor-based quantum dots made by self-organized growth are the subject of intensive studies due to the particular nature of their electronic states [1–3]. Owing to their important optical properties close to the infrared wavelength regime, Stranski-Krastanov InAs/GaAs QDs are considered among the most promising candidates for the fabrication of intermediate band solar cells as well as for optoelectronic devices [4–6]. The QD structures are also of great interest in the exploration of bio-imaging and pharmaceutical applications [7–9]. Such applications require the growth of QDs produced from natural organic materials which can generate a strong optical response compared to some semiconductor QD systems [7]. It is well known that the performance of such devices strongly depends on the thermal effects that relate the spontaneous optical emission to the non-radiative recombination processes [10, 11]. Generally, the increase of the PL measurement temperature in the semiconductor-based QD systems causes an energy reduction in the

inter-band radiative transitions, a gradual quench of their optical signals, and a broadening of their line-widths. These behaviors are owing to the accumulation of several thermal effects such as the expansion of the crystal lattice and the coupling between electrons and phonons [12]. However, many studies [13–15] have indicated that some other abnormal thermal effects could cause significant changes in the temperature dependence of the photoluminescence characteristics for such QD structures. Indeed, the presence of spatially localized deep levels in these structures can induce a sigmoidal dependence of the emission peak energy (S-shape feature), which is often accompanied by an improvement in the PL signals with increasing temperature [13, 16]. This unusual temperature dependence of the PL characteristics has been attributed to the thermal redistribution of charge carriers between the localized states and the ground states of the dots [15–17]. Several published experimental and theoretical works have demonstrated that the LS could coexist with the intrinsic quantum states of the InAs dots [18–21]. Based on deep level transient spectroscopy (DLTS)

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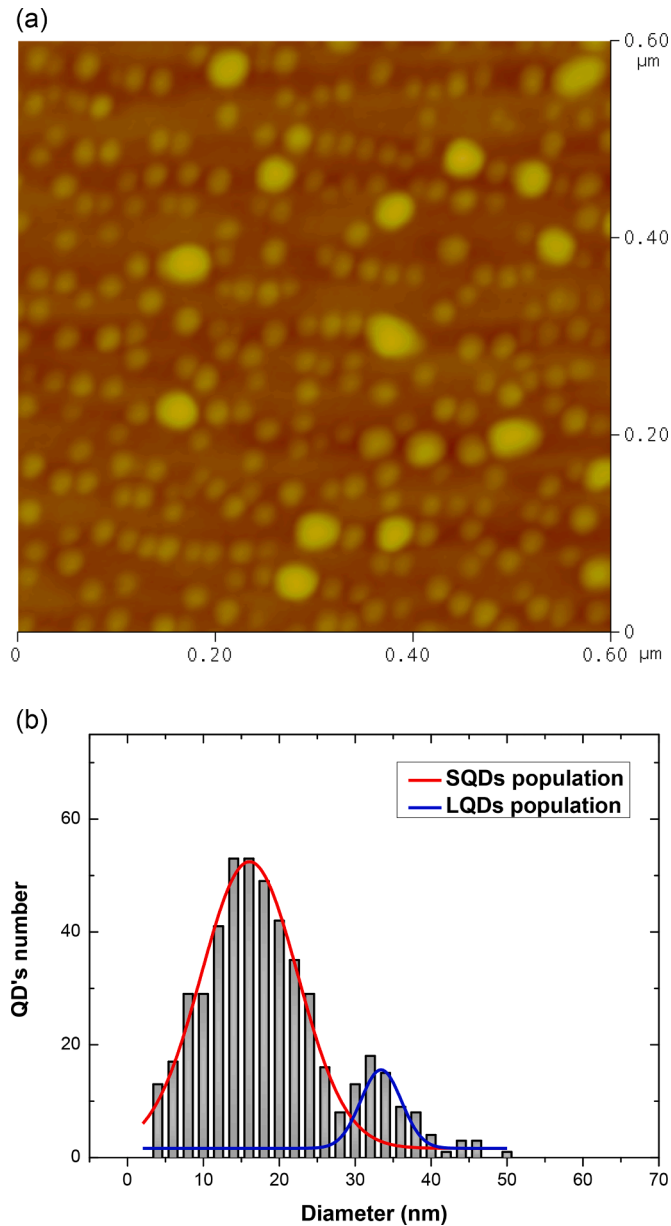


