Evidence of one-dimensional excitons in GaAs V-shaped quantum wires

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We have investigated the optical properties of excitons in GaAs quantum wires grown on nonplanar patterned substrates (V-shaped quantum wires) by photoluminescence and magnetophotoluminescence. The experimentally observed transitions are in agreement with the theoretical evaluation of the quantized energy levels based on TEM measurements. The clear evidence of the $n_y=1$ and 2 excitonic recombination in the photoluminescence spectra is exploited to directly measure the anisotropy of the optical matrix elements.

I. INTRODUCTION

The direct growth of a bent quantum well on a V-shaped etched substrate has been demonstrated to be very promising for the achievement of quantum-wire lasers.[1,2] It is thus very important to get a clear understanding of the optical and electronic properties of these quantum wires, to provide the necessary feedback for the improvement of their optoelectronic performances. In this work we have studied the excitonic properties of high quality GaAs quantum wires grown on nonplanar V-shaped substrates.

The investigated samples were fabricated by molecular-beam-epitaxy growth of a 3-nm (nominal thickness) GaAs quantum well cladded by $(GaAs)_8/(AlAs)_4$ superlattice barriers on a GaAs nonplanar substrate (the subindices indicate the number of monolayers). The substrate was patterned by holographic lithography[3] (UV line at $\lambda=257$ nm) and etched by wet chemical etching to form an array of adjacent V grooves of depth 110 and width 250 nm, oriented along the [110] direction. From TEM observations (see Fig. 1) one can see the bent GaAs quantum well, varying in thickness from about 10 nm at the bottom of the V groove, to 2.2 nm at the top of the sidewalls. No flat quantum wells between adjacent grooves are observed[4] due to the short period of the groove array ($\sim$250 nm). In each sample, part of the substrate is not patterned to get a flat quantum-well region as a reference.

Temperature- and intensity-dependent photoluminescence (PL) experiments were performed by using cw He-Ne, Ar^+, and $Al_xGa_{1-x}As$ diode lasers as exciting sources, up to power densities of the order of 5 W cm^{-2}. Magnetoluminescence experiments were performed by using the blue line of the Ar^+ laser and the samples immersed in liquid He, at 3.5 K in a superconducting cryostate (Oxford-Spectromag 4000) providing magnetic field up to 9.5 T. The spectral resolution was better than 0.8 Å.

II. THEORY

In the investigated structures the lateral confining potential originates from the shrinkage of the quantum well on the sidewalls of the grooves. Following Ref. 5, the lateral confining potential was obtained by a parabolic interpolation of the band gap of the lateral quantum well at different distances from the V-groove bottom. The actual thickness of the well was obtained from TEM micrographs (for the sample shown in Fig. 1 a well width $L_z \simeq 9.3$ nm is observed at the bottom of the groove, and 2.5 nm at the top of the sidewalls). This potential was used in the one-dimensional Schrödinger equation to obtain the confinement energies and to determine the

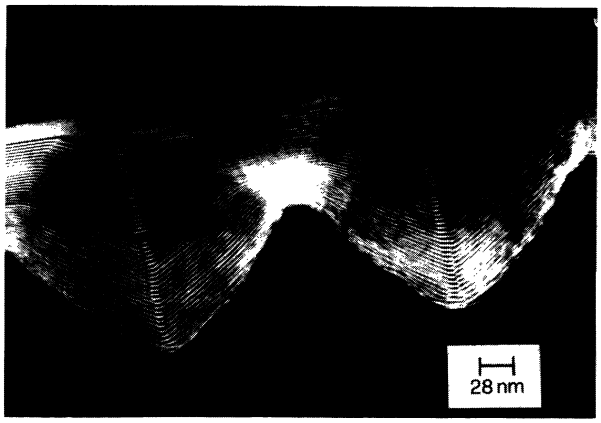

FIG. 1. TEM micrograph of a section of the investigated QWR structure.

