Effect of interfacial states on the binding energies of electrons and holes in InAs/GaAs quantum dots

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The interface between an InAs quantum dot and its GaAs cap in ''self-assembled'' nanostructures is nonhomogeneously strained. We show that this strain can lead to localization of a GaAs-derived X_{1c}-type interfacial electron state. As hydrostatic pressure is applied, this state in the GaAs barrier turns into the conduction-band minimum of the InAs/GaAs dot system. Strain splits the degeneracy of this X_{1c} state and is predicted to cause electrons to localize in the GaAs barrier above the pyramidal tip. Calculation ~present work! or measurement ~Itskevich *et al.*! of the emission energy from this state to the hole state can provide the hole binding energy, $D^{(h)}_{\rm dot}$. Combining this with the zero-pressure electron-hole recombination energy gives the electron binding energy, $D^{(e)}_{\rm dot}$. Our calculations show $D^{(h)}_{\rm dot}$; 270 meV ~weakly pressure dependent! and $D^{(e)}_{\rm dot}$; 100 meV at P 5 0. The measured values are $D^{(h)}_{\rm dot}$; 235 meV ~weakly pressure dependent! and $D^{(e)}_{\rm dot}$; 50 meV at P 5 0. We examine the discrepancy between these values in the light of wave-function localization and the pressure dependence of the hole binding energy. @S0163-1829~98!06635-1#

The interest in potential *optical* applications of semiconductor quantum dots has concentrated attention almost entirely on their direct gap electronic states, i.e., those derived from the G point of the bulk band structure, the G_{1c}-derived electron states and G_{15v}-derived hole states. There are however, several interesting quantum dot situations in which the lowest energy electron states of dots are derived from the X_{1c} point. These include ~i! Si quantum dots,[1] and SiGe nanostructures embedded in Si,[2] ~ii! InP nanostructures embedded within GaP ~Refs. 3 and 4!, and ~iii! InAs nanostructures embedded in GaAs at a hydrostatic pressure above the $G_{1c}\,2\,X_{1c}$ transition.[6] In some of these cases, @e.g., ~ii! and ~iii!#, the resulting band alignment can be type II ~indirect! in both reciprocal and real space, with holes confined to G-like states of the dot and electrons in X-derived states of the barrier. When a lattice mismatch between the dot and the barrier materials also exists, the resulting strain field can lead to localization of these X-derived electron states.[2,4] This can be seen qualitatively by considering the simple case of an isotropic *spherical* inclusion in an isotropic matrix as originally derived in 1956 by Eshelby[5] to first order in the lattice mismatch, $e_m\,5\,(a_i\,2\,a_m)/a_m$, where a_i and a_m are the lattice constants of the inclusion and the matrix, respectively. Eshelby showed that inside the sphere, only uniform hydrostatic strain exists,

$$e_{in}\,5\,e_m\left\{\frac{1}{g}\,2\,1\right\}, \qquad \text{~1!}$$

where $g\,5\,1\,1\,2B_m$

The method of Itskevich *et al.* assumes that above the critical pressure P_c, the emission takes place from the X_{1c} level in the GaAs barrier, to the confined hole state, h^{InAs}, with an emission energy, $E_{\mathrm{dot}}(X_{1c}^{\mathrm{GaAs}} 2\, h^{\mathrm{InAs}};P)$. Combining this value with the bulk GaAs indirect transition energy at this pressure, $E_{\mathrm{bulk}}(X_{1c}^{\mathrm{GaAs}} 2\, \mathrm{G}_{15v}^{\mathrm{GaAs}};P)$ the hole binding energy within the dot, $\mathrm{D}_{\mathrm{dot}}^{(h)}(P)$, can be obtained from Fig. 1~a!

$$\mathrm{D}_{\mathrm{dot}}^{\sim h!}\sim P!\ 5\ E_{\mathrm{bulk}}\sim X_{1c}^{\mathrm{GaAs}}\ 2\ \mathrm{G}_{15v}^{\mathrm{GaAs}};P! \\ 2\ @E_{\mathrm{dot}}\sim X_{1c}^{\mathrm{dot}}\ 2\ h^{\mathrm{InAs}};P!\ 1\ d_{\mathrm{loc}}\sim P!\#. \qquad \sim 3!$$

This approach assumes that the emitting state X_{1c}^{dot} has precisely the energy of the threefold degenerate X_{1c} level in bulk GaAs under pressure P, i.e., that it is an extended Bloch state. This neglects the Eshelby strain that could exist at the GaAs-InAs interface, and the ensuing wave-function localization at the interface. This localization could shift the emitting state @Fig. 1~a!#. An extra correction term $d_{\mathrm{loc}}(P)$, is therefore included in Eq. ~3! to allow for emission from the ''split X_{1c} state,'' called X_{1c}^{dot}, rather than from bulk X_{1c}

$$d_{\mathrm{loc}}\sim P!\ 5\ E\sim X_{1c}^{\mathrm{GaAs}}\ 2\ X_{1c}^{\mathrm{dot}};P!. \qquad \sim 4!$$