Fig. 1. (a): $0.6 \times 0.6 \mu\text{m}^2$ AFM image of the uncapped QD sample. (b): Lateral size distribution of the dots corresponding to the AFM image.

Table 1

Structural parameter values of the investigated QD sample. $\bar{\sigma}$ is the areal density, \bar{d} is the average diameter and \bar{h} is the average height of the dots.

AFM details	SQD class	LQD class
$\bar{\sigma} (\times 10^{10} \text{ cm}^{-2})$	6.9	0.8
$\bar{d} (\text{nm})$	15 ~ 16	33 ~ 34
$\bar{h} (\text{nm})$	4 ~ 5	8 ~ 9

measurements of self-assembled InGaAs/GaAs QDs system [18], T. Asano et al. have shown the presence of deep-level traps located around the GS of the InGaAs/GaAs dots. In this reported work, the activation

energies, the spatial profile and the estimated densities of the deep-trap levels have been theoretically determined. On the other hand, using Bias-dependence calculations of DLTS spectra [19], S. W. Lin et al. have presented direct experimental evidence of the existence of such deep LS within the InAs/GaAs dots themselves. Although it was predicted that the localized states might exist in the vicinity of the InAs dot's barrier, only a few optical works have reported the sigmoidal temperature-dependent variation of the bandgap energy for self-assembled InAs QDs [13, 14]. In addition, spectroscopic studies of PL reporting the presence of deep LS in a single layer of InAs/GaAs QDs are extremely limited.

In this work, temperature-dependent PL measurements are investigated for self-assembled InAs/GaAs QDs grown by molecular beam epitaxy. A sigmoidal temperature-dependent variation of the PLPE is observed. This sigmoidal dependence of the PLPE is accompanied by an anomalous enhancement of the PL signals. Such behaviors are related to the presence of deep LS within the studied structure. PL measurements as a function of the excitation density are carried out to illustrate the eventual existence of the LS within the InAs dots.

2. Experimental details

The structure studied in this work contains a single layer of InAs QDs (wetting layer) sandwiched in a GaAs matrix [22]. The Stranski-Krastanov QDs were grown by molecular beam epitaxy on (001)-oriented GaAs substrates. Gallium and indium fluxes were supplied from thermal effusion cells, As_2 species from a cracking source. The growth procedure for the QDs deposition is the following. First, a $0.3 \mu\text{m}$ thick GaAs buffer was grown at 600°C with a rate of 0.2 nm/s . Subsequently, the substrate temperature was decreased to 490°C in order to allow deposition of the wetting layer. The thickness of the deposited InAs was fixed to 2.4 monolayers with a growth rate of 0.04 nm/s . A 100 nm thick GaAs cap was immediately grown after, while the growth temperature was increased to 600°C . Another sample was grown for the atomic force microscope (AFM) measurements using the same conditions but without the cap layer. Morphology analysis was performed using the tapping mode of a Topometrix *TMX 2000 Explorer* atomic force microscope. PL measurements were carried out using a closed-cycle He cryostat and a 514.5-nm line of an Ar^+ laser. Optical spectra were collected with a $f = 0.25 \text{ m}$ spectrometer with a 1200 l/mm grating and focused onto a cooled GaInAs photodiode detector.

