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Electron-hole alignment in InAs/GaAs self-assembled quantum dots: Effects of chemical composition and dot shape

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We investigate theoretically the effects of chemical composition and shape on the electronic states in InAs/GaAs self-assembled quantum dots, by using an eight-band strain-dependent $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian. For a number of InAs dots with different shapes, and especially with different gallium concentration profiles, we found various ranges of separation between electrons and holes. We show that gallium diffusion changes the confining potential for both electrons and holes through the strain profile in the dots, leading to totally different hole states from those in pure InAs dots. We also compute the electron-hole separation as a function of electron and hole energy levels. For the same gallium concentration profile, pyramidal dots exhibit the inverted alignment with the largest electron-hole separation compared with other two types, truncated-pyramidal and lens-shaped dots. Our calculations agree well with recent experiments [P.W. Fry *et al.*, Phys. Rev. Lett. **84**, 733 (2000)].

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As a typical example of zero-dimensional semiconductor structure,[1] InAs/GaAs self-assembled quantum dots (SAD's) have attracted considerable interest for the investigation of new physics[2] and potential applications in optoelectronics.[3–5] Although a large amount of effort has been expended at both experimental studies[6] and theoretical modeling,[7] there are still many uncertainties, such as chemical composition[8] and shape,[9] which play an important role in the electronic properties of SAD's. Recent experiments on Stark effect spectroscopy on SAD's (Ref. 6) investigated the effect of concentration distribution of gallium on the electronic states in InAs SAD's, and established a relation between the Stark shift and the electron-hole vertical separation. In particular, an inverted electron-hole alignment, i.e., the hole is localized towards the dot apex above the electron, was found. This surprising result was attributed to the influence of gallium diffusion in the SAD base, which squeezes the hole states towards the dot apex, while barely modifying the electron states.

In this work, we address the issue of the universality of electron-hole alignment inversion in SAD's. For this purpose, we perform a theoretical study of the electron-hole vertical separation in InAs/GaAs SAD's, concentrating on the effects of gallium diffusion into InAs islands, and on the shape of the structures. For a number of InAs dots, we found various ranges of vertical separation between electrons and holes, depending on their shapes, sizes, and especially the amount of gallium diffusion in the dots. All three types of SAD's considered in this study, namely, pyramidal, truncated pyramidal, and lens-shaped, could exhibit an inverted electron-hole alignment, contrarily to predictions based on a one-band model.[10]

Our model implements an eight-band strain-dependent $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian,[11] for which the strain is calculated by using linear continuum elastic theory.[12] The electronic structure is solved in the framework of the envelope function formalism by using the Lanczos algorithm.

First, we consider a truncated pyramidal structure, shown in the inset of Fig. 1, with a 17.4 nm base and 8.4 nm height, that is similar to SAD's studied in the Stark effect experiment.[6] Although gallium could diffuse in any portion of InAs SAD that is embedded in the GaAs matrix, we assume this most likely occurs at the base. The structure contains five $In_{1-x}Ga_xAs$ layers at its base, with x decreasing linearly from 50% at the bottom with intervals of 10%. The band structure along the growth direction through the center of the SAD is shown in Fig. 1. For comparison, the various band edges of a pure InAs SAD with the same size are represented in dashed lines.

In pure InAs SAD's, the confining potential for heavy holes (higher valence band) increases linearly from the base to the apex, while for electrons it is relatively flat throughout

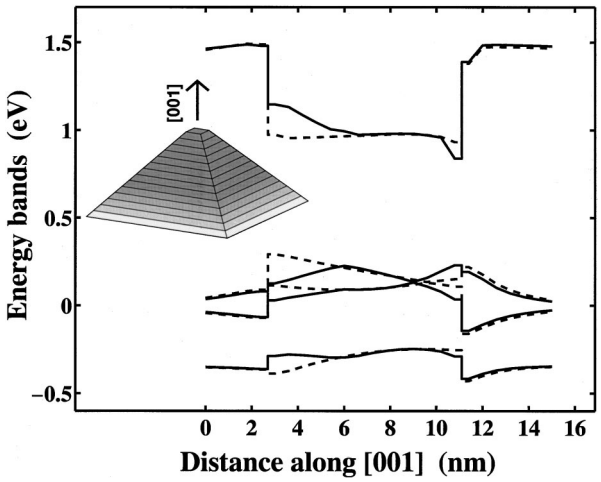

FIG. 1. Band structure along the [001] direction through the center of the dot. The dot bottom and top are at 2.7 and 11.1 nm, respectively. Dashed lines are for a pure InAs dot and solid lines are for the dot with gallium diffusion as shown in the inset. Inset: schematic view of a truncated pyramidal dot characterized by five graded $In_{1-x}Ga_xAs$ layers, with x decreasing linearly from 50% at the base by intervals of 10%.

