Conduction-band crossover induced by misfit strain in InSb/GaSb self-assembled quantum dots

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We address the occurrence of conduction-band crossover in III-V self-assembled quantum dots solely due to misfit strain. Band structure analysis in terms of standard deformation-potential theory shows that Γ-*X* crossover can occur in the dot, while both Γ-*X* and Γ-*L* crossovers are possible in the matrix at the interface. Crossover changes the nature of the fundamental band gap in the heterostructure, which may dramatically affect the optical properties. The implications of this are studied for a realistic InSb/GaSb (001) heterostructure, where Γ-*L* crossover renders the ground-state optical transition indirect in **k** space. Our calculations and photoluminescence data are in remarkable agreement.

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Self-assembled quantum dots (SAQDs) offer unique opportunities both in fundamental and applied research. While possessing properties of an "artificial atom," SAQDs are fully compatible with traditional semiconductor technology and provide a prospective material for active media in optoelectronic devices. To cover a wider optical wavelength range, SAQD heterostructures using novel material combinations are continually being developed.

A specific feature of SAQDs is the large lattice-mismatch strain, which is inherent to growth by the Stranski-Krastanov method.[1] The significance of the effect of strain on the electronic band structure in SAQDs was realized a decade ago. In particular, it was shown[1] that due to the large lattice misfit ($\geqslant 2\%$), strain-induced shift and/or splitting of the band edges occurs, which is comparable in magnitude to the optical band gaps in the dot and/or matrix materials. However, modeling of the electronic band structure in III-V SAQDs has been largely limited to the Γ point of the Brillouin zone, which is the location of the fundamental band gap in the unstrained materials.[2] Specifically, the effect of misfit strain on the gaps *between the conduction bands of different symmetry* has not been analyzed. Such an analysis is particularly important because the strain can induce a crossover of the Γ, *L*, and/or *X* valleys. In this case, the ground electron level in the SAQD heterostructure would be of different symmetry from that in the unstrained dot material, which critically affects the optical properties.

In this Brief Report, we analyze the conditions for crossover between the Γ, *L*, and *X* conduction-band minima as a result of the misfit strain. In particular, we calculate the band profile for a realistic InSb/GaSb (001) SAQD system. We compare results of the calculations with our photoluminescence (PL) data, which provide experimental evidence for the Γ-*L* crossover in these SAQDs.

From the point of view of the effect on the band edges, it is convenient to decompose the strain in the hydrostatic and shear components. Analysis of the strain profile in (001) SAQDs has shown[3] that compressive hydrostatic strain is typically concentrated inside the dot, whereas the strongest shear components are observed in the matrix near the heterointerface. The major effect on the band structure inside the dot is the hydrostatic-strain-induced uplift of the Γ-valley edge, which may result in a crossover with the *X* valley. (This is similar to the Γ-*X* crossover in III-V SAQDs under *external* pressure.[4]) The shear strain inside the dot is predominantly tetragonal. It lifts the degeneracy of the *X* valley, but does not affect the *L* band. The tetragonal shear is small in taller dots, but increases strongly in flatter dots at the expense of the hydrostatic component,[3] providing another possible cause for the Γ-*X* crossover.

In the matrix, hydrostatic strain is generally low, but large values of either tetragonal or trigonal shear components can be found at various locations along the heterointerface.[3] Shear deformation potentials for the *L* and *X* bands are comparable to each other ($\approx$15 eV), but larger than the hydrostatic counterpart for the Γ valley ($\approx$10 eV).[5] Assuming that the *L* band has a lower energy in the unstrained material than the *X* band, one can therefore expect the primary crossover effect to be due the *L*-valley splitting (i.e., the Γ-*L* crossover) and for this to be observed *in the matrix* near the heterointerface. Indeed, an evaluation of realistic Γ-*L* gaps ($\geqslant 0.1-0.3$ eV) against shear deformation potentials shows that the crossover can be observed at a shear strain of a few percent, comparable to the typical misfit strain in SAQDs of 2%–7%. We would like to note that it is very difficult to achieve shear strain[6] of more than 1% in bulk semiconductors.

Of all III-V binary compounds, GaSb has the smallest Γ-*L* gap of $\approx$70 meV.[5,7] Hence, one can expect the most prominent Γ-*L* crossover to be observed in SAQD heterostructures with GaSb as a matrix material. In fact, the Γ-*L* crossover has been observed in bulk GaSb under uniaxial stress along the ⟨111⟩ direction.[7] This effect was mainly due to the *L*-valley splitting. It is illustrative to note that the required shear strain was as small as 0.25%, so the crossover effect ought to be expected in SAQD heterostructures.

