

Electron minibands and Wannier-Stark quantization in an $\text{In}_{0.15}\text{Ga}_{0.85}\text{As-GaAs}$ strained-layer superlattice

B. Soucail, N. Dupuis, R. Ferreira, and P. Voisin

Laboratoire de Physique de la Matière Condensée de l'Ecole Normale Supérieure, 24 rue Lhomond, F75005 Paris, France

A. P. Roth, D. Morris, K. Gibb, and C. Lacelle

Laboratoire des Sciences des Microstructures Conseil National de Recherches, 100 Promenade Sussex, Ottawa, Ontario, Canada K1A 0R6

(Received 15 December 1989)

We have investigated the electronic properties of an $\text{In}_{0.15}\text{Ga}_{0.85}\text{As-GaAs}$ strained-layer superlattice using photoluminescence excitation and photocurrent spectroscopies. Flatband spectra show transitions at the center and edge of the Brillouin minizone, and photocurrent spectra at finite bias show the effects of Wannier-Stark quantization. The heavy-hole transitions evidence the importance of the excitonic interaction between spatially separated carriers. The light-hole transitions show a qualitatively different behavior resulting from their weak confinement in the GaAs layers. Our data agree with a numerical calculation of the electro-optical absorption spectra.

In the last few years, there has been a considerable interest in strained-layer superlattices (SL) and quantum wells (QW), because of their fundamental interest and their potential for devices.¹ Although a number of studies have been devoted to the $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ heterostructures, the electronic properties of this system are still a matter of controversy.^{2,3} More recently, novel electro-optical properties of semiconductor SL's have been discovered.⁴ The investigation of these Wannier-Stark effects in the $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ system is interesting in many respects. In particular, the strain-induced enhancement of the heavy-hole to light-hole splitting allows a complete separation of the corresponding absorption bands, which makes the observation of the zone-edge or "saddle-point exciton"⁵ transitions much clearer than in unstrained SL's. Also, at least for large enough indium concentrations, the light holes must be confined in the GaAs layers, which gives an opportunity to observe the Wannier-Stark effects in the type-II SL configuration, for which a qualitatively new behavior is predicted. In addition, short-period SL's can be accurately characterized, and the study of the electro-optical properties of such $\text{In}_x\text{Ga}_{1-x}\text{As-GaAs}$ SL's is likely to bring a definitive answer to the controversy on the band offsets in this system. In this Rapid Communication, we report investigations by photoluminescence excitation (PLE) and photocurrent (PC) spectroscopies of the electronic structure and electro-optical properties of a $\text{In}_{0.15}\text{Ga}_{0.85}\text{As-GaAs}$ SL in which the well and barrier thicknesses are small enough to ensure a strong coupling of the wells in the flatband conditions and, at least for the heavy-hole transitions, a negligible intrawell Stark effect.⁶⁻⁸

Our sample was grown by low-pressure metal-organic vapor-phase epitaxy on a Si-doped GaAs substrate. It consists of a 10-period SL grown on top of a $1.8\ \mu\text{m}$ thick buffer layer of undoped GaAs. All the layers were undoped with a residual carrier concentration $n \approx 5 \times 10^{14}\ \text{cm}^{-3}$. An important advantage of periodic structures,

with respect to characterization, is the easy evaluation of their structural parameters by x-ray diffraction. Here, the analysis of the high-quality x-ray rocking curve indicated that no plastic relaxation occurred.¹ An In concentration of 15% and thicknesses of 31 and 90 Å for the $\text{In}_x\text{Ga}_{1-x}\text{As}$ and GaAs layers, respectively, were precisely determined, in fair agreement with the target values. The PC measurements were carried out with a semitransparent gold Schottky front contact and an indium Ohmic contact to the substrate. In both PLE and PC experiments, the excitation was provided by a quartz-halogen lamp dispersed by a monochromator. All the spectra were recorded with the sample at 2 K.

