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Influence of intrinsic internal field on recombination kinetics of high coverage (N11) InAs/GaAs quantum dots

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Abstract. We present, by means of cw and time–resolved photoluminescnce, a detailed experimental study of the optical properties of a large set of InAs self–assembled quantum dots grown on (N11)A/B GaAs substrates with different InAs coverages. Large variation of the external PL efficiency is observed, with a strong asymmetry between the A and B substrate termination. The analysis of PL time evolution leads us to exclude that the reduction of PL intensity would be associated to an increase of the non radiative recombination rates. The PL efficiency and decay times of the complete series of samples can be understood as a consequence of a large built-in electric potential associated to piezoelectric field and permanent dipole moment inside the QDs.

PACS. 78.67.-Hc Optical properties of quantum dots – 77.65.Ly Strain-induced piezoelectric fields

1 Introduction

The III–V semiconductors are piezoelectric because of the lack of inversion symmetry and when a lattice mismatched heterostructure is grown on substrates oriented along suitable crystallographic directions built–in piezoelectric field is generated. The internal field can dramatically modify the optical and electrical properties of the heterostructures, *via* the spatial separation of the electron and hole wavefunctions. The knowledge and the control of the piezoelectric field in III–V heterostructures are therefore of the utmost relevance for both application and fundamental physics. Increasing attention has been recently devoted to the role of internal field on the electronic structure and optical properties of the wurzite III–V nitride semiconductor systems [1,2]. However in GaN based heterostructures, spontaneous and piezoelectric polarizations are comparable in magnitude and the discrimination between the two contributions is indeed matter of a current debate [2].

The higher crystallographic symmetry simplifies the physics in the zincblende III–V systems. The spontaneous polarization is zero, while the piezoelectric polarization, originated by the shear strain, can be controlled by growing on proper substrates. Piezoelectric effects in InGaAs/GaAs quantum wells (QW) grown on (N11) substrates have been largely investigated in the last few years, leading to a well–established understanding of the problem [3,4]. Also, the extra degree of freedom in the band gap engineering of semiconductor heterostructures, provided by the piezoelectric field, has been fruitfully exploited in various optical and electronic QW devices [4]. Much less is known on the piezoelectric effects on self–aggregated InAs/GaAs quantum dots (QD), despite the large current interest on their electro–optical properties driven by the goal of very efficient lasers. Only very recently this problem has been investigated in few letters addressing different peculiar aspects. The reverse quantum confined Stark (QCS) effect has been reported in reference [5] in a series of (N11) InAs/GaAs QDs by means of cw photoluminescence (PL) measurements. The direct QCS effect has been investigated by means of photocurrent spectroscopy in a (311)B InGaAs/GaAs QD in reference [6]. The dynamic QCS effect has been analyzed by means of time–resolved PL (TR-PL) in reference [7].

The aim of this paper is to extend the previous studies [5,7] performed on QDs close to the 2D–3D growth transition (1.8 ML of InAs coverage) to QDs with higher coverage and therefore leading to larger QDs, where the QCS effects should be increased. We present and discuss the effects of piezoelectric fields on both optical and dynamic properties of the carrier recombination of (N11) InAs/GaAs QDs with 2.3 and 2.8 ML InAs coverage. We have analyzed a large set of samples grown on several (N11) GaAs substrates with both A and B termination,

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by means of cw and time–resolved PL spectroscopy. Large variation of the external PL efficiency is observed, with a coverage dependence and a strong asymmetry between the A and B substrate termination. The analysis of PL time evolution leads us to exclude that the reduction of PL intensity is be associated to an increase of the non radiative recombination rates. In agreement with the results of references [5,7] we attribute the observed phenomenology to the combined effects of the piezoelectric field and permanent dipole moment in InAs (N11) QDs.

