Microphotoluminescence spectroscopy of vertically stacked $In_xGa_{1-x}As/GaAs$ quantum wires

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Disorder and spectral broadening of vertically stacked InGaAs/GaAs V-grooved quantum wires have been investigated by means of microprobe luminescence. We show that the main spectral broadening mechanism originates from monolayer fluctuations at the bottom of the wire. A direct evidence of monolayer height islands of area 40×40 nm formed at the bottom of the grooves is provided by atomic force microscopy. Lateral and vertical wire-to-wire fluctuations are found to be negligible on the micron scale. [S0163-1829(98)06827-1]

I. INTRODUCTION

Size fluctuations and inhomogeneous size distribution cause important broadening in the optical spectra of nanostructures, particularly in conventional spatially integrated experiments, in which a large laser spot, of typical 100 μm diameter, excites simultaneously thousands of wires and dots. Broadening of the order of 10 meV or more are usually obtained even in state of the art nanostructures having size fluctuations reduced to less than 10%. Microprobe spectroscopy is the method of choice for the selective investigation of very few (eventually single) nanostructures. This allows one to drastically reduce the effects of the size distribution and to study the optical transitions of individual wires and dots, thus measuring the intrinsic linewidth of the structure. The resulting optical spectra are therefore dominated by the homogeneous linewidth of the state and by the size fluctuation occurring in the profile of the individual nanostructure. For the specific case of quantum dots it is nowadays demonstrated that the single dot spectrum obtained by microphotoluminescence (MPL) spectroscopy exhibits atomiclike lines with sub-meV linewidth whereas the conventional photoluminescence (PL) spectra exhibit broad bands given by the envelope of the emission spectra of many dots.[1] In quantum wires the situation is somewhat more complicated than in quantum dots. In addition to the wire-to-wire fluctuations, one has to consider the presence of one degree of freedom for the electron motion which allows the diffusion of carriers in the wires on the micron scale.[2] In this work we have studied high-quality vertically stacked V-shaped InGaAs/GaAs quantum wires by submicron spatially resolved luminescence. The aim of the work is to elucidate the impact of wire to wire fluctuations (either vertical and lateral) and of diffusion on the broadening of the optical spectra. The experimental details are described in Sec. II. The results are presented in Sec. III and discussed in Sec. IV. In Sec. V we draw our conclusions.

II. EXPERIMENTAL

The samples investigated in this paper were vertically stacked InGaAs/GaAs quantum wires grown by subatmospheric pressure metal-organic chemical vapor deposition (MOCVD) on patterned (100) undoped GaAs. The substrates were patterned by holographic lithography followed by anisotropic wet chemical etching in a 0.75 mol/liter $KHCO_3$:H_2O_2 solution (1:1). The resulting surface consisted of an array of V-shaped grooves parallel to the [01-1] crystallographic direction with pitch periodicity (lateral periodicity) of 700 nm and depth of about 250 nm. The angle of the grooves was varied between 90° and 130° [corresponding to (111)*A* and (411)*A* crystallographic planes] by a suitable choice of the etching solution. This allowed us to fabricate V-shaped quantum wires either with crescent profiles or with almost constant thickness. The nominal width of the $In_{0.10}Ga_{0.90}As$ layers and GaAs barriers was 6 and 20 nm, respectively. A typical cross section of the wires is shown in Fig. 1. Confinement occurs at the vertex of the V-shaped structure due to the discontinuity in the profile and to the local increase of thickness at the vertex. The structure was cladded by a 0.5 μm thick GaAs layer. Single InGaAs wires were also grown for reference. Due to the small In content these quantum wires are rather shallow. The lateral confinement potential is of the order of 60 meV for the electrons, and the intersubband splitting is of the order of 6 meV. However, the excellent morphology of these structures results in extremely low injection current for the quantum wire electroluminescence (below 300 $\mu A/cm^2$) and in very narrow luminescence linewidth [below 6 meV (Ref. 3)] at low temperatures.[4] This makes these samples very interesting systems for the study of disorder by microprobe spectroscopy. Larger subband splitting [up to 25 meV (Refs. 4–7)] and deep confinement potential were obtained in wires with larger In content. However, under this condition we have clear evidence that In interdiffusion and alloy broadening are the main disorder sources, and it is very difficult to investigate selectively the effects of the wire to wire fluctuations.