We have examined the electronic structure and PL spectra from the InSb/GaSb (001) SAQD heterostructures. The numerical modeling was performed within the continuum-elasticity approximation and standard deformation potential

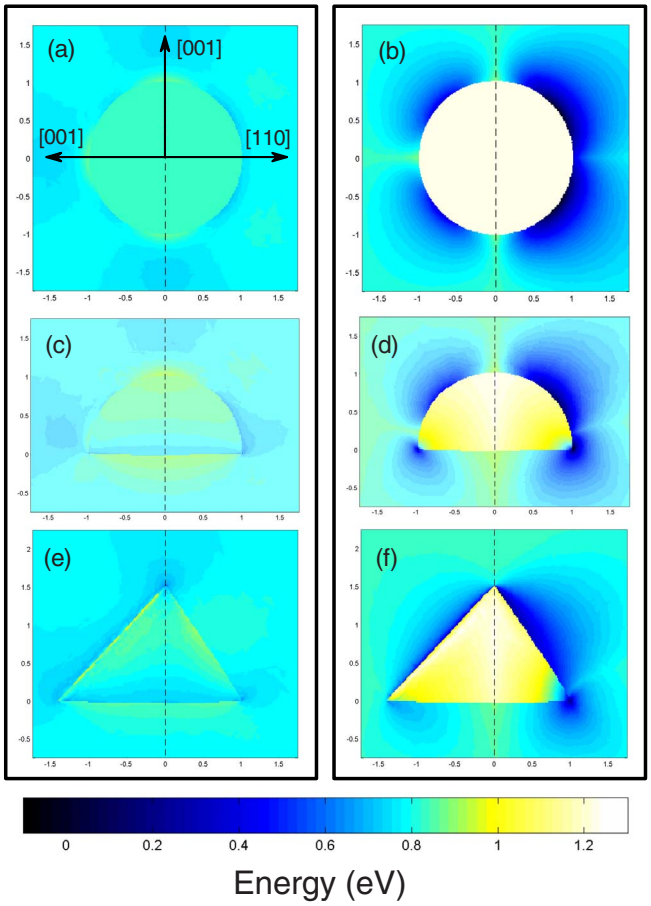

FIG. 1. (Color online) Conduction-band-edge profiles for InSb/GaSb dots of different shapes: [(a) and (b)] sphere, [(c) and (d)] semisphere, and [(e) and (f)] square-based pyramid. Left-hand column shows the profiles for the Γ valley; right-hand column shows the profiles for the L valley.

theory; for details, see Ref. 3. The strain-induced shift of L and X valleys was calculated using the Herring-Vogt notation.[8] The piezoelectric field was also taken into account. The energies of electron levels were calculated using the effective mass tensor, while the hole levels were modeled using the Luttinger Hamiltonian within the four-band $\boldsymbol{k}\cdot\boldsymbol{p}$ approximation. The material parameters were taken from Refs. 5 and 7. The samples for the experimental studies were grown using molecular beam epitaxy and extensively characterized using transmission electron microscopy.[9] The PL spectra were excited by a semiconductor diode laser emitting at 650 nm, and recorded using a Fourier-transform infrared spectrometer and a liquid-nitrogen-cooled InSb detector; for details, see Ref. 9.

Figure 1 presents a color map of the band profile for the Γ valley and the lowest L valley for three representative dot shapes. For the Γ valley, average energies in the dot and in the matrix are close to each other. On the other hand, the L-band-edge profile strongly varies. The most prominent feature of the profile is the deep L "pockets" (dark blue), which are formed in the matrix near the heterointerface. One can see that for *all* the shapes, the band edges in the pockets are far below the Γ-valley edge. Therefore, the pockets provide

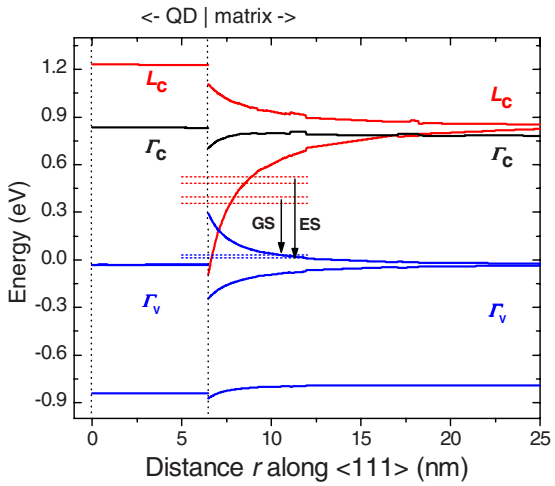

FIG. 2. (Color online) Band profile along the ⟨111⟩ direction for a spherical InSb/GaSb dot. Continuous lines show band edges; dashed lines show energy levels. Arrows indicate optical transitions from the ground (GS) and first excited (ES) states.

local potential minima for electrons of different symmetry to the bulk material.

