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# Strain-Induced Quenching of Optical Transitions in Capped Self-Assembled Quantum Dot Structures

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Strain-induced quenching of optical transitions has been found in capped self-assembled quantum dot structures. Light absorption at the $E_1$ and $E_1 + \Delta_1$ critical points of InSb islands buried in InP disappears for nominal InSb thicknesses lower than 10 monolayers as a consequence of the strain produced inside the islands by the cap layer. Certainly, this strain increases as the InSb deposition diminishes, changing the band lineup of the system from type-I to type-II and therefore drastically reducing the oscillator strengths of the island-related $E_1$ and $E_1 + \Delta_1$ transitions. [S0031-9007(97)05086-2]

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Strain has been shown to play a relevant role not only in the formation [1] and positioning [2,3] of islands in self-assembled quantum dot (QD) structures, but also in the physical properties and, in particular, in the optical properties of such systems [4–8]. All previous studies considered only the strain-induced change of the energies of the island-related optical transitions, but strain also modifies the energies of the electronic states of the dots with respect to those of the matrix. In this paper we present a new effect related to the builtin strain characteristic of self-assembled QD structures: *The quenching of optical transitions produced by a strain-induced modification of the band lineup of the system.* InSb dots grown on top of InP and InSb dots grown deeply buried in InP are the objects of study of this paper. Different modulation spectroscopies were used for optical characterization and transmission electron microscopy (TEM) and Raman scattering for structural characterization.

Uncapped (dots on top) and capped (dots deeply buried) samples were grown by molecular-beam epitaxy [9], under identical conditions, on semi-insulating (001) InP substrates. Growth details are reported elsewhere [10]. The nominal InSb thicknesses were 2.2, 3.2, 5, and 7 monolayers (ML) in the uncapped samples and 3.5, 5, 10, and 15 ML in the capped samples, in which the cap layer thickness was 150 ML. Doubtless due to the large lattice mismatch existing between InSb and InP (≈10.4%), the onset of the island growth mode was observed after growing only 1.2 ML of InSb. In consequence, well-developed dots were obtained in every sample, as TEM results presented below demonstrate. The typical island size was of the order of several tens of nanometers.

Figure 1 shows photoreflectance spectra from uncapped [1(a)] and capped [1(b)] samples recorded at 80 K paying attention to the island-related $E_1$ and $E_1 + \Delta_1$ transitions, which are associated with the corresponding critical points of the dots. Measurements on both kinds of samples were carried out under identical conditions in order to obtain comparable results. As can be observed, in uncapped samples the studied transitions are perfectly detectable even for nominal InSb thicknesses as small as 3.2 ML (some signal was barely detected in the 2.2 ML sample). Strikingly, in capped samples the studied transitions are not detectable by any means for nominal InSb thicknesses lower than 10 ML (also, no signal was detected in the 3.5 ML sample). The less InSb grown the smaller the oscillator strengths of both transitions (note the decrease of the signal-to-noise ratio), as expected. However, the striking disappearance of these transitions in capped samples cannot be related to the low content of InSb, as compared to the uncapped samples with similar amounts of island material. On the other hand, light absorption due to the cap layer is not significant because of its small thickness. Also, it can be observed that in uncapped samples the energies of the studied transitions do not depend on the InSb deposition, being the same as those corresponding to InSb bulk material (indicated by vertical dashed lines). However, both transitions, maintaining the same energies in the 15 ML capped sample, shift appreciably (≈30 meV) towards higher energies in the 10 ML capped sample.

Since they were all grown under identical conditions, the distinct behavior of uncapped and capped dots may be related to structural differences caused in the buried islands by the capping. In order to determine such differences, TEM was performed with a Philips CM-30 microscope operating at 300 kV. Uncapped dots [Fig. 2(a)] are relaxed by a misfit dislocation array placed at the dot-substrate interface. Most of such dislocations have a 90° Burgers vector and are separated by the mean distance (≈4.5 nm) that completely relaxes the InSb/InP system. This result is confirmed