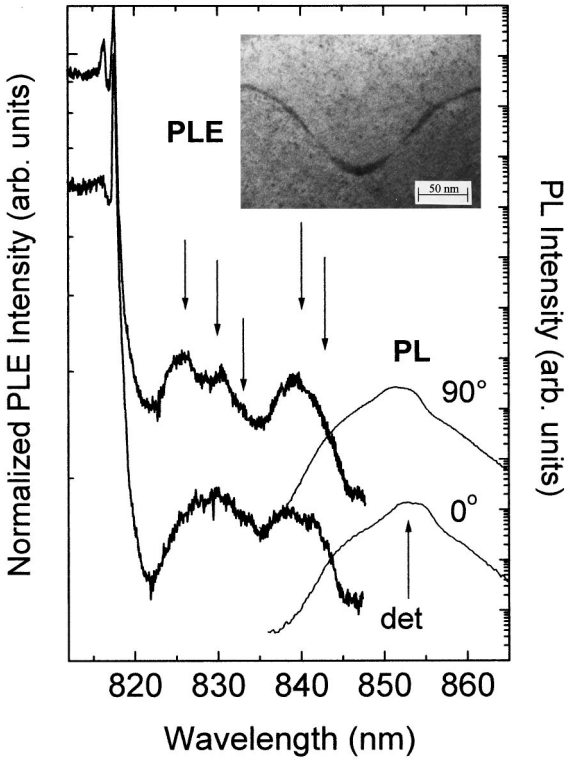

FIG. 1. Spatially integrated PLE and PL spectra recorded for polarization parallel and perpendicular to the wire axis. The temperature is 10 K. The arrows indicate the quantized states. Inset: TEM cross section of the wires.

The microphotoluminescence setup consisted of a 50× objective (numerical aperture 0.42) and a collecting lens system providing a spatial resolution of 0.8 μm under He-Ne excitation. The spatial resolution was just enough to excite the individual grooves of periodicity 0.7 μm, containing one or more vertically stacked dots. The collected luminescence was analyzed by a monochromator coupled to an InGaAs photomultiplier tube. A 30 μm pin hole in the image plane of the system was scanned across the magnified image of the luminescence spot with a resolution of 0.3 μm in order to probe the density profile across the excited area and the diffusion of carriers far away from the spot center. The focusing system was also equipped with a high precision translator (resolution 0.5 μm) allowing us to laterally map the luminescence originating from neighboring grooves. The spectral resolution was always better than 0.6 meV. The samples were kept at 10 K in a vibration insulated closed cycle cryostat.[2]

The samples were excited with the light propagation vector parallel (top excitation) or perpendicular to the growth axis (edge excitation). The latter configuration has been obtained by exciting the sample on the cleaved edge. In all measurements the power density was kept carefully constant (equivalent to about 100 W cm^{-2}) so that the energy position of the luminescence energy was not changed by the screening of the internal piezoelectric field (causing a well resolved blueshift with increasing carrier density).[5]

III. RESULTS

In Fig. 1 we show the photoluminescence excitation (PLE) spectra of the vertically stacked wires taken for polarization parallel and perpendicular to the wire axis. The spectra exhibit a sharp GaAs exciton from the barrier layer, and a weaker absorption continuum in the quantum wire region with distinct quantum wire resonances. The $n=1$ and $n=2$ transitions of the wires were deconvoluted by a Gaussian line-shape fitting procedure, and were found to have a splitting of about 5 meV. Furthermore, they exhibit about 15% relative intensity variation when the laser polarization is changed from parallel to perpendicular to the wire axis. Such polarization is due to the well known mixing of the valence states induced by the lateral $k_y=\pi/d$ momentum shared at $k_z=0$ and is usually considered as a direct consequence of the one-dimensional properties of the structure. The spectral broadening of the PL spectra amounts to about 6 meV at 10 K, and is due to both the wire-to-wire fluctuations and to the microscopic disorder within each individual wire. In our structures, we have to distinguish between the lateral size inhomogeneity (size fluctuations of wires in adjacent groves) and vertical inhomogeneity (differences between stacked wires in the same groove).

