Piezoelectric effect in elongated (In,Ga)As islands on GaAs(100)

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The piezoelectric (PZ) effect is demonstrated for the elongated three-dimensional (In,Ga)As islands grown on a GaAs (100) substrate. The photoluminescence (PL) spectrum is studied as a function of excitation intensity. With increasing excitation intensity, a blue shift and a linewidth reduction of the PL peak from the (In,Ga)As islands are observed. The observed phenomena are attributed to the screening of the internal strain-induced PZ field in the (In,Ga)As islands.

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Since Smith first predicted a large piezoelectric (PZ) effect in strained III-V or II-VI semiconductor heterostructures, investigations have focused on the study of the PZ effect in semiconductors. One reason for this interest is that the strain-induced PZ field can modify the band structure of semiconductors, and therefore, offers another design parameter for fabricating novel optical and electronic devices. For pseudomorphic heterolayers, the PZ polarization is determined by the symmetry of the lattice of the substrate and the orientation of the surface. For example, for zincblende III-V semiconductor materials, the PZ polarization is along the growth direction for pseudomorphic layers grown on the (111)-oriented substrate, while it vanishes for those grown on the (100)-oriented substrate. For other non-(100) substrate orientations, the PZ polarization and PZ field usually can have both vertical (growth direction) and lateral (perpendicular to growth direction) components. This is probably the reason why the overwhelming majority of previous investigations of the PZ effect in semiconductors were performed on structures grown on the (111)-oriented surface and on high index substrates in general. However, there have been some recent efforts in realizing the PZ effect on the (100)-oriented substrate for applications. For example, one approach to generate the PZ effect on the (100)-oriented substrate is to pattern the as-grown structure along the [01̅1] direction while another is to apply surface acoustic waves. In this paper, we discuss a different approach based on the fabrication of three-dimensional (3D) structures, such as, quantum dots and quantum wires.

Quantum dot and wire structures have raised considerable interests because of their unique characteristics for optoelectronic and electronic device applications. Self-organized 3D islands grown on the Stranski-Krastanov (SK) growth mode have proved to be a successful approach to fabricate quantum dot and wire structures. The shape of these SK islands can be controlled by changing the growth conditions making it possible to directly engineer quantum dot and quantum wire structures. For example, long and uniform (In,Ga)As/GaAs quantum wires based on elongated islands have been fabricated using a superlattice (SL) growth scheme plus an annealing process. Recently, the PZ effect has been observed in self-organized quantum dots grown on different high index surfaces. However, these same studies also demonstrate that self-organized quantum dot structures grown on the GaAs (100) substrate do not show the PZ effect. In this work, we demonstrate that the PZ effect is in fact observed in elongated 3D (In,Ga)As islands grown on a GaAs (100) substrate. The evidence for this observation is based on characterization of a sample with elongated islands by excitation intensity dependent photoluminescence (PL) spectroscopy. Specifically, a blue shift and a linewidth reduction of the PL peak from the (In,Ga)As islands are observed to take place with increasing optical excitation intensity. We attribute both the shift and the linewidth reduction to the screening of the internal PZ field in the elongated islands. As a comparison, a similar but more symmetrical shaped quantum dot structure was investigated in the same way and a band filling effect rather than the PZ effect was observed.

A typical sample was grown on a semi-insulating GaAs (100) substrate by molecular beam epitaxy (MBE). After the sample was introduced into the MBE growth chamber, the native oxide was desorped at 580°C followed by overheating the substrate to 600°C. After keeping the substrate at 600°C in As4 atmosphere for 10 min, the sample was cooled down to 580°C and a 150-nm-thick GaAs buffer layer was deposited. The substrate was then cooled down to 540°C and a 15-period (In,Ga)As/GaAs superlattice was grown. Each time, immediately after the deposition of the (In,Ga)As layer, three monolayers of GaAs was grown without interruption to suppress In segregation and 20.3-nm GaAs was then added with growth interruption of 10 sec. Finally, a top (In,Ga)As layer was grown in order to characterize the surface morphology. The whole growth process was monitored by in situ reflection high-energy electron diffraction (RHEED). The RHEED pattern became spotty after the growth of (In,Ga)As layer indicating the appearance of 3D islands. The structural characterization was performed ex situ by x-ray diffraction (XRD) and atomic force microscopy (AFM).

