Pressure dependence of the electronic structure of a [311] piezoelectric Ga_{0.85}In_{0.15}As/AlAs superlattice

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We have studied the electronic subband structure of a piezoelectric [311] $Ga_{0.85}In_{0.15}As/AlAs$ superlattice by means of high-hydrostatic pressure and excitation-power-dependent photoluminescence at 78 K. In particular, we unraveled the origin of two optical transitions at around 1.96 and 2 eV at ambient pressure, which were recently found to give rise to an unexpectedly strong resonant enhancement of the acoustic-phonon Raman scattering for such samples with permanent built-in piezoelectric fields [G. Rozas *et al.*, Phys. Rev. B **77**, 165314 (2008)]. Here we demonstrate that these transitions are *doubly indirect*, in real and reciprocal space, corresponding to radiative recombination processes between electrons at the X valleys of the AlAs barriers and heavy holes at the Γ point of the Brillouin zone but confined to the GaInAs quantum wells. In addition, the partial screening of the piezoelectric field induced by carrier photoexcitation under illumination becomes largely suppressed for pressures above 1.1 GPa due to conduction-band Γ -X crossover effects.

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I. INTRODUCTION

Phonon engineering at the nanoscale has been demonstrated to be achievable in a large number of experiments on III-V superlattices (SLs) and other phononic devices such as mirrors, cavities, and monochromatic phonon sources.^{1–5} Nevertheless, a main drawback of these systems concerning the electron-phonon coupling is that the deformationpotential interaction, which couples acoustic phonons to electrons is relatively weak. This fact introduces an intrinsic limitation for the development of multifunctional acoustic devices designed to act on electronic or optical properties. An alternative solution to this problem involves the use of the piezoelectric electron-phonon coupling in noncentrosymmetric crystals exhibiting permanent built-in piezoelectric fields as is the case of III-V semiconductors strained heterostructures epitaxially grown on substrates oriented in nonprincipal direction.^{6–13} For instance, huge efficiencies for coherent acoustic-phonon generation have been reported in GaInN/GaN SLs.9,10 A related effect was recently observed in a piezoelectric GaInAs/AlAs superlattice,14 where a strong resonant effect on the Raman scattering by acoustic phonons was obtained only for two optical transitions which appear to be activated by the piezoelectric field. Based on the electronic-structure data available so far, these transitions were tentatively assigned to the $e_1 \rightarrow hh_2$ forbidden ones, i.e., between the lowest confined electron state and the first excited heavy-hole level of the GaInAs quantum wells.14 Within this interpretation, however, the unusually large quantum-confinement Stark shift observed for these transitions of up to about 60 meV remained unexplained, raising some doubts about the assignment. In order to attain a deeper understanding of electron-acoustic-phonon interactions in piezoelectric nanostructures, the origin of these transitions must be fully addressed.

In this work we present a systematic study of the electronic subband structure of the piezoelectric [311] Ga_{0.85}In_{0.15}As/AlAs superlattice that exhibits a strong electron-acoustic-phonon coupling.¹⁴ To trace back the origin of the electronic transitions responsible for the resonant enhancement of Raman processes mediated by this coupling we make explicit use of the fact that the conduction-band minima at the Γ , L, and X points in the Brillouin zone of semiconductor compounds exhibit well defined but different hydrostatic pressure coefficients.¹⁵ Pressure appears also as a natural variable to change the piezoelectric field of a structure, which combined with screening effects due to photogeneration of carriers make out of the photoluminescence technique a powerful tool for this kind of investigations.^{16,17} Here we show that these transitions which become optically active in the presence of piezoelectric fields are doubly indirect both in reciprocal $(X \rightarrow \Gamma)$ and real space $(e_{AlAs}^- \rightarrow h_{GaInAs}^+)$. In addition, an abrupt reduction in the partial screening of the piezoelectric field was observed above 1.1 GPa. At around this pressure, the Γ -X conduction-band crossover takes place between the lowest states of the GaInAs quantum wells at the Brillouin-zone center and the X minima of an etch-stop layer (ESL), onto which the SL was grown. This pressure-induced crossing leads to a suppression of the drift of photoexcited electrons through the superlattice,

diminishing the effects of illumination on the screening.

