Light-heavy hole mixing and in-plane optical anisotropy of InP-$Al_xIn_{1-x}As$ type-II multiquantum wells

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We report investigations of the giant in-plane polarization anisotropy of InP-$Al_xIn_{1-x}As$ type-II multiquantum wells. An absorption polarization rate up to 30% is observed over a very broad (>100 meV) spectral range, in contrast with type-I quantum wells where similar effects are observed in a much narrower range. This property results from the heavy- and light-hole band mixing due to the interface symmetry reduction, combined with the lifting of parity selection rule due to carrier spatial separation in type-II quantum wells. Calculations within the framework of the H_{BF} model (i.e., the envelope function theory with C_{2v} perturbations at the interfaces) yield fair agreement with the experimental results.

There is increasing interest in the large in-plane anisotropy of the optical properties of semiconductor heterostructures[1–4] because this recently discovered phenomenon is forbidden in the classical envelope function theory (EFT), and appears to have important implications in different fields of semiconductor science such as quantum-well electro-optics[5–7] or quantum magnetotransport.[8,9] As discussed independently by several authors,[10–12] the point group symmetry T_d of the zinc-blende lattice is reduced by the presence of a sharp interface to the much lower C_{2v} symmetry. It loses not only the translational invariance along the [001] growth axis, but also an element of rotational symmetry, namely the fourfold roto-inversion around z. As a consequence, the Hamiltonian must contain some interface term proportional to the lowest symmetry invariant of the C_{2v} group,[10] i.e., the operator $L_xL_y+L_yL_x$, which appears to be identical to the $(F\text{-}B)$ operator we have introduced in the ''H_{BF} model.'' (H_{BF} denotes the exact C_{2v} Hamiltonian built with the backward (B) and forward (F) operators which are defined in Ref. 12.] This term clearly couples the heavy and light hole states $|3/2,3/2\rangle$ and $|3/2,-1/2\rangle$ (hereafter denoted H^+ and L^-) and, similarly, H^- and L^+. By taking into account that the symmetry reduction is localized in the interface unit cell, the mixing matrix element associated to a single interface takes the simple form[10–12]

$$M_{HL}=\frac{a_0}{4}V_{int}f_H(z_{int})f_L(z_{int})\langle H^+|(F-B)|L^-\rangle, \quad (1)$$

where a_0 is the lattice constant (introduced here for dimensionality convenience), $V_{\rm int}$ is an interface potential whose relation to band-structure parameters is still a matter of discussion, z_{int} is the interface position and f_H, f_L the envelopes of the heavy- and light-hole states. The actual coupling in a quantum-well (QW) structure is the sum of the different (at least two) interface contributions, and it depends on the overall QW symmetry. This k-independent coupling of heavy and light hole states causes the in-plane optical anisotropy,[7,12] and also has a variety of consequences on the detailed properties of the quantum-well band structure (like the lifting of parity degeneracy of the band dispersion[13,14]), which are a matter of present interest. In the case of a nominally symmetrical, common anion quantum well such as GaAs-$Al_xGa_{1-x}As$, the two interface potentials have opposite signs, and they couple only heavy- and light-hole states of opposite parities (for instance, H_2 and L_1): In accordance with the D_{2d} point-group symmetry of such a symmetrical QW, this restricted coupling does not generate any in-plane optical anisotropy. In general however, the quantum well has some asymmetry (modulation doping, applied electric field, asymmetrical composition profile) and retains the low C_{2v} symmetry of a single interface. A case of particular interest is that of ''no-common-atom'' (NCA) heterostructures[7,12] like $In_xGa_{1-x}As$-InP, where nominal interfaces involve half-monolayers of different specific interface bonds, namely (In,Ga)-P and In-As. This NCA situation is also encountered in the InP-(Al,In)As system, which is in addition the best example of regular type-II configuration: Electrons are confined in the InP layers with a conduction-band offset[15] of $\Delta_c=400$ meV and holes in the (Al,In)As layers with a valence band offset of $\Delta_v=300$ meV, as shown schematically in the inset of Fig. 1. From the consideration of Eq. (1), it may be anticipated that the optical properties of such type-II quantum wells should be particularly sensitive to the interface symmetry reduction effect[16] because they result entirely from the overlap of conduction and valence eigenstates in the interface region.[17] Here, we report experimental investigations of the optical anisotropy in InP-$Al_xIn_{1-x}As$ multiple quantum wells (MQW's) and discuss the result in the framework of the H_{BF} model where the corrective matrix elements M_{HL} are treated as a perturbation in the classical EFT.