2 Sample growth and experimental details

The samples under investigation were fabricated by molecular beam epitaxy using a Varian Gen–II system on liquid encapsulated Czochralski semi–insulating GaAs substrates with the following seven orientations: (100), (211)A/B, (311)A/B and (511)A/B. In order to reduce the influence of extrinsic effects (such as impurity contamination and so on), each growth run was carried out simultaneously on the three (N11) GaAs substrates with the same termination. The structures consisted in a 0.3 μm thick undoped GaAs buffer followed by a 15 × (3.8 nm $Al_{0.33}Ga_{0.67}As$ + 3.4 nm GaAs) superlattice, a 0.2 μm undoped GaAs, a few 10^{15} InAs molecules cm^{-2} (the QD region) and a 30 nm undoped GaAs capping layer. For each orientation two growths were performed with $\approx 1.2 \times 10^{15}$ and $\approx 1.5 \times 10^{15}$ InAs molecules cm^{-2}. These values correspond, respectively, to 2.3 and 2.8 monolayers (ML) on the (100) surface and we will use this denomination from here after. The growth temperature was 630 °C except during the growth of InAs and capping layer when the temperature was lowered to 500 °C, the samples were rotated during growth to improve uniformity.

We performed cw PL spectra by using an Ar^+ laser as excitation source on samples immersed in superfluid ^{4}He ($T = 2$ K). PL emission was analyzed with a Fourier transform spectrometer operating with an InGaAs photodetector. Time resolved measurements have been performed by using a ps Ti–sapphire laser (2 ps pulse duration, 760 nm excitation wavelength, 80 MHz repetition rate) at $T = 10$ K with an average excitation power density in the range of 20–200 Wcm^{-2}. The PL, analyzed with a double monochromator, was detected by a streak camera apparatus (20 ps time resolution, 2 ns time window). PL spectra, in both cw and time resolved experiments, were corrected for the spectral response of the detection systems.

3 Results

Figure 1 reports the PL spectra of all the investigated samples at $T = 2$ K. The excitation power was 30 Wcm^{-2}, except for the case of (211)A and (311)A at 2.8 ML samples where 1 $kWcm^{-2}$ was used, due to the extremely low PL efficiency of these samples. The observed PL bands correspond to the emission from the QD fundamental transitions and their broadening reflects the inhomogeneous size fluctuations in the QD ensemble. Independently of the substrate orientation the emission energy decreases as InAs coverage increases due to an average increase of dot size with InAs coverage. The PL bands tend to become structured with pronounced shoulder indicating a bimodal size distribution of the QDs. In fact, in most cases QDs show, for a given substrate orientation, the same two components in the PL spectra. The analysis of the relative weights suggests that, in those cases, the increase of the InAs coverage does not change the QDs size but only increases of the number of larger dots. In addition we find that, in general, the emission energy increases with $1/N$, showing that the QD size depends on the substrate orientation. Minor differences are observed between A and B terminations, even if the QDs grown on B substrates results slightly larger in size.

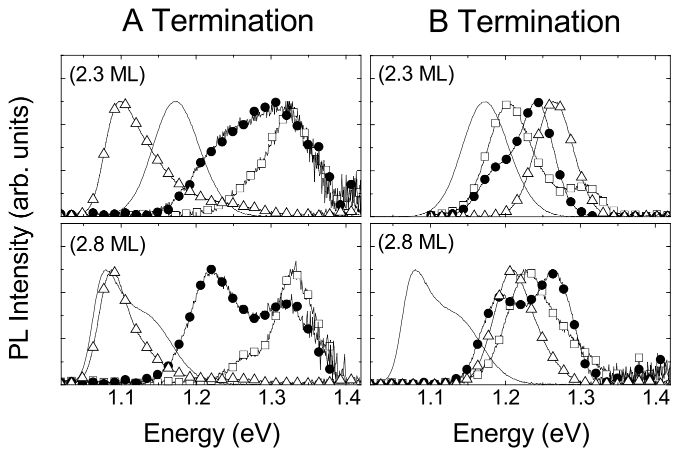

Fig. 1. PL spectra of the samples at $T = 2$ K with cw excitation power density of 30 Wcm^{-2}, except in the case of (211)A and (311)A at 2.8 ML samples where 1 $kWcm^{-2}$ was used. Different symbols refer to different substrate orientations: (211) squares, (311) circles, (511) triangles. (100) is indicated by a solid line. Each spectrum is normalized to its maximum.

