Imaging the Wave-Function Amplitudes in Cleaved Semiconductor Quantum Boxes

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We have investigated the electronic structure of the conduction band states in InAs quantum boxes embedded in GaAs. Using cross-sectional scanning tunneling microscopy and spectroscopy, we report the direct observation of standing wave patterns in the boxes at room temperature. Electronic structure calculation of similar cleaved boxes allows the identification of the standing waves pattern as the probability density of the ground and first excited states. Their spatial distribution in the (001) plane is significantly affected by the strain relaxation due to the cleavage of the boxes.

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Semiconductor zero-dimensional (0D) quantum structures, or quantum boxes (QB), exhibit a three dimensional confinement with a δ-function-like electronic density of states. In the past years, self-assembled QB have shown very rich spectroscopic signatures [1]. As their optical properties depend on the wave functions of the electron and hole confined in the box, the knowledge of the shape, the extent, and the overlapping of the different wave functions is therefore of prime interest. So far, to glean details of the wave functions in such 0D structures, a growing number of theoretical works have been achieved. The electronic structure of semiconductor boxes with different sizes and facet orientations, embedded or not in an overlayer, have been calculated [2]. But the charge densities associated with the confined wave functions have not yet been resolved experimentally. Even though some experimental results, in electroluminescence and magnetophotoluminescence, have been obtained on the average spatial extent or symmetry of the hole or electron wave functions for an array of boxes [3,4], a detailed analysis of the wave functions in a single box is still missing.

In recent years, scanning tunneling microscopy (STM) and spectroscopy have provided unique means to characterize low dimensional structures. These techniques have allowed the determination of the energy level structure and the observation of the charge densities in artificial structures like the quantum corral [5] or on natural scattering centers like surface steps [6] and nanoscale islands [7]. Since the temperature imposes a limit of the spectroscopic resolution of the STM, most of the experiments were achieved at low temperatures. Indeed, at room temperature, the energy separation between electron levels must be in the range of a few kT, to resolve each of them individually. In the case of 0D semiconductor nanostructures, this requirement is fulfilled.

Such nanostructures can be formed by the controlled growth of InAs on a GaAs substrate in the Stransky-Krastanow growth mode. Recently, cross-sectional STM studies of InAs QB buried in GaAs have been achieved on the (110) face of cleaved samples to determine the shape and size of individual boxes at an atomic resolution [8]. Here, we investigate the conduction band (CB) states of InAs quantum boxes embedded in GaAs with the spectroscopic ability of the STM. We observe at room temperature, for the first time, standing wave patterns in the InAs boxes associated with the lowest CB states. For comparison, within the single band effective mass approximation, we calculate the electronic structure of the box according to its shape given by the STM image. This calculation enables us to determine unambiguously the spatial distribution of the states in the (110) plane of a truncated box.

The InAs quantum boxes were grown by molecular beam epitaxy on a (001) oriented GaAs substrate, with a residual p-type concentration. The active part of the samples consists of 15 arrays of InAs boxes separated by 15 nm GaAs barriers. The whole structure is covered by a 140 nm GaAs overlayer. In order to build each box array, 2.3 monolayers of InAs were deposited on the GaAs layer within 20 s at a temperature of 520 °C. They were immediately buried with GaAs. Samples cut from the wafer were cleaved *in situ* at a pressure below 5×10^{-11} Torr. Polycrystalline tungsten tips were prepared by electrochemical etching. The tips were then cleaned by heating and sharpened by a self-sputtering process in UHV. Topographic STM images were acquired with a 120 pA current and positive sample biases.

In Fig. 1, we display a cross-sectional topographic STM image of a stack of self-aligned QB along the [001] growth direction. The QB appear bright and the GaAs layers dark. The four boxes are lying on bright layers which correspond to the wetting layers. We may expect the boxes that show the largest sizes and the highest contrast to be cleaved near the dot center, leaving one-half of the boxes underneath the cleavage plane. We will now focus on such