The samples are 50-period InP-$Al_xIn_{1-x}As$ multiple-quantum wells grown by atmospheric pressure metal oxide chemical vapor deposition (MOCVD). X-ray diffraction reveals an astonishingly good structural quality.[18] Their basic optical properties (including low threshold lasing) have been reported in Refs. 15 and 20–22. The discussion will be focused on a sample with layer thicknesses InP (40 Å) and (Al,In)As (100 Å), but equivalent results have been observed on samples having different parameters. The absorption spectrum obtained from direct optical transmission measurements using unpolarized light is shown in Fig. 1, together with the corresponding EFT calculation. With respect to the

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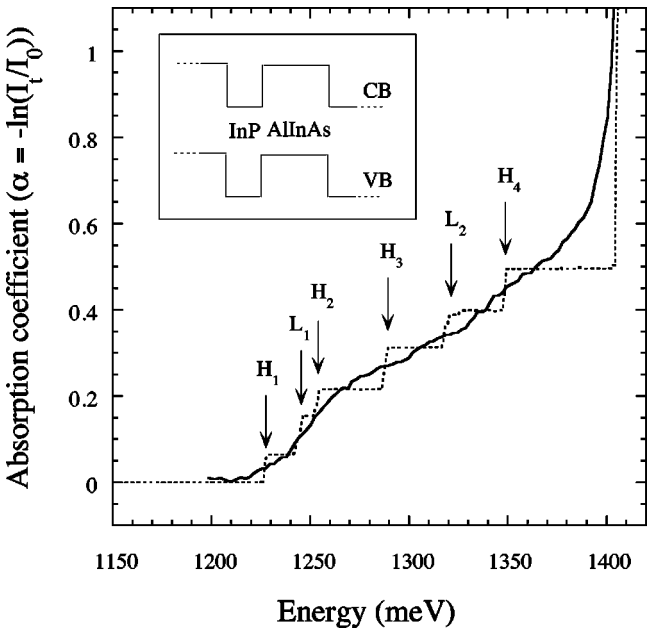

FIG. 1. Unpolarized optical absorption spectrum recorded at 77 K in a 50-period InP (40 Å)–$Al_xIn_{1-x}As$ (100 Å) MQW, and calculated spectrum in the classical envelope function theory. All the steplike transitions end in the first confined state E_1 of the InP QW, so we only label the starting hole level. The strong absorption which appears at about 1405 meV comes from a 1000-Å-thick InP cap layer. The inset shows schematically the band alignment in this type-II system.

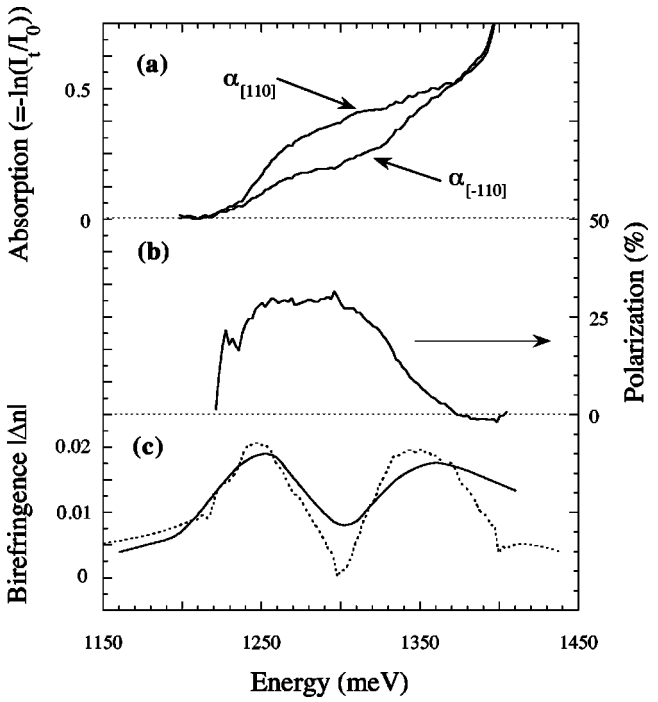

FIG. 2. (a) Polarization-resolved absorption spectra, and (b) their relative difference or polarization spectrum. Light is polarized parallel to the [110] and $[\bar{1}10]$ directions. The lower panel (c) shows the anisotropy of the optical index $|\Delta n|$ (solid line) together with the calculated Kramers-Kronig transform of the absorption difference (dashed line).