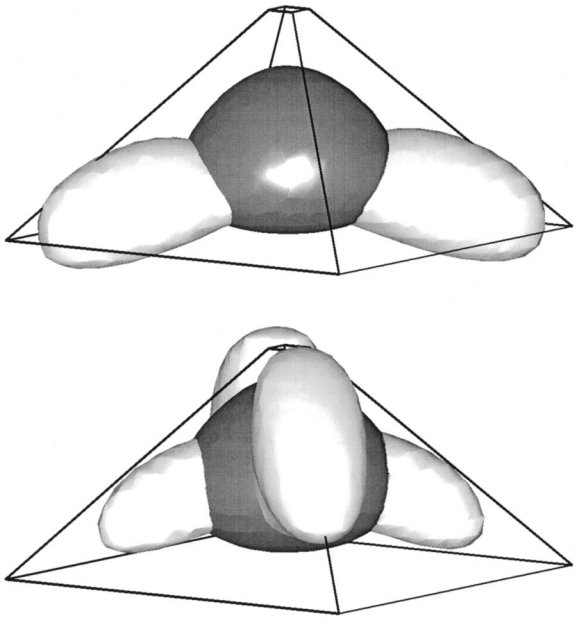

FIG. 2. Probability density isosurfaces of the ground states of electrons (dark) and holes (light gray). The upper panel is for the pure InAs dot and the lower panel for the dot with gallium diffusion as shown in the inset of Fig. 1.

the structure, which results in stronger hole confinement in the dot bottom. As seen in the figure, the presence of diffused gallium shifts the band edges at the bottom of the structure for both electrons and holes. As the offset between valence bands in the dot is much shallower than between the conduction bands, the gallium diffusion imposes a more remarkable effect on hole confinement than on electron confinement.

Figure 2 shows the wave functions of the ground electron and hole states. The upper panel is for the pure InAs SAD, and the lower panel for the structure with gallium diffusion. The overlap between the wave functions of electrons and holes is hidden by the dark isosurfaces of electron wave functions. The calculated vertical separation between the electron state and the hole state is 11.1 Å (upper panel) and -4.8 Å (lower panel), respectively. Here, we adopt the rule that the electron-hole separation is positive if the ground electron state is localized towards the dot apex above the ground hole state and negative in the opposite situation. For the pure InAs SAD, it is found the hole state is localized closer to the dot bottom, in agreement with Fig. 1, but, this is due to the different influences of strain on electrons and holes rather than to the heavier hole effective mass: electron states are more sensitive to hydrostatic strain components, while hole states are more affected by biaxial strain components. In SAD's with gallium diffusion, both electron and hole states are pushed away from the bottom towards the dot apex. However, as the hole state cannot occupy the space near the apex around the central axis, its wave function develops new ''wings'' around the facet edges because of the strong local confinement. These wings are responsible for the inversion of the electron-hole alignment, found experimentally.[6]

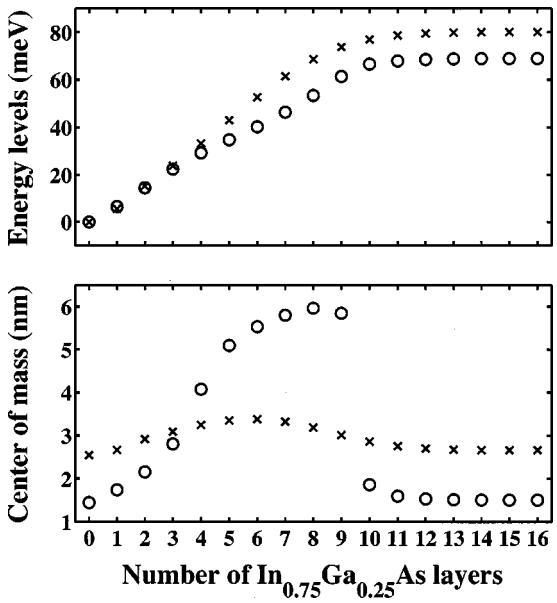

FIG. 3. Top: calculated energy level shifts, bottom; center of mass as a function of the number of $In_{0.75}Ga_{0.25}As$ layers in the bottom of the dot. The cross markers are for electrons and circles for holes.

