# Excitons in ultrathin InAs/InP quantum wells: Interplay between extended and localized states

P. Paki and R. Leonelli[a)]
*Département de Physique et Groupe de Recherche en Physique et Technologie des Couches Minces (GCM), Université de Montréal, Case Postale 6128, Succursale Centre-ville, Montréal, Québec H3C 3J7, Canada*

L. Isnard and R. A. Masut
*Département de Génie Physique et Groupe de Recherche en Physique et Technologie des Couches Minces (GCM), Ecole Polytechnique, Case Postale 6079, Succursale Centre-ville, Montréal, Québec H3C 3A7, Canada*

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We have performed detailed optical measurements of ultrathin InAs/InP quantum wells grown by metalorganic vapor phase epitaxy. Absorption and photoluminescence excitation spectra reveal the excitonic resonances associated with two- and three-monolayer-thick InAs layers. Photoluminescence spectra also show an emission band at intermediate photon energies which is associated with excitons localized in thin InAs quantum dots. Polarization-dependent measurements clearly show the heavy-hole or light-hole nature of the quantum well resonances. Such an identification of both type of transitions provides a test for electronic structure models. We find that the energy positions of the excitonic transitions in ultrathin InAs/InP quantum wells are not consistent with calculations based on the envelope function model. © *2000 American Vacuum Society*. [S0734-2101(00)01903-5]

Ultrathin InAs-based quantum wells (QW) are characterized by high radiative efficiencies and thus have significant potential for optoelectronic devices.[1,2] Moreover, since their optical properties are strongly affected by the presence of monolayer width fluctuations, they can serve as test structures to study exciton localization.[3–12] However, the electronic states in ultrathin QWs are still not well understood. They are usually described with effective mass models within the envelope-function approximation (EFA). But EFA is highly questionable in the case of QWs which are only a few monolayers (ML) thick,[13] as indicated by recent tight-binding calculations on the InAs/GaAs system which depart significantly from the EFA approach.[14] Nevertheless, all experimental results on InAs ultrathin QWs have been interpreted to date using EFA models. In the case of the InAs/GaAs system, the energy position of both heavy-hole (hh) and light-hole (lh) excitonic resonances agree with the predictions of EFA models.[4,7,12] However, because of the small value of the critical thickness induced by the high lattice mismatch between InAs and GaAs, these measurements were performed in the limited range of 0–1.6 ML. In this respect, the InAs/InP system is more interesting since InAs layers can in principle be grown pseudomorphically on InP with thicknesses of up to eight MLs. The photoluminescence (PL) spectra of this system generally show multiple lines which are associated with InAs layers of integer ML thicknesses.[3,6,10,11,15,16] The energy position of heavy-hole excitonic transitions deduced from the PL spectra can also be well reproduced by EFA. Emission lines in PL spectra give, however, only a first approximation to the position of excitonic transitions since interface roughness can result in significant Stokes shift in ultrathin QWs.

In this article, we present detailed optical measurements of InAs/InP single QWs. The samples were grown by low-pressure metallorganic vapor phase epitaxy on (001)-oriented semi-insulating InP:Fe. The growth procedure involved the deposition of InAs layers between a 300-nm-thick InP buffer and a 120-nm-thick InP cap layer. Tertiarybutylarsine (TBAs), trimethylindium (TMI), and pure phosphine were used as source materials. All the samples were grown under a reactor pressure of 40 Torr and at a substrate temperature of 600 °C. More details of the system and growth procedure have been reported in Refs. 17 and 18.

The thickness of the InAs layers can be accurately determined from the relative positions between the main InP Bragg peak and the Pendellösung fringes observed in high-resolution x-ray-diffraction (HRXRD) measurements.[4,6,19] HRXRD was performed with a Philips five-crystal diffractometer. The (004) rocking curves were analyzed assuming a tetragonal deformation[19,20] and no relaxation of InAs. They gave average thicknesses of $0.7\pm0.1$ nm for sample A and $0.8\pm0.1$ nm for sample B, in agreement with the values expected from the growth conditions. Within the continuum elastic theory, the InAs lattice parameter along the growth axis is 0.626 nm, yielding nominal thicknesses of $2.2\pm0.3$ and $2.6\pm0.3$ MLs for samples A and B, respectively.

For the optical measurements, the samples were mounted strain free in a helium flow cryostat and cooled to 5 K. The PL and PLE spectra were obtained using a Ti:sapphire laser tunable in the range 1.18–1.38 eV. It thus allowed us to study the excitonic resonances of InAs QWs having thick-

a)Author to whom correspondence should be addressed; electronic mail: richard.leonelli@umontreal.ca.