The photoluminescence (PL) and the PLE spectra are shown in Fig. 1(a). The luminescence is dominated by a very narrow line (full width at half maximum, 4 meV) at 1445 meV. There is a low-energy tail, which saturates with increasing excitation intensity, and which is attributed to electron-acceptor recombination, with a small (15 meV) acceptor binding energy⁹ due to the light in-plane effective mass of the heavy-hole band.¹⁰ On the high-energy side, there is a shoulder at about 5 meV above the intense peak. We believe that this high-energy shoulder appearing above the SL absorption edge (see below) betrays the presence of a $\text{In}_x\text{Ga}_{1-x}\text{As}$ well (most likely the first one) about one monolayer thinner than the others. This assignment is also supported by the analysis of the PC spectra. A remarkable feature of the PLE spectrum shown in Fig. 1(a) (recorded with the detection on the electron-acceptor transition at 1425 meV) is the absence of Stokes shift of the exciton with respect to the PL peak, which confirms the excellent quality of the structure. The heavy-hole (hh1-e1) excitonic peak at 1445 meV is followed by an absorption band over about 18 meV, and the onset of the light-hole absorption (lh1-e1) is observed at 1480 meV, also followed by an absorption band. Finally, a large increase of signal occurs around 1510 meV due to the absorption in the GaAs buffer layer.

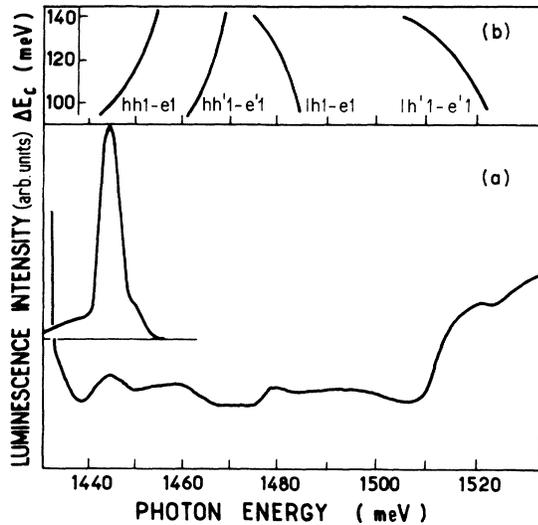


FIG. 1. (a) Low-temperature PL and PLE spectra of our sample, and (b) band-offset dependence of the calculated band structure.

To interpret these data, we calculate the SL band structure using the three-band envelope-function formalism, taking into account the strain in $\text{In}_x\text{Ga}_{1-x}\text{As}$.¹ The careful choice of material parameters is crucial for this system, where the strain induced effects compare in magnitude with the band-gap difference. For GaAs ($\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$), we have used the following values:¹¹

$$S_1 = 2(S_{11} + 2S_{12}) / (S_{11} + S_{12}) = 1.094(0.978)$$

and

$$S_2 = (S_{11} - S_{12}) / (S_{11} + S_{12}) = 1.905(2.022),$$

where the S_{ij} 's are the elastic compliance constants; $a = -9.80$ eV (-9.08 eV) and $b = -1.76$ eV (-1.766 eV) for the hydrostatic and shear deformation potentials; $E_g = 1.519$ eV (1.291 eV) for the unstrained material band gap, and $m_H = 0.37m_0$ ($0.34m_0$) for the heavy-hole mass along (100). The value of Kane's matrix element, chosen to give the correct conduction mass in GaAs, was 24 eV. The evolution of the calculated minibands with the conduction band offset ΔE_c is shown in Fig. 1(b), where the zone-boundary transitions are labeled $hh'1-e'1$ and $lh'1-e'1$, respectively. Clearly, an excellent fit of the data is obtained for $\Delta E_c = 115 \pm 5$ meV, if one takes into account exciton binding energies of the order of 5 meV. This value agrees with the original determination of Marzin *et al.* for multiple quantum wells with the same composition.¹² It corresponds to a type-I heavy-hole band offset of 56 meV and to a type-II configuration for the light holes which are marginally confined in GaAs with a light-hole band offset of -11 meV. The calculated conduction subband width of 17 meV is in excellent agreement with the observation.