3. Results and discussion

3.1. Structural details

Fig. 1a depicts the AFM image of the uncapped QD sample and Fig. 1b shows the corresponding lateral size histogram of the dots. The structural details of the dots are summarized in Table 1. A careful analysis reveals two families of large and small dots that differ from the point of view of surface density and average size. A population of large quantum dots (LQDs), with diameter around 33 nm and height $8\text{--}9 \text{ nm}$ is observed on the surface with a density of $8 \times 10^9 / \text{cm}^2$. In comparison, the small quantum dots (SQDs) group with $15\text{--}16 \text{ nm}$ diameter and $4\text{--}5 \text{ nm}$ height has a much higher surface density ($7 \times 10^{10} / \text{cm}^2$) and a therefore smaller lateral spacing. Such non-homogeneity of the dot size may be due to the coalescence of a fraction of the InAs dots during the growth of the wetting layer at 490°C . Optimization of growth conditions in molecular beam epitaxy helps to reduce this effect [23].

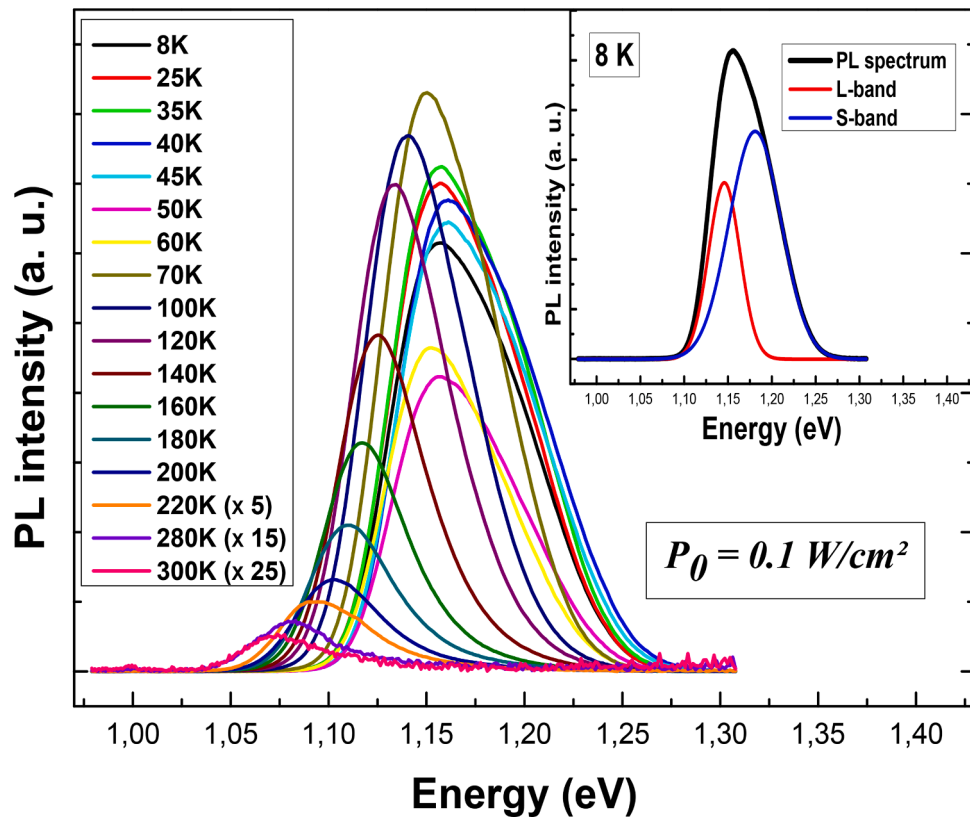


Fig. 2. PL spectra of the studied sample taken at different temperatures under an excitation density of about 0.1 W/cm^2 . The inset shows that the 8K-PL spectrum of the studied sample can be well fitted with two-Gaussian line shapes (S and L).