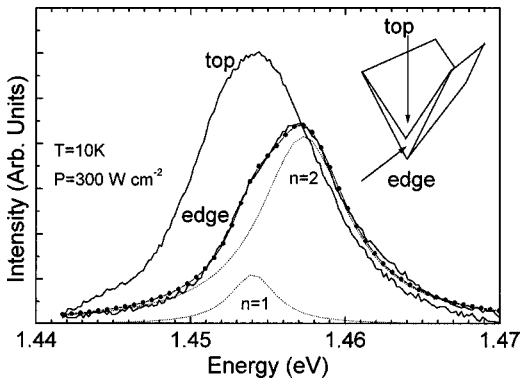

FIG. 2. MPL spectra recorded under edge or top excitation. The dotted curves represent the Lorentzian deconvolution of the MPL spectrum collected under edge excitation. The dashed-dotted curve is the total line-shape fitting. The temperature is 10 K.

The results of the MPL experiments are exemplified in Fig. 2, where we compare the luminescence spectra of the vertically stacked wires taken under top and edge excitation. The main results are as follows.

(i) There is no substantial narrowing of the MPL linewidth even though the laser spot excites a single groove. The spectra indeed exhibit the same linewidth of the spatially integrated PL, both under top and edge excitation. We should mention that clear single dots lines were observed in a reference sample containing luminescent quantum dots[8] under identical excitation and alignment conditions, thus ruling out possible malfunctions of the microprobe setup.

(ii) There is no change in the MPL linewidth of the single and vertically stacked wires (see Fig. 3).

(iii) A clear shift of the MPL band is seen depending on the propagation direction of the exciting beam (top or edge excitation). In particular in edge configuration the luminescence of the $n=2$ excited state is dominant (this point will be discussed at the end of Sec. IV).

Points (i) and (ii) indicate that both the lateral and the vertical wire-to-wire fluctuations are irrelevant on the micron scale. This is clearly demonstrated in Fig. 3 where we show the energy position and the linewidth of the MPL taken from

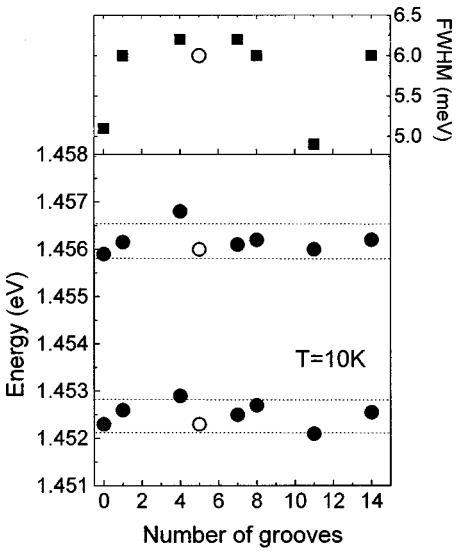

FIG. 3. Energy of the $n=1$ and $n=2$ transitions of the wires (full dots in the bottom panel and left axis) measured in different individual grooves by scanning the sample surface with the microprobe setup. The empty dots represent the energy position of the spatially integrated luminescence bands. The top panel displays the full width at half maximum (FWHM) of the ground level emission band measured in each individual groove (full squares and right axis). The empty symbol is the FWHM of the conventional PL.