The evolution of PC spectra with applied voltage is shown in Fig. 2(a). The flatband PC spectrum obtained at a 0.8-V forward bias coincides with the PLE spectrum. As soon as the bias is reduced from this value, the spectrum changes and new features can be seen: a peak appears and rapidly develops at 1450 meV, i.e., close to the

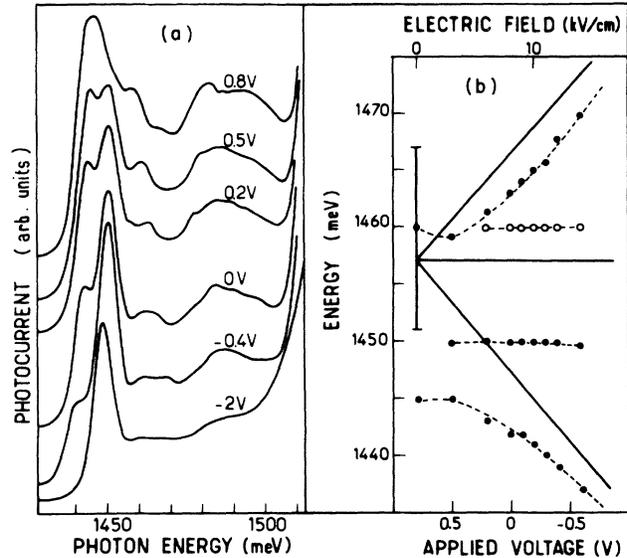


FIG. 2. (a) Low-temperature PC spectra at various applied voltages, and (b) plot of the absorption maxima (dots) vs the applied voltage. The dashed lines are guides to the eye. The solid lines show the calculated transitions between Wannier-Stark states, and the vertical bar the zero-field absorption miniband.

miniband center, while the $hh'1-e'1$ and $hh'1-e'1$ excitons transform into transitions splitting away from the central one and vanishing as the electric field F increases. This behavior is characteristic of the quantization of the energy spectrum into Wannier-Stark ladders, as already observed in GaAs- $\text{Al}_x\text{Ga}_{1-x}\text{As}$ (Refs. 13 and 14) or $\text{Ga}_{0.47}\text{In}_{0.53}\text{As}-\text{Al}_{0.24}\text{Ga}_{0.24}\text{In}_{0.48}\text{As}$ (Ref. 15) SL's. However, the small bandwidth combined with the strain-induced enhancement of the heavy-hole to light-hole splitting make the observation of the zone-edge singularity particularly clear in the present case, as the heavy-hole and light-hole absorption bands do not overlap. In Fig. 2(b), we have plotted the bias dependence of the absorption maxima, and the calculated transitions between Wannier-Stark levels, which obey the simple relation

$$E_p = E_g^{\text{QW}} + p eFd, \quad p = 0, \pm 1, \dots; \quad (1)$$

$E_g^{\text{QW}} = 1457$ meV is the band gap of the isolated quantum well. Note that the zero-field miniband indicated by the vertical bar at 0.8 V (flatband bias) is not exactly centered at E_g^{QW} , as a result of the interaction with upper lying minibands. However, the transitions between Wannier-Stark levels are still precisely given by Eq. (1), as can be checked by a numerical solution of the problem (see below). The calculation shown in Fig. 2(b) is made with a conduction-band offset equal to 120 meV, which turns out to be the value giving the best fit of the whole set of data.

The nearly field independent transition at 1450 meV, which corresponds to the dominant feature in the high-field spectra, is closely fitted by the $p = 0$ "vertical" transition (between Wannier states centered in the same QW), corrected by the 7 meV calculated QW exciton binding energy.¹⁶ The splitting of the $p = \pm 1$ "oblique" transitions (between states centered in adjacent QW's) in the

reverse-bias region should be exactly $2eFd$, as the excitonic corrections on these two symmetrical transitions are expected to be the same. This gives a precise determination of the electric field, which we have used to draw the corresponding theoretical lines in Fig. 2(b). Then, the comparison of theory and experiment indicates that the binding energy of the “oblique” exciton formed by an electron and a hole localized in adjacent QW’s is about 3 meV, which coincides with the calculated value.¹⁶ At lower electric field (positive-bias region), the situation becomes more complicated as the exciton binding energies become field dependent,¹⁷ and as more oblique transitions ($p = \pm 2$, etc.) come into play and finally merge into the zero-field miniband. The open circles near 1460 meV in Fig. 2(b) correspond to the weak but clearly resolved absorption following the dominant “vertical” exciton. The intensity of this absorption is essentially field independent. The 10 meV splitting seems definitely too large for this transition to be the onset of the absorption continuum. On the other hand, the exciton in a 28-Å-thick QW is precisely expected at 1460 meV. We thus argue that the presence of such a QW, narrower than the others by one monolayer, plausibly explains the transition at 1460 meV in the PC spectra, while electron-acceptor recombination in this well would account for the high-energy shoulder in the luminescence.

