Effects of interface-layers composition and strain distribution on the optical transitions of InAs quantum dots on InP

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In the present work we have investigated the optical properties of the self-assembled quantum dots (SAQD's) on an InP substrate. The dots are grown by gas source molecular-beam epitaxy (GSMBE) and characterized by photoluminescence (PL) and atomic force microscopy. The energy of the fundamental optical transitions measured by PL present a redshift compared to calculated values. Two hypotheses have been tested to explain this apparent difference: the existence of an intermediate $InAs_yP_{1-y}$ layer, with a composition depending on the experimental conditions, changes the value of the transition energy, and the strain induced in the InP confinement barrier by the dot as pointed out by Tersoff, has the same effect. The present study concludes with a discussion of the presence of a thin $InAs_yP_{1-y}$ interface layer originating from As/P exchange kinetic energy at the second interface on top of the dots.
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I. INTRODUCTION

The self-assembled quantum dots (SAQD's) have emerged as an attractive subject from both an experimental and theoretical point of view. The dot properties are promising for optoelectronic applications. For example, their use in place of quantum wells may theoretically help to reduce threshold current and thermal sensitivity and to increase quantum efficiency of laser structures. Controlling and understanding physical properties of SAQD's is a key to new optoelectronic applications. Photoluminescence (PL) measurements are widely used to investigate both qualitative and quantitative physical parameters,[1,2] and are particularly well suited to studying fundamental transitions.

As we are interested in controlling dot size and composition down to a monolayer scale, we need to theoretically predict or explain energies observed by photoluminescence. We present here a simple but detailed model based on the envelope function approximation (EFA) and the *kp* approximation to investigate composition and strain distribution effects on the optical properties of InAs SAQD's grown on InP by the analysis of their photoluminescence spectra.

Many experimental and theoretical studies[3–8] have pointed out the existence of strain around dots. These strain situated in the barrier layer in the region where the dot/barrier transition is not a high-symmetry crystallographic plane may induce some modifications on energetic properties of the dots. However, another problem that may influence the energy level values of dots is the existence of an interface layer with variable composition and size,[9–14] depending on the growth conditions and composition difference between the dots and the surrounding barrier. This situation is encountered especially when we are interested in the InAs/InP interfaces where the interface change occurs via the element *V* (As to P) transition. In the present work we have analyzed luminescence experimental results using these two hypothesis.

II. SAMPLE GROWTH AND PL MEASUREMENTS

InAs island morphology is very sensitive to growth techniques, temperature and substrate orientation.[15–22] Our samples are grown by gas source molecular-beam epitaxy (GSMBE) on a (100) semi-insulating InP:Fe substrate at $T = 490\,^{\circ}C$. They are made of six layers of InAs SAQD, the nominal thickness of which is 2.5 ML. The SAQD layers are separated by a 20-nm-thick InP barrier. At each InAs/InP interface, a 15-sec growth interruption is performed. The two-dimensional–three-dimensional (2D-3D) morphology during this interruption is observed on the reflection high-energy electron diffraction (RHEED) pattern. SAQD morphology is very sensitive to interruption time.[14] Transmission electron microscopy studies on similar structures reveal low dislocation density.[16]

Atomic force microscopy (AFM) measurements[23] show that dot diameters vary from 30 to 50 nm and their density is about 50 μm^{-2}, which is in good agreement with other published values.[20,22] The reason leading to the selection of this particular sample for the interpretation of photoluminescence measurements is related to the quality of the PL results. The PL peaks on this sample are well defined with a relatively good energy width. A sample with only one plane of InAs dots has been grown first, but the PL peak, situated at the same energy position as the present sample, is too large and the different peaks related to different dot thicknesses cannot be well resolved. Samples with many dot planes are known to give better results because of the improvement of the self-organization leading to less dispersion in dot dimensions (ordering effect). This ordering effect is probably the cause of the better PL peaks in the present sample.

The photoluminescence experiments have been performed

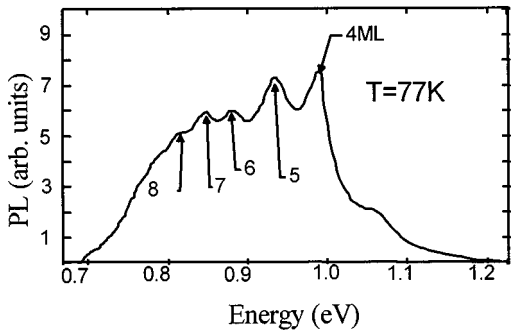

FIG. 1. Photoluminescence spectra of InAs/InP SAQD's measured at $T=77$ K. The different peaks are referenced by their relative thicknesses in (ML).