In Figure 2 we report, as a function $1/N$ ($N = \infty$ is the (100) plane), the PL integrated intensity of the (N11) QDs at $T = 2$ K at low excitation power density, normalized to the 2.3 ML (100) sample (PL efficiency from now on). We find a strong reduction of the PL intensity for large values of $1/N$ and the effect is more pronounced in the 2.8 ML set with respect to the 2.3 ML set. Also an asymmetry is found between A and B termination. The PL efficiency of (N11)A samples drops by more than two order of magnitude when the QD size is increased, while on the (N11)B side the PL quenching is definitely less pronounced.

Additional pieces of information are obtained by TR-PL measurements. Typical time evolution of the emission at the peak energy of the PL bands for two (211)A samples – showing the largest PL quenching – with different InAs coverage is reported in Figure 3a. In agreement with cw data, we find a strong decrease of the PL intensity when increasing the InAs coverage. On the contrary, only minor differences are found in the time evolution of the two samples. The PL lifetime and risetime show a much less

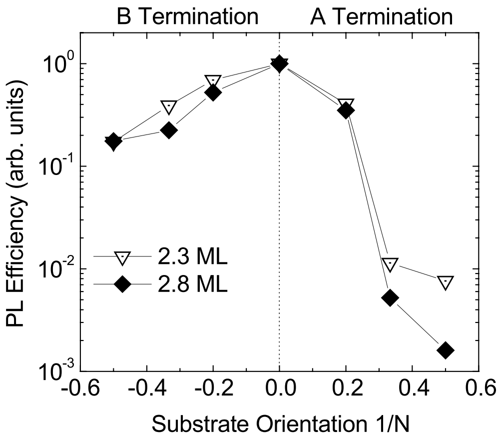

Fig. 2. PL integrated intensity normalized to the 2.3 ML (100) QD emission as a function of the substrate orientation and termination. Different symbols refer to different InAs coverages: 2.3 ML (triangles) and 2.8 ML (diamonds).

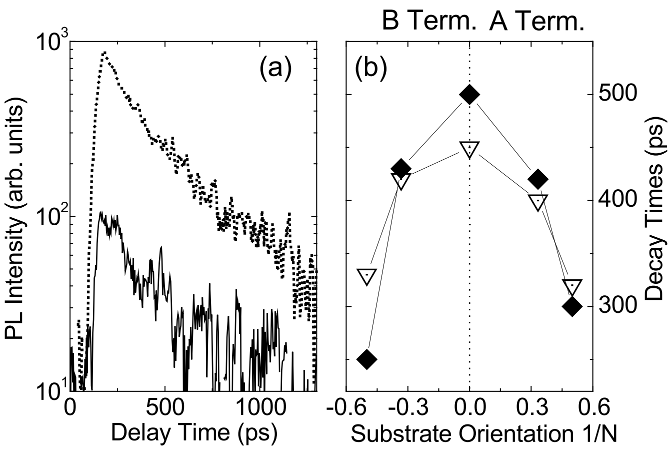

Fig. 3. (a) Time evolution of the emission at the peak energy of the PL bands for two (211) A samples with different InAs coverages: 2.3 ML (dotted line), 2.8 ML (continuous line). (b) Summary of the decay times as a function of the substrate orientation and termination. Different symbols refer to different InAs coverages: 2.3 ML (triangles), 2.8 ML (diamonds).

pronounced reduction with respect to the PL intensity. The data have been fitted with a phenomenological model based on the difference between two exponential decays after a convolution with the experimental response function. The measured risetime of the PL from the QD is quite fast (30 ps) and almost constant for all the structures investigated. Such value of the QD risetime is in agreement with previous findings in InAs/GaAs(100) QDs [8]. A summary of the decay times is reported in Figure 3b. The estimated error on the presented data is of the order of 10%. The decay times show a slight decrease when decreasing the Miller index N. However the variations are quite small: all the decay times are in the range of 380 ± 120 ps, in striking contrast with the dramatic decrease of the PL efficiency when increasing the InAs coverage on high Miller index substrates, reported in the previous section.