As can be seen in Fig. 2(a), the behavior of the light-hole absorption band is much less spectacular: the excitonic character of the absorption onset rapidly disappears, while vague structures can be observed in the absorption coefficient. For the largest bias, the light-hole absorption band seems to shrink and it finally vanishes. The Wannier-Stark quantization in a type-II SL is expected to give electro-optical properties qualitatively different from the type-I case:⁴ indeed, in the high-field regime, one ends up with a localized electron optically connected to two hole states in the adjacent QW’s, and separated in energy by eFd . This gives a double-step absorption at the energies $E_g^{\text{QW}} \pm eFd/2$. However, in the present case, the situation is complicated by two factors: (i) The relatively thick GaAs layers and the very small light-hole band offset combine to produce important intrawell corrections (Stark effect). (ii) The gap separating the lh1 and lh2 subbands is very small, so that the electro-optical features associated with the lh1-e1 and lh2-e1 transitions (both are parity allowed) interfere. Furthermore, the band-structure calculation shows that the width of the lh1 miniband is about twice as large as the barrier height. Thus, the qualitative behavior of the spectra, even at zero field, cannot be predicted from a simplified model such as the tight-binding analysis of the envelope functions. To overcome this difficulty, we have directly calculated the electro-optical absorption spectra in this system by solving numerically the Schrödinger equation for the actual potential distribution in the sample in presence of the external bias. The results are shown in Fig. 3 for $F=0, 5$, and 10 kV/cm. The predicted characteristics of the heavy-hole transition are easily recognized in Fig. 3: the absorption at $F=0$ consists of 10 steps closely approaching the miniband profile, and at 10 kV/cm, it reduces to a main step at the QW band-gap energy (sizably distinct from the miniband center), and accompanied by symmetrical satel-

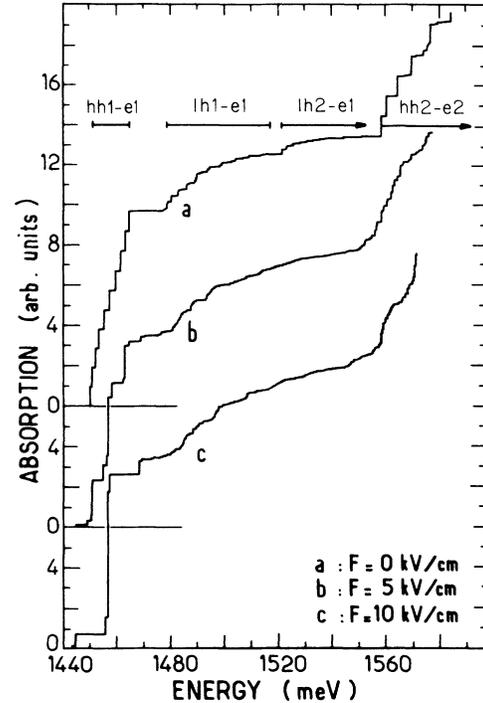


FIG. 3. Theoretical electro-optical absorption spectra calculated by a numerical solution of the Schrödinger equation for $F=0, 5$, and 10 kV/cm.

lites at $E_g^{\text{QW}} \pm eFd$. On the opposite, the lh1-e1 absorption at $F=0$ shows the expected shape for a $\Delta n=0$ transition in a type-II SL,¹⁸ i.e., it is allowed at the zone center and forbidden at the zone boundary, which washes out the singularity in the density of states. The lh1-e1 transition ends at 1517 meV and is followed, at 1522 meV, by the onset of the lh2-e1 transition, which displays the same behavior. When F is increased up to 10 kV/cm, the calculated light-hole absorption shows no spectacular modification, which agrees qualitatively with our observation. This calculation does not take into account the excitonic interaction, which is clearly enhanced by the field-induced localization in the case of the hh1-e1 transition, while it is presumably destroyed by the field-induced ionization in the case of the light-hole transition.

In conclusion, PLE and PC spectroscopies have provided a measurement of the first electron miniband width in a strained-layer $\text{In}_{0.15}\text{Ga}_{0.85}\text{As-GaAs}$ SL, allowing a precise determination of the controversial electronic properties of this system. The absence of overlap between the heavy-hole and light-hole absorption bands permits a clear observation of the zone-edge “saddle-point” exciton and of its electric-field dependence. In the large field regime, we find both experimentally and theoretically that the binding energy of the “oblique” exciton is close to 3 meV, while that of the “vertical” exciton is 7 meV. While the type-II nature of the light-hole band lineup in this system is confirmed by our results, the light-hole confinement is too weak to allow the observation of the specific electro-optical properties for type-II SL’s, as clearly shown by the numerical calculation of the electro-optical absorption spectra.