using an Ar ion laser (514 nm) for the sample excitation. The optical density is about 1 W/cm^2. The sample sits into a gas flow cryostat operating in a controlled temperature range between 2 and 300 K. The PL emission is detected by liquid-nitrogen cooled Ge detector using conventional lock-in technique. These results are shown in Fig. 1 where the experimental temperature is 77 K. The PL excitation beam diameter is 50 μm, leading the number of dots taking part in PL spectra to be about 4×10^5. Since the power excitation is low, only the ground-electron to ground-hole states transition ($Ee1\rightarrow Ehh1$) contributes to PL spectra.[22,24,25]

We have observed that on the thickest dots, increasing temperature from 2 to 300 K has little effect on integrated PL intensity. For the same experiment, the value of the integrated PL intensity of a lattice matched $In_xGa_{1-x}As/InP$ quantum well has decreased by more than a factor of 10 above 120 K. This is a clear evidence of the lateral spatial localization effect[25,26] of the dots which prevents the lateral diffusion and the trapping of the carriers on nonradiative centers.

III. MODEL, ASSUMPTIONS AND LIMITATIONS

Due to its relative simplicity, the most popular calculation method is the envelope function approximation in conjunction with the k.p. Hamiltonian model of Kohn-Luttinger. In the present work, the eight-band k.p. theory and envelope function approximation (k.p.+EFA) is used.[27,28] This model is well suited to our problem since a fundamental transition occurs near the Brillouin-zone center ($k=0$). The strain effect is treated using a Pikus-Bir Hamiltonian.[29] All the parameters used for our calculations come from Ref. 28.

The present SAQD's are treated as isolated quantum disks presenting a revolution symmetry along the z growth axis. For this reason the 0D Schrödinger equation can be solved using a cylindrical geometry that helps to separate the equation into two different parts (lateral and perpendicular). The energy and wave-function calculations are treated as a first-order perturbation to the quantum well problem. As the disk diameter ($\sim$40 nm) is greater than its height ($\sim$2 nm), we first performed a z axis quantum confinement energy calculation for electrons and heavy holes.

We then calculated in plane confinement using effective mass resulting from a k.p.+EFA band structure calculation. The wave-function solutions in the lateral plane are analytic Bessel functions. Due to the relatively large diameter of our dots ($\sim$40 nm),[23] the lateral contribution to the confined energy levels is quite small (14.8 meV for electrons and 1.6 meV for the heavy holes) compared to the z confinement energy ($>$100 meV).

We then include the Coulomb interaction between electrons in a first-order perturbation approach, as mentioned above. We took a simple 2D exciton variational model introducing in plane confinement via electron and hole wave functions as in Ref. 30.

This Coulomb interaction energy varies from 35 to 5 meV for, respectively, 20 to 70 nm disk diameters. These results are comparable with Refs. 30–33 which are given for other materials and geometries. For our particular dimension (40 nm disk diameter), the Coulomb interaction energy for all disk heights approaches 16 meV. Finally, for our purpose, the lateral confinement effect (16.4 meV) is almost cancelled by Coulomb interaction (16 meV).

We have also examined the experimental dispersion of the diameter, ranging from 30 to 50 nm, on the PL peak positions. The energy correction of the addition of the lateral confinement and Coulomb interaction on a 6-ML-wide 2D quantum well calculation varies from $+5$ to -3 meV for, respectively, 30 and 50-nm diameters. This result shows that the lateral size distribution of our dots has little effect on PL peak positions. Thus, in our particular case, the pertinent variable is the disk height and assigning a PL peak position to a finite number of monolayers height is rather rigorous.

The precision of the calculated energies is affected by the dispersion of the published data values (Luttinger γ parameters, band offsets, deformation potentials . . . [28]) incorporated in the model. The estimation of this dispersion on our calculated energy level values is in the range of 10 meV. This dispersion is small enough compared to the main effects shown in this work.

IV. EFFECT OF INTERFACE COMPOSITION

The first examined assumption is the existence and the influence of the interfacial layer on optical transitions. The composition and size of this thin layer is not well controlled in growth techniques using gases for group-V elements[9,10] GSMBE, and chemical beam epitaxy (CBE). To obtain highest crystalline quality and better physical properties, during the growth of III-V compounds layers, special conditions are used. The growth occurs under element V surface stabilization with V/III partial pressure ratio usually approaching or higher than 10. The growth rate, chosen to keep a good control on interface quality for thin wells and to avoid unwanted impurity incorporation is about 1 ML/sec for InP and 0.3 ML/sec for InAs. In our case for the first interface (InAs on InP), a growth interruption under As flux of a sufficient duration to saturate the surface by As atoms is used. For this reason the first interface is considered to be P free, almost perfect and planar.[9] The growth of the InAs dot is started by the deposition of the required number of InAs monolayers on a planar surface. The growth is completed by an interruption time under As flux to let the physical process of island formation occur. Then the As flux is interrupted and simultaneously the P flux is switched on. During the time where the dot is under P flux stabilization the exposed dot surface is