well-known steplike absorption spectrum of a type-I two-dimensional heterostructure, there are three major differences: (i) the lifting of parity selection rules makes the number of allowed transitions much larger than in a supposedly type-I, 40-Å-thick InP QW where only H_1-E_1 and L_1-E_1 would show up in the displayed energy range; (ii) the absorption per transition is much smaller than the 1% per well usually observed in type-I QW's, as a consequence of the small overlap of spatially separated envelope functions; and (iii) the extreme sensitivity of the $Al_xIn_{1-x}As$ band gap to composition fluctuations, 27 meV/%, is directly reverberated in the broadening of the optical transitions, which ultimately washes out the steplike density of states, in spite of the high structural quality. We can also notice that due to the strong confinement of conduction states in the 40-Å InP layers, all the optical transitions that may be observed below the gap of InP end in the first conduction state E_1. Polarization-resolved results are displayed in Fig. 2 where the absorption spectra obtained with light linearly polarized along the [110] and $[\bar{1}10]$ directions (i.e., parallel to the sample cleaved edges) are shown in the upper panel, their relative difference (or absorption polarization spectrum) in the second panel, and the birefringence spectrum in the lower panel. The latter is obtained by combining the polarization-resolved absorption spectra with the transmission spectrum measured between crossed polarizers, which measures the squared modulus of the complex refractive index $|\Delta n|^2$. The birefringence spectrum is nearly perfectly fitted by the Kramers-Kronig transform of the absorption difference [dashed line in Fig. 2(c)], according to the formula

$$\Delta n(\omega_0)=\frac{\lambda}{4\pi L}\omega_0 \mathrm{p}\int_0^\infty \frac{\Delta\alpha(\omega)}{\omega^2-\omega_0^2}d\omega \qquad (2)$$

where L is the total width of the MQW region, $\Delta\alpha=\alpha_{[110]}-\alpha_{[\bar{1}10]}$ and the notation p stands for the principal value of the integral. This indicates that the ''causality'' of the optical anisotropy in these samples is entirely contained in the near-band-gap region, with essentially no influence from the absorption near the $E1$ band gap (L point of the Brillouin zone), which governs the main part of the refractive index. Note that similar trend has been observed in type-I $In_xGa_{1-x}As$-InP quantum wells.

In order to illustrate the specific role played by the type-II nature of this system, we show for comparison the results obtained in a 113-Å-thick $In_xGa_{1-x}As$-InP QW.[6] The latter shows indeed a comparable sequence of hole levels, hence the consequences of the symmetry reduction in the valence band [Eq. (1)] should be similar. However, as shown in Fig. 3 the polarization spectra in the two samples differ strikingly by the maximum value of the polarization rate (30% compared to 10%) and by the width of the polarization spectrum (more than 100 meV compared to 16 meV). These differences are mainly the consequence of the partial lifting of parity selection rules[17] in type-II MQW's: In the presence of asymmetrical interfaces, all the hole bands are coupled by the C_{2v} component of the interface potential [Eq. (1)], and all of them contribute with equivalent strength to the optical absorption towards the first conduction state confined in the InP layers. Therefore, it may be anticipated that the anisotropy features are observed over the whole spectral range in-