4 Discussion

The reverse photoinduced quantum confined Stark effect has been already observed in (N11) InAs/GaAs QDs [5,7] with 1.8 ML of InAs coverage and related to the presence of an intrinsic built-in field due to the combined effect of the piezoelectric field and permanent dipole moment. The total field depends on the Miller's index N [9] and turns out to be larger on the A terminated QDs – where the two contributions have the same sign- with respect to the B terminated QDs – where the two contributions are opposite.

Within this framework we can also give an explanation to the optical properties of the larger size QDs with 2.3 and 2.8 ML InAs coverage here investigated. On one side we found a large decrease of the PL efficiency when increasing $1/N$, with a large asymmetry between A and B termination. Also the PL reduction is larger in the samples with higher InAs coverage. On the other side the PL time evolution after pulse excitation is similar for any substrate and InAs coverage. This clearly does not support a relevant increase of the non radiative recombination mechanisms either in the relaxation or in the recombination of the photogenerated carriers involved in the QD emission (as a consequence of an increase of defects inside the QDs) when the InAs coverage is increased on high Miller index substrates. Also changes in the QD emission efficiency due to possible barrier deterioration can be ruled out. The lack of dependence of the QD risetimes on the (N11) substrates and their agreement with the literature values [8] indicate a good barrier quality and homogeneity in the whole sample set. This was expected, since each growth run was carried out simultaneously on the three (N11) GaAs substrates with the same termination, in order to minimize different extrinsic contamination in the investigated samples.

The comparison between PL efficiency and PL lifetime suggests the prevalence of a nearly constant non–radiative recombination associated with a large decrease of the radiative recombination rate when increasing the InAs coverage. This will strongly quench the PL efficiency keeping the PL decay time almost constant. A reduction of the oscillator strength is indeed found in the case of a large built-in electric field, as expected in (N11) QDs. The electric field leads to a spatial separation of the electron hole pair reducing the overlap integral and then increasing the radiative lifetime. The electron-hole spatial separation is larger in bigger QDs corresponding to higher InAs coverage, in agreement with the reduction of the PL efficiency in the 2.8 ML structures with respect to the 2.3 ML QDs. The same argument cannot explain the reduction of the PL intensity when increasing $1/N$, since we find that the QD size increases with N. We therefore conclude that the internal field increase with $1/N$ as expected for piezoelectric fields. Finally, in agreement with [5,7], the asymmetry found in the PL efficiency between A and B terminations can be explained by assuming that the total built–in electric field is larger on the A side with respect to the B side.

5 Conclusions

We have studied the optical properties and carrier kinetics of a large set of (N11) InAs QDs with different substrate orientations, terminations and InAs coverages. We found a strong reduction of the PL intensity for large values of $1/N$. The effect is more pronounced for QDs of larger sizes and for A termination. TR-PL measurements does not support a relevant increase of the non radiative recombination mechanisms neither in the relaxation or in the recombination of the photogenerated carriers when the InAs coverage is increased on high Miller index substrates. The observed phenomenology can be accounted by the presence of a large internal electric field produced by the combined effects of piezoelectric polarization and permanent dipole moment in (N11) QDs, recently demonstrated in [5,7]. The induced spatial separation of the electron hole pair, which is larger in larger QDs, reduces the overlap integral. This gives rise to a strong increase of the radiative lifetime which eventually becomes larger than the non-radiative recombination rate, leading to a reduction of the PL efficiency. Therefore the optical properties on InAs QDs grown on high Miller index substrates are strongly influenced by the presence of internal built-in field. We believe that a deep understanding of the nature of this built in field and therefore its control could be used to engineer new QD based devices. The possibility of designing contactless QDs structures with large, controlled internal electric field indeed provides a further powerful degree of freedom in tailoring the band structure and the electro–optical properties of QD semiconductor heterostructures.

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