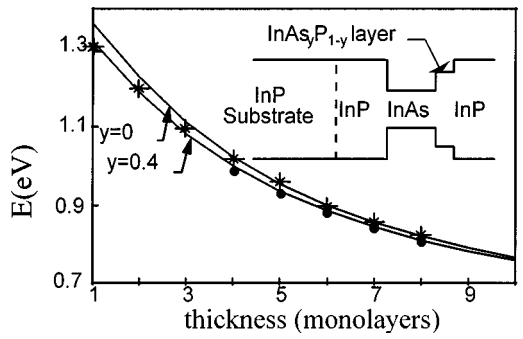

FIG. 2. Plot of calculated (solid lines) and measured PL energy levels vs InAs dot thickness at $T=77$ K. Our experimental results (solid dots) are presented with published results (stars) of Ref. 20. In the inset is presented the band diagram of the dot structure with the $InAs_yP_{1-y}$ interface layer. For clarity, only two calculated curves with two interface layer compositions ($y=0, y=0.4$) are presented.

nonplanar and presents a high degree of strain.[34] The thermodynamic law governing the dot formation remains active and induces a certain P/As atomic exchange on the shaped dot surface. This hypothesis is confirmed by the difference observed experimentally between the shape of uncovered and covered InAs dots by an InP capping layer.[23] Because of this 3D growth process, the final quantum dot top layer must be $InAs_yP_{1-y}$ with uncontrolled composition instead of pure InAs. This is this picture of the dot formation that has been used to explain and to evaluate the optical transitions.

In our experimental work, the number of layers of InAs/InP SAQD's does not exceed the value of 10. The calculation of optical transition energy between the first heavy-hole level and first electron level Ee_1hh_1 has been obtained using a simple picture of the SAQD. The dot is represented by a disk with a thickness equal to an integer number of InAs monolayers followed by one $InAs_yP_{1-y}$ interface layer with variable composition from $y=0$ to $y=1$. For this simulation we assume that the total strain localized inside the dot is used.

In Fig. 2 our PL experimental results for thick wells are reported as well as published experimental results[35] measured at the same temperature of 77 K. On the same figure are also reported the calculated optical transition energies. The result of the simulation shows that the optical transition energy of the well becomes less and less sensitive to this interfacial layer when its height increases as observed experimentally.

When no $InAs_yP_{1-y}$ transition layer is taken into account ($y=0$), the calculated optical transition energies always overestimates the experimental ones especially for the thinner dots. A good agreement between PL experimental and calculated energies is reached when the simple picture of a quantum disk $InAs_yP_{1-y}$ interfacial monolayer is used for the simulation. The average As composition of this $InAs_yP_{1-y}$ monolayer is found to be around 40%. A slight difference can be observed between our experimental results and the measurements of Ref. 35 for the same dot thickness. The different growth techniques (GSMBE, in our case, versus CBE) used to grow the dots can be invoked to explain this discrepancy. This can be also related to the difference between the As, P interchange kinetic energies on the shaped dot surface during the growth in the two chambers. To explain the difference observed between experimental PL and calculated optical transition energies the authors[35] suggest the existence of an excessive surface roughness during the growth of the dots. For this reason they consider mean values of dot thicknesses which are not an integer multiple of a monolayer to evaluate their transition energies. However, numerous experimental results (x-ray diffraction,[14] x-ray photoelectron diffraction,[36] Raman spectroscopy[37]), have been interpreted using the existence of this interface in the growth of quantum well arsenide compounds on InP. In our particular case, transmission electron microscopy (TEM) as well as AFM experiments performed on similar samples (<10 ML) with one InAs dot plane on InP does not help us to observe the interfacial layer. On these samples, TEM experiments are difficult to analyze because of the lack of precision due to strain effect and composition variation around the dot. However, for an InAs islands with greater heights (~ 23 ML), AFM, and TEM size measurements[16,23] in addition to photoluminescence experiments are in very good agreement with the present model.