II. EXPERIMENTAL

Cubic phases such as the zinc blende do not present spontaneous electric fields due to their high degree of symmetry. Nevertheless, a piezoelectric field is induced in the structure by strain in any crystallographic direction departing from the principal axes.¹⁸ The sample studied here consists of a SL with 24 Ga_{0.85}In_{0.15}As/AlAs periods grown by molecularbeam epitaxy on a [311] oriented GaAs substrate. The thicknesses of the Ga_{0.85}In_{0.15}As and AlAs layers are 23 Å and 84 Å, respectively. In addition, a $1-\mu$ m-thick Al_{0.56}Ga_{0.44}As ESL was grown between the SL and the GaAs substrate to eventually allow for the chemical etching of the substrate. The sample is terminated at a GaInAs layer. For a detailed structural characterization of the sample, showing its high quality, we refer to the work of Ref. 13. Whereas the AlAs barriers are only weakly strained (less than 0.05%), the Ga_{0.85}In_{0.15}As layers are under a biaxial compressive strain which amounts to about 1%, considering the lattice mismatch to GaAs. The piezoelectric superficial charge induced by this strain at the interfaces of the GaInAs layers can be readily calculated following Ref. 18 to be $\approx 6.5 \times 10^{-8} \text{ C/cm}^2$.

The high-pressure photoluminescence (PL) measurements were performed at liquid-nitrogen temperature in a bath cryostat specially designed to fit in its cold bore a diamondanvil cell (DAC). The sample was thinned to about 30 μ m by mechanical polishing and placed into the DAC together with a ruby sphere for pressure calibration.^{19,20} Helium was used as pressure medium and nonhydrostatic conditions were avoided by changing the pressure always above the He melting temperature. The PL spectra of the sample were excited using the 514.5 nm line of an Ar⁺-ion laser and collected using a LabRam HR800 spectrometer equipped with a charge-coupled device detector. All the measurements were done at 78 K and the power of the incident light was varied between 0.5 and 4.4 mW.

III. RESULTS AND DISCUSSION

Figure 1 shows three representative PL spectra taken at different pressures of 0.5, 1.1, and 1.9 GPa. The main peak labeled e_1 - hh_1 corresponds to the fundamental radiative recombination between the electron- and heavy-hole ground states of the GaInAs quantum wells. The peaks above 2.02 eV arise from the indirect $\Gamma \rightarrow X$ recombination in the thick AlGaAs etch-stop layer. The doublet denoted as $X_{1,2}$ corresponds to the optical transitions which lead to the fieldactivated resonant enhancement of the Raman scattering by acoustic phonons, as discussed in Ref. 14, and whose assignment is the main purpose of this high-pressure work. All spectra have been normalized to the intensity of the e_1 - hh_1 peak at 0.5 GPa. For the sake of clarity the spectra have been multiplied by different factors and vertically shifted. Here we point out that the spectral region of the $X_{1,2}$ transitions above 1.9 eV was always measured by defocusing the image of the sample on purpose (that is the reason for the different mul-



FIG. 1. Photoluminescence spectra of the [311] GaInAs/AlAs SL sample measured at different pressures of 0.5, 1.1, and 1.9 GPa. Dashed curves represent the Gaussian functions used for fitting the spectral line shape, corresponding to the three main emission peaks of the SL (see text for the assignment). Peaks observed at higher energies than 2.02 eV correspond to the emission from the etch-stop layer. The spectra were multiplied by appropriate factors, as indicated, and vertically displaced for clarity. The sharp peaks at around 1.8 eV correspond to the ruby lines for pressure calibration.

tiplication factors of parts of the same spectrum in Fig. 1). Only in this way we were able to partially suppress the extremely strong signal from the etch-stop layer that hampered the observation of the faint $X_{1,2}$ peaks in the pressure cell, due to the loss of confocality of the optics in *macro* mode.

A striking result concerns the evident different pressure behavior observed for the e_1 - hh_1 and the $X_{1,2}$ emission bands, i.e., as pressure increases the e_1 - hh_1 peak shifts strongly to higher energies, whereas the X_{1.2} bands slightly decrease in energy. In order to obtain the linear pressure coefficient for each transition, we performed a line-shape analysis of the spectra using an asymmetric and symmetric Gaussian functions for e_1 - hh_1 and $X_{1,2}$ PL peaks, respectively. The asymmetry toward higher energies of the fundamental quantum well transition originates from the overlap with the emission peak corresponding to the ground-state light-hole recombination $(e_1 - lh_1)$ since at 78 K both peaks cannot be resolved from each other. We note that the full width at half maximum of all three Gaussian peaks was kept fixed to its ambient-pressure value for all pressures since no dependence is expected. This allowed us to precisely follow the evolution of each peak in the pressure region of spectral overlap between 1.8 and 2.5 GPa. The dashed curves in Fig. 1 are representative for the results of the fitting procedure.