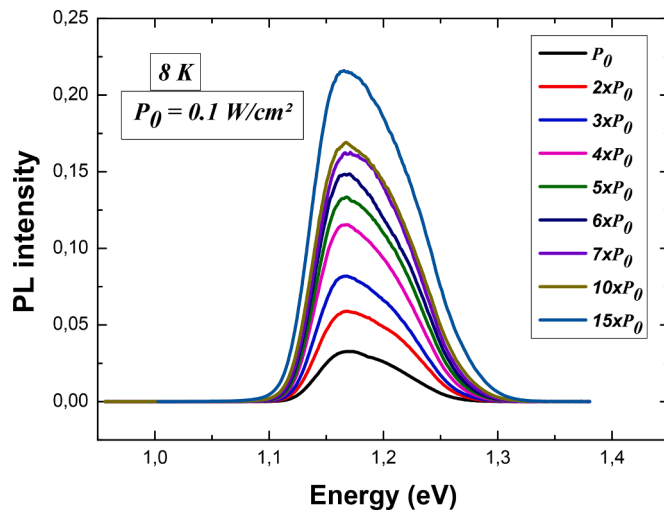


Fig. 3. 8K-PL spectra from the studied sample taken at different excitation densities.

3.2. Temperature PL study

Fig. 2 exhibits the PL spectra of the studied sample in the 8–300 K temperature range under an excitation density of about P_0 ($P_0 = 0.1 \text{ W/cm}^2$). The inset of this figure shows that the 8K-PL spectrum of the QD

sample highlights the presence of two well-defined sub-bands, namely S and L. The energy difference between the peak positions of the two sub-bands is about 40 meV. Referring to the structural details which demonstrate a bimodal QD size distribution within the studied sample and considering, on the other hand, the dependence on the dot size of the quantum confinement potentials [24], the S and L bands are attributed to the radiative recombination from the SQDs and LQDs, respectively [25]. This assignment is also supported by the pump power-dependent 8K-PL measurements (see Fig. 3), which show no PL emissions from the excited states of the dots. It should be noted that the average sizes of the two families of dots are sufficiently distinct (see Table 1), so that their emission bands are distinguishable. This is consistent with many theoretical studies that predict an energy shift of the InAs QD bandgap of about 60 meV by increasing the dot size twice [24].

The PL spectra in Fig. 2 are fitted with two-Gaussian line shapes. The evolutions of the integrated PL intensity and the PL peak energy as a function of the temperature are determined from fitting results and are represented in Figs. 4a and 4b, respectively. For the two bands, it can be seen from Fig. 4a that the dependence on the temperature of the integrated PL intensity exhibits an anomalous enhancement in the low-temperature region. Meanwhile, Fig. 4b shows that the PL peak energy of the two bands exhibits a sigmoidal temperature-dependent variation. Indeed, with the increase of the temperature from 8 K to 30 K, the PLPE of the two bands remains constant (see Fig. 4b), then quickly increases till 45 K, and then decreases with a further increase in the temperature. This sigmoidal temperature dependence of the PLPE presents a large deviation from the band gap change of the InAs material

