JOURNAL OF APPLIED PHYSICS VOLUME 95, NUMBER 3 1 FEBRUARY 2004

Photoluminescence from self-assembled long-wavelength InAs/GaAs quantum dots under pressure

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(Received 14 August 2003; accepted 29 October 2003)

The photoluminescence from self-assembled long-wavelength InAs/GaAs quantum dots was investigated at 15 K under hydrostatic pressure up to 9 GPa. Photoemission from both the ground and the first excited states in large InAs dots was observed. The pressure coefficients of the two emissions were 69 and 72 meV/GPa, respectively. A nonlinear elasticity theory was used to interpret the significantly small pressure coefficients of the large dots. The sequential quenching of the ground and the excited state emissions with increasing pressure suggests that the excited state emissions originate from the optical transitions between the first excited electron states and the first excited hole states. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1635988]

I. INTRODUCTION

In recent years InAs/GaAs self-assembled quantum dots (QDs) have been attracting continuous and popular interest due to their potential and feasible applications in electronics, optoelectronics, photocommunication, and quantum information due to their prominent zero-dimensional properties, as well as convenient and economical growth process.[1] Application to optical fiber communication systems requires 1.3–1.55 μm long-wavelength emissions from the dots at room temperature. Many approaches, such as the overgrowth of an InGaAs strain-reducing layer and the decrease of the growth rate, have been introduced to extend the room-temperature emission wavelength to 1.3 μm. The InGaAs layer can reduce the surface strain in the dots induced by a GaAs capping layer, and the low growth rate favors the formation of large dots,[2–5] both of which give rise to the redshift of the dot's emission energy. The large InAs/GaAs dots studied in this article are fabricated in this way.

Photoluminescence (PL) measurement under high hydrostatic pressure has proved to be an effective tool for exploring the electronic structures and optical transitions in bulk crystals or microstructures. To data PL observations under pressure have been mostly focused on small dots, i.e., InAs dots less than 5 nm in height and 20 nm in base size, the PL energy of which is always more than 1.1 eV.[6–8] The measured pressure coefficients of these dots are about 75–90 meV/GPa, 17%–30% smaller than that of the band gap of GaAs. They are also smaller than the commonly accepted pressure coefficient (PC) of bulk InAs. It is, therefore, interesting to look for the root cause of the smaller PC. Monjón *et al.* have recently studied the pressure dependence of the PL from large InGaAs dots, which are 6–8 nm in dot height.[9] They found that the PC of the large dots was only 65 meV/GPa, smaller than those of the small dots. Their explanation was based on an early work for bulk InAs, which gave a considerably small PC of about 48 meV/GPa.[10] However, the reason the QDs have so small PCs is still an open issue. In this article we report on a PL investigation of the large InAs/GaAs QDs under hydrostatic pressure. Photoemissions from both the ground and the first excited states were observed. Similar to Monjón's results, the PCs of the ground-state-related and excited-state-related PL peaks measured here are also smaller than the previous results for small dots. In virtue of a nonlinear elasticity theory developed by Frogley *et al.*,[11] we demonstrate here that the built-in strain in InAs dots may be the main reason for the much smaller PC.

II. EXPERIMENTAL DETAILS

The sample was grown by molecular-beam epitaxy in the Stranski–Krastanow mode on a (100)-oriented n^+ GaAs substrate. The grown structure consisted of a 510 nm GaAs buffer layer, then three 2.5-monolayer (ML) InAs QD layers, and finally a 50 nm GaAs capping layer. Each InAs QD layer was covered by a 3 nm $In_{0.1}Ga_{0.9}As$ strain-reducing layer and a 50 nm GaAs protecting layer. There were growth interruptions during the growth of the InAs QDs: each 0.1 ML InAs deposition was followed by a 20 s As_4 exposure. The whole growth process and sample structure were similar to those presented in Ref. 5.

Atomic force microscopy (AFM) measurements before the overgrowth of the InGaAs layer show that the QDs have an average lateral size of 78 nm and height of 7.3 nm. The QDs could undergo changes in the dot size and shape during the overgrowth. The density of dots is not very high, and a few sparsely distributed smaller dots are also found in the AFM image.