The PL peak positions obtained from the line-shape analysis are plotted in Fig. 2 (symbols) as a function of hydrostatic pressure. The solid and dashed-dotted lines represent the results of least-squares fits to the data points using a linear relation. The pressure coefficient of e_1 - hh_1 was found to be (101 ± 5) meV/GPa, which is typical for direct optical transitions at the Γ point of the Brillouin zone in III-V semiconductors¹⁵ and is very close to value of



FIG. 2. Pressure dependence of the e_1 - hh_1 and $X_{1,2}$ transitions. The solid and dashed-dotted lines represent the results of least-squares fits to the data points using linear functions. The dashed line corresponds to the pressure-dependent energy position of the X conduction-band minima of the AlGaAs ESL relative to the energy of the heavy-hole ground state hh_1 of the quantum wells.

 (108 ± 3) meV/GPa measured in bulk GaAs.²¹ On the contrary, the $X_{1,2}$ emission bands exhibit a negative and much smaller pressure coefficient. For X_1 and X_2 we obtained a slope of (-3 ± 2) meV/GPa. The magnitude and sign of the pressure coefficient of the X1,2 peaks is the main result of this work since such a dependence on pressure is the signature of an *indirect* optical transition that takes place between electron states at the X conduction-band minima and holes at the Brillouin-zone center.¹⁵ For comparison the Γ -X indirect gap of GaAs decreases at a pace of (-13.5 ± 1.3) meV/GPa (Ref. 21) with increasing pressure. This is a key piece of information to rule out the possibility that the $X_{1,2}$ peaks are related to the forbidden, direct optical transition $e_1 \rightarrow hh_2$ within the quantum wells, as presumed earlier.¹⁴ We notice that the magnitude of the pressure coefficient is surprisingly small, for instance, compared to that of bulk GaAs. At first glance, this might be an indication that the piezoelectric field diminishes under pressure, leading to a pressure-induced blueshift of the X1,2 states that compensates in part the downward variation in their energy. Such an effect is expected from the reduction in the built-in strain of the lattice-matched GaInAs layers upon compression. Although the applied pressure is hydrostatic, the lattice mismatch between GaInAs layers and GaAs substrate decreases with pressure since the bulk modulus of both materials is different. Nevertheless, a simple calculation shows that the contribution of this effect to the pressure coefficient of the $X_{1,2}$ energies is about one order of magnitude too small. We rather believe that this is a consequence of the nature of the conduction-band states involved in the $X_{1,2}$ transitions, which result from a mixing of states with wave vectors in the vicinity of the conduction-band X point of the Brillouin-zone edge. These states have small admixtures of Bloch factors with atomic s-like character,²¹ which, in turn, exhibit a different, positive pressure coefficient than the states with pure p-like character as for the X conduction-band minima in the bulk.²²

The next step is to investigate which is the spatial location of the electrons involved in the $X_{1,2}$ optical transitions, i.e., if



FIG. 3. Sketch of the electronic band structure of the piezoelectric [311] GaInAs/AlAs SL. The optical transitions between different confined states of the SL are represented by broken arrows and their assignment is indicated. In the sketch, the free surface of the heterostructure is toward the right-hand side.

their wave function belongs to the X states of the GaInAs quantum wells or those of the AlAs barriers. For that purpose we have performed electronic band-structure calculations within the envelope function approximation (EFA) in order to construct the sketch of the conduction and valence-band profiles of the piezoelectric GaInAs/AlAs SL, displayed in Fig. 3. Here we used available literature data for the composition dependence of the band gaps in $Al_rGa_{1-r}As$ (Ref. 23) and $Ga_xIn_{1-x}As$ (Ref. 24) alloys and considered the band alignment given by the rule of calculating the conduction-(valence-) band offsets as 50% of the band-gap difference between well and barrier material.²⁵ We have also taken into account the temperature dependence of the gaps as given for GaAs (Ref. 26) and the energy shift of the direct and indirect gaps of the Ga_{0.85}In_{0.15}As quantum wells due to the large built-in strain of these layers.²⁷ The electric field distribution in the sample has been estimated by considering the piezoelectric surface charge induced by 1% compressive strain in the GaInAs layers,¹⁸ the different dielectric constants,²⁸ and the boundary condition imposed by the buildup of a negative band bending at the surface of the sample due to accumulation of electrons at the surface states. The latter results from the assumption that the Fermi energy is pinned close to the bottom of the conduction band at the surface of an In rich layer,²⁹ whereas in the bulk the Fermi energy should lie close to the top of the valence band of the AlGaAs etch-stop layer since the carbon impurities present in any molecular-beam epitaxy chamber typically lead to residual p doping of the grown layers. We estimate the potential drop across the superlattice to range between 1.3 and 1.5 eV, resulting roughly in the field distribution sketched in Fig. 3, i.e., the fields almost cancel each other in the GaInAs quantum wells but the AlAs barriers are subjected to fields between 65 and 75 kV/cm in magnitude.

