

## Piezo-optical effects in GaAs with interspersed (211)-InAs lattice planes

M. Ilg, O. Brandt, A. Ruiz,\* and K. Ploog†

*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Germany*

(Received 21 January 1992)

We study piezoelectric field effects in the ultimate atomic limit. The insertion of piezoelectrically active (211)-InAs lattice planes in GaAs gives rise to optical phenomena not observed in analogous (100) structures. The InAs-related luminescence exhibits a pronounced blue shift with increasing excitation density. The nonlinear dependence of the corresponding luminescence efficiency is a manifestation of the internal piezoelectric field in the strained InAs sheets.

Currently, molecular-beam epitaxy (MBE) allows routine fabrication of III-V semiconductor heterostructures with abrupt interfaces and high structural quality.<sup>1</sup> It even is possible to exert control on the chemical composition down to the subnanometer regime.<sup>2,3</sup> However, most of this work was carried out on (100)-oriented substrates. The departure from the highly symmetric and therefore exceptional [100] direction is expected to give rise to novel phenomena. In particular, strong piezoelectric fields of the order of  $10^5$  V/cm are present as soon as the growth axis of strained-layer heterostructure is different from certain directions such as [100] and  $[\bar{1}\bar{1}0]$ .<sup>4-6</sup> The presence of strong internal electric fields modifies the electronic properties of strained-layer heterostructures.<sup>7</sup> It shifts the bound states in the quantum well to lower energies<sup>8</sup> analogous to the quantum-confined Stark effect (QCSE). The photoluminescence signal is therefore redshifted in comparison to a hypothetical structure otherwise identical but without the internal field. Photogenerated carriers screen this internal field and thus cause a blueshift of the luminescence signal with increasing excitation density. Matrix elements and oscillator strengths vary significantly with the strength of the field and thus also with the concentration of free carriers. For these reasons the existence of an internal piezoelectric field results in nonlinear optical properties of strained-layer heterostructures.<sup>6,9</sup>

The InAs/GaAs system is an excellent candidate to study these phenomena because the magnitude of the piezoelectric field is proportional to the strain in the layers. The huge lattice mismatch between InAs and GaAs is expected to cause strong internal fields. Predicted piezoelectric effects are largest along the [111] and [211] directions.<sup>5</sup> We opted for the [211] direction because it is less difficult to handle in MBE growth than the [111] direction. The purpose of this paper is twofold. First, we demonstrate that highly strained InAs quantum sheets consisting of isolated isoelectronically substituted planes that exhibit excellent structural quality can be grown along the [211] direction. Second, the study of the optical properties and comparison with (100) oriented samples reveals the novel optical and electronic properties of the (211)-oriented structures which can be understood as manifestation of an internal piezoelectric field present in the strained monolayer.

The structure under investigation is synthesized in a three-vacuum-chamber molecular-beam epitaxy system,

equipped with elemental solid sources providing atomic species of Ga, In, and tetrameric As<sub>4</sub>. A semi-insulating (211B) substrate is mounted on a molybdenum block by soldering with In. The growth process is monitored *in situ* by means of reflection high-energy electron diffraction (RHEED) using an electron gun operating at 20 keV under a glancing-angle incidence of 1°. X-ray diffraction is performed with a high-resolution double-crystal x-ray diffractometer in Bragg geometry. A rotating-anode 12-kW generator with a copper target ( $\lambda_{\text{Cu } K\alpha_1} = 0.1540562$  nm) is employed as the x-ray source, and an asymmetrically cut (100) Ge crystal serves as monochromator and collimator. The diffraction pattern was recorded in the vicinity of the symmetrical (422) reflection. For the photoluminescence experiments the sample is mounted in an optical flow-through cryostat at 6 K. As an excitation source the red line (647.6 nm) of a Kr<sup>+</sup> laser is employed. The excitation density is varied between 0.1 mW/cm<sup>2</sup> and 1 kW/cm<sup>2</sup> by means of calibrated neutral density filters. The emitted light is focused on the entrance slit of a 1-m single-grating monochromator and detected by a photomultiplier operating in the photon-counting mode.

