Photoluminescence polarization of single InP quantum dots

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The linear polarization dependence of photoluminescence emission was measured on single self-assembled InP quantum dots. The dots were obtained by Stranski-Krastanow growth on $Ga_{0.5}In_{0.5}P$. The highest-intensity emission occurred for light polarized parallel to the elongation of the dots in agreement with theoretical calculations. The excitation intensity was varied to obtain the polarization dependence of higher (state-filled) levels.

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Measurements involving large numbers of quantum dots are subject to ensemble averaging, and hence important information about the dots is lost. By growing samples with low quantum dot densities where the average spacing is larger than the optical resolution (around 1 μm), individual dots can be investigated using micro photoluminescence (μPL). Such single-dot studies have revealed sharp lines[1–6] and few-particle effects,[6] as well as unexpected behavior, such as emission intermittency[7–10] and phonon-assisted absorption.[11] It has even been possible to measure the emission lifetimes of single InP quantum dots.[2,12]

While PL provides information about electronic energy levels, the polarization of the emitted light reveals additional information about the electronic states. In particular, the polarization depends on the symmetry of the wave function, and thus provides indirect information about the geometric symmetries of the dot. Polarized PL measurements have been reported for individual $GaAs/Al_xGa_{1-x}As$ quantum dots consisting of monolayer thickness fluctuations in a quantum well.[13,14] Also, polarized PL measurements on InAs/GaAs dots have been reported for ensembles of dots[15,16] as well as for single dots.[17] Comparison with calculations show the observed polarization anisotropy to be consistent with the dots being elongated along the $[1\bar{1}0]$ direction.[15] InAs dots grown on (311) surfaces have been reported with arrowheadlike shapes and corresponding macro-PL polarization.[18] In contrast to InAs/GaAs dots, the shape of metal-organic vapor phase epitaxy (MOVPE) grown $InP/Ga_xIn_{1-x}P$ dots is well characterized.[19] Polarized PL measurements are needed, however, to probe the structure of the valence band states, which are expected to be localized near the bottom of the dot, and in the barrier above it.[20]

In this report, we give measurements and calculations of the luminescence polarization of single InP quantum dots. We find that the emission is mainly polarized along the elongation axis of the quantum dot. The magnitude of the polarization anisotropy is similar to calculated values. We have looked for contribution from ordering in the $Ga_xIn_{1-x}P$ to the polarization anisotropy of the dots but find this to be negligible.

The sample was grown by MOVPE at 580 °C, below the optimal temperature for ordering (around 650 °C).[21] First, a 300 nm thick layer of $Ga_xIn_{1-x}P$ was deposited. The quantum dots (QD's) were obtained by depositing 2.4 monolayers of InP, a growth interrupt of 12 s followed, the sample was then capped by 300 nm of $Ga_xIn_{1-x}P$.

Atomic force microscopy (AFM) imaging of similar uncapped InP quantum dots shows an elongation in the [110] direction[22] and is confirmed on capped samples by transmission electron microscopy.[19] AFM shows that about 90% of the QD's are elongated in the [110] direction. The fully developed dots are typically 15 nm high and 60×40 nm at the base. The growth method and conditions affect the orientation of the InP quantum dots,[23,24] with CBE grown dots showing elongation in the $[1\bar{1}0]$ direction and MOVPE-grown dots showing elongation in the [110] direction.

The sample was placed in a liquid helium cryostat, and the luminescence was collected using a microscope objective; the excitation source was a frequency-doubled Nd:yttrium aluminum garnet (YAG) laser emitting at 532 nm and was focused on the sample to a diameter of about 100 μm. All measurements were obtained at a temperature of 7 K. For polarization-dependent PL, a birefringent calcite crystal was placed between the microscope and a monochromator, resulting in a sufficient displacement between the two emerging polarized beams to allow easy simultaneous measurement of the emission intensity in the [110] and $[1\bar{1}0]$ directions, corresponding to the short and long axis of the dots. The spectral resolution of the system was about 0.1 meV. Conventional polarizers were used to study the angular dependence. The spectra were detected with a cooled charge-coupled device (CCD) camera. The excitation power density was of the order of 3 $W\,cm^2$ for below state-filling experiments, yielding a typical integration time of 120 s. The system response was carefully calibrated using an unpolarized light source in the cryostat. In addition, the sample was rotated 90° and the experiment was repeated on the same dots. It was therefore confirmed that the polarization anisotropy was not dependent on the sample orientation in the cryostat.