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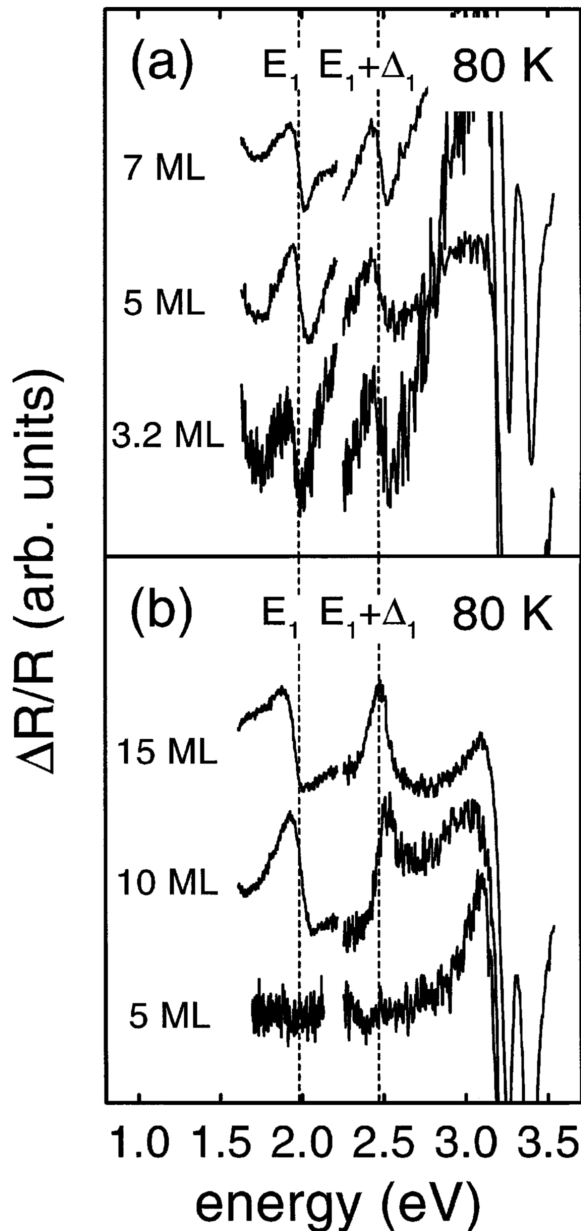


FIG. 1. Photoreflectance spectra taken at 80 K from (a) uncapped and (b) capped samples in which features associated with the island related as well as the InP related $E_1$ ($\approx$2.0 and 3.2 eV, respectively) and $E_1 + \Delta_1$ ($\approx$2.5 and 3.4 eV, respectively) transitions can be observed. Unlike uncapped samples, in capped samples the island related transitions shift from the energies corresponding to InSb bulk material (indicated by vertical dashed lines) towards higher energies as the InSb deposition decreases, disappearing for amounts of island material lower than 10 ML.

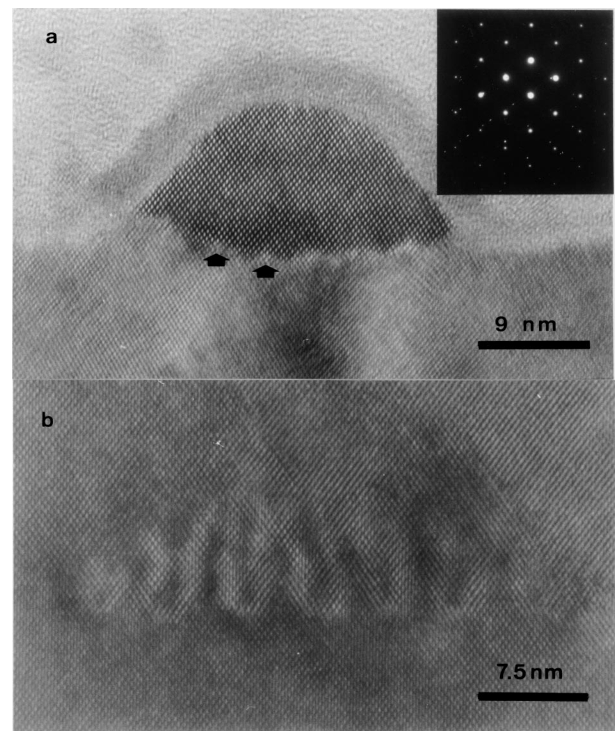


FIG. 2. High resolution cross-section TEM images of single dots. (a) Free-standing island completely relaxed by a misfit dislocation array, as mean separation between dislocations and spot splitting in selected area diffraction demonstrates. The arrows indicate two contiguous dislocations. (b) Buried island showing Moiré fringes due to relaxation.

by selected area diffraction [inset in Fig. 2(a)], which shows spot splitting due to the difference in the lattice parameters of dots and substrate. The ratio of the distances that separate main spots and splitted spots is approximately 10%, a value very close to the lattice mismatch existing between InSb and InP. Like uncapped dots, capped dots [Figs. 2(b), 3(a), and 3(b)] are relaxed by a misfit dislocation array placed at the dot-substrate interface. Certainly, several islands exhibit Moiré fringes separated by the mean distance ($\approx$4.5 nm) that corresponds to a complete degree of relaxation. Nevertheless, contrast at the boundaries of the islands related to strain can be clearly seen in the plan-view images. As the InSb deposition lowers, the strained fraction of the dots increases. Since the less InSb grown the smaller the islands, this strained fraction increases as the island size decreases.

