Pressure dependence of photoluminescence spectra of self-assembled InAs/GaAs quantum dots

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Photoluminescence (PL) measurements have been performed in InAs/GaAs self-assembled quantum dots (QDs) under high excitation conditions at low temperatures and under high hydrostatic pressures up to 10 GPa. Mechanically polished samples for high pressure experiments exhibited PL emission from the QD ground state but not from the excited states. Instead, a new broad band is observed in the energy range of the first excited state, which is tentatively attributed to emission from smaller dots formed during the mechanical thinning of the sample. With increasing pressure we found a similar blue shift for the PL maxima of the QD ground state (65 meV/GPa) and of the new broad band (69 meV/ GPa). These pressure coefficients are 20% and 40% lower than those reported for dots of less than half the height as in our case and for the wetting layer, respectively. Our results point to a systematic reduction of the pressure coefficient of the InAs QDs with the increase of the dot height.

Introduction Photoluminescence (PL) measurements of InAs/GaAs self-organised quantum dots (QDs) studied under high excitation conditions at low temperatures and under high hydrostatic pressures showed narrow bands (30–40 meV in width) coming from the ground and first excited states of the dots [1]. In that work, the QDs were 15×15 nm^2 in base size but only 3 nm in height. All PL lines showed an initial blueshift with increasing pressure up to the Γ–X conduction band crossover occurring above 4 GPa, as previously reported for monolayer-thick InAs/GaAs quantum dots [2]. The sequential quenching of the dot emission lines in Ref. [1] indicated that each PL peak originates from optical transitions between confined electron and hole states with the same quantum number.

Here we report PL measurements of thicker (6–8 nm in height) InAs/GaAs dots as compared to those of Ref. [1] studied under similar high-excitation conditions. Our as-grown sample is part of a series of QD samples annealed at different temperatures up to 700 °C for systematic change in dot size and In composition [3, 4]. At ambient pressure the dot sample considered here showed up to four narrow bands corresponding to the ground and excited state emission. From magnetoluminescence data a distinct magnetic-field splitting behavior was obtained with σ^+ and σ^- circular polarization for the emission of the first excited state [3, 4], which was taken as evidence of a strong coupling between hole levels depending upon dot size and composition. Therefore, our original motivation was to study the evolution of the PL signal corresponding to the excited dot states as a function of pres-
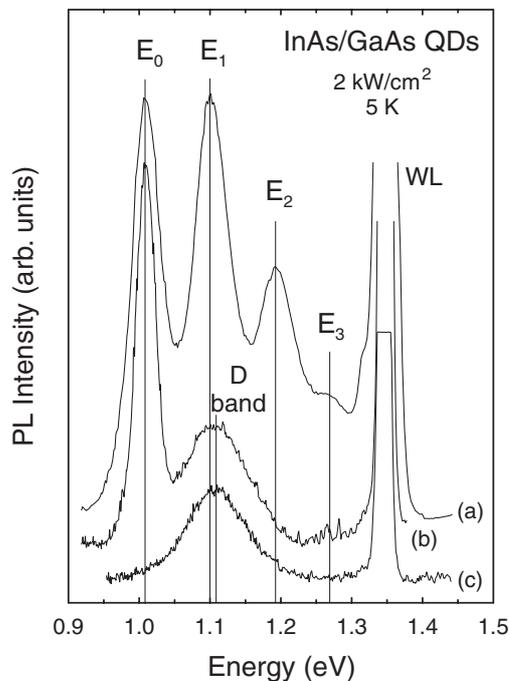
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sure. For this purpose, as-grown samples were thinned down by mechanical polishing and inserted into a diamond anvil cell. After polishing, however, samples exhibited PL signal of the dot ground state but not from the excited states. Instead, a new broad band (labelled as D for defect band) becomes apparent in PL spectra at around the energy of the first-excited state emission. In this work we discuss the pressure dependence of the dot emission in the mechanically thinned sample and the possible origin for the D band.

Experiment The as-grown sample consists of a single layer of InGaAs dots embedded in GaAs grown by low-pressure metalorganic chemical vapor deposition (MOCVD) at a temperature of 485 °C in the Stranski-Krastanow mode. The nominal In concentration of 42% of the InGaAs layer is expected to be substantially higher in the QDs due to In segregation and migration effects [4]. A plate-let-shape crystal 100×100 μm^2 in lateral size and 30 μm thick was fitted into a diamond anvil cell for high-pressure experiments. Low-temperature ($T = 5$ K) photoluminescence measurements were performed in a continuous-flow helium cryostat. Helium was used as pressure medium and the change of pressure was always performed above the He melting temperature in order to avoid non-hydrostatic conditions. The ruby luminescence method was used for pressure calibration [5, 6] with temperature correction according to Ref. [7]. The 514.5 nm line of an Ar$^+$-ion laser was used for excitation of the sample luminescence. The emitted light was analysed by a 0.64 m single-grating spectrometer equipped with a liquid N-cooled Ge detector.