Our calculations confirm that the pockets originate from the shear strain, which lifts the degeneracy of the L valleys. Spatial positions of the L pockets reflect the fact that the trigonal shear strain comprises contraction along the ⟨111⟩ direction (or equivalent) and compensating expansion in the perpendicular directions. In the particular case of a spherical dot, it creates eight identical pockets centered along ⟨111⟩-type directions, in agreement with the symmetry of the elastic problem.

Figure 2 presents the profile for both conduction and valence bands for a particular example of the *spherical* dot along the radial ⟨111⟩ direction. One can see that in addition to the L valleys, shear strain splits the valence band in the matrix. As a result, deep L pockets are formed for electrons, with the band gap becoming even negative at some specific points close to the heterointerface. One can also see that the size of the pockets is comparable to that of the dots. Therefore, taking into account heavy effective masses of L electrons, one can expect the ground electron states to originate from the localized states in the pockets, instead of the Γ-valley-related states.

Figure 2 includes the size-quantized levels of electrons and holes and also the corresponding optical transitions. Energy levels were calculated for spherical InSb/GaSb dots with a diameter of 13 nm. This appears to be the best approximation for the geometry of the dots in our samples.[9] The reported range of L-valley effective masses in GaSb is very wide, from $0.08m_0$ to $0.28m_0$ for the transverse mass m_t; it is typically assumed that the longitudinal mass $m_l=10m_t$.[5] This results in an uncertainty of ≈100 meV for the ground-state energy. However, despite the uncertainty, the electron levels are clearly localized in the L pockets well below the Γ-valley edge. (The levels are split into doublets due to the piezoelectric field, which is discussed later.) Therefore, the ground-state optical transition becomes indirect in **k** space. Our analysis shows that this is a general feature of

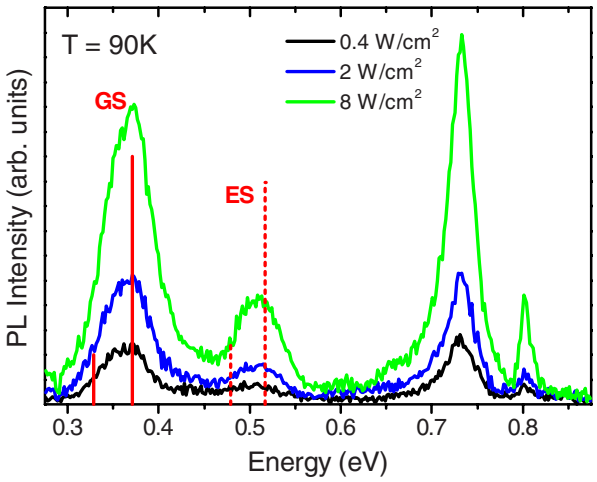

FIG. 3. (Color online) Experimental PL spectra for different excitation densities at 90 K. The spectra are not offset. Vertical bars show calculated peak positions.

InSb/GaSb SAQDs. One can see in Fig. 2 that the hole ground state is also localized in the matrix near the heterointerface. This is due to a combination of the shallow localization potential inside the dot and the valence-band bending in the matrix. This particular feature is specific to the spherical dot; for flatter dots, the hole localization potential inside the dot is deeper. Still, the overall localization of holes in the heterostructure is generally shallow.

We note that the effect of the piezoelectric field on the L pockets is much stronger than on the electronic states inside the dot. This occurs because the piezoelectric potential peaks *outside* the dot near the heterointerface in the $\langle 111 \rangle$-type directions, exactly where the L pockets are located. As a result, eight equivalent L pockets around the spherical dot split into two subsets (four and four), according to noninversion spatial symmetry of the piezoelectric effect. The splitting between the band edges in the subsets amounts to $\approx$100 meV. In addition, the piezoelectric field modifies the valence-band profile around the L pockets in a different way for the two subsets. The latter has a dramatic effect on the electron-hole overlap. In particular, negative (positive) piezoelectric potential pushes an L pocket up (down) in energy and simultaneously enhances (diminishes) the amplitude of the hole wave function around the pocket. As a result, the overlap is different for the two subsets. Incidentally, this effect is highest for the spherical dot, because for this shape, the hole tends to be localized at the heterointerface. However, the effect does not vanish for other shapes. While in the latter case, the *overall* electron-hole overlap may decrease substantially, the difference in the overlap for the two subsets is still significant.