For the MBE growth of the structure we adopted a heating-cooling cycle which has recently been shown to yield excellent results on (100) substrates.<sup>10</sup> For the use on (211) substrate, however, we have to modify the employed growth temperatures which are all raised by 40°C. The Ga and In fluxes used correspond to growth rates of 2.8 and 0.2 Å/s, respectively, as determined by means of RHEED intensity oscillations. The [As<sub>4</sub>]/[Ga] flux ratio is measured by arsenic induced RHEED oscillations and adjusted to a value of 1. First, a GaAs buffer of 1 μm thickness is grown at a temperature of 620°C. Then the substrate is cooled down at a rate of 40°C/min to a temperature of 460°C where 2 (211) monolayers (ML) of InAs are deposited in two steps. Each step is followed by an annealing period of 120 s. After the deposition of a thin intermediate GaAs layer of about 12 Å thickness the substrate is heated up to a temperature of 580°C. At this temperature a GaAs barrier of 200 Å is grown. This cooling-heating cycle allows for the evaporation of segregated In floating on top of the intermediate GaAs layer and thus results in abrupt interfaces.<sup>10</sup> This procedure is repeated for each of the ten periods of the InAs/GaAs superlattice.

The x-ray-diffraction pattern obtained from the sample

is shown in Fig. 1(a). Satellite peaks due to the superlattice structure can be seen up to the third order. *Pendellösung* fringes appear around the substrate and zeroth-order epitaxial peaks. We used the dynamical theory of x-ray diffraction to obtain the simulated diffraction pattern shown in Fig. 1(b). The simulation was carried out under the assumption of a barrier with a thickness of 217 Å and an InAs sheet with an In atom density of  $6 \times 10^{14} \text{ cm}^{-2}$ . This coverage corresponds to 1.2 (211) monolayers. It was also assumed that the strain state of the InAs layers coincides with the prediction by linear elasticity theory. The agreement between simulation and experiment shows the excellent structural quality of the sample. Deviations of the experimental from the calculated pattern in the vicinity of the main epitaxial reflection is due to inevitable diffuse scattering at point defects in the substrate. This is especially evident in this case of low In content in the sample.

In Fig. 2 we show the photoluminescence (PL) spectra from the sample under consideration obtained at different excitation densities. At the lowest excitation density three peaks at 1.4857, 1.4900, and 1.4930 eV are resolved in addition to the free exciton transition at 1.5150 eV. The last two peaks quickly saturate when the excitation density is increased slightly, revealing their extrinsic nature. Their spectral position identifies them as carbon-related ( $D^0$ ,  $C^0$ ) and ( $e, C^0$ ) transitions. The third peak, however, does not saturate and gains even more in intensity than the free exciton transition. Therefore this transition at 1.486 eV is clearly intrinsic in nature and has to be ascribed to the insertion of the InAs layers.

Two more features of these spectra deserve attention.

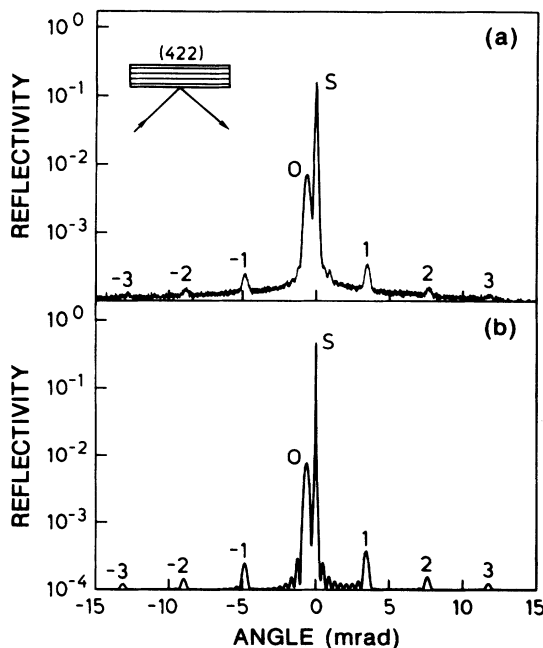


FIG. 1. (a) Experimental and (b) simulated x-ray-diffraction pattern in the vicinity of the symmetrical (422) reflection with Cu  $K\alpha_1$  radiation for a (211) MQW structure with ten periods and an InAs thickness of 1.2 ML.