For pressure experiments the samples were mechanically thinned to a total thickness of 20 μm, and then cut into pieces of 100×100 μm^2 in size. High-pressure PL measurements were performed at 15 K by using a diamond-anvil cell (DAC) to generate pressures up to 9 GPa. Condensed argon was used as the pressure-transmitting medium. The pressure was determined from the shift of the ruby R_1 fluorescence

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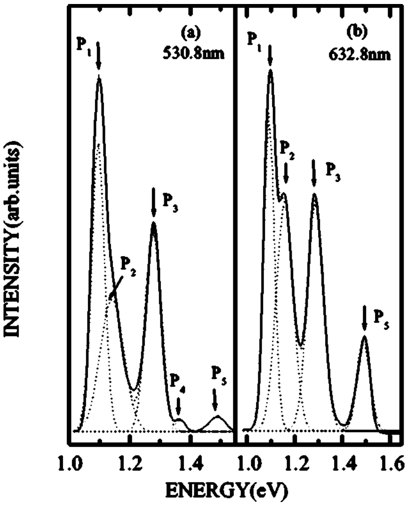

FIG. 1. PL spectra of the InAs/GaAs QDs at 0.14 GPa and 15 K excited by: (a) 530.8 nm and (b) 632.8 nm lines. The dotted curves represent the fitted Gaussian line profiles.

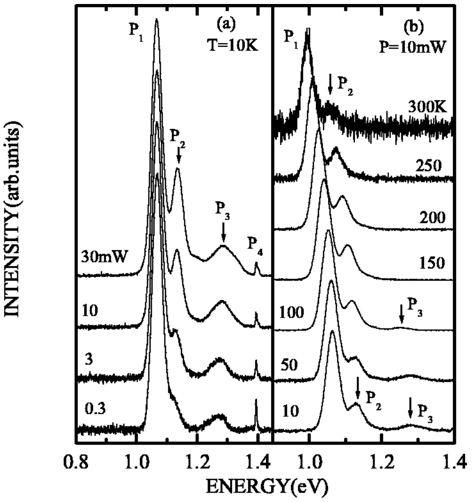

FIG. 2. PL spectra of the InAs QDs at ambient pressure excited: (a) under different excitation levels at 10 K and (b) at different temperatures under 10 mW excitation. The spectra have been normalized according to the respective strongest peak.

line and was always changed at room temperature so as to ensure the best possible hydrostatic conditions.

Both the 632.8 nm line (red) of a He–Ne laser and the 530.8 nm (green) line of a Kr^+ laser acted as the excitation sources for the PL measurements. The excited luminescence was analyzed by using a 0.5 m single-grating monochrometer equipped with a cooled Ge detector. In addition, the variations of the PL spectra of the samples without mechanical thinning with excitation power and temperature were measured on a Nicolet FTIR760 Fourier spectrometer at ambient pressure.

III. RESULTS AND DISCUSSION

Figure 1 shows the low-temperature (15 K) PL spectra measured at nearly zero pressure (0.14 GPa) and excited by the 530.8 nm (a) and 632.8 nm (b) lines, respectively. Up to five peaks are observed in the 530.8 nm line-excited spectra, and four in the 632.8 nm line-excited one. These peaks are labeled P_1–P_5 as shown in Fig. 1. The dotted curves represent the fitted Gaussian line profiles. The energy discrepancy between the same peaks in the two spectra is within experimental uncertainty.

P_5 at around 1.493 eV is much weaker in intensity under the 530.8 nm line excitation than under the 632.8 nm line one. It is attributed to an impurity-related emission from the GaAs substrate. Due to more absorption of the green 530.8 nm line in the samples, the GaAs substrate-related emissions excited by this line get weaker, while those related to the InAs structures vary little in intensity.

The three peaks, P_1, P_2, and P_3, can be attributed to the luminescence from the QDs. Similar to Ref. 5, the most dominant feature, P_1, is attributed to the ground state transitions of the InAs QDs. P_2 at the higher energy side of P_1 has proved to be an emission from the excited states in the QDs. We measured the PL spectra under different excitation levels and temperatures, which are illustrated in Fig. 2. With increasing excitation power, P_2 gets more and more distinct with respect to P_1. That is to say, the intensity ratio between P_2 and P_1 increases with the excitation power. This is a typical feature of the excited state transitions in QDs.[4,12–14] In contrast to P_2, P_3 at 1.29 eV undergoes little change in its intensity relative to P_1 when the excitation power increases by 2 orders. So P_3 is independent of P_1 and impossible to stem from the excited states of the P_1-related QDs.[12,15] We tentatively attributed P_3 to the PL from the smaller InAs QDs in the sample. This assignment is also consistent with the changes of PL spectra with temperature. As shown in Fig. 2(b), P_3 quenches at about 120 K, while P_1 and P_2 exist until room temperature. It is known that smaller dots have larger localization energy,[14,16–18] which leads to smaller activation energy for the thermal escape of carriers. Therefore the PL from smaller dots will disappear at a lower temperature. Actually, as can be seen in the AFM image, some smaller dots scatter among the crowd of larger dots. Their size distribution is much more sparse and inhomogeneous than that of the larger dots; consequently, P_3 is far weaker than P_1, and its full width at half maximum is broader.