Figure 1(a) shows the ω-2θ scan of a double crystal XRD around the GaAs (400) reflection measured with an open detector. Eight satellites appear within the measurement range indicating the good structural quality of the sample. The SL period of the sample was determined to be 23.7 nm from the spacing between the satellites. Figure 1(b) shows the simulation by the XRD dynamic theory assuming a coherent growth of the (In,Ga)As on GaAs. Good agreement between the experimental and the simulated curves is observed. The obtained thicknesses of (In,Ga)As and GaAs layers are 2.5 and 21.2 nm, respectively, and the In composition 0.28, which is in agreement with the values from RHEED.
oscillation measurements. Since the growth time of GaAs and \( \text{In,Ga}_x\text{As} \) layers are known, the growth rate of GaAs and InAs are then obtained to be 0.81 and 0.32 Å sec\(^{-1}\), respectively.

Figure 2 shows the AFM image of the sample measured in air in the tapping mode, revealing elongated islands along the \([0\bar{1}1]\) direction. The islands are uniform, although elongated with a width of about 50 nm. The main difference of the growth condition between this sample and the quantum wire sample described in Ref. 13 is that, for the latter case, the substrate was heated to 580°C after the growth of the \( \text{In,Ga}_x\text{As} \) layers and the three-monolayer thick GaAs, while for the former case, the SL structure was continuously grown at 540°C. During the annealing process, Ostwald ripening has occurred making the islands much more elongated. Therefore, the islands of this sample are only slightly elongated as opposed to forming quantum wires as in Ref. 13. Due to the relatively thin GaAs spacer layer thickness, vertical correlation probably occurs in this sample enhancing the uniformity of the islands, as revealed in the transmission electron microscopic image in Ref. 13.

Figure 3 gives the PL spectra of the sample measured at 10 K. Two peaks are observed. The peak located at the lower energy is attributed to the \( \text{In,Ga}_x\text{As} \) islands while the broad peak centered around 1.50 eV is related to carbon impurity in the substrate and in the epitaxial GaAs layer. When changing the excitation intensity from 5 to 500 W/cm\(^2\), the peak from the \( \text{In,Ga}_x\text{As} \) islands blue shifts about 12 meV while the position of the higher energy side peak stays unchanged. We believe that this blue shift is caused by the screening of the internal PZ field by photogenerated carriers. A larger excitation intensity results in more photogenerated carriers, and the screening of the internal PZ field becomes more pronounced. As the optical excitation intensity is further increased from 500 to 5000 W/cm\(^2\), the peak blue shift becomes less pronounced as can be seen from Fig. 3. This indicates that the screening effect is saturating at the excitation intensity from 500 to 5000 W/cm\(^2\). Although a band filling effect can also account for an observed blue shift, another concurrent characteristic of a band filling effect is a corresponding broadening of the PL peak, as has been observed, for example, for an InAs/InP quantum wire structure. On the other hand, when an electric field is applied to a quantum confined structure, the linewidth of the PL peak will increase. Therefore, if the blue shift of the PL peak is due to the screening of the PZ field, the linewidth of the PL peak should decrease with increased excitation intensity and further screening. In order to obtain an accurate value of the peak position and the full width at half maximum (FWHM) of the PL data measured at low excitation intensities, we fit the two PL peaks with a Gaussian shape. As shown in the inset of Fig. 3(a)–3(c) correspond to the fitted PL spectra measured at excitation intensity of 150, 100, and 5 W/cm\(^2\), respectively. For each of them, the dashed line represents the two deconvoluted PL peaks; the dotted line represents the fitted curve and the black line is the measured original spectrum. The error of the fitted PL peak position is smaller than 5% of the FWHM of the corresponding PL spectrum. Therfore, the fitted PL peak position for the (a), (b), and (c) in the inset of Fig. 3 has a uncertainty smaller than 3.5, 1, and 1 meV, respectively.