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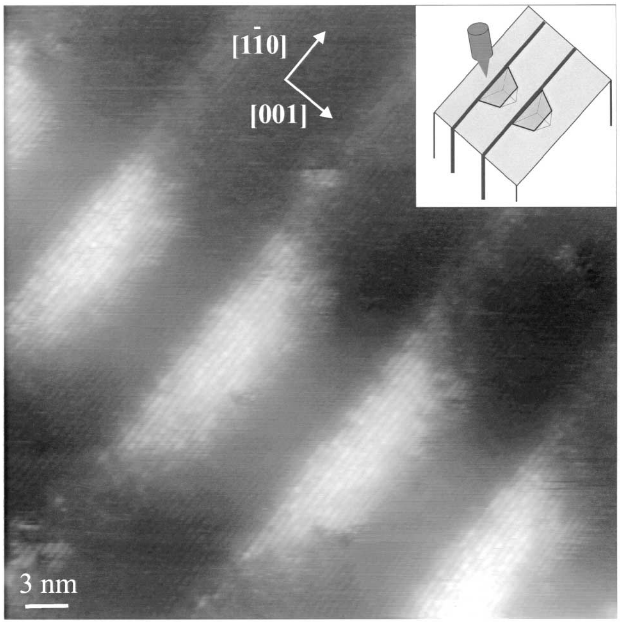

FIG. 1. STM image of the (110) face of an InAs box-stack layer in GaAs. The image was acquired at a sample bias of 1.86 V. Inset: Schematic diagram of the cleave.

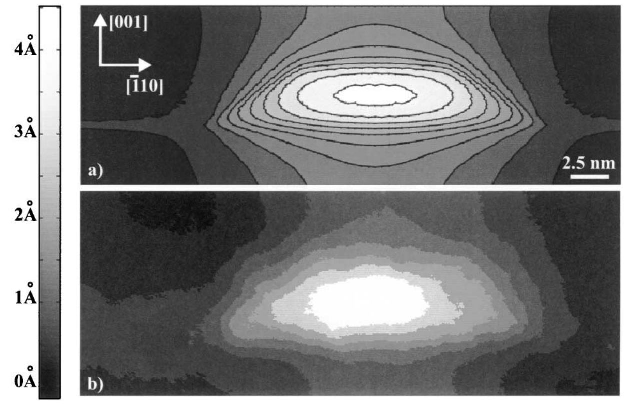

FIG. 2. Comparison of the height variation between (a) a simulated topographical image and (b) a STM image of the cleaved InAs box located in the center of Fig. 1. A low pass filter was used to remove the atomic corrugation from (b). For clarity, a contour line plot is displayed.

boxes, where the confined electron states can easily be resolved. In general, these boxes have a length around 20 nm and their height ranges from 4 to 5.6 nm.

As the sensitivity of the spectroscopy measurements depends strongly on the tip-sample separation, we first focus on the contrast mechanism of topographic STM images. To estimate the relative contributions of the electronic effects and the strain relaxation on the contrast, we have performed computations on cleaved InAs boxes embedded in GaAs. A truncated pyramidlike InAs box, with a 20 nm $[100] \times [010]$ square base and $\{110\}$ faces, was considered in the calculation. The truncated pyramid lies on a 0.4 nm thick wetting layer. The calculated box was cleaved perpendicular to the [110] direction, along the main diagonal of the square base. Strain relaxation at the cleaved edge was calculated with a finite difference method within continuum elasticity theory, following the work of Grundmann *et al.* [9].

Figure 2(a) shows the calculated structural height variation at the cleavage surface due to the strain relaxation. For comparison, a contour line plot obtained from the central dot on the STM image in Fig. 1 is plotted in Fig. 2(b). Both the computation and the experimental observation give a height variation of 4 Å. At voltages higher than +1.86 V, the strain relaxation is thus the major source for the contrast between the box and the surrounding layer. This result is similar to the one found for InGaAsP heterostructures imaged at sufficiently large positive sample bias [10].