In Fig. 1, we present emission spectra of single dots for different polarization directions, obtained under low excitation power density. We note that even under low excitation the dots emit more than one single line, as previously reported.[2,12] The separation between the lines is in agreement with the expected separation of the electron states but much

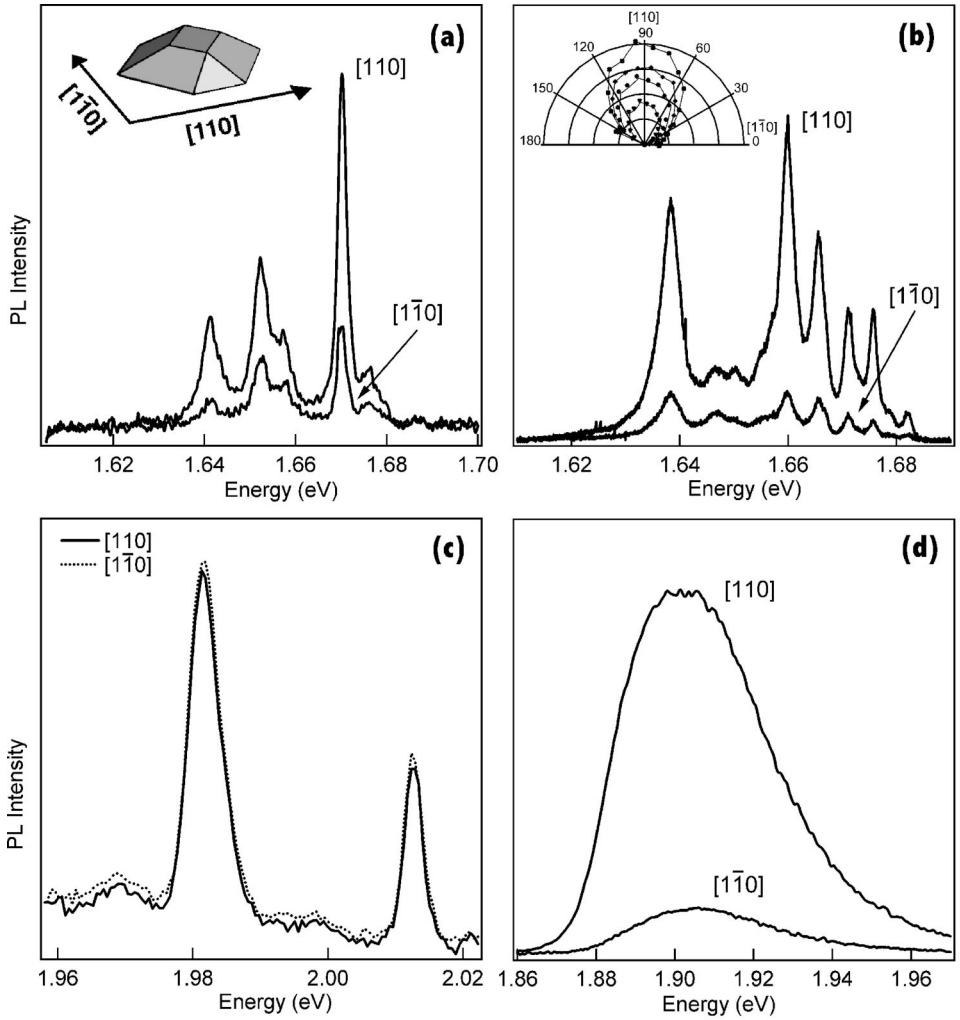

FIG. 1. (a) Single-dot polarized photoluminescence spectra along the [110] and $[1\bar{1}0]$ direction. The inset is a sketch of a typical fully grown InP quantum dot elongated in the [110] direction. (b) Another single-dot spectra taken in a different region of the same sample. The inset shows the polarization of different lines. (c) $Ga_xIn_{1-x}P$ and wetting layer PL from the region where (a) was measured. (d) $Ga_xIn_{1-x}P$ PL from the region where (b) was measured.