The optical properties of QD structures are governed by both quantum confinement and strain effects. However, in our case, quantum confinement is not expected to be important due to the large dimensions of the islands as well as the large effective masses of the charge carriers at the $E_1$ and $E_1 + \Delta_1$ critical points [11]. Therefore, in order to explain the distinct behavior of uncapped and capped dots only strain has to be considered. According to the TEM results, Raman scattering measurements indicate that uncapped dots are completely relaxed, while in capped dots some residual strain exists, doubtless due to the presence of the cap layer. Figure 4 shows the island-related peak of Raman spectra from capped samples measured at 80 K in the backscattering configuration $z(x,y)\bar{z}$, where $x$, $y$, and $z$ denote the [100], [010], and [001]

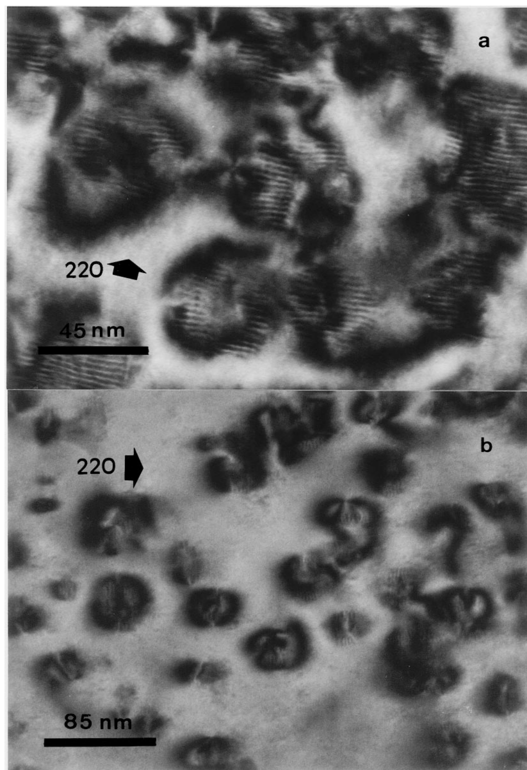


FIG. 3. Plan-view TEM images from the (a) 10 ML and (b) 3.5 ML capped samples. Moiré fringes inside the dots due to relaxation, and contrast at the boundaries of the dots because of residual strain, can be observed. Notice that the strained fraction increases as the island size diminishes.

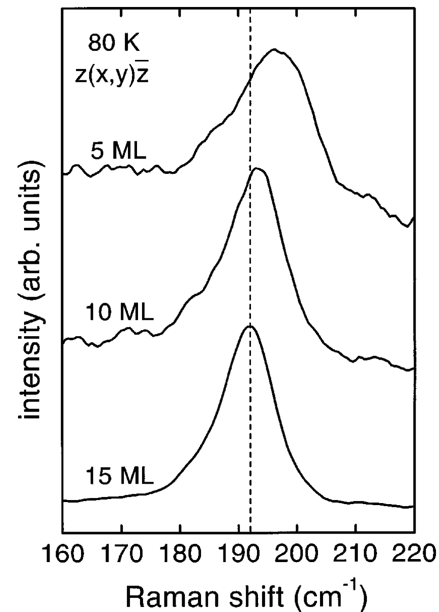


FIG. 4. Island related peak of Raman spectra measured at 80 K in the backscattering configuration $z(x,y)\bar{z}$ from capped samples. The shift from the energy corresponding to the LO(Γ) phonon of InSb bulk material (indicated by a vertical dashed line) towards higher energies as the InSb deposition decreases is attributed to augmentation of the strain produced inside the dots by the cap layer as the island size diminishes. In the 15 ML sample the dots are supposed to be completely relaxed.

directions, respectively. This peak is located, in the 15 ML sample, at the same frequency as that corresponding to the LO(Γ) phonon of InSb bulk material (indicated by a dashed vertical line), but shifts towards higher frequencies as the InSb deposition diminishes. Since quantum confinement is negligible in our case, such a shift must be due solely to an augmentation of the strain inside the dots by decreasing the island size [12]. In the 15 ML sample the dots are supposed to be completely relaxed.