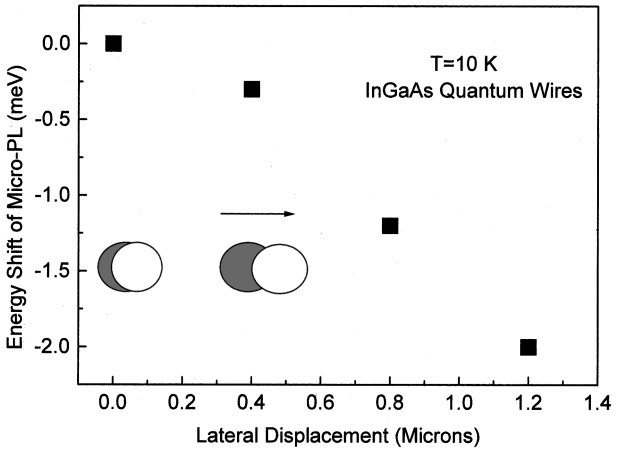

FIG. 4. Redshift of the quantum wire ground level emission measured by scanning the detection pin hole (empty circle in the inset) far away from the center of the excitation spot (full circle in the inset) along the infinite quantum wire axis.

different individual grooves of a single wire structure (under top excitation). The scanning resolution of 0.5 μm allowed us to probe adjacent individual grooves as well as distant grooves (up to about 15 grooves far away from the origin). The energy position of the first and second quantum wire subbands were obtained by the deconvolution of the MPL spectra (see Fig. 2). In most cases a Lorentzian line-shape fitting was found to reproduce quite well the MPL bands.[9] Both the $n=1$ and $n=2$ states exhibit a variation of less than 1 meV over several tens of investigated grooves. Similarly the full width at half maximum (FWHM) of the single wires ranges between 5 and 6.2 meV (top panel of Fig. 3). The measured FWHM values are very similar to those obtained in the vertically stacked wires or by the conventional spatially integrated PL (empty symbols in Fig. 3) demonstrating the excellent lateral homogeneity of the investigated samples. We can conclude that the wire-to-wire fluctuations should introduce a spectral broadening of the order of 1 meV in the spatially integrated PL spectra.

Another important piece of information obtained by the MPL concerns diffusion. The possibility of mapping the emission spectrum across the excitation area allowed us to probe the diffusion of carriers through the spread of the luminescence. The main results about this point are as follows.

(iv) A clear redshift of the luminescence was observed when the pin hole was displaced far away from the spot center (see Fig. 4).

(v) An overall redshift of the luminescence was observed without a pin hole in the image plane.

Point (iv) indicates that carriers diffuse along the infinite axis of the wire, as already clearly observed in Ref. 2. Assuming a mean thermal velocity of carriers of 10^6 cm/sec and a recombination time of 350 ps (measured in Ref. 7) we obtain an upper limit for the diffusion length of electrons of the order of 3.5 μm. This value reduces to about 1 μm if we account for the heavy-hole diffusion and the eventual formation of excitons. This is clearly seen by displacing the pin hole far away from the spot center along the wire axis, where the carrier density reduces progressively due to diffusion. Under this condition the screening of the internal piezoelectric field is also reduced and the luminescence is redshifted due to the stronger Stark shift induced by the built-in field. For displacements of about 1 μm away from the spot center we indeed measure a redshift of the MPL of about 2 meV. Moreover, removing the pin hole from the image plane allows us to collect all the luminescence signal from the center of the spot and from the diffusion region. The overall shift of the luminescence in this case corresponds to about 1.5 meV, and is related to the average density of carriers distributed across the diffusion area.

These results indicate that diffusion along the wire axis spreads the carrier distribution over an area which is larger than the excitation spot. In the diffusion process carriers undergo localization at disorder, resulting in a broadening of the optical spectra, somehow limiting the effective spatial resolution of the experiment. In order to suppress the effect of diffusion, we have also fabricated isolated square mesas of submicron size by lithography and wet chemical etching. Since the size of the mesa was comparable to the laser spot, a homogeneous carrier density without diffusion was obtained under optical excitation. Under this condition the luminescence spectra again showed a peak at the ground level energy with a linewidth of the order of 6 meV. This experiment confirmed that the spectral broadening does not originate from the localization of carriers spread by the diffusion in different regions of the quantum wires.