At low temperature, the PL from the InAs wetting layer is always several orders weaker than the QD's emission, and its peak energy is about 1.4 eV.[19,20] In Fig. 2(a), a sharp and weak peak is well resolved at about 1.4 eV. Furthermore, the energy position and the line shape of this peak change little with respect to P_1 during the increase of the excitation level. All these suggest that this peak may be an emission from the InAs wetting layer. There is also a weak peak, P_4, at 1.37 eV in the 530.8 nm line-excited spectra shown in Fig. 1(a). In view of the similar energy and intensity, this feature may also be the emission from the InAs wetting layer. The linewidth of P_4 in Fig. 1(a) measured in the DAC is much larger than that in Fig. 2(a) measured before the mechanical thinning of the sample. It may be the result of the strain relaxation and defects induced during the mechanical thinning.

The evolution of the PL spectra with increasing pressure is illustrated in Figs. 3 and 4. All the spectra have been normalized according to the respective strongest peak. In the pressure range below 4 GPa, all the features in the spectra

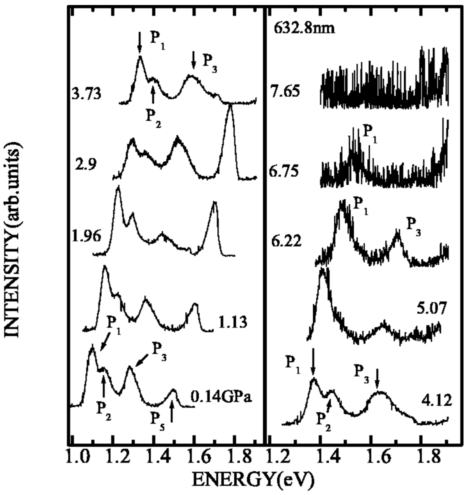

FIG. 3. PL spectra of the InAs QDs excited by 632.8 nm line under different pressures. The spectra have been normalized with respect to the respective strongest peak.

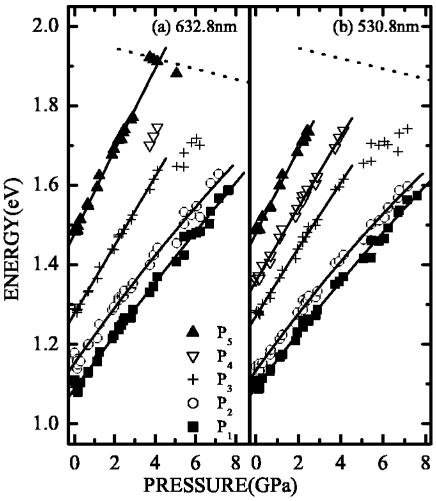

FIG. 5. Pressure dependence of the PL peak energies of the InAs/GaAs QDs under the excitation of: (a) 632.8 nm and (b) 530.8 nm lines. The solid lines represent the results of least-squares fits to the experimental data. The dotted lines are the pressure dependence of the indirect transition from the *X* valley to the valence band in bulk GaAs (see Ref. 13).

shift to higher energy with increasing pressure. At about 4 GPa, P_5 weakens abruptly and then quenches beyond detection. Above 4 GPa, other peaks keep on shifting to higher energy positions with increasing pressure until P_3 disappears at nearly 6.7 GPa. P_1 remains up to 6.8 GPa in the red-line-excited spectra and up to 7.7 GPa in the green-line-excited one.