The closed circles in Fig. 4 represent the FWHM of the PL peak versus the excitation intensity for our experiments. When the excitation intensity is raised from 5 to 150 W/cm\(^2\),
the FWHM significantly decreases from about 69 to 23 meV. The decrease in the FWHM also becomes saturated for a further increase of the excitation intensity. When taken together, the behavior of the observed blue shift and linewidth reduction support a screening explanation and is inconsistent with a band filling picture. In fact, at high excitation intensity, the PL FWHM of the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands is about 21 meV which is only a little bit larger than the best results for the self-organized quantum dots, demonstrating good uniformity of the islands.

Further evidence for a PZ field and a screening effect can be found by paying attention to the relative PL intensity change between the lower-energy peak and the higher-energy peak. If there were no PZ field in the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands, the excited carriers would try to populate the lower-energy peak first, as observed in an InAs/GaAs quantum dots structure. Therefore, by increasing the excitation intensity, the higher-energy peak would become stronger and stronger and correspondingly, the relative PL intensity ratio between the lower-energy peak and the higher-energy one should decrease. However, we observed an opposite tendency, as shown with closed triangles in Fig. 4. This can be explained by the influence of the PZ field on the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands. Since the PZ polarization charges are distributed at the interface of the heterostructure, the PZ field is, therefore, localized in the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ layers. While there is no PZ effect for the (100) wetting layer, a PZ field will exist in the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands. The strong PZ field separates the electrons and holes thus making the recombination efficiency of excitons much lower. When increasing the excitation intensity, the screening of the PZ field by the photogenerated carriers partially recovers the recombination efficiency and the relative PL intensity ratio between the lower-energy peak to the higher-energy one should decrease. Therefore, the behavior of the relative PL intensity is also consistent with the existence of the PZ effect.

As a comparison, an $\text{In}_{x}\text{Ga}_{1-x}\text{As}/\text{GaAs}$ quantum dot structure was investigated in the same way. For a fixed growth temperature, the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands with larger In mole fraction favor the isotropic shape. Therefore, the In composition of the quantum dot structure was adjusted to be 0.36 and the thickness of the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ and GaAs layers is 2 and 17 nm, respectively. The growth procedure of the quantum dot structure is the same as that of the sample with the elongated island shape. Figure 5 shows the AFM image of the sample revealing 3D $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands with isotropic shape. The isotropic islands are laterally aligned along the $[0\overline{1}1]$ direction although with a wiggle. Figure 6 shows the PL spectra of the quantum dots sample measured at different excitation intensity at 5 K. When the excitation intensity is increased from 15 to 450 W/cm$^2$, the PL peak due to the $\text{In}_{x}\text{Ga}_{1-x}\text{As}$ islands shifts from 1.287 to 1.317 eV with a change of the FWHM from 70 to 96 meV while the shape of the PL peak remains symmetric. A further increase of the excitation intensity from 1500 to 5000 W/cm$^2$ already makes the PL peak asymmetric. In fact, the asymmetric PL peak measured at the high excitation intensity can be well fitted by two separate peaks with a Gaussian shape. For example, when the excitation intensity is 5000 W/cm$^2$, the dotted line in Fig. 6 shows the fitted curve and the dashed lines depict the two deconvoluted peaks centered at 1.340 and 1.398 eV with the FWHM of about 120 and 70 meV, respectively. As an approximation, we treat the quantum dot structure as a quantum well struc-

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**FIG. 4.** The closed circles depict the FWHM of the PL peak at the lower-energy side versus excitation intensity. The closed triangles show the relative PL intensity ratio between the lower-energy side peak and the broad higher-energy side one. The lines connecting the circles and the triangles are guide to the eyes. $I_0 = 5000$ W/cm$^2$.

**FIG. 5.** AFM top view of the quantum dots. The black-to-white height contrast is 5 nm.

**FIG. 6.** Excitation intensity dependent PL spectra of the quantum dots measured at 5 K. The spectra are shifted vertically for clarity. The dotted line is the fitted curve of the PL spectrum measured at excitation intensity of 5000 W/cm$^2$ and the dashed lines show the two deconvoluted peaks.
ture and numerically solve the one-dimensional Schrödinger equation by using the parameters shown in Ref. 18 and assuming the conduction to valence-band offset ratio of 60:40. It is found that the energy separation of the interband transition between heavy hole and light hole is 53 meV. Therefore, the peak centered at 1.398 eV is likely due to the light hole. As discussed previously, the observed blue shift and FWHM broadening of the PL peak of the quantum dots with increasing excitation intensity are typical of a band filling effect. Nevertheless, in principle, we cannot rule out the PZ effect and can only conclude that the blue shift of the PL peak is dominated by the band filling effect. However, recent studies have shown that the PZ effect is absent from the quantum dots grown on GaAs (100) substrate, which is consistent with our observations.