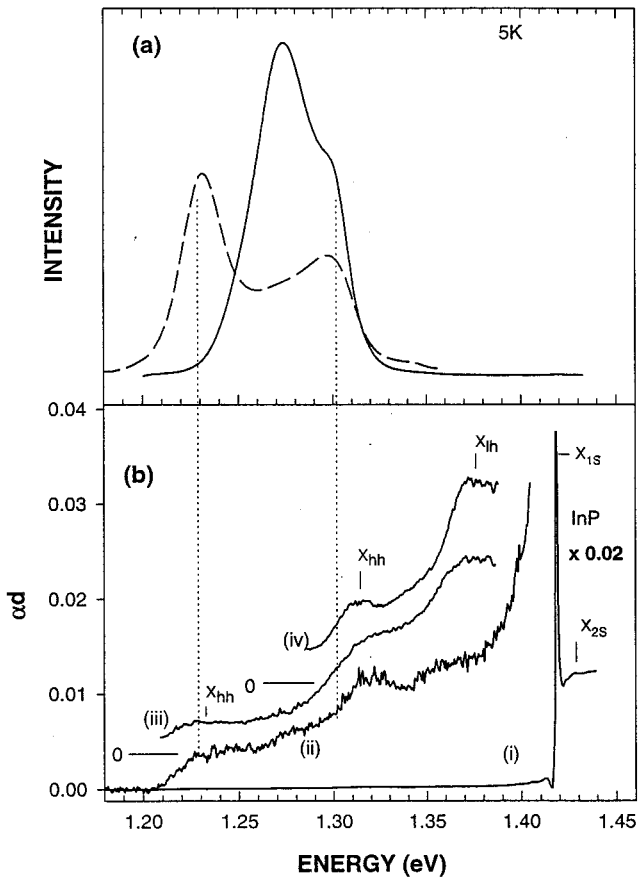


FIG. 1. Panel (a) shows the PL spectra from sample A (solid line) and sample B (dashed line). Panel (b) shows the absorption spectrum from sample B [curve (i)], its magnification [curve (ii)], and the PLE spectra from samples B [curve (iii)] and A [curve (iv)]. The spectrometer was set at 1.19 and 1.26 eV for curves (iii) and (iv), respectively.

nesses up to three MLs.[6] PLE spectra were measured in the standard configuration, where the exciting laser light impinges on the front surface of the sample, and in the edge-excitation mode, where the laser is focused on a cleaved edge of the sample and the emission is collected from the surface close to the edge. The PL was analyzed with a 1 m spectrometer and detected by a liquid-nitrogen-cooled Ge photodiode using conventional lock-in techniques. The absorption spectra were obtained with a Fourier transform interferometer. The time-resolved pholuminescence (TRPL) experiments were performed using a mode-locked Ti:sapphire laser operating at a wavelength of 860 nm. The TRPL signal was detected with a cooled InGaAs photomultiplier using the time-correlated photon counting technique.

The PL spectra of the samples are shown in Fig. 1(a). The spectrum of sample A is dominated by a broad emission at 1.27 eV followed by a shoulder at 1.30 eV, while the spectrum of sample B shows two peaks centered at 1.23 and 1.30 eV. Curves (i) and (ii) of Fig. 1(b) show the absorption spectrum of sample B.[21] The two main structures of the spectrum, namely an absorption edge at 1.23 eV and a resonance at 1.32 eV, are present in the standard PLE spectrum of this sample [curve (iii)], which also reveals another resonance near 1.37 eV. Moreover, the two high energy resonances *are also observed in the PLE spectrum of sample A* [curve (iv)]. These resonances have full width at half maximum (FWHM) of $\sim 20$ meV.

Confining potential fluctuations naturally occur in ultrathin quantum wells. These fluctuations arise from 1-ML-thick islands randomly formed at the interfaces. Islands whose characteristic size is much smaller than the exciton Bohr radius broaden the excitonic resonances[22] while islands with sizes comparable to the exciton Bohr radius form quantum dots (QDs) which localize the excitons.[23] Such simple considerations are sufficient to explain our PL, PLE, and absorption spectra. The shoulder at 1.30 eV on the PL spectrum of sample A is Stokes shifted by 20 meV from the corresponding resonance observed in the PLE spectrum. Given the nominal thickness of 2.2 ML of sample A, we assign the resonance at 1.32 eV to heavy-hole free exciton emission ($X_{hh}$) from a 2 ML InAs QW, while the PLE peak at 1.37 eV is attributed to the light-hole exciton resonance ($X_{lh}$). The more intense emission at 1.28 eV is associated with 3-ML-thick QDs on a 2 ML QW. The fact that these two resonances are also observed in the spectra of sample B (nominal thickness of 2.6 ML) indicates that 2-ML-thick InAs layers are present in this sample. However, the presence of 3 ML regions in sample B is evidenced by the absorption edge at 1.23 eV and its associated emission band which thus corresponds to 3 ML $X_{hh}$ transitions.