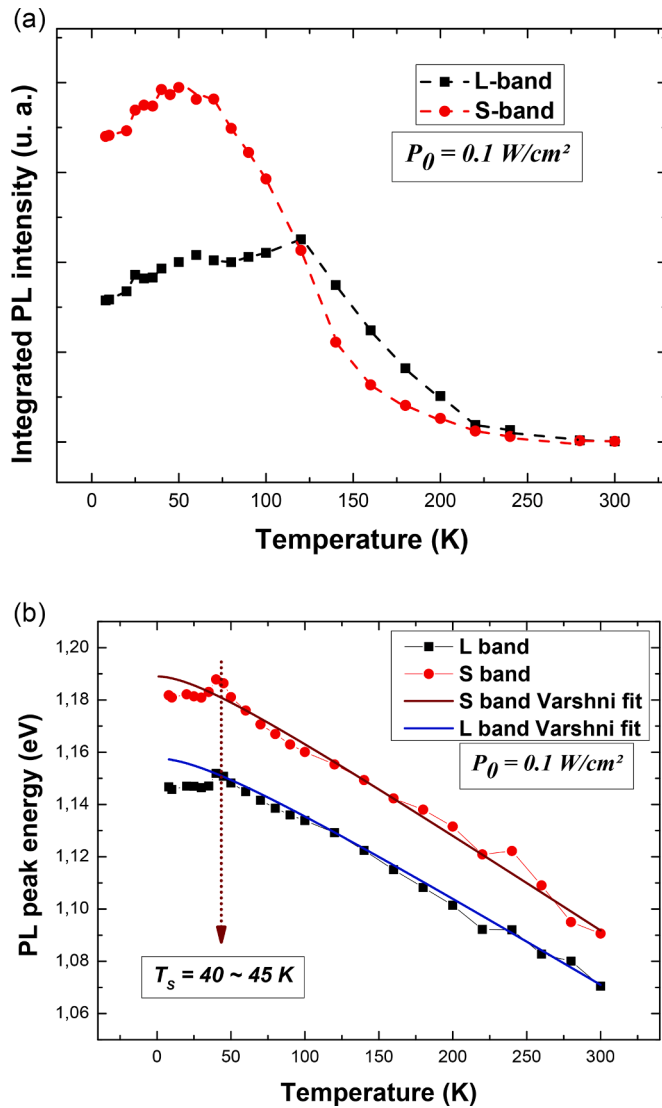


Fig. 4. (a): Temperature dependence of the integrated PL intensity for both S and L sub-bands under an excitation density of P_0 . (b): Temperature dependence of the PL peak energy for both S and L sub-bands under an excitation density of P_0 . The fit of the experimental data to the Varshni law [23] (solid line) is also added in this figure.

according to the Varshni law [26]. Such a sigmoidal dependence of the PLPE is characterized by two interesting parameters, namely T_s and ΔE_s . T_s is the temperature of turning out to red-shift of the PLPE, and ΔE_s is the value of the energy difference between the PLPE at T_s and that at 8 K. As pointed out by many optical studies [13–17], the unusual blue-shift occurring in the temperature dependence of the PLPE for self-assembled QD structures is originating from the presence of deep localized states that exist within the QD bandgap. Compared to that of the electronic GS of the dots, the localized states should have shallower confinement levels and a much weaker density of states [19] (see Fig. 5). The value of the energy difference between the GS and the LS is usually defined as the activation energy for the temperatures under study. The recorded abnormal deviation of the temperature dependence of the

PLPE from that of the InAs bandgap change (see Fig. 4b) can be explained by the thermal redistribution of the confined carriers between the LS and the GS [13–17]. Indeed, with the increase in temperature, the carriers trapped in the LS are thermally re-emitted towards the GS where they will take part in the radiative emission (blue-shift of the PLPE) [16].