We summarize the pressure dependence of the PL peak energy in Fig. 5. The solid lines represent the results of the least-squares fits to the experimental data by using quadratic relations. Above 4 GPa, the blueshift of P_5 in the red-line-excited spectra is replaced by a relatively small redshift with increasing pressure. The data points are located on the dashed line, which is a commonly accepted pressure dependence of the *X*-related emission in GaAs.[13] The energy positions of the P_3 peaks deviate significantly from the linear extrapolation when the pressure increases beyond 4 GPa. Thus, the fits to the energy data of P_3 only cover the pressure range below 4 GPa. There is no fit to the data of P_4 in the 632.8 nm line-excited spectra due to insufficient data. The corresponding first- and second-order coefficients are listed in Table I. The fitting parameters in Table I are obtained from the 632.8 nm line-excited spectra except those of P_4. The PCs obtained under the two excitations are all but the same.

By and large, no PL peaks except P_1 and P_2 exhibit an anomalous PC. The PC of P_5 is almost the same as that of the direct gap of bulk GaAs at low temperatures, 108 meV/GPa.[21,22] The PC of P_4 agrees well with the results reported recently for the wetting layer,[9] and that of P_3 also lies in the range of 75–102 meV/GPa,[6–8,23] the results of small InAs dots measured before. Surprisingly, the PCs of P_1 and P_2 are only 69 and 72 mev/GPa, respectively, which are 35% lower than that of bulk GaAs. The PC of the InAs QD is sensitive to the size of the QD. Li *et al.* demonstrated that the PC measured for the 1 ML thick InAs QDs grown on a slightly misoriented (terraced) GaAs substrate is 102(2) meV/GPa, only 5% smaller than that of GaAs.[23] Lyapin *et al.* reported the PCs of InAs/GaAs QDs, which are 16 nm in lateral size and 1.6 nm in dot height, to be 80(2) meV/GPa.[6] The PCs of the InAs dots measured by Itskevich *et al.*, which exhibit a 15 nm lateral size and 3 nm thickness, are only 75(2) meV/GPa.[8] Each of these results is larger than ours. On the other hand, the recently reported PC of large

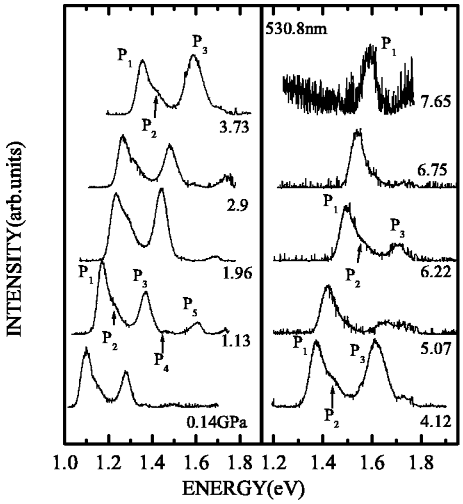

FIG. 4. PL spectra of the InAs QDs excited by 530.8 nm line under different pressures. The spectra have been normalized with respect to the respective strongest peak.

TABLE I. Coefficients describing the dependence on pressure of the PL peaks of the InAs/GaAs QDs obtained from least-square fits to the experimental data by using $E(p)=E_0+A_1\times P+A_2\times P^2$.

Peak	E_0 (eV)	A_1 (meV/GPa)	A_2 (meV/GPa^2)
P_1	1.088(4)	69(3)	−0.5(4)
P_2	1.152(6)	72(4)	−1.1(7)
P_3	1.278(3)	85(3)	0.1(9)
P_4	1.359(3)	95(4)	−0.9(11)
P_5	1.479(3)	106(4)	−0.7(11)

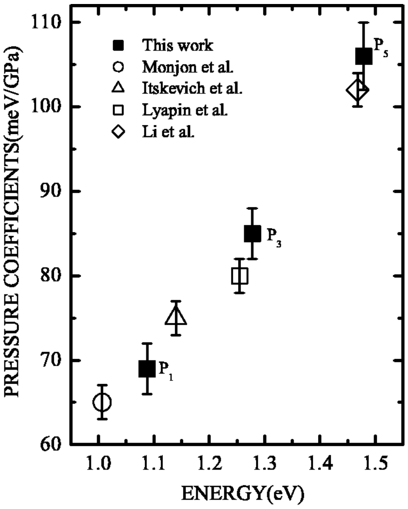

FIG. 6. Pressure coefficients of the InAs/GaAs QDs as a function of the PL peak energies. The solid square are the results of the peaks P_1, P_3, and P_5 in this work. The open circle is from Manjón *et al.* (see Ref. 21), the open triangle from Itskevech *et al.* (see Ref. 8), the open square from Lyapin *et al.* (see Ref. 6), and the open diamonds from Li *et al.* (see Ref. 23).