The oscillator strength per unit surface of excitonic resonances in QWs can be obtained from[4]

$$f = \frac{4m_0\varepsilon_0 nc}{e^2\hbar\pi}\int p(\hbar\omega)d(\hbar\omega), \tag{1}$$

where $n$ is the index of refraction and $p$ is the absorption probability given by

$$p = 1 - T \approx \alpha d. \tag{2}$$

The oscillator strength of the 2 ML $X_{hh}$ resonance obtained numerically from the absorption spectrum of Fig. 1 is $f \sim 4 \times 10^{12}$ cm$^{-2}$. This value, which is necessarily an underestimate since the 2 ML QW coverage in sample B is smaller than one, is close to what is observed in standard GaAs/AlGaAs QWs.[24]

Information on exciton dynamics in sample B was obtained from TRPL experiments. Figure 2 shows the decay times of the 1.23 eV band and the 1.28 eV band as a function of temperature. The gradual increase of the decay time of the 1.23 eV band is characteristic of delocalized excitons in quantum wells, while the near constant decay time of the 1.28 eV band corresponds to what is expected from localized, isolated excitons.[6]

We have also performed polarization-dependent PLE measurements since the hh and lh character of the optical transitions in QWs is experimentally accessible on the basis of their polarization dependence. In particular, the oscillator strength of heavy- and light-hole related transitions is predicted to be $\frac{3}{4}$ and $\frac{1}{4}$ for excitation polarized perpendicular to the growth axis and 0 and 1 for a parallel polarization.[4,25,26]

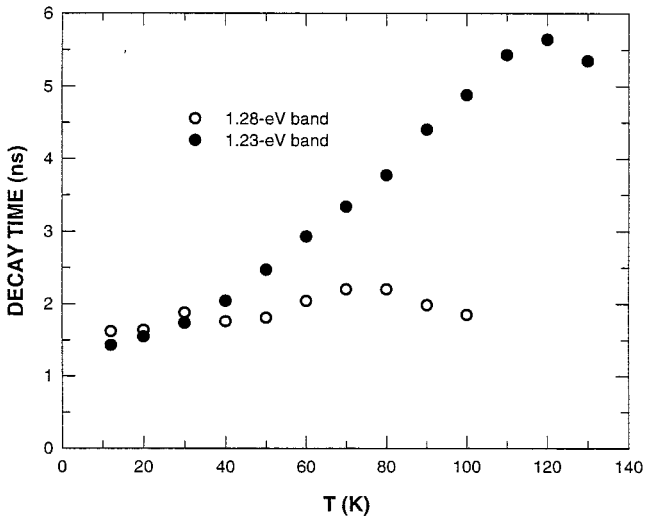


FIG. 2. Average emission decay time for sample B as a function of temperature.

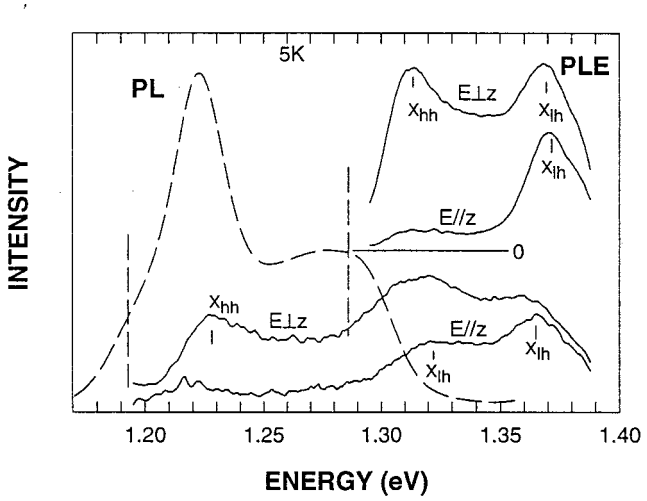


FIG. 4. PL and polarization-dependent PLE spectra from sample B. The two upper PLE curves were taken with the spectrometer set at 1.29 eV, while the two lower PLE curves were taken at 1.19 eV.

In Fig. 3, we present polarization-dependent PLE spectra of sample A obtained in the edge-excitation mode. It is to be noted that, in this configuration, the onset of absorption from the semi-insulating InP substrate reduces the high photon energy signal with respect to what is measured in the standard configuration. Nevertheless, as is evident from the spectra, the transition assigned to $X_{\mathrm{hh}}$ does in fact exhibit a distinct heavy-hole character.