As can be seen from Fig. 4, both the sigmoidal dependence of the PLPE and the anomalous enhancement of the IPLI occur in the same temperature region, they are probably arising from the same origin. With increasing temperature, the improvement in the IPLI observed in Fig. 4a suggests that the loss of carriers via non-radiative processes is compensated by some mechanism providing carriers to the GS of the dots. Generally, for a bimodal structure of InAs QDs, the redshift of the PL transition of the SQD class with increasing temperature is much faster than that of the bandgap change of the InAs material [27, 28]. In addition, for such a structure, the increase of the temperature can induce an improvement in the PL signal of the LQD class [29]. These behaviors are due to the carrier redistribution between the QDs in different size distributions [28, 29]. However, with the increase of the temperature from 8 to 110 K, Fig. 4a shows that the two families of dots have a similar improvement of their integrated PL intensities. This suggests that the thermal redistribution of carriers via neighboring dots is not appreciable in the studied structure. As a consequence, the carrier dynamic processes that involve the PL emission are probably independent of the QD's size. This is consistent with the behaviors of the integrated PL intensity and the PL peak energy shown in Fig. 4. Indeed, the two families of dots exhibit practically the same tendencies in the temperature dependence of their PL characteristics. It should be noted that the anomalous enhancement of the integrated PL intensity observed in Fig. 4a cannot be related also to the supply of carriers via the thermal transport of electrons through the wetting layer and/or the thermal release of carriers trapped in non-radiative centers. In fact, these mechanisms should not induce the blue-shift of the PLPE [27]. In view of these reasons, the anomalous enhancement of the IPLI could be attributed to the presence of deep LS within the studied structure [13–17]. Therefore, the present optical results are explained as follows (see Fig. 5). For temperatures smaller than T_s , the lower energies are favored, the trapping process of photo-generated carriers in the deep LS is therefore much more efficient than that in the GS. With increasing temperature from 8 K to T_s , the GS are gradually repopulated via the thermally escaped carriers from the LS. This aspect simultaneously induces a gradual improvement in the PL signal of the QDs and a blue-shift of the energy bandgap (see Fig. 4). With a further temperature increase ($T > T_s$), most of the carriers trapped in the LS are thermally emitted towards the GS. Then, as can be found in III-V semiconductor materials, conventional thermal effects that deteriorate the PL yield (such as the crystal lattice expansion and the electron-phonon coupling) lead to regular evolutions of the PL peak energy and the integrated PL intensity (bandgap reduction and quench of the PL signals, respectively).

3.3. Pump-power PL study

In order to illustrate the presence of the LS within the studied structure, pump power-dependent PL measurements at different temperatures were carried out. The experiment results extracted from the PL measurements are plotted in Fig. 6 which depicts the temperature dependence of the PL characteristics under several excitation densities. Compared to the GS, the LS should have a much lower density of states [19]. Due to the band filling effects, the increase in the excitation density at 8 K should quickly saturate the deep LS [15, 17]. Then, the