Strain inside capped dots is not uniform and, therefore, difficult to calculate. However, a mean value can be estimated roughly from the blueshift of the island-related Raman peak with respect to that of InSb bulk material. Certainly, each fraction of the buried islands characterized by a given value of strain may contribute to the width of such a feature. Assuming that the capped dots are subjected only to a hydrostatic compression produced by the matrix and using the deformation potential for the LO(Γ) phonon of InSb bulk material given in Ref. [13], the values of the mean strain inside the islands ($\Delta a/a$) 0, $-3.7 \times 10^{-3}$, $-9.6 \times 10^{-3}$, and $-12.6 \times 10^{-3}$ are obtained for the 15, 10, 5, and 3.5 ML capped samples, respectively. Notice that, despite the presence of the cap layer, capped dots are quite relaxed.

The uncapped samples must have the band lineup of the unstrained InSb/InP system because they contain completely relaxed islands. However, in the capped samples, in which the mean strain inside the islands augments as the InSb deposition decreases, the band lineup must depend on this mean strain. Continuing with the above realized assumption, the valence (VBO) and conduction (CBO) band offsets at the $E_1$ critical point shown in Fig. 5 can be estimated for each case. The band alignment of the uncapped samples is obtained from the unstrained VBO at the Γ critical point [14] and the relative positions of the conduction band edges for InSb and InP [15]. The band alignment of the capped samples is obtained by supposing that under a hydrostatic compression, due to the large difference existing between the valence and conduction hydrostatic deformation potentials [16–18], the valence band top energy does not change, whereas the conduction band bottom energy increases according to the equation,

$$\Delta E_L^C(\Delta a/a) = 3\epsilon_{HL}\Delta a/a\,,$$

where $\epsilon_{HL}$ is the hydrostatic interband deformation potential [19]. The result is a band lineup of type-I with CBO's of about 50 and 15 meV for the 15 and 10 ML capped samples, respectively, and a band lineup of type-II that confines holes but not electrons with CBO's of

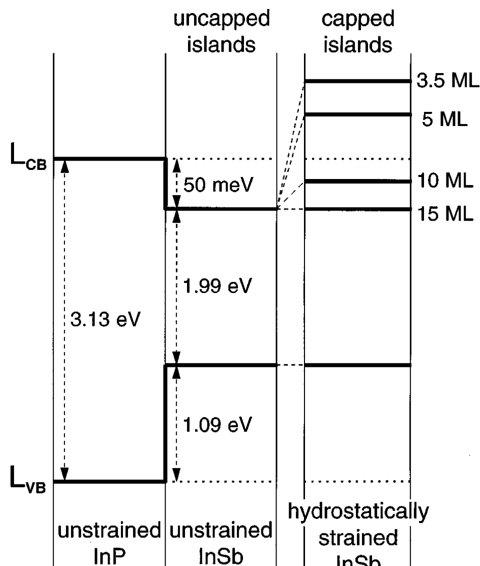


FIG. 5. Schematic diagram of the band alignment at the $E_1$ critical point proposed for islands of InSb grown on top of InP, as well as grown deeply buried in InP. In buried islands, observe the rise of the lower energy conduction electronic state of the dots with decreasing island size (i.e., augmenting the strain produced inside the dots by the cap layer) and the consequent change of the band lineup of the system from type-I to type-II.

approximately 45 and 75 meV for the 5 and 3.5 ML capped samples, respectively. This result reveals a change of band alignment in the capped samples from type-I to type-II as the mean strain inside the islands increases. In a band lineup of type-I, quantum confinement produces a strong overlap of the charge carrier wave functions and, therefore, appreciable oscillator strengths of optical transitions. However, in a band lineup of type-II, the overlap of such wave functions is rather weak and, consequently, optical transitions have oscillator strengths extremely small and cannot be detected by modulation spectroscopies in any way. Notice that the rise of the lower energy conduction electronic state with augmenting the mean strain inside the islands also accounts for the shift towards higher energies of the $E_1$ and $E_1 + \Delta_1$ transitions in the 10 ML capped sample. The surprising good agreement (in view of the simplicity of the theoretical model) existing between calculations and experimental results suggests that the main effect of the cap layer on the islands in self-assembled QD structures lies in increasing the hydrostatic component rather than the shear component of the strain, as suggested in previous works [5,6], but here found in incoherent islands.

In conclusion, strain-induced quenching of optical transitions has been found in capped self-assembled QD structures. Such a quenching is related to a change of the band alignment of the system from type-I to type-II due to the strain caused inside the islands by the cap layer.

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