Results and discussion Figure 1 displays low-temperature PL spectra of the InGaAs/GaAs QD sample taken at ambient pressure with a high excitation density of 2 kW/cm^2. The spectrum labeled as (a) corresponds to PL emission of the as-grown sample before mechanical polishing. At that laser powers and for the low dot density of our sample up to four peaks denoted E_0 to E_3 are apparent from the PL spectrum below the band gap energy of GaAs. These peaks are attributed to optical transitions between confined states of the dots. The strong peak at about 1.34 eV corresponds to the emission from the InGaAs wetting layer. After the sample was thinned down mechanically for high-pressure experiments the spectrum changed drastically, see curve (b) of Fig. 1. The ground-state emission is still observed but none from the excited states. Instead a new broad band labeled D appeared near the energy of the first excited state recombination. The overall intensity of the luminescence also decreased by an order of magnitude after polish, indicating a clear degradation in radiative recombination efficiency of the QD sample.

We tentatively attribute the D band to emission from small QDs formed due to fracture of the original dots induced by tensions during the mechanical thinning of the sample. Magnetoluminescence measurements [3] and atomically-resolved cross-sectional

Fig. 1 Ambient-pressure PL spectra under high excitation power of (a) the as-grown InAs/GaAs QD sample before polishing, (b) after mechanical polishing but before applying pressure, and (c) after a pressure cycle through the zincblende to rocksalt transition. WL stands for the PL emission from the wetting layer.

scanning-tunneling microscopy [8] indicate that the In concentration in the dots is in excess of 90% due to In segregation effects during growth. Hence, the QDs are largely stressed due to the 7% lattice mismatch between InAs and GaAs. The mechanical polishing procedure might favor strain relaxation by formation of dislocations, thus reducing the effective size of the dots by fracture, or it may even activate In migration with the consequent increase in Ga content in the dots. Both effects would result in a blueshift of the emission energy in a manner similar to the effect of thermal annealing [3, 9]. Dislocations and defects open up additional nonradiative channels such that many dots will no longer effectively emit light, which explains the by far less intense PL of the polished samples. Photoexcited carriers would now be trapped by defects recombining nonradiatively. The used excitation power density is not sufficiently high to lead to saturation of the ground-state emission, while the QD excited states being higher in energy than the D band, become sparsely populated and no luminescence is observed from them.

Further evidence for the origin of the D band is obtained from the ambient-pressure PL spectrum after a pressure cycle through the zincblende to rocksalt transition which for bulk InAs occurs between 7 and 8 GPa [10, 11]. Above 7.2 GPa the luminescence of the sample is fully quenched. After releasing the pressure below 3 GPa only the wetting layer (WL) and the D band emission reappears although the overall intensity is again a factor of 10 weaker than during the pressure upstroke. Curve (c) of Fig. 1 represents an example of the PL spectrum obtained outside the pressure cell after pressure cycle. The ground-state PL peak of the self-assembled dots is completely absent. The original dots show a higher propensity to relax the large built-in strain by structural defect formation as compared to the wetting layer (1–2 monolayers thick) and to the dots related to the D band, thus the former do not seem to withstand the pressure-induced phase transition.
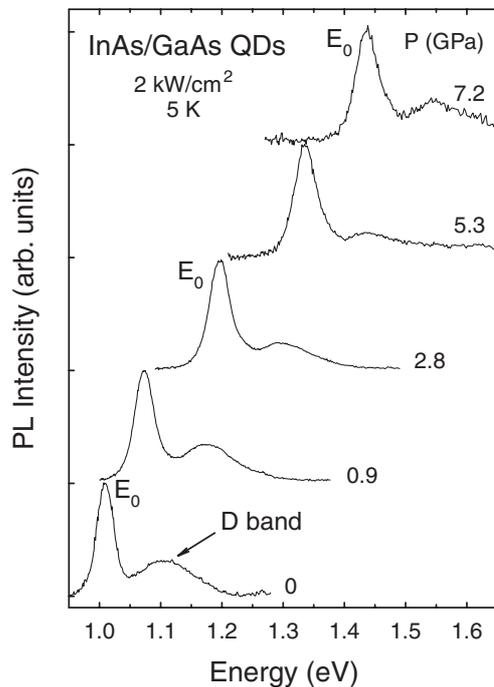