larger than the expected separation of hole states.[20] We thus attribute the lines to transitions involving different electron states. The emission is polarized along the [110] direction. We note, however, that a few dots did not follow this pattern, in agreement with the AFM results, which shows that about 10% of the dots have irregular shapes. The polarization of the luminescence from the $Ga_xIn_{1-x}P$ barrier was found to be strongly dependent on the location on the sample [Figs. 1(c) and 1(d)], indicating strong local fluctuation in the $Ga_xIn_{1-x}P$ ordering. It is commonly observed that disordered $Ga_xIn_{1-x}P$ has a narrower luminescence linewidth than ordered $Ga_xIn_{1-x}P$ and that the emission energy is lower for ordered $Ga_xIn_{1-x}P$.[21] This is usually explained by domain formation in the ordered phase, giving fluctuations in the transition energy.[21] These strong ordering fluctuations enabled the study of similar single quantum dots in different environments [Figs. 1(a) and 1(b)]. The $Ga_xIn_{1-x}P$ polarization anisotropy was not found to be correlated with the polarization anisotropy of the quantum dots.

Figure 2 shows single-dot spectra obtained under low and high excitation intensity, with clearly visible state-filling effects. We observe that the state-filled levels have a somewhat lower degree of polarization anisotropy than the lower states.

We have performed calculations of the dipole matrix element between the lowest hole state and different electron levels for different polarization directions of the emitted light. The calculations shown in Fig. 2(b) were made with a six-band $\mathbf{k}\cdot\mathbf{p}$ theory taking strain, piezoelectric polarization, and the exact dot geometry into account.[20] The calculations show the existence of two types of hole states, denoted *A* states and *B* states.[20] The *A* states are localized near the base of the pyramid while the *B* states are localized near the top of the pyramid (and have a higher energy than the *A* states). The electrons are localized centrally in the dots. As can be seen in Fig. 2(b) the polarization of transitions involving *A* states is mainly along the [110] direction, in agreement with the experiment, while the *B* states are polarized in the orthogonal direction. Thus we conclude that *A* states are involved in the observed transitions.

In Fig. 3, we show the macrophotoluminescence polarization for a large number of quantum dots measured on the same sample. The bulk GaAs signal (not shown) is unpolarized while the QD emission along the $[1\bar{1}0]$ polarization direction has a lower intensity (4 times weaker) than in the [110] direction. This polarization dependence is in agreement with the single-dot results obtained on the same sample and shown in Fig. 1.

Calculations show that electrons are confined inside the QD, while the lowest-energy hole state is located at the base

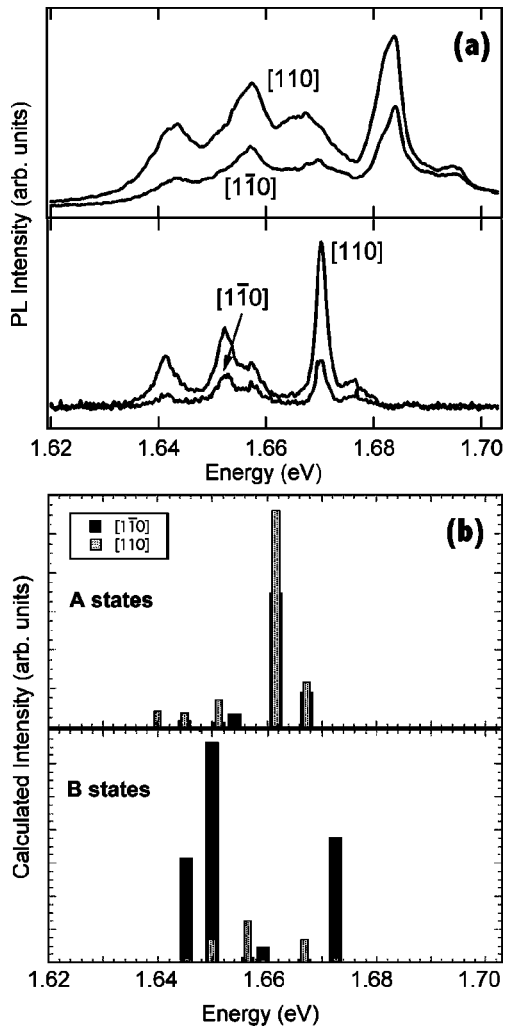

FIG. 2. (a) Single-dot spectra taken under different excitation intensities. (b) Calculations: *A* states show a dominant polarization along the $[1\bar{1}0]$ direction, while the *B* states show a dominant polarization along the [110] direction. The hole state involved in the transition is thus attributed to *A* states.