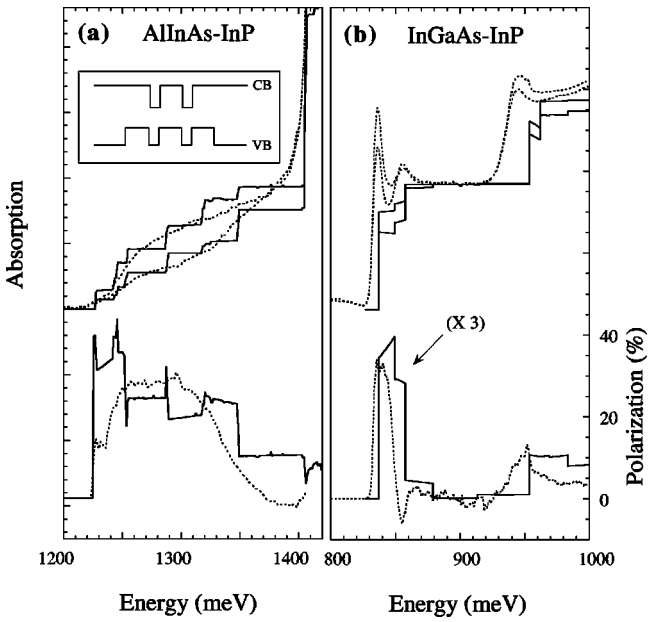

FIG. 3. (a) Polarization-resolved calculated spectra (solid lines) and corresponding measurements (dashed lines) in the type-II InP-$Al_xIn_{1-x}As$ MQW compared with (b) equivalent spectra in a 113 Å (In,Ga)As-InP type-I MQW. The inset in (a) shows the model potential used in the calculation.

stead of being restricted to the intervals between heavy- and light-hole transitions of same index. Also, the volume of material tested by the optical absorption is restricted to the interfacial region and it seems natural that the effects are proportionally larger than in the type-I configuration where optical absorption tests the whole QW volume. These simple-minded ideas are in fact supported by the calculations also displayed in Fig. 3. The procedure follows the general line of the H_{BF} model: we first calculate the hole and conduction wave functions, using the classical EFT (solved by a transfer matrix routine) in the model structure given as a schematic in the inset of Fig. 3. It corresponds to three (Al,In)As quantum wells separated by two InP layers, and is embedded in a virtual material forming a barrier for both the electrons and the holes. Then we estimate the coupling matrix elements M_{HL} between all the confined hole states (four H and two L states for each well), using different potential strengths for the $Al_xIn_{1-x}As$-InP and InP-$Al_xIn_{1-x}As$ interfaces. The 18×18 coupled valence-band Hamiltonian is then diagonalized numerically, and the new eigenstates are finally used to calculate the optical absorption toward the two conduction states forming the first ''conduction miniband.'' The subtle symmetry selection rule of type-II superlattices makes this computationally nontrivial procedure necessary, while the case of type-I QW's is satisfactorily handled analytically with a mere 2×2 matrix. In practice (according to the H_{BF} representation of the C_{2v} potential), we choose to write the interface potentials $(V_1+\Delta_v)/\sqrt{3}$ and $(V_2-\Delta_v)/\sqrt{3}$, where Δ_v is the valence band offset. The theoretical spectra in Fig. 3(a) are obtained with $V_1=1000$ meV, $V_2=200$ meV. Noticeably, these values fall in the same range as those used to fit the data in the $In_xGa_{1-x}As$-InP system ($V_1=1300$ meV, $V_2=300$ meV). It is noteworthy that we have neglected in these calculations the noncommutativity of the band offsets that had been evidenced in these systems[20,23,24] and would slightly affect the symmetry of the f_H and f_L envelope functions. However, in absence of experimental data on the electric field dependence,[19] the fit is not very sensitive (within 200 meV variations) to the values of V_1 and V_2. Numerically we find that the field dependence of the anisotropy varies strongly with the interface potentials and more specifically with their difference V_1-V_2. Using the above-mentioned parameters we calculate that the polarization rate should increase by an average factor of 1.5 over the spectral region of interest under an electric field of 50 kV/cm and decrease by a factor of 0.8 if the field is reversed. Besides, no drastic change in the shape of the anisotropy spectrum is predicted. Although the general trends of the results seem to be well explained by the present calculation, there are still some discrepancies. In particular, the experimental polarization spectrum vanishes above 1350 meV while the calculated one remains significantly polarized. It seems that the discrepancy cannot be explained by the choice of band-structure parameters (including V_1 and V_2). One possibility is that the restriction of the basis to quantum-well bound states ignores a significant coupling of these states to those lying above the barrier, which may affect the calculated spectra. An even more likely cause of discrepancy is the use of an ideally sharp interface model, which cannot be realistic and must reach some limits, especially when coming to excited states with rapidly varying envelopes: The possible use of polarization-resolved measurements as a spectroscopic tool for the investigation of semiconductor interfaces would require a careful examination and modeling of these discrepancies between ideal model predictions and observations.