InGaAs QDs, 6–8 nm in height,[9] is slightly smaller than our results. Figure 6 displays the measured PCs as a function of the PL peak energy. It can be seen clearly that the PC is reduced with the decrease of the PL peak energy. In other words, the increase of the dot size (mainly the dot height) results in the decrease of the PC, provided that the PL peak energy is negatively correlated to the dot size/height.

It is interesting to compare the PCs of the InAs QDs with that of bulk InAs even though the PC for the band gap of bulk InAs is still a controversial issue. The PC value, 96 meV/GPa, which is a little smaller than that of bulk GaAs, is obtained from most theoretical calculations and experimental measurements.[24] Although Edwards reported that the PC of the absorption edge of bulk InAs was only 48 meV/GPa,[10] it does not seem well accepted. If we take 96 meV/GPa as the PC of bulk InAs, a majority of the measured PCs of InAs/GaAs QDs is smaller than those of both bulk GaAs and bulk InAs. Note that 96 meV/GPa corresponds to the PC value of strain-free InAs bulk materials. It is well known that self-assembled InAs/GaAs QD structures are characterized by huge built-in strains due to the large difference between the lattice constant of InAs and that of GaAs. The variation of the internal strain with pressure then seems certainly to affect the PC of the InAs band gap. Itskevich *et al.* mentioned the possible influence of the strain in the dots on the PCs, but neither detailed qualitative analysis nor quantitative estimate was made in their paper.[7] Frogley *et al.* estimated quantitatively the strain's effect on the PC of an InGaAs strained layer grown on a GaAs substrate. They concluded that the change of misfit strain and elastic constants with pressure accounted for the anomalously low band gap PCs of strained InGaAs layers.[11] Following their treatment, we can also evaluate the PC of an InAs strained layer. According to Frogley's model, the heavy-hole band gap of the InAs strained layer grown on a (001)-oriented GaAs substrate, E_g^{hh}, is expressed as

$$E_g^{\mathrm{hh}}=E_g+(a_{\mathrm{CB}}-a_{\mathrm{VB}})\epsilon_{\mathrm{vol}}-b_{\mathrm{VB}}\epsilon_{\mathrm{ax}}, \tag{1}$$

where E_g is the band gap of unstrained InAs, a_{CB} and a_{VB} are the hydrostatic deformation potentials of its conduction band and valence band, respectively, and b_{VB} is the axial deformation potential of the valence band. ϵ_{vol} denotes the volumetric strain, while ϵ_{ax} the axial strain. In the biaxial strain condition the two types of strain are given by

$$\epsilon_{\mathrm{vol}}=(2-\nu_{2\mathrm{D}})\epsilon \tag{2a}$$

and

$$\epsilon_{\mathrm{ax}}=(1+\nu_{2\mathrm{D}})\epsilon, \tag{2b}$$

respectively, where $\epsilon=(a_l-a_s)/a_l$ is the misfit strain induced by the difference between the lattice constant of the substrate, a_s, and that of the epilayer, a_l. $\nu_{2\mathrm{D}}=2C_{12}/C_{11}$ is the biaxial Poisson ratio. C_{12} and C_{11} represent the elastic constants. Under applied pressure the lattice constants of the substrate and the epilayer vary, as does the misfit strain. The misfit strain $\epsilon(P)$, deduced from Murnaghan's equation of state and then expanded to the first order in pressure, is

$$\epsilon(P)\approx\frac{a_l^0-a_s^0}{a_l^0}+\frac{1}{3}\frac{a_l^0}{a_s^0}\left(\frac{1}{B_s}-\frac{1}{B_l}\right)P=\epsilon_0+\epsilon' P, \tag{3}$$

where B_l and B_s are the bulk modulus of the InAs epilayer and that of the GaAs substrate, respectively. Similarly, the Poisson ratio also varies with pressure. Assuming that the elastic constants, C_{ij}, depend linearly on pressure and taking C'_{ij} as the linear coefficients, one obtains the biaxial Poisson ratio

$$\nu_{2\mathrm{D}}(P)=2\frac{C_{12}^0+C'_{12}P}{C_{11}^0+C'_{11}P}\approx\nu_{2\mathrm{D}}^0+\nu'_{2\mathrm{D}}P. \tag{4}$$