IV. DISCUSSION

The results presented in the previous section unambiguously indicate that (1) lateral and vertical wire-to-wire fluctuations are not responsible for the spectral broadening of the quantum wire luminescence and (2) diffusion does not influence the spectral broadening of the emission. This suggests that the actual disorder occurs on a scale length considerably

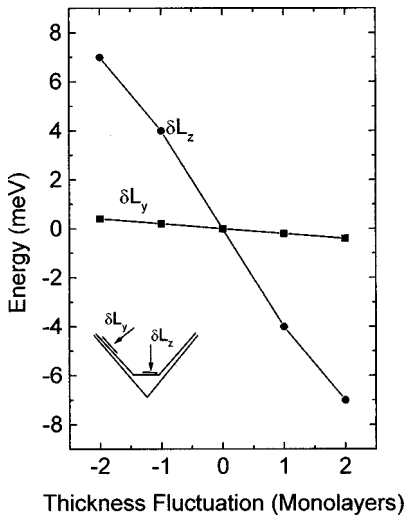

FIG. 5. Spectral broadening of the luminescence versus the thickness fluctuation (in number of monolayers) at the bottom of the groove (circles) and along the side walls (squares). The inset shows schematically the thickness fluctuations on the wire cross section.

shorter than 1 μm in quantum wires, so that submicron luminescence of individual grooves does not provide a reliable identification of the disorder mechanism. Compositional disorder has been carefully studied in our InGaAs quantum wells, and is found to be controlled within $\pm 0.1\%$ in the range $x=0.1$.[3] This is supported by the observation of typical linewidths below 2 meV in the absorption and luminescence spectra of our $In_{0.1}Ga_{0.9}As$ quantum wells. Moreover, our results have been confirmed by additional MPL observations performed on other InGaAs wires and on GaAs wires grown on patterned substrates and/or on SiO_2 matrices.[5,10] Therefore other broadening mechanisms such as the thickness fluctuations in the single wire profile have to be accounted for in order to understand our results. Since the lateral potential profile originates from the local variation of thickness occurring at the bottom of the groove with respect to the side walls, we can write a simple one-dimensional model in which the lateral potential $V(y)=f[L_z(y)]$ is a function of the thickness of the bent quantum well at different lateral positions in the grooves $[L_z(y)$, y being the lateral coordinate]. The fluctuations in the wire profile are reflected in random variations of L_z at different y positions $[\delta L_z(y)$, see inset of Fig. 5]. The disorder induced change in the potential is thus given by

$$V_{\mathrm{dis}}(y)=V(y)\pm\frac{dV(y)}{dL_z}\,\delta L_z(y). \tag{1}$$

Putting V_{dis} in the Schrödinger equation we obtain the variation of the ground level eigenstates due to the size fluctuations. In order to further simplify the calculation we separate the effect of size fluctuations at the bottom of the groove $(y=0)$ and along the side walls $(y\neq 0)$. For $y=0$, the problem is equivalent to the well known case of the well width fluctuations along the growth axis. For δL_y variations occurring at lateral y positions in the groove, we note that these cause local variations of the lateral potential height, which in turn cause only small variations of the ground level. A rough estimate of these contributions is given in Fig. 5, for δL_z values of the order of 1–4 monolayers (i.e., less than 1.2 nm thickness fluctuation) occurring at the vertex of the groove $(y=0$, dots) and along the side walls $(y>0$, squares). Clearly the effect of disorder at the bottom of the groove is more relevant as compared to fluctuations in the V profile along the side walls. Though with a strong approximation, we might expect that 2 monolayers fluctuations in the wire thickness at the bottom of the groove originate a spectral broadening of the order of 6 meV, i.e., comparable to the measured value. Similar thickness variations along the sidewalls would affect the MPL linewidth much less. These results indicate that broadening in the optical spectra of our quantum wires mainly comes from thickness fluctuations of few monolayers height and lateral extension well below 1 μm, occurring at the bottom of the bent quantum well.[11] The existence of monolayer steps is inherent to the step-flow nature of the MOCVD growth adopted for the fabrication of our wires. In order to confirm our assumptions we have thus performed atomic resolution atomic force microscopy (AFM) experiments on a single quantum wire structure grown without the topmost GaAs barrier. Under this condition we have directly studied the monolayer planarity of the InGaAs wire at the bottom of the groove. Atomic force microscopy measurements were taken on the surface of a single $In_{0.1}Ga_{0.9}As$ wire capped by only 1 nm of GaAs to avoid surface reconstruction. Measurements performed with atomic