and is elongated in the [110] direction (*A* states).[20] It can therefore be expected that luminescence will be polarized in the [110] direction. In the case of InP dots, the polarization can be attributed to the elongation of the hole wave functions. Since the dielectric constants are nearly the same for the dot and the barrier material, a depolarizing field induced by charges at the interfaces cannot be the cause of the polarization.[25] A possible origin of the polarization anisotropy could be the barrier material. The $Ga_xIn_{1-x}P$ alloy is often ordered in a CuPt structure.[26] Experiments by Sugisaki *et al.*[23] have indeed shown a strong correlation between dot polarization and $Ga_xIn_{1-x}P$ ordering. In contrast, we did not observe any correlation between the ordering of the $Ga_xIn_{1-x}P$ and the polarization anisotropy of the dots (Fig. 1). The experiments are not directly comparable since the growth technique used by Sugisaki *et al.* was CBE, which produces differently shaped dots. The shapes of CBE grown dots may be governed by the ordering characteristics of the $Ga_xIn_{1-x}P$ barrier layer. One difficulty in experiments on quantum dots is the lack of knowledge about the degree of intermixing in the quantum dots, which may be of importance. The dots measured by Sugisaki *et al.*[23] had a lesser degree of shape anisotropy than our dots and had a height of 5 nm in contrast to our dots, which are 15 nm in height. From this comparison we draw the conclusion that if both the shape anisotropy and the sizes are small, polarization anisotropy of the photoluminescence may be induced by ordering in the matrix but not otherwise.

The geometrical anisotropy of self-assembled quantum dots is thus reflected in the photoluminescence and could prove useful for optimization of lasers with quantum dots incorporated as the active material.[27,28] Six-band $\mathbf{k}\cdot\mathbf{p}$ calculations are in agreement with the measurements and reveal that the holes are confined in the quantum dots.

In summary, we have measured the photoluminescence polarization on single InP quantum dots. The polarization is attributed to geometrical effects, related to the elongation of the dots, in agreement with calculations.

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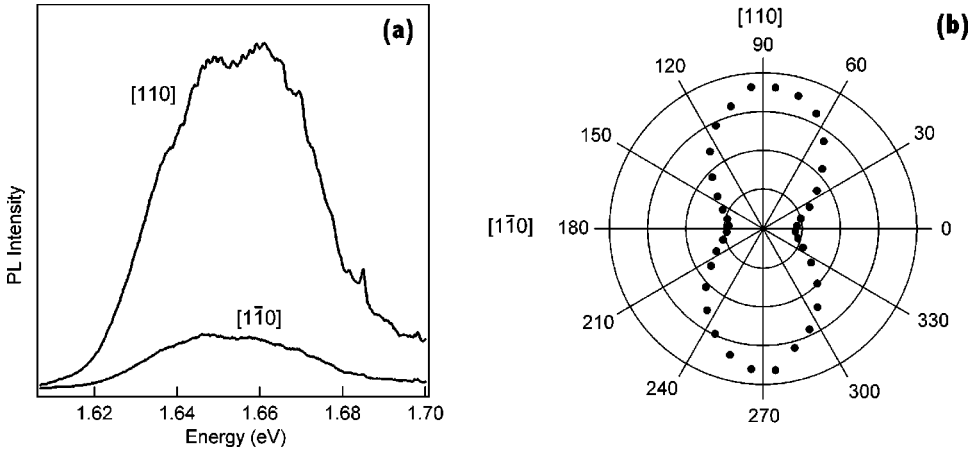

FIG. 3. (a) Polarized PL from a large number of dots with polarization along the $[1\bar{1}0]$ and [110] directions. (b) Polar plot of the polarized PL peak intensity as a function of the polarizer angle.

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