In conclusion, we have shown that the polarization-resolved spectroscopy of type-II NCA MQW's reveals qualitatively new aspects of the interface symmetry reduction effect. The large polarization ratio observed over a broad spectral range also make the type-II configuration more interesting for the use of the anisotropy properties in the transparency region, since it results in an index anisotropy comparatively larger than in type-I NCA QW's.

[1] C. Gourdon and P. Lavallard, Phys. Rev. B **46**, 4644 (1992).
[2] O. Krebs, W. Seidel, J. P. André, D. Bertho, C. Jouanin, and P. Voisin, Semicond. Sci. Technol. **12**, 938 (1997).
[3] B. Koopmans, P. V. Santos, and M. Cardona, Phys. Status Solidi A **170**, 307 (1998).
[4] Y. H. Chen, Z. Yang, Z. G. Wang, Xu Bo, and J. B. Liang, Phys. Rev. B **60**, 1783 (1999).
[5] S. H. Kwok, H. T. Grahn, K. Ploog, and R. Merlin, Phys. Rev. Lett. **69**, 973 (1992).
[6] O. Krebs, D. Rondi, J. L. Gentner, L. Goldstein, and P. Voisin, Phys. Rev. Lett. **80**, 5770 (1998).
[7] E. Ivchenko, A. Toropov, and P. Voisin, Phys. Solid State **40**, 1748 (1998).
[8] W. Pan, R. R. Du, H. L. Stormer, D. C. Tsui, L. N. Pfeiffer, K.

W. Baldwin, and K. W. West, Phys. Rev. Lett. **83**, 820 (1999).
[9] M. P. Lilly, K. B. Cooper, J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **83**, 824 (1999).
[10] E. Ivchenko, A. Kaminski, and U. Rössler, Phys. Rev. B **54**, 5852 (1996).
[11] B. A. Foreman, Phys. Rev. Lett. **81**, 425 (1998).
[12] O. Krebs and P. Voisin, Phys. Rev. Lett. **77**, 1829 (1996).
[13] T. Guettler, A. L. C. Triques, L. Vervoort, R. Ferreira, Ph. Roussignol, and P. Voisin, Phys. Rev. B **58**, R10 179 (1998).
[14] L. Vervoort, R. Ferreira, and P. Voisin, Phys. Rev. B **56**, R12 744 (1997).
[15] E. Lugagne-Delpon, P. Voisin, J. P. Vieren, M. Voos, and J. N. Patillon, Semicond. Sci. Technol. **7**, 524 (1992).
[16] A. V. Platonov, V. P. Kochereshko, E. L. Ivchenko, G. V. Mikhailov, D. R. Yakovlev, M. Keim, W. Ossau, A. Waag, and G. Landwehr, Phys. Rev. Lett. **83**, 3546 (1999).
[17] P. Voisin, G. Bastard, and M. Voos, Phys. Rev. B **29**, 935 (1984).
[18] Up to 15 superlattice replica on each side of the 004 substrate diffraction line are observed in double diffraction, with their intensities closely fitted by a model considering abrupt interfaces [P. Auvray, J. P. André, and P. Voisin (unpublished)].
[19] Due to the presence of a thick semi-insulating InP top layer, it was not possible to apply a reliable electric field in these samples.
[20] E. Lugagne-Delpon, J. P. André, and P. Voisin, Solid State Commun. **86**, 1 (1991).
[21] E. Lugagne-Delpon, P. Voisin, M. Voos, and J. P. André, Appl. Phys. Lett. **60**, 3087 (1992).
[22] W. Seidel, P. Voisin, J. P. André, and F. Bogani, Solid-State Electron. **40**, 729 (1996).
[23] R. G. Dandrea, C. B. Duke, and A. Zunger, J. Vac. Sci. Technol. B **10**, 1744 (1992).
[24] W. Seidel, O. Krebs, and P. Voisin, Phys. Rev. B **55**, 2274 (1997).