effective wire width L_y, defined as the distance between the points where the wave function of the fundamental quantized level ($n_y=1$) decays exponentially. Throughout this paper we will label the quantum-well growth direction as z, the quantum-wire axis direction as x, and the lateral confinement direction as y. The quantum numbers of the eigenstates will be indicated by $n_{y,z}$ depending on the confinement direction. In Table I we quote the confinement energies obtained for the two investigated samples. In a more refined model a full two-dimensional potential has been used to describe the realistic quantum-well-wire structure.[6] From this we found that the higher-index states become closer in energy and approach a continuum distribution due to the finite height of the barrier ($\simeq 105$ meV for electrons and 50 meV for holes). Further, they become resonant with the states of the lateral quantum well. Both these effects should prevent the observation of pure one-dimensional (1D)-transitions involving states with quantum numbers higher than $n_y=3$. However, the $n_y=1$ and 2 transitions predicted by the two models are in good agreement, indicating the reliability of the simplified one-dimensional approach.

The quantization energies listed in Table I deserve some comment. Unlike the case of quantum wells, where the $m_J=\pm 3/2$ and $\pm 1/2$ valence states are evaluated separately, in quantum wires the valence subbands should be evaluated starting from the exact k_{xy} dispersion of the bands, to account for the heavy- and light-hole mixing occurring even at the zone center. This results in a unique set of discrete valence states of quantum number n_y with no definite heavy-hole or light-hole character. The dominant heavy-hole or light-hole character of each subband depends on the degree of band mixing occurring between different valence states.[7–9] Such mixing is obviously expected to be more important for higher-index subbands ($n_y>1$) and/or for narrow wires, where a larger valence-band mixing occurs due to the relevant lateral momentum $k_y=\pi/L_y$ available at $k_z=0$. In fact, deeper valence states are expected to have dominant heavy-hole character, whereas an increasing light-hole character is found for the higher-index valence states having energy closer and closer to the light-hole band of the quantum well from which the quantum wires were fabricated.[9]

In our quantum wires the full k_{xy} dispersion of the valence subbands is unfortunately not known (somewhat similar systems like the serpentine superlattices or triangular quantum wires have been recently discussed in Refs. 10 and 11, respectively). We have thus used the bulk heavy-hole and light-hole masses of GaAs to solve the one-dimensional Schrödinger equation for the lateral potential defining the wires. The heavy-hole states ($m_{hh}=0.34m_0$) around $k=0$ are found to be split by about 4 meV, whereas the first light-hole state ($m_{lh}=0.094m_0$) has a confinement energy of the order of 25 meV. Though very rough, this calculation indicates that at least the deepest quantized states in Table I should have predominant heavy-hole character, whereas states with $n_y>5$, being quasiresonant to the quantum-well light-hole band, should be predominantly light-hole-like. In terms of electronic transitions we thus expect that $\Delta n_y=0$ transitions should occur, involving the $n_y=1$, the $n_y=2$, and the $n_y=3$ conduction and valence subbands, the latter having dominant heavy-hole character. Throughout the paper these transitions will be labeled $n_y e$-$n_y h$. The selection rule is defined to conserve the total parity in the process. The parity being defined with respect to the quantum-wire axis x. In order to observe a predominantly light-hole transition in our samples one should excite the transition between the $n_y=1$ conduction state and the $n_y=7$ valence subband. This transition is expected to couple subbands with the same parity of the envelope functions. This situation is somewhat analogous to the calculation reported in the paper of Bockelmann and Bastard,[9] in which the 1*e*-1*h* and 2*e*-2*h* transitions are observed with excitation polarized in the x-y planes, whereas the lowest-energy light-hole-like transition (1*e*-6*h*) is observed with excitation polarized along the z direction.

We should mention that the light-hole character of the valence subbands of higher quantum numbers results in a polarization anisotropy of the matrix elements of the optical transition (with respect to the ground-level transition).[9] Therefore some polarization anisotropy should be observed between the ground-level transition and the 2*e*-2*h* transition in the optical spectra. Such anisotropy is expected to be intermediate between the quantum-well case (no x-y anisotropy) and the case of the light-hole and heavy-hole transitions in ideal quantum wires.[9]

TABLE I. Lateral electron and hole confinement energies (in meV) obtained by the simplified one-dimensional potential for the investigated quantum-wire structures. The ground-state energy level, having quantum number $n=0$ for a paraboliclike confining potential, has been indicated with $n_y=1$.