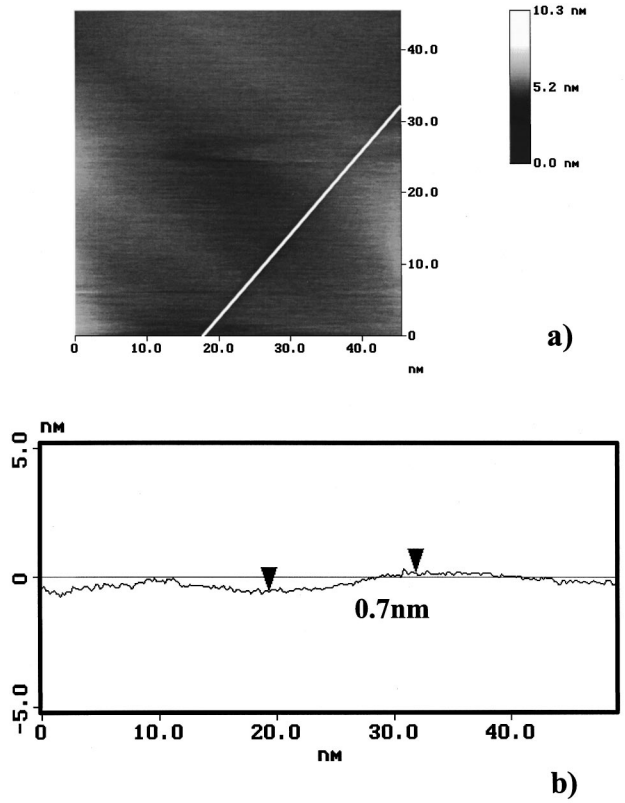

FIG. 6. Surface topography of the bottom of a single InGaAs quantum wire. The measurement was performed in atomic resolution by a Digital Nanoscope IIIa microscope equipped with an *A* scanner (*X* sensibility 2.757 nm/V, *Y* sensibility 3.113 nm/V, *Z* sensibility 1.77 nm/V). The cross section profile (b) reveals the formation of distinct islands of monolayer height extending over about 40 nm×40 nm.

resolution evidence the presence of 1 and 2 monolayers high islands in the bottom of the grooves extending over areas of the order of 40 nm×40 nm. A typical image of the wire surface at the bottom of the groove and of its cross section is shown in Fig. 6. Steps of the order of 0.35 and 0.72 nm are clearly observed thus confirming the interpretation given above. Clearly the lateral size of the island cannot be resolved individually by the submicron probe used in our experiment, and can be considered as a form of microscopic disorder which broadens the optical spectra already on the micron scale (which adds to compositional disorder).

Before concluding we would like to briefly comment on point (iii) of Sec. III. The unique possibility of exciting a single groove along the cleaved edge of the sample revealed that the final state of recombination changes with the direction of the exciting beam in our wires. The Lorentzian deconvolution of the spectra shown in Fig. 2 demonstrates that for top excitation the strongest emission comes from the ground level state $n=1$ of the wires and a weak $n=2$ emission is observed at higher energy. For excitation along the infinite axis of the wires (edge excitation) the $n=1$ emission is strongly reduced whereas the $n=2$ state is enhanced. By applying the selection rules of the quantum wells,[12] we note that the increasing light-hole character of the $n=2$ wire state causes the enhancement of the corresponding dipole matrix element for propagation along the basal plane of the quantum well (parallel to the infinite wire axis), whereas the heavy-hole-like ground-state is favored for propagation along the growth axis. This result evidences the increasing heavy-hole–light-hole mixing occurring in the higher index states of quantum wires. A much stronger anisotropy should be observed in strongly confined wires with larger intersubband splitting and mixing. Work in this direction is presently underway in our laboratories.