The resulting band structure is as follows: the X conduction-band states of the AlAs barriers are lower in energy than their counterparts of the GaInAs quantum wells, which leads for the SL to a type-II band alignment between the conduction-band edge at the X point and the top of the valence band at Γ . Since the holes are always confined to the GaInAs wells due to the large valence-band offset of \approx 0.45 eV, the X_{1.2} optical transitions are doubly indirect, in real and reciprocal space, as illustrated in Fig. 3. The EFA calculations further indicate that the splitting between the X_1 and X₂ transitions corresponds to that of the discrete conduction-band states of the AlAs barriers. The splitting arises from the combined effect of quantum confinement and electric field on X states which are characterized by largely anisotropic effective masses, parallel and perpendicular to the [311] direction. In fact, the latter contribution to the splitting almost vanishes at very large laser powers for which the piezoelectric field becomes largely screened.14,17 As indicated in Fig. 3, the radiative recombination takes place from the X electron states of an AlAs barrier to the same heavyhole level of the adjacent quantum well to the left-hand side, which are of smaller energy due to the electrostatic potential drop in the piezoelectric superlattice. Although the transitions to the other well at the right-hand side of the barrier are expected to be more intense due to the much larger overlap of the electron and hole wave functions, they are estimated to appear at energies larger than 2 eV, such that their observation is completely hampered by the much stronger emission from the etch-stop layer. Moreover, the EFA calculations seem to indicate that these higher energy transitions to the right well should exhibit a redshift with increasing laser power, i.e., with increasingly screened piezoelectric fields.³⁰ As discussed below, the X_{1,2} PL peaks shift to the blue with laser power, as calculated transitions to the left well do. Finally, we remark that there is no evidence either of an energy anticrossing between the e_1 - hh_1 and $X_{1,2}$ emission peaks or an intensity transfer between them,²⁷ speaking against the possibility of the X states of the GaInAs wells being involved in the observed optical transitions. We would like to point out that the agreement between calculated and measured splittings and shifts is within a factor of two, which is not really surprising in view of the uncertainties in several [311] band-structure parameters such as band offsets, effective masses, etc. Nevertheless, the overall phenomenology is satisfactorily described with the assignments performed here.

We now turn to the discussion of the energy shifts due to the laser-power variation in the quantum-confined Stark effect on the different electronic states of the SL. As previously reported for this sample,¹⁴ the quantum-confined Stark shift, resulting from the permanent piezoelectric field, diminishes under illumination due to the partial screening of the latter by the photogenerated carriers. Hence, PL peaks shift to higher energies at larger laser powers. Such shifts, though, are much larger for the X_{1,2} transitions (up to 60 meV) (Ref. 14) than for the direct e_1 - hh_1 one (less than 10 meV). Actually, in the latter case, we do not expect any shift at all due to the extremely small well thickness and the partial cancelation of the electric fields in the GaInAs layers. The observed influence of laser power on the energy of the direct e_1 - hh_1 emission is most likely to be related to an increasing wave-



FIG. 4. (a) Laser power dependence of the energy shift of the $X_{1,2}$ and e_1 - hh_1 optical transitions for three different pressures of 0.6, 1.2, and 1.9 GPa. The energy shift is referred to the energy of each peak at 4.4 mW of excitation power. (b) Dependence on pressure of the laser-induced energy shift of the $X_{1,2}$ transitions for the lowest illumination power of 1.1 mW. Solid and dash-dotted lines are guides to the eye.

