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

A. J. Williamson and Alex Zunger
National Renewable Energy Laboratory, Golden, Colorado 80401
~Received 24 April 1998!

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, $D_{\rm dot}^{(h)}$. Combining this with the zero-pressure electron-hole recombination energy gives the electron binding energy, $D_{\rm dot}^{(e}$

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, $E_{\mathrm{dot}}(X_{1c}^{\mathrm{GaAs}} - h^{\mathrm{InAs}};P)$. Combining this value with the bulk GaAs indirect transition energy at this pressure, $E_{\mathrm{bulk}}(X_{1c}^{\mathrm{GaAs}} - \Gamma_{15v}^{\mathrm{GaAs}};P)$ the hole binding energy within the dot, $\Delta_{\mathrm{dot}}^{(h)}(P)$, can be obtained from Fig. 1(a)

$$\Delta_{\mathrm{dot}}^{(h)}(P) = E_{\mathrm{bulk}}(X_{1c}^{\mathrm{GaAs}} - \Gamma_{15v}^{\mathrm{GaAs}};P) - [E_{\mathrm{dot}}(X_{1c}^{\mathrm{dot}} - h^{\mathrm{InAs}};P) + \delta_{\mathrm{loc}}(P)]. \quad (3)$$

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 $\delta_{\mathrm{loc}}(P)$, 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}

$$\delta_{\mathrm{loc}}(P) = E(X_{1c}^{\mathrm{GaAs}} - X_{1c}^{\mathrm{dot}};P). \quad (4)$$

The electron binding energy $\Delta_{\mathrm{dot}}^{(e)}(P)$, 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 $E_{\mathrm{dot}}(e^{\mathrm{InAs}} - h^{\mathrm{InAs}};P=0)$, and the zero-pressure hole confinement energy:

$$\Delta_{\mathrm{dot}}^{(e)} = E_{\mathrm{bulk}}(\Gamma_{1c}^{\mathrm{GaAs}} - \Gamma_{15v}^{\mathrm{GaAs}};P=0) - E_{\mathrm{dot}}(e^{\mathrm{InAs}} - h^{\mathrm{InAs}};P=0) - \Delta_{\mathrm{dot}}^{(h)}(P=0). \quad (5)$$

Since Eq. (3) gives the hole confinement energy at $P > P$

$P=0$ the ground electron state is confined *within* the InAs dot [Fig. 2(a)], while at the pressure where the GaAs barrier undergoes a $\Gamma_{1c}\rightarrow X_{1c}$ 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

$$\Delta_{\rm dot}^{(h)}(P=60)=1.850-1.528-0.021=0.301\ \ \text{eV} \qquad (8)$$

and for Eq. (5)

$$\Delta_{\rm dot}^{(e)}(P=0)=1.548-1.180-0.271=0.097\ \ \text{eV}. \qquad (9)$$

For a larger dot (base$=150$ Å, height$=15$ Å) we obtain $\Delta_{\rm dot}^{(h)}(P=60)=0.338$ 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 $P=0$ the natural GaAs/InAs offsets allow InAs to act as a ''well'' for both the conduction-band Γ_{1c} electrons and the valence-band Γ_{15v} holes (a ''type I'' offset). The solid lines in Fig. 3(a) show the offsets subject to the local strain $\epsilon(\mathbf{R})$, 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 $\mathbf{R}$ and then performing 60 bulk band-structure calculations of InAs and GaAs, thus obtaining the bulk eigenvalues $E_{nk}[\epsilon(\mathbf{R})]$ 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 $\Gamma_{1c}\rightarrow X_{1c}$ 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