To investigate the electronic structure of a box, tunneling current voltage curves were acquired simultaneously with the topographic image above individual quantum boxes. At every point of the image, obtained with a sample voltage of +2.15 V, the feedback loop was switched off to measure I-V curves. During an I-V measurement, the vertical position of the tip is held stationary. A current image at a given voltage is then drawn by plotting in each point of the image the value of the current obtained at this point during the I-V measurement. The results are displayed in Fig. 3. Figure 3(a) is the topographic image of a QB. This particular box has a base length of 20 nm and a height of 4 nm. Figure 3(b) shows a typical series of tunneling current voltage curves taken at different locations above the QB. Depending on the positions where the $I(V)$ curves are taken, a significant voltage offset can be seen. The current images are displayed in Figs. 3(c) and 3(d). They were obtained for an applied voltage of +0.69 and +0.82 V, respectively. In these images, regions of high current are bright and regions of low current are dark. While the Fig. 3(a) outlines the box contour, the current image in Fig. 3(c) shows a standing wave pattern in the center of the box. The intensity of this feature varies with voltage: It becomes clearly visible at a voltage of +0.63 V, and its intensity increases up to +0.74 V. At a voltage of +0.74 V, the standing wave pattern suddenly changes. Two new features are now apparent surrounding the central feature in the $[1\bar{1}0]$ direction, as shown in Fig. 3(d). Their intensity increases with voltage up to +0.9 V. For sample voltages larger than +0.9 V, the box becomes brighter and brighter with no other distinct feature visible in the box. For such large voltages, we may expect to tunnel into the empty states of the wetting layer and the GaAs conduction band.

We have previously shown that the tip-sample separation is insensitive to the electronic effects for a voltage of +2.15 V. Therefore, the variation of the tip-sample separation over the quantum box can be considered significantly small and cannot explain the measured 2 orders of magnitude in the current images.

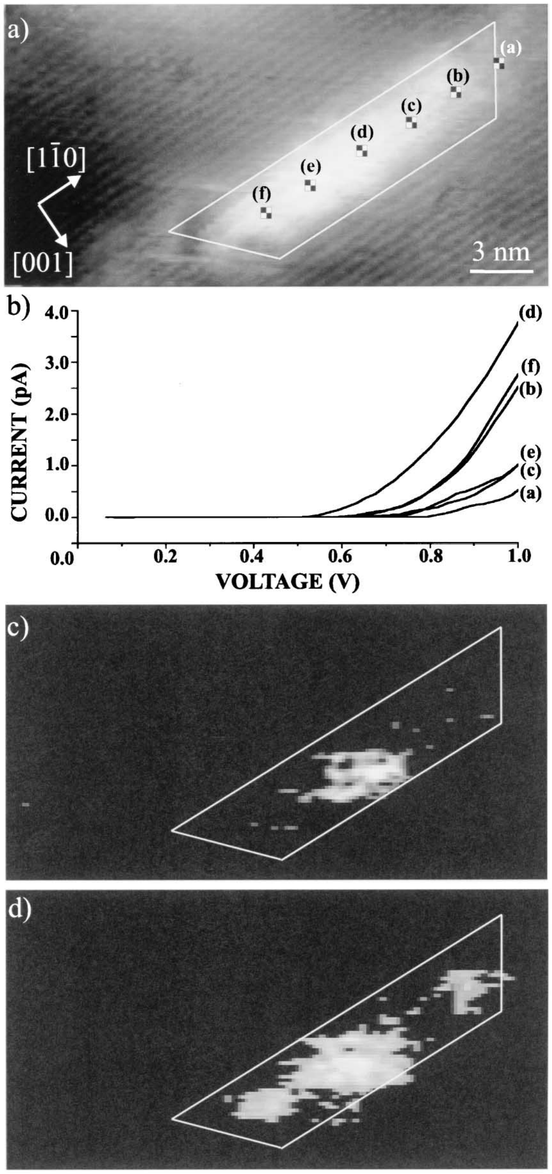

FIG. 3. Simultaneously acquired topographic and current images of an InAs QB: (a) STM topograph with +2.15 V applied to the sample, (b) tunneling current voltage curves for different locations in and near the box as reported in (a), current images at a sample bias of (c) +0.69 V and (d) +0.82 V. The boundaries of the box, determined from Fig. 2(a), are outlined in each image. The grey-scale ranges from 0.01 to 0.8 pA and 1.5 pA for (c) and (d), respectively.

The wave patterns in Figs. 3(c) and 3(d) are obtained without the interference of mechanical contributions and so reflect the spatial distribution of the lowest electron states confined in the box. As only the states lying between the Fermi levels of the sample and of the tip contribute to the tunneling process, the standing wave pattern at a voltage of +0.69 V corresponds therefore to the probability density of the electron ground state in the box, whereas at a voltage of +0.82 V, the standing wave pattern can be regarded as a combination of the probability densities of the electron ground and first excited states [11].