Figure 3 shows a representative set of PL spectra from our samples recorded over a wide energy range. In each spectrum, two sets of broad peaks can be observed in the ranges 0.3–0.55 eV and 0.6–0.85 eV, respectively. Emission in the latter range has been reported previously for InSb/GaSb SAQDs.[10] It was interpreted as a combination of emission from the dots, the wetting layer, and numerous shallow defect states in bulk GaSb. Note that the calculated Γ-Γ band gap (see Fig. 2) falls within this range.[9]

Emission in the 0.25–0.55 eV range was first reported in Ref. 9. Analysis shows that it is in excellent agreement with our calculations. Vertical solid bars in Fig. 3 correspond to modeled PL peaks from the ground states in two subsets of the L pockets. The positions of the bars reflect the calculated transition energies. (We did not take the exciton binding energy into account.) The best agreement with the observed PL peak at 0.37 eV was obtained for effective masses of $m_t = 0.19m_0$ and $m_l = 10m_t$.

Variation of the relative intensity of the peak at 0.5 eV with excitation density suggests that it is due to emission from an excited state. Transitions from the excited states in the two subsets of the L pockets, modeled using the *same* parameters, are shown with dashed vertical bars. They are in remarkably good agreement with experiment.

The height of the vertical bars in Fig. 3 shows the relative values of the electron-hole overlap integral. We suggest that they may be used as an *estimate* of the expected transition intensities. (Calculation of the oscillator strength for these transitions, which requires consideration of the relaxation of the momentum conservation rule, deserves a separate study.) These estimates appear to provide a good description of the spectra, with the peak half-width of $\approx$30 meV being due to inhomogeneous broadening. In particular, the ground-state transition from the lower-energy subset of the L pockets is not resolved at $\approx$0.32 eV. However, it accounts for the asymmetry of the observed PL peak. A transition from the excited state in the same subset can be found in the spectra as a poorly pronounced shoulder at $\approx$0.47 eV.

We note that the large energy difference between the transition from the ground and excited states ($\approx$140 meV) is almost entirely due to a contribution from the electron levels. Such a large separation of quantized levels is another distinct indication of the deep quantization potential. We also note that the values of the effective masses that provide the best fit are close to the upper limit of the reported range. This may reflect a strong nonparabolicity of the L valley in GaSb.[11]

Now we consider additional arguments that support our interpretation of the PL spectra in terms of the L-valley-related emission. First, examination of the band profile for this system, as shown in Fig. 2, reveals no other available electronic states to account for emission in the 0.25–0.55 eV range. We would like to point out that in our samples, the dots are fully and coherently strained.[9] Hence, neither plastically relaxed dots nor dislocations can be expected to contribute to the emission. Second, the overall PL intensity is low, despite high density and excellent structural quality of the dots.[9] The PL signal was estimated to be at $\approx 1/100$ of that from comparable InAs/GaAs SAQD samples under similar experimental conditions. Such low emission is consistent with a ground-state transition which is indirect in $\mathbf{k}$ space, i.e., from the L pockets. Third, we note the high temperature stability of the PL emission. The integrated intensity falls only about four times over a temperature rise from 90 to 300 K. This is indicative of a deep localization of the ground electron state in the L pockets.

Further evidence for the origin of the electron ground state in InSb/GaSb SAQD heterostructures can be obtained from time-resolved or high-pressure PL experiments.

In addition, we would like to comment on the robustness of the prediction of the Γ-L crossover and its effect on the optical transitions with respect to the approximations used. The continuum-elasticity approximation generally shows good agreement with atomistic-elasticity calculations.[12] Thus, the deep L pockets should emerge within any realistic approach to the elastic problem, at least within linear deformation potential theory.

Finally, we note that we have performed a similar modeling for the well-studied system of InAs/GaAs (001) SAQDs. It has also revealed a strong splitting of the L-valley edge at the heterointerface. The ground electron state, however, is still localized within the dot, mainly due to the larger Γ-Γ and Γ-L gaps in bulk GaAs.

To summarize, we have reported that Γ-L (Γ-X) crossover can occur in the conduction band of SAQD heterostructures purely as a result of misfit strain. Due to the resulting crucial effect on the optical properties, this needs to be taken into account in fundamental analysis and band-profile engineering. For InSb/GaSb (001) SAQD heterostructures, we have shown that the Γ-L crossover results in a deeply localized ground electron state that originates from the L valley. Hence, the nature of the resulting ground-state transition becomes incompatible with optical applications of such SAQDs. The experimental PL spectra are in excellent agreement with our predictions.

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