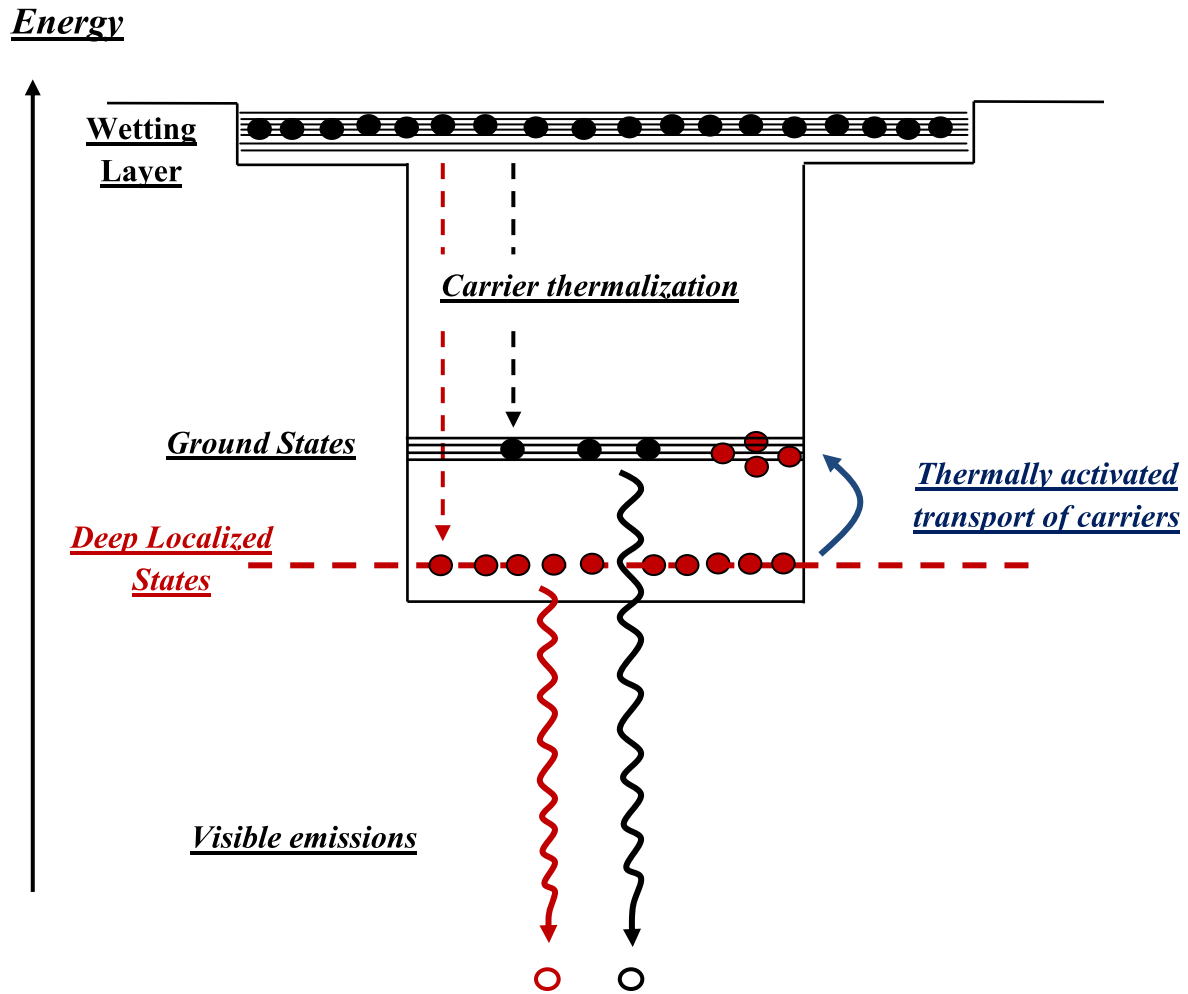


Fig. 5. Schematic representation of the energy band diagram of the studied sample. The diagram includes the energetic location of the localized states with respect to the QDs.

contribution of the LS to the PL emission should be progressively masked by that coming from the GS of the dots [13–17]. Consequently, the sigmoidal temperature-dependent variation of the PLPE seen in Fig. 4b should gradually transform into a monotonic variation with increasing excitation density [15, 16]. In the high excitation regime, a typical bandgap reduction of the PL emission with increasing temperature should be evident [16, 17]. It can be clearly seen from Fig. 6b that the value of ΔE_S decreases with increasing excitation density. The abnormal blue-shift of the PLPE is completely disappeared for an excitation density of approximately $15 \times P_0$. These optical observations illustrate that at pumping levels for which the filling of the localized states is complete, the anomalous evolution of the PL peak energy versus temperature seen in Fig. 4b is transformed into a monotonic variation. As pointed out by many studies [15–17], the decreasing behavior of the ΔE_S value with the increase of the excitation density is attributed to the gradual saturation of the LS due to the band filling effects. Therefore, the present optical results provide clear evidence for the existence of deep LS within the studied structure. It can be also noted that due to the presence of the LS, the anomalous improvement in the IPLI with increasing temperature is much stronger at higher excitation densities (see Fig. 6a) [15, 17]. This

is possibly due to the fact that the GS have a higher radiative recombination efficiency than that of the LS.

The coexistence of deep LS with the intrinsic quantum states of the InAs dots has been related to several causes which depend on the growth processes [18, 21]. The localized states can result from the migration of Ga adatoms to the InAs wetting layer during the growth of the QD structure [18]. They can also result from the formation of point defects in the volume of the dots which are due to the growth processes of lens-shaped dots [20]. Deep levels within the dots themselves could also be caused by the lattice mismatch between InAs and GaAs [19]. Defects, such as vacancies and interstitials, which are due to the incomplete relaxation of the residual compressive strain in the vicinity of the dots, can also lead to the appearance of the LS [18]. On the other hand, from DLTS measurements [19], S. W. Lin et al. have suggested that the presence of deep LS in self-assembled InAs QDs cannot be related to the misfit dislocations, nor to the Stranski-Krastanov QD formation process. The coexistence of the LS with the intrinsic states of the InAs dots is of great interest for the design of memory devices and/or single-photon emitters as well as for the fabrication of high-efficiency photovoltaic devices [30, 31].