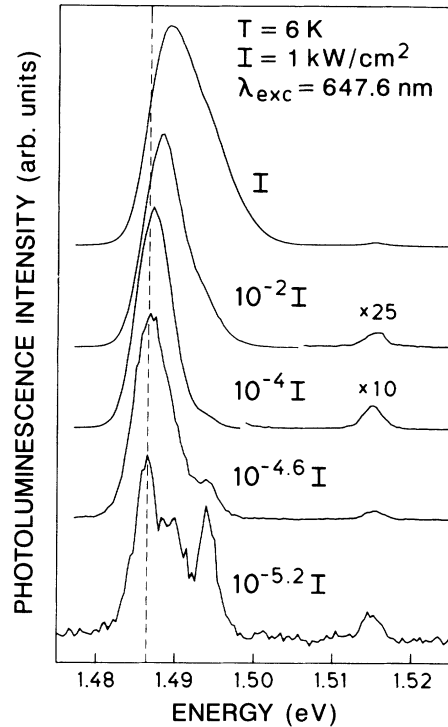


FIG. 2. PL spectra obtained at 6 K and different excitation densities for the sample in Fig. 1. The dashed vertical line emphasizes the blueshift with increasing excitation density.

First, there is a clear blueshift of the PL peak with increasing excitation density. At the lowest excitation density we observe the maximum at 1.4857 eV. This has to be compared to a peak position of 1.4884 eV at the highest excitation density which yields a shift of the PL maximum by 2.7 meV [Fig. 3(a)]. Second, the increase of the excitation density not only causes this blueshift but also results in a strong asymmetric broadening of the peak. The full width at half maximum (FWHM) increases from 3 meV at the lowest to about 10 meV at the highest excitation density. These two observations are in contrast to experiments carried out on the reference structure grown along the (100) direction, where neither a blueshift nor a line broadening can be seen.<sup>11</sup>

The data of Fig. 2 also show that the peak due to the InAs layers gains more in intensity than the free exciton transition when the excitation density is increased. This nonlinear dependence of the luminescence efficiency on the excitation density is depicted in Fig. 3(b). The open circles represent the results for the (211) sample. For excitation densities between 0.01 and 1 W/cm<sup>2</sup> there is a nonlinear behavior which then switches to a linear dependence for densities above 1 W/cm<sup>2</sup>. For comparison we show results of a measurement (solid circles) carried out on a (100) structure with an InAs thickness of 0.8 (100) monolayer. There we clearly observe a linear dependence of the luminescence efficiency over the whole measurement range which spans 6 orders of magnitude. The (211) sample, however, shows a completely different qualitative behavior. Above an excitation density of 1 W/cm<sup>2</sup> it mimics the curve of the (100) sample and also exhibits a

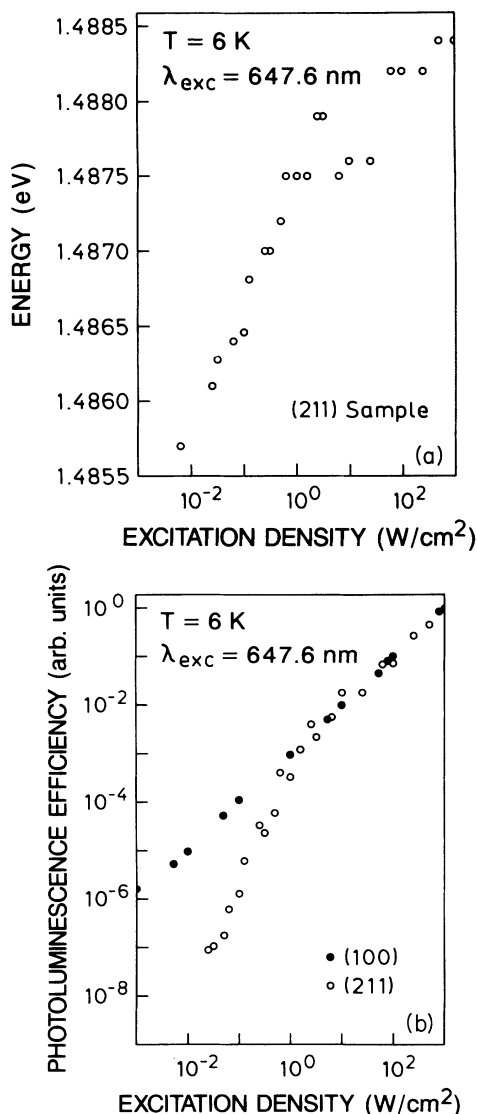


FIG. 3. (a) Photoluminescence peak position vs excitation density for the (211) sample. (b) Photoluminescence efficiency for sample of Figs. 1 and 2 (open circles) and for a (100) InAs/GaAs MQW samples with ten periods and an InAs thickness of 0.8 ML (solid circles). This figure does not allow a comparison of the absolute luminescence efficiency of the (100) with that of the (211) sample.

linear behavior. Below this density, however, the slope increases from 1 to 2.6 and we see a superlinear dependence of the luminescence efficiency on the excitation density. Note that these two domains of different behavior are also reflected in the plot of PL peak position versus excitation density of Fig. 3(a).