The superscripts or subscripts 0 in Eqs. (3) and (4) indicate the values of the parameters at ambient pressure. Applying the expressions (3) and (4) to Eq. (1) gives the PCs of the strained layers as

$$\begin{aligned}\frac{\partial E_g^{\mathrm{hh}}}{\partial P}\approx{}&\frac{a_{\mathrm{CB}}-a_{\mathrm{VB}}}{B_l}+\{(a_{\mathrm{CB}}-a_{\mathrm{VB}})[(2-\nu_{2\mathrm{D}}^0)\epsilon'-\epsilon_0\nu'_{2\mathrm{D}}]\}\\&-b_{\mathrm{VB}}[(1+\nu_{2\mathrm{D}}^0)\epsilon'+\epsilon_0\nu'_{2\mathrm{D}}],\end{aligned} \tag{5}$$

where $(a_{\mathrm{CB}}-a_{\mathrm{VB}})/B_l$ is the PC of the band gap of unstrained bulk InAs. Based on Eq. (5) and the values of the concerned parameters given in Ref. 11, we can calculate the PCs of the fully strained InAs layers grown on (100)-oriented GaAs substrates. The calculated result is 77 meV/GPa, much lower than that of unstrained bulk InAs, 96 meV/GPa. Though the strain in InAs QDs may be somewhat different from that in two-dimensional InAs layers, the strain effect is one of the main reasons for the low PCs of such large flat InAs QDs as studied here.

In addition to the effect of the internal strain on the band-gap PC of InAs, the variations of the confined energy of electrons and that of holes with pressure work on the PCs of the QDs. The decrease of the PC with the increase of the dot size, as shown in Fig. 6, may be largely due to the change of the confined energy. In fact, the PL peak energy of the dots can be written as

$$E_{\mathrm{PL}}(P)=E_g^{\mathrm{hh}}(P)+E_{1e}(P)+E_{1h}(P)-E_{\mathrm{ex}}(P), \qquad (6)$$

where E_g^{hh} is the heavy-hole band gap of strained InAs, E_{1e} and E_{1h} are the confined energy of electrons and that of holes, respectively. E_{ex} is the exciton binding energy. Then the PC of the PL peak energy of the QDs is

$$\frac{dE_{\mathrm{PL}}}{dp}=\frac{dE_g^{\mathrm{hh}}}{dp}+\frac{dE_{1e}}{dp}+\frac{dE_{1h}}{dp}-\frac{dE_{\mathrm{ex}}}{dp}. \qquad (7)$$

The four terms on the right side of the above equation can be treated in different ways. The first term, which is 77 meV/GPa in the two-dimensional and fully strained InAs layer, has been intensively discussed above. The fourth term can be neglected on account of the small magnitude of E_{ex}. As a result, the second and third term play an important role in the PCs of the emission from InAs QDs.

Three main factors: barrier height, effective mass, and dot size (usually referred to as dot height) have influence on the confined energy of electrons and holes. First of all, since the PC of InAs is smaller than that of GaAs, the barrier height increases rapidly with increasing pressure.[25] Hence, the confined energy of electrons and that of holes grow higher, which results in a positive contribution to the PCs. For narrow and shallow InGaAs/GaAs quantum wells (QWs), the smaller the well width, the more the change of the barrier height affects the PCs.[25] This effect is still true in the case of the InAs QDs and is the major reason for the increase of PC with the blueshift of the PL peak corresponding to the reduced dot size. Second, owing to the pressure-induced nonparabolic effect,[26] the electron effective mass grows heavier with increasing pressure giving rise to the drop of the electron-confined energy and thereby a negative contribution to the PCs.[6,7,27] If the influence of the increased effective mass prevails over that of the elevated barrier height, the PC of the PL from the QDs will be even smaller than that of the InAs band gap. Finally, similar to the well width in QWs, the dot size, especially the dot height, decreases with increasing pressure. As a result, the confined energy becomes larger leading to the increase of the PC. However, this positive contribution was proved to be small in the InGaAs/GaAS QWs.[25]

The above discussion is still qualitative. The formation of InAs QDs will somewhat relax the strain in the dots.[28,29] Thus, the QDs may not be so fully strained as two-dimensional InAs layers. Moreover, a more detailed calculation is required to ascertain the confined energy of the electrons and that of the holes in the dots with different sizes and their variations with pressure. In other words, it is necessary to perform a further theoretical study to obtain the exact value of the PC of the PL from the QDs and the PC's dependence on dot size.