The electron binding energy $\mathrm{D}_{\mathrm{dot}}^{(e)}(P)$, can then be determined @Fig. 1~b!# by subtracting from the zero-pressure direct gap of GaAs the measured zero-pressure electron-hole recombination energy $E_{\mathrm{dot}}(e^{\mathrm{InAs}}\ 2\ h^{\mathrm{InAs}};P\ 5\ 0)$, and the zero-pressure hole confinement energy:

$$\mathrm{D}_{\mathrm{dot}}^{\sim e!}\ 5\ E_{\mathrm{bulk}}\sim \mathrm{G}_{1c}^{\mathrm{GaAs}}\ 2\ \mathrm{G}_{15v}^{\mathrm{GaAs}};P\ 5\ 0!\ 2\ E_{\mathrm{dot}}\sim e^{\mathrm{InAs}}\ 2\ h^{\mathrm{InAs}};P\ 5\ 0! \\ 2\ \mathrm{D}_{\mathrm{dot}}^{\sim h!}\sim P\ 5\ 0!. \qquad \sim 5!$$

Since Eq. ~3! gives the hole confinement energy at $P\ .\ P$

P 5 0 the ground electron state is confined *within* the InAs dot @Fig. 2~a!#, while at the pressure where the GaAs barrier undergoes a $G_{1c} \rightarrow X_{1c}$ transition, the GaAs/InAs interfacial strain produces a strain-split, *localized* X_{1c} state @Fig. 2~b!# localized *outside* the dot, above its tip. The energy of this state differs in energy by 0.024 eV from a Bloch-extended bulk GaAs X_{1c} state obtained in our calculation at a position far away from the dot, where the Eshelby strain @Eqs. ~1!–~2!# has decreased to zero. The *hole* states do not significantly change their character with pressure and are always localized within the dot. Our directly calculated values at 60 kbar ~see Table I! for Eq. ~3! are

$$D_{dot}^{~h!}~P\ 5\ 60!\ 5\ 1.850\ 2\ 1.528\ 2\ 0.021\ 5\ 0.301 \text{ eV} \qquad ~8!$$

and for Eq. ~5!

$$D_{dot}^{~e!}~P\ 5\ 0!\ 5\ 1.548\ 2\ 1.180\ 2\ 0.271\ 5\ 0.097 \text{ eV}. \qquad ~9!$$

For a larger dot ~base 5 150 Å, height 5 15 Å! we obtain $D_{dot}^{(h)}(P\ 5\ 60)\ 5\ 0.338$ and earlier. In Fig. 3, we use atomistic calculations to illustrate the effects on the band offsets of applying hydrostatic pressure to a GaAs embedded, strained InAs *pyramidal* quantum dot. The *unstrained* band offsets between GaAs and InAs are shown as dashed lines at zero pressure in Fig. 3~a!. They show that at P 5 0 the natural GaAs/InAs offsets allow InAs to act as a ‘‘well’’ for both the conduction-band G_{1c} electrons and the valence-band G_{15v} holes ~a ‘‘type I’’ offset!. The solid lines in Fig. 3~a! show the offsets subject to the local strain e~**R**!, plotted along a @001# direction down through the tip of the InAs pyramid ~see inset!. We obtain the position-dependent strained offsets by discretizing the GaAs/InAs nanostructure into ‘‘cells’’ with position vector **R** and then performing 60 bulk band-structure calculations of InAs and GaAs, thus obtaining the bulk eigenvalues E_{nk}@e(**R**)# for band n at wave vector k within each cell, using the strained In-As or Ga-As bond geometry in that cell.[14] These solid lines in Fig. 3~a! show that far from the dot where the strain is small, the offsets approach the unstrained value, however the compressive strain within the InAs dot, increases the valence-band offset from 0.11 to 0.41 eV ~allowing more confined hole states! and decreases the conduction-band offset from 1.01 to 0.55 eV ~reducing the number of confined electron states!.

Figure 3~b! shows the band offsets under 60 kbar of pressure. The GaAs barrier material has already undergone a conduction band $G_{1c} \rightarrow X_{1c}$ crossing, while the InAs remains direct. We observe that the strained band offset for X_{1c} electrons has developed *local minima* ~indicated by arrows! just above the tip and below the base of the InAs dot. The development of these minima is principally due to the splitting of the triply degenerate X_{1c}-derived states by the epitaxial strain at the interface between the dot and the barrier as predicted by Yang *et al.*[2] It is within these minima that the lowest energy conduction state localizes. This leads to trapping of the electron wave function at the interface, and to a lowering of the energy level relative to the bulk GaAs X_{1c} level.

In conclusion, our pseudopotential calculations show that the InAs/GaAs interfacial strain leads to the development of a trough in the X