V. EFFECT OF STRAIN REPARTITION

The second assumption we have used to treat the problem of discrepancy between the measured and the calculated optical transition energies is the presence of less strain in the InAs dot compared to the value estimated from the lattice parameter difference between InAs and InP. This hypothesis is based on a recent work on SiGe dots on an Si substrate.[3] In this work the effect of strain in the barrier is found to help the dots to organize themselves and to reduce the dispersion of their dimensions. After the growth of several dot planes, the experimental results show that the organization tends to be enhanced and the dispersion to be reduced.[3,5]

Raman spectroscopy experimental results have also been interpreted by the presence of strain in the barrier and the well.[4] Another work by Cusack, Briddon, and Jaros[8] shows theoretically that the strain effect is also present in the barrier within a limited distance from the dot interface and has the consequence of the reduction of the strain in the well. These results tend to show that the global strain is reduced in the dot by the interaction with the surrounding barrier and this effect may explain the reduction of the optical transition energy. We have then incorporated this assumption in the evaluation of the transition energy using the model described previously.

For strain repartition, we consider that both barrier and dot are biaxially strained on an a_0 lattice parameter substrate; since our dots are wide enough, shear strain can be neglected.[7,8] The in-plane lattice parameter in and around the dot can then be represented by a very simple equation:

$$a_0 = a_{0\mathrm{InP}} + Cx(a_{0\mathrm{InAs}} - a_{0\mathrm{InP}}),$$

where C is a coefficient varying from 0 to 1. We also considered a more sophisticated strain distribution (Gaussian shape) but this does not modify the trend of change in calculated energies. To simplify the discussion we do not present these results.

The calculated and experimental results are presented in Fig. 3. In the inset of the same figure the strain distribution

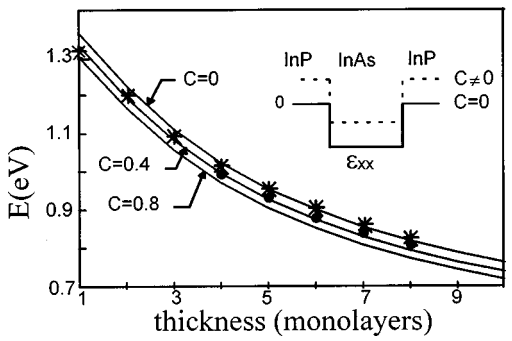

FIG. 3. Plot of calculated (solid lines) and measured PL energy levels vs InAs dot thickness at $T=77$ K. Our experimental results (solid dots) are presented with published results (stars) of Ref. 20. In the inset is presented the strain component ε_{xx} in the well ($C=0, C\neq 0$) and when it exists in the barrier ($C\neq 0$). For clarity, only three curves are presented with the parameters $C=0.0,0.4,0.8$.

inside the structure is represented, as well as its repartition between the well and the barrier for the usual case ($C=0$) and for the picture used in the model ($C\neq 0$). With the help of this simple model, it is clear that the calculated optical transition energies can be made equal to PL measured experimental values. However, a comment can be addressed to the simulation using the lowering of the strain in the dot and its presence in the barrier. When the dot thickness decreases we need to increase the strain in the barrier and to decrease it in the dot comparatively to found calculated energies equal to PL experimental energies. This conclusion leads to a contradiction because it shows that as the dot becomes thinner, its effect on the surrounding barrier becomes larger. For this reason the discrepancy between experimental and theoretical energies cannot be explained by the presence of the strain in the barrier. The explanation that can be invoked for the absence of the influence of the strain in the barrier is the lateral dimensions of our dots. In the evaluation of strain distribution Cusack, Briddon, and Jaros[8] show that the strain is maximum around low-symmetry crystallographic planes at the edges of the dot. The strain effect in the barrier nearly disappears at a distance of a few nm from the dot.[34] In our case, the dot diameter approaches the 50-nm range in the lateral plane perpendicular to the growth direction. For this reason, the presence of some strain at the far edges of the dot has little influence on the properties of its center. This description means that our dots have the properties of isolated disks, and their properties depend little on what is occurring on the edges.

VI. CONCLUSION

We have compared our calculations of fundamental transition levels to PL spectra on InAs quantum dots grown on a (100) InP substrate. Calculations, even for thin layers, show quite good agreement with experiments. We investigated theoretically the interfacial layer composition and strain effect on these fundamental transitions and showed their relative importance. We were able to predict a redshift that can be deduced either from a strain or gradient composition effect. Both effects must be taken into account for calculating energies if a high accuracy is needed. The interpretation of the redshift of optical transitions in InAs/InP SAQD's is related mainly to the existence of an $InAs_yP_{1-y}$ interfacial layer on the second growth interface. The existence of this interface is related to the special growth mode used to obtain the SAQD's and to the exchange kinetic between group-V elements at the surface of the dot. This conclusion is valid for dots with large in-plane dimensions assimilated to isolated quantum disks.

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