Figure 7 depicts the changes of the integral intensities of various PL peaks with pressure. For clarity, the intensities of P_3 and P_5 are multiplied by the factors 0.1 and 0.01, respectively. Consistent with the previous measurements,[8,21,22] the intensity of the GaAs-related peak, P_5, decreases abruptly by about 2 orders of magnitude above 4 GPa, which indicates the $\Gamma-X$ crossover in the GaAs substrate. P_3 begins to weaken at about 5 GPa, not so dramatically as P_5. Both the intensity drop and the significant deviation from the extrapolated linear pressure dependence at 4 GPa, as mentioned above, suggest that the $\Gamma-X$ state mixing effect is evident in the small dots.[23] Without X-related experimental data available, the corresponding interaction potential has not been estimated. With increasing pressure, the PL lines P_2 and P_1 sequentially quench at around 6.2 and 7.8 GPa, respectively, and P_2 drops more rapidly than P_1 in intensity. Itskevich *et al.* have pointed out that if all optical transitions are from the same ground electronic state in the QD, all the corresponding PL lines should quench simultaneously at the same pressure.[8] Therefore, we ascribe P_2 and P_1 to the transitions from different electron states. That is to say, P_2, the excited-state emission in the dots, is related to the optical transition from the first excited electron state to the first excited hole state, rather than from the ground electron state to the excited hole state, as has been predicted in small InAs/GaAs dots.[16,14] Theoretical calculation confirms that there is more than one bound-electron state in InAs/GaAs dots if their bases are large enough.[17] Here the InAs QDs are 7.3 nm thick and 78 nm long on average; therefore, it is probable that excited electron states exist in them.

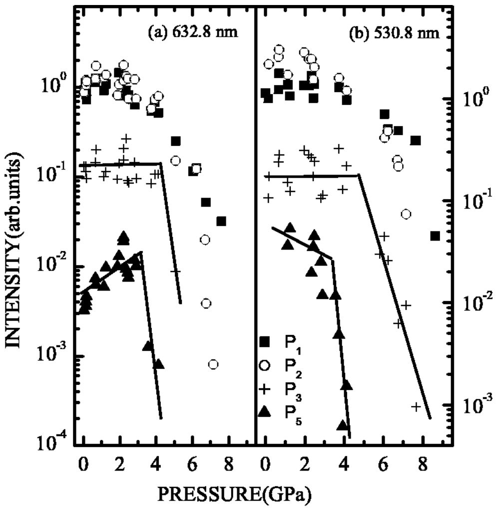

FIG. 7. Peak intensities vs pressure of the features in the PL excited by: (a) 632.8 nm and (b) 530.8 nm lines. The solid lines are guides for the eye.

IV. SUMMARY

To conclude, we measured the PL from large, flat, and strained InAs/GaAs QDs under hydrostatic pressure up to 9 GPa at 15 K. The five features observed in the PL spectra are attributed to the emissions from the ground and the excited states of the large dots, the PL from the small strained dots and the wetting layer, and the impurity-related emission in the GaAs substrate, respectively. They all exhibit a blueshift with increasing pressure below the crossover. The PCs of the first two peaks are only 69 and 72 meV/GPa, respectively, which are smaller than those of small InAs/GaAs dots. The analysis based on the nonlinear elasticity theory reveals that the change of the misfit strain and elastic constants with pressure can significantly reduce the PC of the band gap of a strained InAs layer. It may be the main reason for the considerably low PCs of the large dots studied here. The sequential quenching of the PL peaks P_2 and P_1 suggests that the excited-state emissions from the large QDs are related to the transitions from the first excited electron states to the first excited hole states.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Contract Nos. 60076012, 60176006), the special funds for Major State Basic Research Project of China (No. G2001CB3095), and the Nanometer Science and Technology Program of the Chinese Academy of Sciences. The authors are grateful to Professor Conboy (Timothy Joseph Conboy) for the revisions of the article. B.S.M. would like to thank Professor Han, Professor Tan, Dr. Liu, and Dr. Bian for their kind help.

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