Figure 4 shows polarization-dependent PLE spectra from sample B. The two upper PLE spectra were obtained with the spectrometer set at 1.29 eV. They are similar to the spectra of Fig. 3 and thus they confirm the presence in sample B of InAs QDs on 2 ML QWs. The two lower curves were obtained with the spectrometer set at 1.19 eV. In the edge-excitation mode, the exciton resonance at 1.23 eV is more apparent and its hh character confirmed by an eightfold reduction in intensity between the perpendicular and parallel configurations. On the other hand, the strength of the resonance at 1.32 eV decreases only by a factor of 2. This result unambiguously shows that the $X_{\mathrm{lh}}$ resonance of 3 ML QWs is also located near 1.32 eV and thus nearly degenerate with the $X_{\mathrm{hh}}$ resonance of 2 ML QWs.

In Fig. 5, these results are compared to standard EFA calculations including strain effects using the eight-band $\mathbf{k}\cdot\mathbf{p}$ model,[27,28] a square confining potential, and the material parameters given in Ref. 29. It is to be noted that the standard EFA model assumes that the material parameters and the gap offset are independent of well thickness. Estimated values found in the literature for the conduction band offset $\Delta E_c$ in the InAs/InP or $\mathrm{InAs}_x P_{1-x}$/InP systems are either close to 70%[6,29,30] or to 40%.[15,31] EFA calculations performed using both values of $\Delta E_c$ are presented in Fig. 4. Neither band offset value nor any intermediate one can satisfactorily reproduce our experimental results. The strong

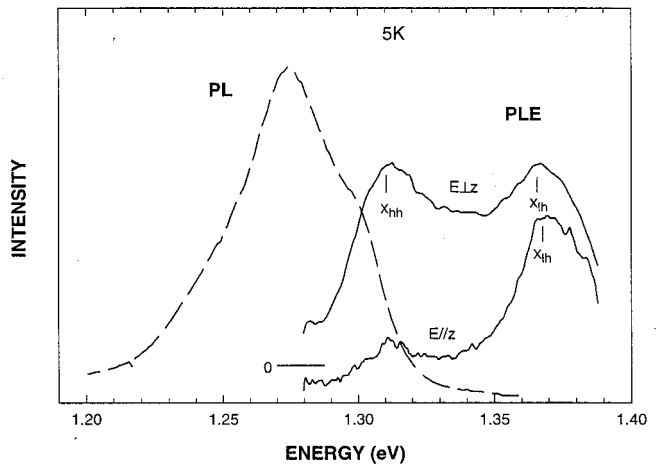


FIG. 3. PL and polarization-dependent PLE spectra from sample A. The curve with $E\perp z$ has been shifted for clarity.

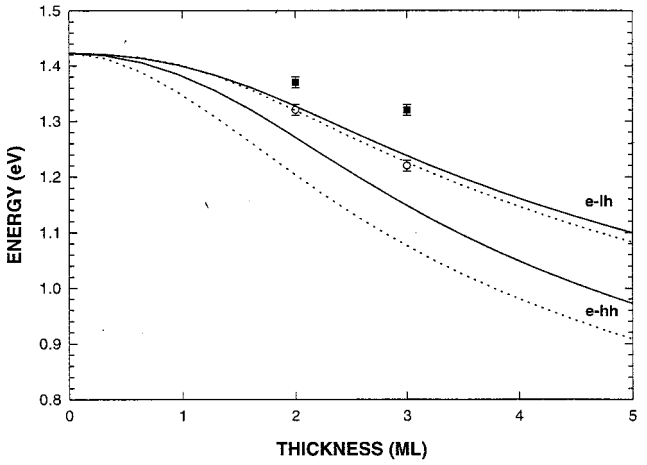


FIG. 5. Transition energy as a function of well thickness for samples A and B. Circles: $X_{\mathrm{hh}}$; squares: $X_{\mathrm{lh}}$. Also plotted are the calculated dependence using EFA with $\Delta E_c = 75\%$ (solid line) and $\Delta E_c = 40\%$ (dotted line).

constraint imposed by the values of the energy positions of both $X_{hh}$ and $X_{lh}$ thus allows us to conclude that, contrary to what was previously found on the basis of PL spectra alone, standard EFA calculations cannot reproduce the energy positions of the excitonic resonances observed in our samples.

In summary, we have studied the excitonic resonances and determined the physical origin of the PL signal in ultrathin InAs/InP single quantum wells. Depending on coverage, hh-exciton transitions from 2 or 3 ML quantum wells can be observed together with the emission from excitons localized in quantum dots. The heavy- and light-hole character of the quantum well transitions is revealed by selective excitation of the states by linearly polarized light. Such an identification of heavy-hole and light-hole excitonic resonances provides a test for electronic structure calculations. We find that standard effective-mass models based on the envelope function approximation are not appropriate to describe the electronic structure of our samples.

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