L_y		n_y = 1	2	3	4	5
22 nm	electrons	6.5	19.26	31.08	42.03	52.1
	holes	1.83	5.43	8.9	12.21	15.4
20 nm	electrons	7.36	21.52	34.58	46.52	57.34
	holes	2.03	6.01	9.83	13.47	16.93

III. EXPERIMENTAL RESULTS AND DISCUSSION

Before proceeding in the discussion of the optical data, we briefly comment on the main spectral features of the investigated samples. In Fig. 2 we show the photoluminescence spectra collected from the patterned region of the sample (curve *a*). As previously observed by Kapon *et al.*[12] three distinct lines can be observed: the superlattice barrier emission at 673 nm, the lateral quantum-well emission (QWL line at 791 nm), and a weaker luminescence from the single quantum wire at 790 nm (QWR line). The energy position of the QWL line is consistent with the confinement energy of the 2.5-nm-thick quantum well observed by TEM around the top of the groove sidewalls (about 80 nm far away from the bottom). The spectra collected from the unpatterned

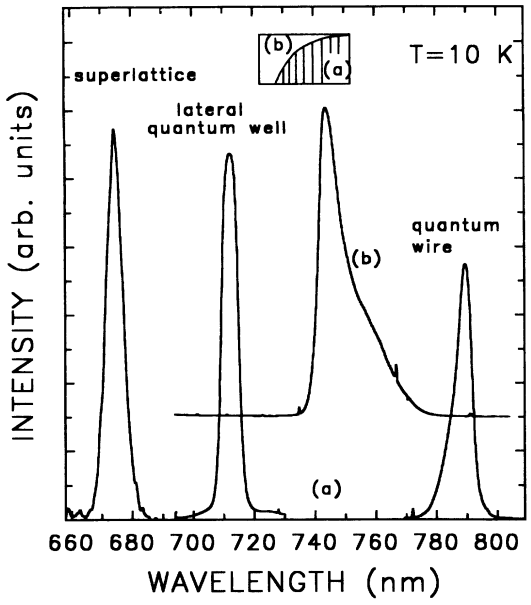

FIG. 2. Photoluminescence mapping of the samples under investigation: (a) patterned region (quantum wires), (b) unpatterned region.

reference region (curve *b*) exhibit only the emission corresponding to the flat 3-nm GaAs quantum well (around 750 nm). The broad low-energy tail is due to residual impurities which are readily saturated at excitation intensity of the order of few tens of m W cm^{-2}. The flat quantum-well emission at 750 nm is clearly absent in the patterned area. The QWR line is related to the quantum-wire ground-level transition calculated as described before (1*e*-1*h* transition). In the following we concentrate on this spectral region where the quantum-wire emission occurs, and we will neglect the higher-energy emissions originating from other parts of the heterostructure.

The patterned areas of the investigated samples are as large as 0.5 cm^2. Nevertheless, the quantum-wire luminescence measured at different positions on the patterned area shows a maximum spectral shift of the order of ± 1 meV and linewidth variations smaller than 15%. This indicates the good uniformity of the grown quantum wires. The typical linewidth of the luminescence lines amounts to 6–8 meV at 10 K, indicating the presence of some microscopic disorder.[13]

In Fig. 3(a), we report the intensity dependence of the QWR band for the sample with $L_y \sim 20$ nm (around 790 nm) excited by the 514.5-nm line of an argon laser. With increasing the photogeneration rate, the spectra exhibit a striking bandfilling, with a sharp emission rising 18.5 meV higher than the 1*e*-1*h* line. The lowest-energy band grows proportionally to the exciting power over more than a four order of magnitude increase of the exciting power. This rules out the possibility of any extrinsic emission. Further increase of the power density under cw excitation is prevented by the sample heating [see the redshift of the topmost spectrum in Fig. 3(a)].