V. CONCLUSIONS

In conclusion we have investigated the optical properties of vertically stacked InGaAs/GaAs quantum wires by microprobe luminescence. We demonstrated that the broadening of the optical spectra comes from thickness fluctuations of monolayer height and lateral extension of about 40 nm, mainly occurring at the bottom of the groove. The systematic analysis of several samples either with single or vertically stacked wires in each groove, revealed that the wire-to-wire fluctuations, either lateral or vertical, are not responsible for the observed spectral broadening. The effect of carrier diffusion along the infinite axis of the wires has been monitored through the changes in the screening of the internal piezoelectric field (resulting in a redshift of the MPL) occurring along the diffusion path of carriers. Presumably, near field luminescence methods should be applied in order to resolve the microscopic morphology of our wires and to resolve the homogeneous linewidth of the transitions.

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[1] J. Y. Marzin, J. M. Gerard, A. Izrael, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994); U. Bockelmann, W. Heller, and G. Abstreiter, Phys. Rev. B **55**, 4469 (1997); D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).

[2] Y. Nagamune, H. Watabe, F. Sogawa, and Y. Arakawa, Appl. Phys. Lett. **67**, 1535 (1995).

[3] A careful structural study of our $In_xGa_{(1-x)}As$ quantum wells demonstrated that the compositional fluctuation in our structures amounts to about $\pm 0.2\%$ at $x=10\%$. See, M. DiDio, M. Lomascolo, A. Passaseo, C. Gerardi, C. Giannini, A. Quirini, L. Tapfer, P. V. Giugno, M. DeVittorio, D. Greco, A. C. Convertino, L. Vasanelli, R. Rinaldi, and R. Cingolani, J. Appl. Phys. **80**, 482 (1996).

[4] R. Rinaldi, A. Passaseo, M. DeVittorio, M. Lomascolo, G. Coli', and R. Cingolani, Solid-State Electron. (to be published).

[5] R. Rinaldi, R. Cingolani, L. DeCaro, M. Lomascolo, M. DiDio, L. Tapfer, U. Marti, and F. K. Reinhart, J. Opt. Soc. Am. B **13**, 1033 (1996).

[6] R. Cingolani, R. Rinaldi, M. Lomascolo, G. Coli', A. Passaseo, V. Emiliani, and M. DeVittorio, Nonlinear Opt. **18**, 347 (1997).

[7] M. Lomascolo, R. Rinaldi, A. Passaseo, and R. Cingolani, J. Appl. Phys. **83**, 302 (1998).

[8] R. Rinaldi, P. V. Giugno, R. Cingolani, H. Lipsanen, M. Sopanen, J. Tulkki, and J. Ahopelto, Phys. Rev. Lett. **77**, 342 (1996).

[9] In our experiments, the line shape of the MPL has been deconvoluted by Lorentzian bands, with χ^2 values smaller than 10^{-6}. No convergence was obtained by using Gaussians. On the contrary, the spatially integrated PL spectra, though with similar broadenings, could not be deconvoluted by Lorentzians. Convergence was obtained only by using Gaussians with χ^2 values smaller than 10^{-5}.

[10] Y. Nagamune, T. Tanaka, T. Kono, S. Tsukamoto, Y. Arakawa, K. Uchida, and N. Miura, Appl. Phys. Lett. **66**, 2502 (1995).

[11] Recently a clear evidence of carrier localization at monolayer islands has been reported in GaAs quantum wires by MPL. In this case, however, the PL linewidth was almost twice that of our wires. See J. Bellessa, V. Voliotis, R. Grousson, X. L. Wang, M. Ogura, and H. Matsuhata, Appl. Phys. Lett. **71**, 2481 (1997).

[12] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (les Editions de Physique, Paris, 1987).