Fig. 2 Photoluminescence spectra of the polished InAs/GaAs QD sample for different pressures at 5 K and at high excitation density. Spectra were measured during a first upstroke up to 8.2 GPa.
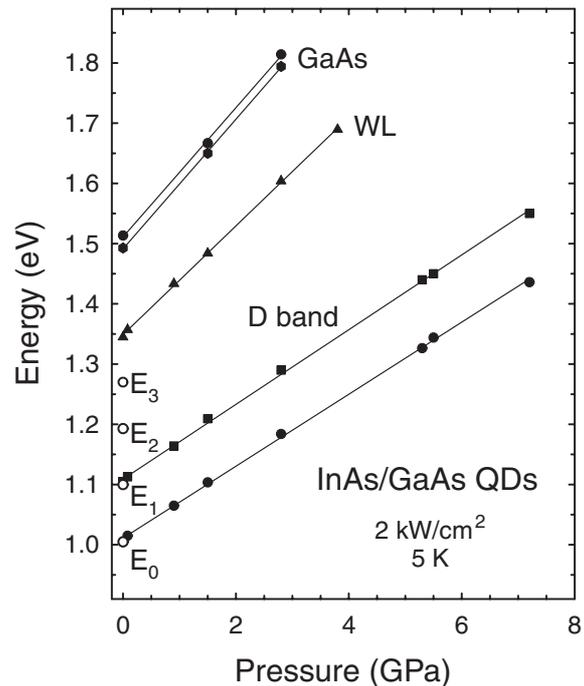
Fig. 3 Energies of PL peaks of the polished InAs/GaAs QD sample as a function of pressure. The solid lines correspond to the results of least-squares fits. Open symbols represent the energy position of the PL peaks in the unpolished sample at ambient pressure.

Table 1 Coefficients describing the dependence on pressure of the PL bands of the InAs/GaAs QDs obtained from fits to the data using $E(P) = E(0) + a_1 P + a_2 P^2$.

Feature	$E(0)$ (eV)	a_1 (meV/GPa)	a_2 (meV/GPa^2)
E_0	1.007(2)	65(2)	$-0.8(2)$
D band	1.104(2)	69(2)	$-1.0(3)$
WL	1.347(2)	95(3)	$-1.2(8)$
GaAs	1.516(2)	108(3)	—

Figure 2 shows several PL spectra of the polished sample for different pressures up to 7.2 GPa at 5 K and high excitation power. With increasing pressure both the E_0 peak corresponding to the QD ground-state emission and the D band shift to higher energies, as expected for zone-center direct optical transitions. The wetting layer peak (not shown in Fig. 2) also exhibits a blueshift up to a pressure of about 5 GPa, at which the Γ−-X conduction band crossover occurs. At higher pressures the intensity of the PL spectra decreases markedly. The dependence on pressure of the energy of the different PL peak maxima observed in our experiments is depicted in Fig. 3. For comparison, we also show the position of the excited-state emission lines of the as-grown sample at ambient pressure (open symbols). The solid lines represent results of least-squares fits of quadratic or linear relations to the data. The corresponding first and second-order pressure coefficients are listed in Table 1.

We point out a systematic trend in the linear pressure coefficients of the direct optical transitions as a function of dot height for QD structures with a large base-to-height aspect ratio (flat dots). The first-order pressure coefficient of the E_0 peak and D band in our samples (see Table 1) is close to the value of 48 meV/GPa determined for bulk InAs [12], which is about one half of that of bulk GaAs (108 meV/GPa) [13]. The InAs/GaAs QDs studied in Ref. [1], which are only about 3 nm in height, exhibit a blueshift of the ground-state emission with pressure at a 20% higher rate (75 meV/GPa) than in our case. The wetting layer displays a pressure coefficient of 95 meV/GPa, which is slightly lower than that measured for a pure InAs monolayer embedded in GaAs [14] and monolayer-thick InAs dots grown on a tilted GaAs substrate [2], both coefficients being about 104 meV/GPa. All these results taken together indicate that the larger the penetration of the exciton wave function into the GaAs barrier material, the higher is the linear pressure coefficient, approaching that of bulk GaAs in the limit of monolayer-thick structures.

In summary, we have measured low-temperature photoluminescence spectra of self-organized InAs/GaAs QDs at high excitation levels under high pressure using mechanically polished samples. We have observed that the mechanical polishing procedure leads to the formation of smaller dots probably by fracture of the original QDs giving rise to the appearance of a broad PL band centred near the energy of the first excited state. In addition, a strong decrease in the PL efficiency of the sample is observed as a consequence of the formation of dislocations and structural defects making impossible the measurement of PL signal coming from the QDs excited states. The linear pressure coefficients of the QD ground-state emission and the new D band are close to that of bulk InAs due to the confinement of the exciton wave function in the dots which are of large size and have high In concentration.

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