A similar behavior can be observed in the temperature-dependent PL spectra of the $L_y \simeq 22$ nm samples displayed in Fig. 3(b). With increasing temperature, the thermal filling of the higher-index states occurs, allowing the clear observation of the 1*e*-1*h* and 2*e*-2*h* states of two broad structures around the 3*e*-3*h* and 4*e*-4*h* transitions at 250 K (a detectable PL signal persists up to room temperature).

As in the case of quantum wells, the relative splitting between the higher-index transitions is significant for the

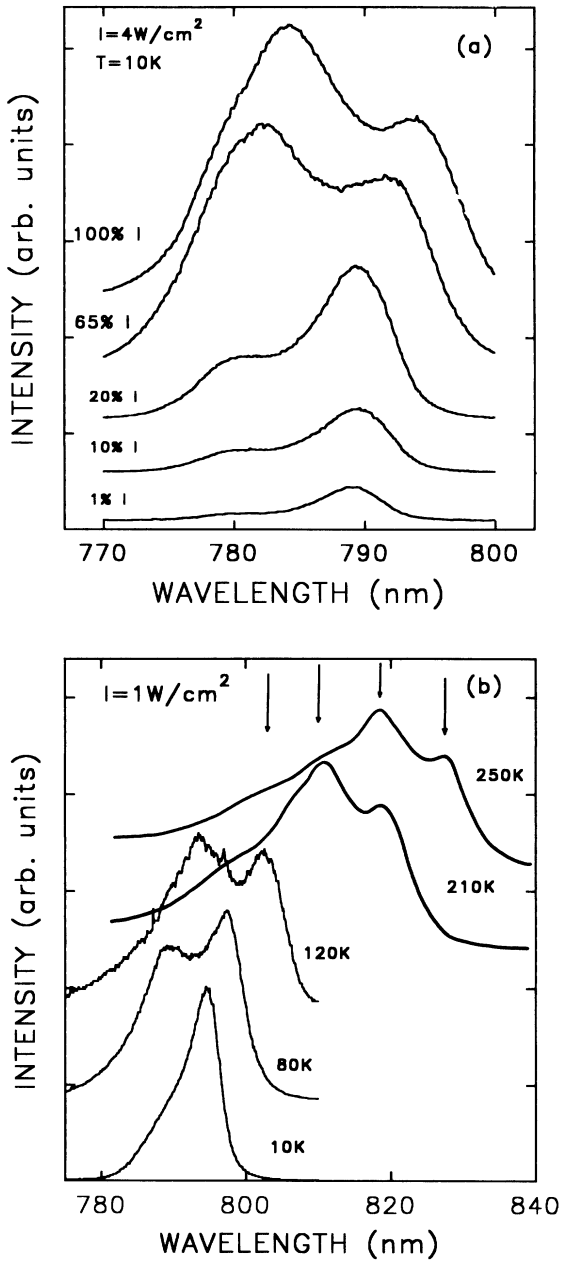

FIG. 3. (a) Intensity-dependent PL spectra of the $L_y = 20$ nm sample. The excitation intensity is 4 W cm^{-2}. All the spectra are on scale. The bottom spectrum is magnified by a factor 10. (b) Temperature-dependent PL spectra of the $L_y = 22$ nm sample. $I_{exc} = 1$ W cm^{-2}. The arrows on the 250-K spectrum indicate the calculated energy splitting between 1D transitions (all the spectra have been normalized to 1 and the topmost spectra have been smoothed).

comparison with the calculated eigenstates. (We did not try to fit the exact energy position of the ground-level transition as the exciton binding energy is unknown.) The vertical arrows reported on the $T=250$ K spectrum of Fig. 3(b) indicate the splitting among the quantum-wire transitions calculated with the simple one-dimensional model outlined in the previous section (Table I) and assuming the selection rule $\Delta n_y=0$. To account for the thermal shrinkage of the gap the ground-level transition energy has been shifted to coincide with the lowest-energy peak in the spectra. The calculated transition energies reproduce the experimental splitting among the PL bands fairly well. According to quantum mechanics, the almost constant energy splitting among the subbands indicates that the lateral potential barrier is effectively paraboliclike (we recall that the confinement energy for a rectangular wire would grow quadratically with the quantum number).