Given the strong evidence for an observed PZ effect in the elongated islands, one can explore the cause of the effect on the (100) surface in zincblende materials since it is somewhat unexpected. For zincblende symmetry, the relationship between the PZ polarization, $P_z$, and the strain is given by $P_z = 2\varepsilon_{ij}E_{jk}$, with $i \neq j, k$, where $\varepsilon_{ij}$ is the PZ constant and $E_{jk}$ the strain tensor referred to the crystalline axes. It can be seen that only shear strain can induce a PZ polarization. For a quantum well structure on the (100) orientation, the PZ effect is not expected because the shear strain vanishes.

While, for elongated islands on the (100) orientation, shear deformation could be present at the edges of the islands causing the PZ effect. However, the shear strain is related to the direction of the elongated islands. If the islands are elongated along the [001] direction, the shear deformation vanishes, while, if the elongation of the islands is along the [011] direction, shear strain survives. This point has been confirmed in quantum well structures which are grown on the [100] substrate orientation, but mesaetched along the [011] direction. The elongated SK islands are along the [011] direction and the PZ effect is, in fact, expected. Following Ref. 8, the PZ polarization in this case is along the growth direction, i.e., the [100] direction. Therefore, the influence of the PZ field is similar to the quantum confined Stark effect. With respect to the crystalline axes, since $P_{[100]} = 2\varepsilon_{14}E_{[010][001]}$, it can be seen that it is the shear strain $\varepsilon_{[010][001]}$ which induces the PZ polarization. If assuming that the PZ field is completely screened by the photo-generated carriers at the high PL excitation intensity, we can then estimate the strain-induced PZ field. As a simplified model, we still treat the structure with elongated In$_{0.28}$Ga$_{0.72}$As islands as a quantum well structure and numerically calculate the Schrödinger equation. It is found that a vertical electric field of $8 \times 10^4$ V/cm can cause a redshift of about 12 meV for the $e_1$-$hh_1$ transition compared to the case without electric field. Therefore, the strain-induced PZ field $E_{[100]}$ is about $8 \times 10^4$ V/cm. The calculation also shows that the influence of electric field on the heavy hole wave function in fact dominates the shift. We then get the PZ polarization $P_{[100]} = \epsilon_0 \epsilon_r E_{[100]} = 9.6 \text{ C/cm}^2$ and correspondingly, $\epsilon_{[010][001]} = 3.8 \times 10^{-3}$, where $\epsilon_0$ and $\epsilon_r$ are the vacuum permeability and the relative dielectric constant, respectively. For the In$_{0.28}$Ga$_{0.72}$As/GaAs system, we use $\epsilon_r = 13.537$ and $\epsilon_{14} = 1278 \text{ C/cm}^2$. On the other hand, for the quantum dots on the (100) orientation, shear strain might not survive due to the isotropic shape of the islands. This could be the reason why the PZ effect is not observed for the self-organized quantum dots on the (100) orientation. However, an accurate understanding about this question needs a complete analysis by taking into account the concrete shape of the quantum dots.

In summary, we have demonstrated the piezoelectric effect for elongated Stranski-Krastanov islands grown on a GaAs (100) surface. The sample is characterized by double crystal x-ray diffraction, atomic force microscopy and excitation intensity dependent photoluminescence spectroscopy. When increasing the excitation intensity, a blue shift and a linewidth reduction of the PL signal from the elongated islands are observed. The phenomena are explained as due to the screening of the internal piezoelectric field in the elongated islands. To compare, a quantum dot structure is investigated in the same way and the band filling effect rather than piezoelectric effect is observed. Our results indicate that the piezoelectric effect is possible for the elongated nanostructures grown on GaAs(100) and can have promising applications in novel devices utilizing the piezoelectric effect.

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