Now, we investigate the dependence of the electron-hole separation on the amount of gallium diffusion in a InAs pyramidal SAD consisting of several $In_{1-x}Ga_xAs$ layers with a constant composition $x=0.25$. The structure has a 18.6 nm base and a 9.6 nm height. Figure 3 shows the calculated energy levels and the corresponding center of mass as a function of the number of diffusion layers. The energy levels are given in reference to the pure InAs SAD. When the number of diffusion layers is $n=16$, the structure is a homogeneous $In_{0.75}Ga_{0.25}As$ SAD. As seen in Fig. 3(a), both energies of electrons and holes increase almost linearly up to $n=10$ and remain unchanged for $n\geqslant 10$. In the latter situation ($n\geqslant 10$), electron and hole states are localized near the bottom of the structure [see Fig. 3(b)], so, the change of composition in the region around the pyramid tip has little effect on the energy levels. This result also implies that, for $n\geqslant 10$, the InAs pyramid could be regarded as a $In_{0.75}Ga_{0.25}As$ SAD with In-rich composition near its apex.

In Fig. 3(b), we show the behavior of the center of mass of the electron and hole states which is calculated from the bottom of the dot. At $n=0$, which corresponds to pure InAs SAD's, the hole state is localized 1.1 nm below the electron state. While both electron and hole states move towards the dot apex with the addition of the first few gallium layers, the shift of the hole state is larger than for the electron state, resulting in inverted electron-hole alignment between $n=3$ and $n=9$. We notice that the electron state begins to move down after $n=6$ while the hole state reverses its trend after $n=8$. Especially, the hole state is found to drop abruptly below the electron state at $n=10$, after which, positive electron-hole separation that converges to its value in homogeneous $In_{0.75}Ga_{0.25}As$ SAD's, is recovered. This result implies that InAs/GaAs SAD's with homogeneous composition throughout the structure could not exhibit inverted electron-

TABLE I. Calculated center of mass of electron states ($\mathbf{P}_e$) and of hole states ($\mathbf{P}_h$), and their separation $\mathbf{P}_e-\mathbf{P}_h$ for a number of InAs SAD's with different shapes, sizes, and gallium concentration profiles. All the length is in units of nm.

Type	Shape	Size	Composition	Pe	Ph	Pe–Ph
1	pyramid	18.6×9.6		3.26	3.89	−0.63
2	pyramid	18.6×9.6		3.22	3.64	−0.42
3	pyramid	18.6×9.6		3.2	4.01	−0.81
4	pyramid	18.6×9.6		3.21	3.3	−0.09
5	pyramid	18.6×9.6		3.49	4.71	−1.22
6	pyramid	18.6×9.6		3.2	3.39	−0.19
7	pyramid	18.6×9.6		3.34	4.69	−1.35
8	pyramid	18.6×9.6		3.24	3.28	−0.04
9	pyramid	18.6×9.6		3.34	3.99	−0.65
10	pyramid	18.6×9.6		3.17	3.96	−0.79
11	pyramid	18.6×9.6		3.12	3.58	−0.46
12	pyramid	18.6×9.6		3.08	3.21	−0.13
13	truncated	17.4×3.6		1.43	1.31	0.12
14	truncated	17.4×3.6		1.8	1.95	−0.15
15	truncated	18.6×8.4		3.23	3.98	−0.75
16	truncated	17.4×8.4		3.29	3.77	−0.48

hole alignment, which was predicted earlier with the one-band model.[10]

Besides gallium concentration profiles with linear grading (Fig. 1) and steplike grading (Fig. 3), we have also considered other possibilities, including the combinations, of linear and steplike grading, and nonlinear grading. Most of these diffusion profiles consist of a number of $In_xGa_{1-x}As$ layers in the bottom of InAs SAD's, with $x<0.5$. The number of diffusion layers does not exceed 8. Table I lists a number of InAs SAD of different shapes, sizes and gallium concentration profiles (along the growth direction) and the corresponding center of mass of the electron state and the hole state. For pure InAs SADs, there is no gallium inside the island, so the corresponding composition profile is flat.

For truncated pyramidal structures, the electron-hole separation is less pronounced because of the stronger confinement along the growth direction. For a truncated pyramid of 3.6 nm height and 17.4 nm base (case 14 listed in Table I), the electron-hole separation is only 1.2 Å without gallium diffusion and −1.5 Å with the same gallium concentration profile as shown in Fig. 1. For pyramidal structures, the dependence of the positions of electron and hole states on their energy levels is shown in Fig. 4. The energy levels are given in reference to the middle of GaAs band gap and the hole energies are given in their absolute values. Electron-hole separation is shown to vary roughly between −30 and 10 Å. The result illustrates such a basic feature as the decreasing separation between the electron and the hole state with increasing electron and hole energies. For both large positive and negative electron-hole separation, an obvious saturation is observed, which sets the upper limit on the separation in the dot.