We thank M. C. Joncourt from the Centre National d'Etudes des Telecommunications for the x-ray evaluation of the sample and M. Voos for his critical reading of the manuscript. This work has been partly supported by NATO through a Cooperative Research Grant No. RG.0709/87. One of us (B.S.) is supported by the Direction des Recherches, Etudes et Techniques and another one of us (R.F.) by Coordenação de Aperfeiçoamento de Pessoal do Ensino Superior-Brazil.

-
- ¹For a review, see J. Y. Marzin, J. M. Gérard, P. Voisin, and J. A. Brum, in *Semiconductors and Semimetals*, edited by T. Pearsal (Academic, New York, 1990), Vol. 32.
- ²D. J. Arrent, K. Deneffe, C. Van Hoop, J. de Boek, and G. Borhs, *J. Appl. Phys.* **66**, 1739 (1989).
- ³M. J. Joyce, M. J. Johnson, M. Gal, and B. F. Usher, *Phys. Rev. B* **38**, 10978 (1988); see also A. Ksendzov *et al.*, in *Proceedings of the Fourth International Conference on Modulated Semiconductor Structures, Ann Arbor, MI, 1989*, edited by R. Merlin [*Surf. Sci.* (to be published)]; X. M. Fang *et al.*, *ibid.*
- ⁴J. Bleuse, G. Bastard, and P. Voisin, *Phys. Rev. Lett.* **60**, 220 (1988).
- ⁵H. Chu and Y. C. Chang, *Phys. Rev. B* **36**, 2946 (1987).
- ⁶D. A. B. Miller, J. S. Weiner, and D. S. Chemla, *IEEE J. Quantum Electron.* **QE-22**, 1816 (1986).
- ⁷P. W. Yu, G. D. Sanders, K. R. Evans, D. C. Reynolds, K. K. Bajaj, C. E. Stuz, and R. L. Jones, *Appl. Phys. Lett.* **54**, 2230 (1989).
- ⁸E. Fortin, B. Y. Hua, and A. P. Roth, *Phys. Rev. B* **39**, 10887 (1989).
- ⁹A. P. Roth, D. Morris, R. A. Masut, C. Lacelle, and J. A. Jackman, *Phys. Rev. B* **38**, 7877 (1988).
- ¹⁰G. C. Osbourn, J. E. Shirber, T. J. Drummond, L. R. Dawson, B. L. Doyle, and I. J. Fritz, *Appl. Phys. Lett.* **49**, 12 (1986).
- ¹¹*Landolt-Börnstein: Numerical Data and Functional Relationship in Science and Technology*, edited by O. Madelung (Springer-Verlag, Berlin, 1982), Group. 3, Vol. 17, Part A; *Landolt-Börnstein Numerical Data and Functional Relationship in Science and Technology*, edited by O. Madelung (Springer-Verlag, Berlin, 1987), Group. 3, Vol. 22, Part A.
- ¹²J. Y. Marzin, M. N. Charasse, and B. Sermage, *Phys. Rev. B* **31**, 8298 (1985).
- ¹³E. E. Mendez, F. Agullo-Rueda, and J. M. Hong, *Phys. Rev. Lett.* **60**, 2426 (1988).
- ¹⁴P. Voisin, J. Bleuse, C. Bouche, S. Gaillard, C. Alibert, and A. Regreny, *Phys. Rev. Lett.* **61**, 1639 (1988).
- ¹⁵J. Bleuse, P. Voisin, M. Avollon, and M. Quillec, *Appl. Phys. Lett.* **53**, 2632 (1988).
- ¹⁶We calculate QW-exciton binding energies using a variational method similar to that described by J. A. Brum and G. Bastard, *J. Phys. C* **18**, L789 (1985).
- ¹⁷R. H. Yan, F. Laruelle, and L. A. Coldren, *Appl. Phys. Lett.* **55**, 2002 (1989).
- ¹⁸P. Voisin, G. Bastard, and M. Voos, *Phys. Rev. B* **29**, 935 (1984).