The temperature and intensity dependence of the measured spectra clearly indicate the intrinsic origin of the luminescence. However, the actual recombination mechanism is unclear. The observed band filling resembles the recombination of hot free carriers, which can hardly be generated at low temperature and under low-power excitation. In fact, a strong proof of excitonic character is given by the diamagnetic shift of the 1*e*-1*h* and 2*e*-2*h* lines in magnetic fields up to 8 T.[14,15] This is shown in Fig. 4 for the $L_y=20$ nm sample. The magnetic-field direction was parallel to the growth direction of the sample (*z* direction). The fundamental exciton line exhibits a diamagnetic shift of 0.025 meV T^{-2}. The 2*e*-2*h* exciton exhibits a larger diamagnetic shift, of the order of 0.061 meV T^{-2}, consistent with the expectation of a more extended wave function for the higher-index excitons in the V-shaped quantum wire.[14] The measured diamagnetic shift is found to change depending on the orientation of the field with respect to the carrier confinement direction, in agreement with the results of Ref. 15. However, the different degree of localization of the excitonic wave functions should be reflected in a greater diamagnetic shift of the 2*e*-2*h* state independent of the field direction. In the same figure we also plot the total Landau shift expected for free-carrier transitions.[14] Clearly, any free-carrier related transition can be ruled out under the adopted experimental conditions. These results demonstrate that even under low-power excitation (as low as 100 m W cm^{-2}) and/or relatively low temperatures, an extremely efficient population of the higher-index excitons occurs. Such phenomenon has never been observed under similar excitation conditions in quantum wells. This is a signature of the reduced 1D density of states, whose $1/\sqrt{\hbar\omega-E_{n_y}}$ dependence originates sharp resonances in correspondence with the 1D quantized levels and few energy states available in the continuum.

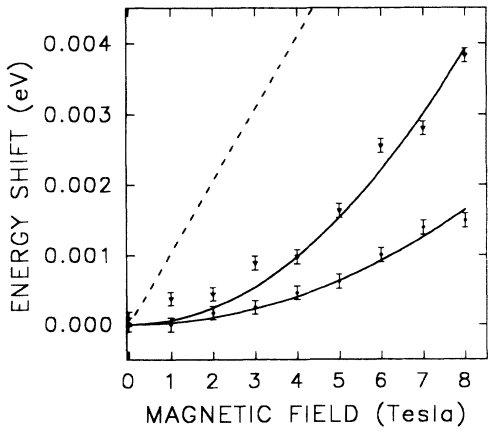

FIG. 4. Diamagnetic shift of the $n_y=1$ (dots) and $n_y=2$ (triangles). The continuous and dashed lines are the theoretically evaluated diamagnetic shift and Landau shift, respectively, for GaAs. The photoluminescence measurements were performed at 4 K, under 2.5-W cm^{-2} excitation intensity and with the magnetic field parallel to the growth direction of the sample.

Exploiting the direct observation of the 1*e*-1*h* and 2*e*-2*h* excitons in the photoluminescence spectra, we have investigated the polarization dependence of the intensity ratio of the two exciton lines [$R=(2e\text{-}2h)/(1e\text{-}1h)$]. As discussed in several theoretical works, quasi-one-dimensional excitons should exhibit clear polarization anisotropy, either in the emission[8,16] or in the absorption spectra,[7,8,9] due to the different polarization dependence of electron–heavy-hole and electron–light-hole squared optical matrix elements. Such anisotropy, though weaker, is expected to be quite effective even in the present samples, where the 1*e*-1*h* and 2*e*-2*h* transitions carry out a different degree of light-hole–heavy-hole mixing, and therefore a different polarization dependence of the respective matrix elements (see Sec. II).