To confirm the origin of the observed standing wave patterns, we performed electronic structure calculations on a cleaved InAs box embedded in GaAs. The box has a similar shape to the one considered in the previous part. The conduction band states of the box were computed with a single band spatially varying effective mass approximation. A finite element method with piecewise-linear interpolation elements was used. The CB strain potential and effective mass tensor were calculated for each cell with an eight band $\mathbf{k} \cdot \mathbf{p}$ model [12,13]. The mean effective mass in the InAs box was found to be $0.039 m_0$, in agreement with other calculations [13,14]. The piezoelectric potential was neglected, its effects being less important [9]. Energies are measured with respect to the conduction band minimum of bulk GaAs. All parameters are taken from Ref. [14]. A 4.07 eV high barrier was assumed between bulk GaAs and vacuum.

The squared ground state $|000\rangle$ [15] and first excited state $|010\rangle$ are plotted in Fig. 4. Their energy is, respectively, $E = -307$ and -190 meV. The ground state is "s-like" (no nodes) and the first excited state is "p_y-like" with one nodal plane. Both states are pushed towards the surface by the strain potential which abruptly decreases at the cleaved edge [16]. Indeed, because of the strain relaxation, a "triangular" well appears along the [110] direction, whose depth with respect to the innermost parts of the box is about 500 meV. This triangular well increases the local density of states near the cleavage surface that is scanned by the STM tip.

Comparing the simulations with the experiments, we find that Fig. 4(a) matches Fig. 3(c). Similarly, a linear combination of Figs. 4(a) and 4(b) gives the probability amplitude distribution of Fig. 3(d). Therefore, the calculated density for the ground state and first excited state appears consistent with the STM current images.

As the geometry of the box remains to a large extent unknown, other geometries (position of the cleaved surface, shape of the base, ...) were investigated. In composition variations were also considered (In rich core with flanks down to $Ga_{0.7}In_{0.3}As$) [17]. Although the eigenenergies may depend on the detailed geometry and alloy composition of the box, we always find an s-like ground state separated from the next p_y-like bound state by about 100 meV, which is consistent with our STM observations. In the case considered here, two other bound states exist with respect to the wetting layer ($E = -31$ meV): the $|100\rangle$ ($E = -131$ meV) and $|020\rangle$ ($E = -60$ meV) bound states. However, the $|100\rangle$ state may be hardly visible, the lobe pointing towards the surface being almost nonexistent.

In conclusion, we have used the STM to study the probability amplitude distribution of conduction states in cleaved InAs/GaAs QB. Tunneling spectroscopy performed on a low dimensional semiconductor structure provides evidence for size quantization at room temperature, due to the large energy splitting. We have observed two bound

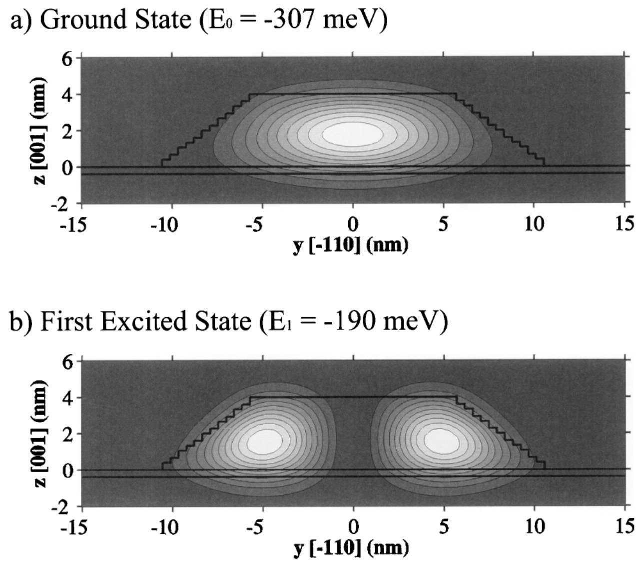

FIG. 4. Isosurface plots of the charge densities of the electron ground and first excited states just below the cleavage face of a cleaved InAs box.

states: the ground state shows an s-like envelope function, whereas a p_y-like envelope function is found for the first excited state. The shape of these envelopes is consistent with our electronic structure calculation of cleaved InAs QB. Our experiment demonstrates that the spatial variation of the probability densities of the electron states of a semiconductor QB can be resolved. It should not be restricted to electron states but could open up the possibility of obtaining the probability densities of the hole states. Such a direct observation of the wave functions could bring strong evidence for the presence of a permanent dipole in InAs/GaAs QB [18].

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