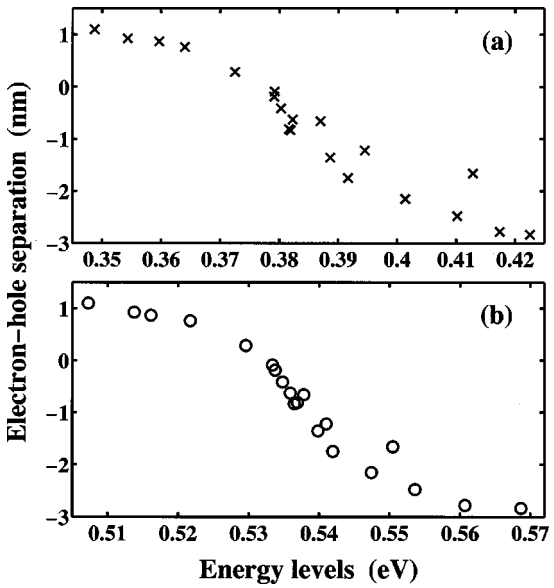

FIG. 4. Electron-hold separation versus energy levels of electrons (a) and holes (b) for a number of gallium diffusion profiles as listed in Table I and shown in Fig. 3.

We also investigated lens-shaped InAs SAD's for which the base diameter and height are the same as those of the pyramid structure in Fig. 3. The center of mass and the electron-hole separation in lens-shaped SAD's as a function of energy levels for a number of gallium diffusion profiles with linear grading and nonlinear grading are shown in Fig. 5. It is seen that lens-shaped SAD's share a similar feature with the pyramidal structures since the center of mass of electron states varies less than hole states. However, lens-shaped structures of similar size as pyramids exhibit smaller electron-hole separations [see Fig. 5(b) and 5(d)]. This can be explained as follows: In pyramidal structure, as seen in Fig. 2, the hole wave function leaks partially outside the structure by developing ''wings'' around the facet edges.

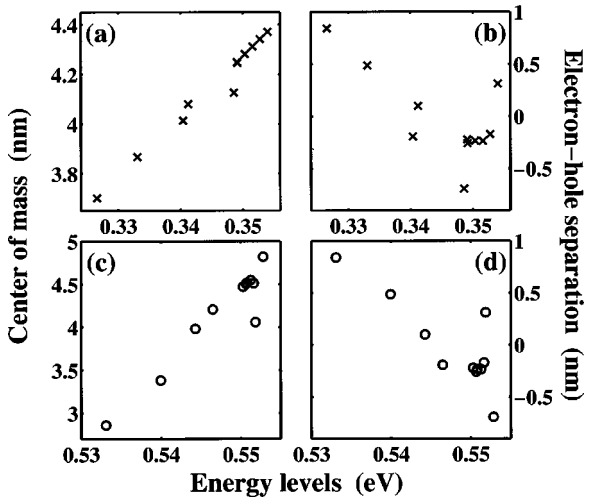

FIG. 5. Center of mass and electron-hole separation versus energy levels of electrons (a),(b) and holes (c),(d) for a number of gallium diffusion profiles of lens-shaped dots.

While the lens-shaped SAD provides a more wider region than in pyramids to accommodate the hole inside the structure, the hole wave function is generally found to stay inside the structure away from the apex. Hence holes are confined in a narrower region, resulting in smaller electron-hole separation in comparison with the pyramidal structure.

Besides the electron-hole separation, the gallium diffusion also affects the optical transition energy, and especially the transition strength. Calculations show that a negative electron-hole separation is associated with larger transition energy compared with positive separation. The result also demonstrates that the optical transition in structures with negative electron-hole separation is not as strong as that with positive separation. For instance, the transition strength becomes three times smaller as the electron-hole separation changes from 1 nm to -1 nm.[13]

In conclusion, we studied the electron-hole separation in InAs SAD's due to the effect of gallium diffusion and dot shape. We found that diffused gallium into InAs SAD's imposes a different effect on the effective confinement of electrons and holes, leading to inverted electron-hole alignment for specific gallium diffusion profiles. We also found that hole states are more sensible to gallium diffusion, especially for pyramidal SAD structures. The separation between electrons and holes is generally larger for higher structures; that is the reason why truncated pyramids have smaller electron-hole separation. Let us point out that our model does not take into account excitonic effects in the calculated electron-hole alignment. Given the attractive nature of this interaction, exciton binding would reduce the particle separation without producing major effects on the electronic states. Indeed, as the Bohr radius is much larger than the dot size, excitons only perturb the electron-hole system without affecting the direction of their alignment. Therefore, based on energetic considerations, we can expect our calculated results to be at most overestimated by 10–20 %. Hence, for structures with similar sizes as SAD's studied experimentally, we found inverted electron-hole alignments that are consistent with the experiment for several gallium diffusion profiles characterized by linear grading or steplike grading. However, the inverted alignment is not a universal property of SAD's, but ultimately depends on diffusion profile and dot size that are imposed by fabrication processes.

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