The emission spectrum of the quantum wires is given by

$$L(\hbar\omega)\simeq\sum_{c,v}|M|^2_{c,v}D(E_c-E_v-\hbar\omega)f_c(E_c)[1-f_v(E_v)]\ , \tag{1}$$

where $D(E_c-E_v-\hbar\omega)$ is the one-dimensional joint density of states and f_c (f_v) is the electron(hole) distribution function. The interaction of the optical field with the quantum wires is governed by the ϵp Hamiltonian, through the dipole matrix element

$$|M|^2_{c,v}=\langle\Psi^c|\epsilon\cdot\mathbf{p}|\Psi^v\rangle\ ,$$

weighted by the overlap integral of the wave functions of the subbands involved in the transition. These quantities give rise to the dependence of the absorption and emission spectra on the polarization vector ϵ of the exciting wave. By rotating the polarization vector of the exciting light in the quantum-wire plane, we thus expect a change in the peak emission intensities of the 1*e*-1*h* and 2*e*-2*h* bands. This can be observed directly in the photoluminescence spectra, provided that the radiative recombination from the 1*e*-1*h* and 2*e*-2*h* excitons can be simultaneously detected, as shown in Fig. 5. The two PL spectra ($L_y=20$ nm sample) were excited by pumping in the continuum of absorption ($\lambda_{exc}=514.5$ nm) with ϵ parallel and perpendicular to the wire axis. The enhancement of the 2*e*-2*h* exciton transition under the perpendicular exci-

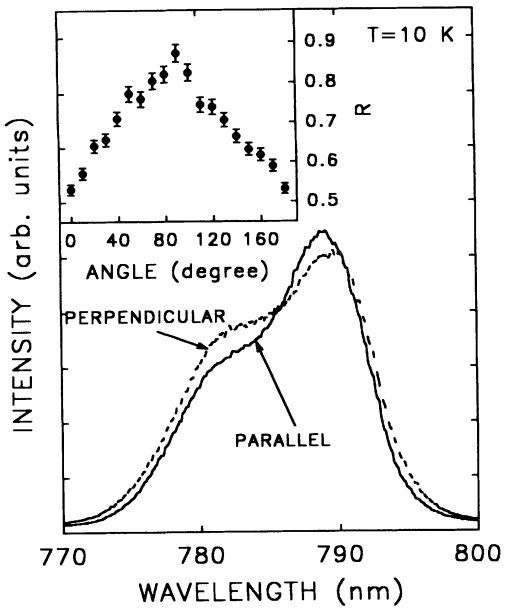

FIG. 5. PL spectra of the $L_y \sim 20$ nm sample recorded under parallel (continuous line) and perpendicular (dashed line) orientation of the polarization vector ϵ with respect to the wires axis. ($\lambda_{exc}=514.5$ nm.) Inset: Ratio of the 2e-2h and 1e-1h excitonic photoluminescence intensity vs the angle between the quantum-wire axis and the polarization vector of the exciting laser beam for the $L_y \sim 20$ nm sample.

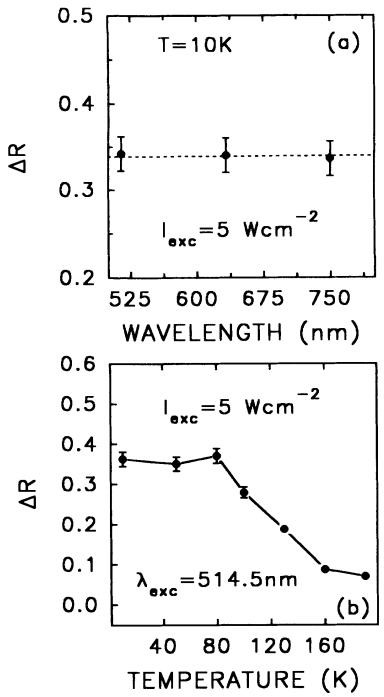

FIG. 6. (a) Polarization anisotropy ($\Delta R=[(2e\text{-}2h)/(1e\text{-}1h)]_{\perp}-[(2e\text{-}2h)/(1e\text{-}1h)]_{\parallel}$) measured in the investigated samples for different pumping energies. (b) Temperature dependence of the polarization anisotropy.