These findings show that the optical properties of (211)-oriented InAs/GaAs quantum sheets are significantly different from those of samples grown along the [100] direction. Since the growth axis is the [211] direction we necessarily have nonzero off-diagonal elements of the strain tensor.<sup>12</sup> Therefore a piezoelectric field in the InAs layers exists. Consideration based on continuum elasticity theory<sup>5</sup> let us expect a field strength of 200

kV/cm. In the case of piezoelectrically active quantum wells the internal field tries to separate positive and negative carriers and thus distorts the excitonic wave function. For this reason optical matrix elements depend strongly on the magnitude of the internal field<sup>6</sup> and therefore also on the carrier concentration. Consequently, it is natural to pinpoint the piezoelectric field as the reason for the observed nonlinearities. The screening of the electric field by photogenerated carriers is also a plausible explanation for the blueshift of the luminescence peak. The pronounced peak broadening, however, cannot undoubtedly be attributed to the existence of a piezoelectric field. Since the broadening already starts at very low excitation densities, the piezoelectric field may have its share in this effect. It has to be recalled that any theoretical considerations available are based on continuum elasticity theory and are likely to break down for the atomic limit of a single monolayer which is under investigation here.

One more observation which only at a first glance seems to be contradictory is that the luminescence peak appears at an energy which is about 10 meV higher than that of (100)-oriented quantum sheets.<sup>11</sup> We are not yet able to assess the significance of this energy shift because the switch to a different growth direction changes a number of quantities influencing the quantum-sheet related energy levels. The effective mass of the holes, for example, is known to be anisotropic and therefore influences the energetic position of the luminescence peak. Up to now there are no results for the (211) heavy-hole mass, but the values suggested in the literature<sup>13,14</sup> for the neighboring [311] and [111] directions tend to be larger than the (100) values and therefore the valence-band anisotropy should express itself in a redshift of the luminescence line with respect to the (100) structure. The band offset also shows anisotropies which are expected to be small.<sup>15-17</sup> The mass anisotropy together with the Stark-effect-like influence of the piezoelectric field nourish the expectation of a redshift of the (211) luminescence signal with respect to the (100) reference structure.

However, the atom density on the (211) lattice planes is smaller than that on the (100) planes and this certainly is of decisive influence on the energy levels. The distance between the (211) lattice planes is by a factor of  $1/\sqrt{6}$  smaller than between (100) lattice planes. This smaller separation between the lattice planes results in larger distances between neighboring atoms populating the same lattice plane. A *Gedankenexperiment* shows that a smaller atom density should lead to a blueshift of the luminescence signal. If we think of a sequence of different lattice planes [for example, (211), (311), (411), . . .] we see that the atom density decreases with increasing sum of indices. For high indices the separation between In atoms is so large that the coupling between neighboring In atoms can be neglected and we can view this limiting case as isolated In atoms in a GaAs matrix. It is well known that isolated In atoms do not form impurity states in the band gap of GaAs.<sup>18</sup> Since our sequence of planes approaches this state we expect an increase of the energy for the higher index planes. This argument shows that the changes connected to the switch from [100] to [211] growth axis do not simply add up but might cancel each other as well as

blueshift or redshift the luminescence line with respect to the (100) reference structure.

To summarize our results we have investigated GaAs with inserted (211)-InAs lattice planes which were shown to have excellent structural properties by means of double-crystal x-ray diffraction. The photoluminescence

spectra show a blueshift of the luminescence peak with increasing excitation density and the corresponding luminescence efficiency increases superquadratically for small but linearly for large excitation densities. We attribute these unusual properties to the existence of an internal piezoelectric field.

\*Present address: Centro Nacional de Microelectronica, Serrano 144, 28006 Madrid, Spain.

†Present address: Fachbereich Materialwissenschaft, Technische Hochschule Darmstadt, D-6100 Darmstadt, Germany.

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