function delocalization at higher excitation levels of the carriers localized by well-width fluctuations. Regarding the high-pressure experiments, Fig. 4(a) shows the energy shifts of the three main PL peaks as a function of laser power for three representative pressures of 0.5, 1.2, and 1.9 GPa. The laser power was varied between 1 and 4.4 mW limited by the weak intensity of the emission and the energy shifts are referred for comparison to the position of the corresponding PL peak at the maximum power of 4.4 mW. Whereas for the direct e_1 - hh_1 transition the shifts are very small and do not depend much on pressure, as expected, a clear distinction can be made in the photoexcitation dependence of the Stark shift for the X_{1.2} transitions depending upon the pressure being lower or higher than 1.1 GPa. Figure 4(b) illustrates the behavior of the laser-induced shift of the X1.2 transition energies for the lowest used power of 1.1 mW. For pressure points above 1.1 GPa there is a much weaker effect of illumination on the Stark shift, meaning that its screening is no longer effective. The change in the Stark-shift screening, though, is fairly abrupt and not gradual. This speaks against any effect on the piezoelectric field induced by a reduction in the built-in strain due to the different compressibility of well and barrier material, which should change in proportion to the applied hydrostatic pressure. It arises the question, what makes this specific pressure so particular? We believe the answer lies in the pressure-induced changes in the electronic band structure of the piezoelectric superlattice. The thick $Al_{0.56}Ga_{0.44}As$ etch-stop layer is an indirect-gap (Γ -X) material. At ambient pressure the zero-phonon line is observed at 2.09 eV, which decreases with increasing pressure at a rate of -20 meV/GPa, as determined in this work (data not shown). The valence-band discontinuity between etch-stop layer and the GaInAs wells is estimated to be approximately 0.32 eV. By considering a confinement energy of 0.07 eV for the hh_1 state, we calculate the pressure at which the Γ -X conductionband crossover between the electron ground-state confined to the well (e_1) of the SL and the X minima of the etch-stop layer occurs to be 1.15 GPa (see Fig. 2). Above this pressure, thus, all the photoexcited electrons generated by absorption in the etch-stop layer would now encounter an energy barrier which hinders their way through the SL to the surface.³¹ In this way, many photoexcited electron-hole pairs would recombine outside the SL, thus, not contributing to the screening of the piezoelectric field, which translates into an insensitivity of the $X_{1,2}$ transition energies upon illumination.

Finally, we would like to address the issue which motivated us for this study about the relation between the fieldinduced X_{1,2} optical transitions and their resonant enhancement of the Raman scattering by acoustic phonons.¹⁴ In the absence of any (piezo)electric field, the X1,2 optical transitions are within the dipolar approximation forbidden by symmetry.³² A similar situation occurs, for instance, for type-II transitions in GaAs/AlAs double barrier quantum well structures.³³ At the origin of this particular selection rule is, on the one hand, the fact that in III-V semiconductors the Bloch factor of the heavy-hole states at the Γ point of the valence band and the electron states at the X conductionband minima have both atomic *p*-like character.^{15,22} On the other hand, both electron and hole envelopes are even functions with respect to the mirror plane in the middle of any GaInAs well. The $X_{1,2}$ transitions, though, become optically active with field due to the breakdown of the parity symmetry for the envelope functions. In fact, as the (piezo)electric field increases, the center of mass of the electron wavefunction shifts gradually toward the AlAs barrier with lower potential, giving rise to an induced optical dipole moment which is proportional to the charge displacement associated with an electronic transition from hh_1 to $X_{1,2}$. This explains why these optical transitions lead to resonant enhancement of the Raman signal only in the piezoelectric [311] SL but never in the conventional [001] samples.

The present assignment also allows for a better understanding of the field-mediated mechanism of acousticthe Raman efficiency for scattering by acoustic phonons in piezoelectric structures contains two terms which depend explicitly on electric field and which stem from the change in electric susceptibility induced by the acoustic phonon. The terms reflect either the phonon modulation of the transition energy or of the transition probability, i.e., the dipolar matrix element. The former is related to the electro-optic constant and accounts for the piezoelectric mechanism in bulk materials. As follows from the huge laser-power dependence of the $X_{1,2}$ transition energies (see Fig. 4), this term may provide an important contribution to the electron-acousticphonon coupling in the resonant Raman process. The latter term might be also of importance in the special case of the $X_{1,2}$ transitions, for their dipole matrix element has an appreciable magnitude, as indicated by the relative strong intensity of the doubly indirect X_{1,2} optical transitions in the piezoelectric [311] sample.

IV. CONCLUSIONS

In conclusion, we have demonstrated that the optical transitions leading to a prominent electron-phonon piezoelectric coupling in a [311] GaInAs/AlAs SL arise from doubly indirect, in reciprocal and real space, recombination processes between the split X conduction-band minima of the AlAs barriers and the heavy-hole ground state of the GaInAs quantum wells. This result is a fundamental step toward a detailed description of this piezoelectric coupling mechanism of importance for the proper design of novel phononic devices making explicit use of an enhanced electron-acoustic-phonon interaction. In addition, by studying the pressure dependence of the changes of the quantum-confined Stark shifts with laser power we have found a curious effect, namely, that the partial screening of the piezoelectric field by carrier photoexcitation becomes inefficient at pressures higher than 1.1 GPa. This effect is explained by the appearance of an energy barrier for the migration of photoexcited electrons from the AlGaAs etch-stop layer hindering their accumulation at the SL surface.

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