tation is clearly observable. In the inset of Fig. 5 we plot the R ratio [$R=(2e\text{-}2h)/(1e\text{-}1h)$] versus the angle formed by the polarization vector of the exciting light and the quantum wire axis. The maximum variation occurs by changing the polarization from parallel ($\parallel$) to perpendicular ($\perp$) to the wires, and amounts to

$$\Delta R \simeq 35\%,$$

where

$$\Delta R = \left[\frac{2e\text{-}2h}{1e\text{-}1h}\right]_{\perp} - \left[\frac{2e\text{-}2h}{1e\text{-}1h}\right]_{\parallel}.$$

These results compare well with those of Ref. 17 and with the theoretical calculations reported in Ref. 7, where the calculated ΔR value is found to be $\sim 40\%$. The observed anisotropy is too great to be ascribed to scattering at step edges of the tilted quantum wells or any other extrinsic effect (disorder or strain). As pointed out in Ref. 18 these effects should give rise to a typical anisotropy of the order of 4%. We emphasize that the observation of optical anisotropy directly in photoluminescence spectra is favored by the extremely efficient band filling occurring in our samples, which allows the simultaneous observation of both the 1e-1h and 2e-2h excitons, even at very low excitation intensities.[19]

Finally, we point out that our experiments are carried out under low-power excitation when intercarrier scattering does not play an important role during relaxation at the band edge. Our results suggest that carriers are efficiently trapped at the $n_y=1$ and 2 subbands while keeping the memory of the polarization of the exciting photon. This is supported by the fact that for low excitation intensities the measured polarization anisotropy is independent on the excitation energy, as shown in Fig. 6(a), where we display the polarization anisotropy (ΔR) of the PL spectra for three different pump energies. Clearly, the optical anisotropy is observed either under off-resonant or quasiresonant excitation, indicating that the PL spectra contain the information on the polarization of the exciting photon, at least when the excitation power is low and the intercarrier scattering is not very important. Consistently, we observe that the polarization anisotropy disappears in the PL spectra recorded with strong pulsed excitation. Analogously, we observe a dramatic reduction of the ΔR value with increasing the sample temperature, a shown in Fig. 6(b). The optical anisotropy persists up to about 100 K, then it strongly decreases for higher temperatures, when intrasubband scattering processes are enhanced by the increased thermal energy of the carriers.

IV. CONCLUSIONS

In conclusion, the optical properties of excitons in V-shaped GaAs quantum wires have been investigated by means of photoluminescence experiments. A strong filling of the higher-index excitonic states has been observed by raising the temperature and/or the excitation intensity even at very-low-power densities. The spectral position of the main emission band is consistent with the confinement energies calculated within a simplified one-dimensional potential model. The excitonic character of

this recombination has been established from the spectral shift of the PL features in high magnetic field. Exploiting the simultaneous observation of the ground level and some excited state in the PL spectra we have measured the polarization anisotropy of the matrix elements in the investigated quantum wires.

ACKNOWLEDGMENTS

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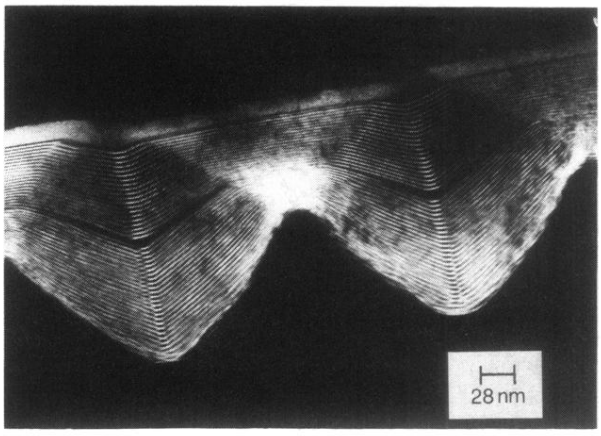

FIG. 1. TEM micrograph of a section of the investigated QWR structure.