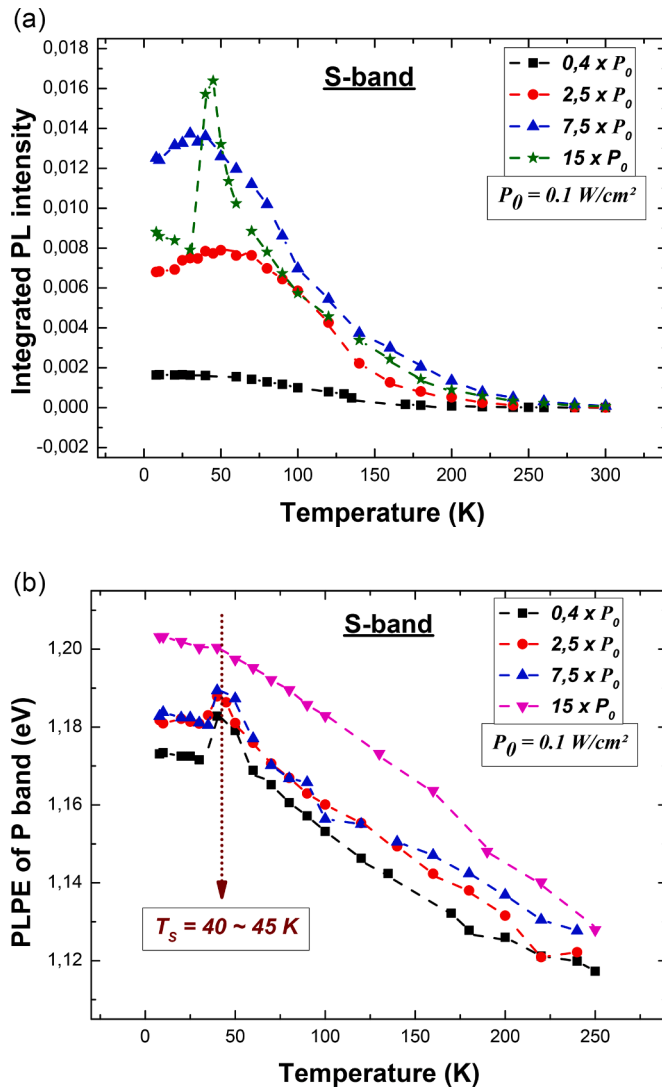


Fig. 6. (a): Temperature dependence of the integrated PL intensity for both S and L sub-bands recorded under several excitation intensities. (b): Temperature dependence of the PL peak energy for both S and L sub-bands recorded under several excitation intensities.

4. Conclusion

In summary, structural and optical properties are investigated for self-assembled InAs/GaAs QDs. The 8K-PL spectrum of the QD sample exhibits two well-defined sub-bands, namely S and L. The S and L bands are identified as the PL emission from a bimodal size distribution of dots within the studied sample. For the two bands, an abnormal blue-shift of the PLPE is observed with increasing temperature. Simultaneously, an anomalous increase in the integrated PL intensity is detected over the temperature range of 10 K to 110 K. Such optical behaviors are related to the presence of deep LS within the studied structure. To illustrate the made interpretation, temperature-dependent PL measurements under several excitation densities were carried out. The abnormal blue-shift of the PLPE is found to gradually disappear with increasing excitation density. This behavior is related to a saturation phenomenon of a weak density of deep localized states. Such optical observations provide clear evidence for the existence of deep levels within the InAs dots.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

There are no financial interests/personal relationships which may be considered as potential competing interests

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