Linear polarization of photoluminescence emission and absorption in quantum-well wire structures: Experiment and theory

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The photoluminescence (PL) and photoluminescence excitation (PLE) spectra of $In_xGa_{1-x}As$ quantum-well wires (QWW's) with wire widths L_x between 10 nm and 1 μm have been found to demonstrate a strong linear polarization parallel to the wires. The polarization is proportional to $1/L_x$ for broad wires and saturates in the case of narrow wires at a level of 20–60 % depending on the QWW structure design. This effect in PL and band-edge PLE transitions is shown to be mainly due to the modification of the electromagnetic-field eigenmodes in the vicinity of the quantum wires.

I. INTRODUCTION

The investigation of low-dimensional semiconductor structures such as quantum-well wires (QWW's) has attracted much attention in recent years.[1,2] The interest is connected with the possibility of the realization one-dimensional (1D) carrier confinement in these structures, which should lead to interesting phenomena. One of the intriguing questions in the optics of QWW structures is the strong linear polarization of the emission spectra,[3] that does not agree with the results of calculations of the dipole matrix element for optical transitions which neglect the lateral modulation of the dielectric constant in the wire gratings.[4]

The aim of the present paper is to demonstrate the crucial role of the lateral modulation of the etched QWW dielectric constant in the polarization dependent absorption/emission phenomena. These polarization-dependent effects are not the only interesting properties of nanostructures with large differences of the adjacent layers' dielectric constants. For example, the image-potential-mediated Coulomb interaction causes a considerable enhancement of excitonic effects in semiconductor/insulator nanostructures,[5] which makes them promising systems for optoelectronics. Such nanostructures have been predicted also to demonstrate striking polarization-dependent optical effects.[6,7]

Semiconductor/semiconductor quantum wells (QW's) and superlattices (SL's) have small differences of the dielectric constants, and these effects are negligible. However, the situation changes in the case of lateral semiconductor/vacuum (S/V) nanostructures, such as etched quantum-well wires (QWW's) and quantum-well dots (QWD's). It is natural to expect these nanostructures to demonstrate marked polarization-dependent optical effects. The basic reason for this is the inhomogeneous distribution of the electric field in S/V structures in the case when the electric field has a component normal to the S/V surface. In etched QWW's this is expected to cause (i) polarization-dependent excitonic electromagnetic wave absorption, (ii) polarized excitonic luminescence, and (iii) anisotropic ac and dc Stark effects on excitons.[8]

We have investigated the linear polarization of the exciton emission for two types of QWW's. The schematic design of our structures is presented in Figs. 1(a) and 1(b). The $In_xGa_{1-x}As/InP$ structures are fabricated by a combination of high-resolution electron-beam lithogra-

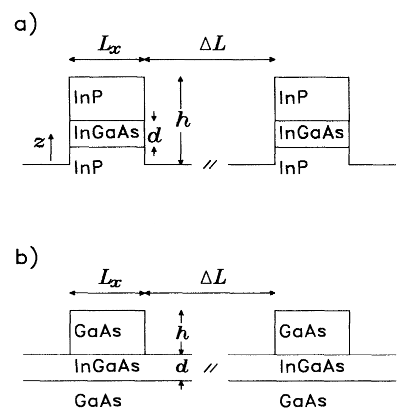

FIG. 1. (a) Schematic cross-sectional view of deep-etched $In_xGa_{1-x}As/InP$ wires. The distance ΔL between the wires is much greater than the wire width L_x. h denotes the etch depth, and z and d the position and thickness of the quantum well, respectively. (b) Schematic design of the cross section of modulated barrier quantum-well wires. Electrons and holes are confined in the $In_xGa_{1-x}As$ layer below the top GaAs wires.

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phy at 200 kV and deep wet chemical etching through the barrier and the QW layers. Details of the fabrication process are reported elsewhere.[9] Down to the smallest widths obtained, the $In_{0.53}Ga_{0.47}As/InP$ wires show a good luminescence efficiency.

Furthermore $In_xGa_{1-x}As/GaAs$ wires defined by lateral top barrier modulation have been investigated [Fig. 1(b)]. In these QWW's the lateral quantum confinement is achieved due to the difference in the quantum-well quantization in the regions with a top semiconductor barrier layer and regions where this layer has been removed by etching. Both the electrons and the holes are confined to the 1D QWW's below the top GaAs layer, which determines the size of the 1D confinement.[10,11]

In both types of structures the wire sidewalls include S/V surfaces. The large difference between the dielectric constants of the semiconductor and the vacuum causes a pronounced variation of the local electric field inside the QWW, if there is a component of the electric field normal to the S/V surface. Both the photoluminescence (PL) and absorption are governed by the same matrix element, which is proportional to the local electric field. Thus, within our model, the PL and the absorption acquire the same dependence on the QW position inside the wire. The local-field distribution depends also on the shape of the quantum wire. Hence one should expect a slight polarization dependence on the quantum wire profile. The results obtained from our theoretical model are in good agreement with the experimental data.

As far as we know, the influence of an electromagnetic wave modulation on the absorption and emission of QWW's has been previously discussed only for a special case of a surface grating with the distance ΔL between wires equal to their width L_x.[12] We would like to stress that within our model the linear polarization of the PL and absorption is not caused by a grating effect. The linear polarization is nearly the same for single and arbitrarily positioned QWW's, provided that $\Delta L, \lambda \gg L_x, h$ (where λ is the wavelength and h the vertical height of the QWW structure, see Fig. 1).

II. EXPERIMENT

The starting material for the fabrication of the deep-etched $In_xGa_{1-x}As/InP$ wires [see Fig. 1(a)] are metal-organic vapor-phase epitaxy (MOVPE) grown $In_{0.53}Ga_{0.47}As/InP$ single-quantum-well structures with a 200-nm-thick InP buffer layer, a 5 nm $In_{0.53}Ga_{0.47}As$ layer, and an InP cap layer with a thickness of 10 nm. The lateral widths of the 5-nm-thick $In_{0.53}Ga_{0.47}As$ layer, which is located between 8 and 13 nm below the top of the structure, range from about 10 nm up to 1 μm.

The $In_{0.16}Ga_{0.84}As/GaAs$ QW structures used for the fabrication of the modulated barrier QWW's [see schematic drawing Fig. 1(b)] were grown by molecular-beam epitaxy (MBE). The 5-nm-thick QW is covered by a 20-nm-thick GaAs top barrier layer. Using high-resolution electron-beam lithography, wire mask patterns with wire widths $L_x > 29$ nm have been defined.[10,11] The quantum efficiency of QWW's fabricated by this technique was found to be considerably higher than that of $In_xGa_{1-x}As/GaAs$ QWW's with open surfaces.

The structures were immersed in a cryostate with superfluid helium. The photoluminescence was excited by a cw Ar^+-ion laser (typical excitation density 10 W/cm^2), dispersed by a 1-m double monochromator and detected by a liquid-nitrogen-cooled germanium detector ($In_xGa_{1-x}As/InP$ QWW's) or a GaAs photomultiplier tube ($In_xGa_{1-x}As/GaAs$ (QWW's). The emission was passed through a polarizer oriented parallel or perpendicular to the wire axis. In order to avoid artifacts due to the polarization characteristics of the monochromator, the luminescence was circularly polarized in front of the monochromator. In the case of $In_xGa_{1-x}As/GaAs$ wires the polarization properties have additionally been investigated by using photoluminescence excitation spectroscopy by a titanium-sapphire laser.

III. OPTICAL STUDIES

We have studied the degree of linear polarization of the photoluminescence from $In_{0.53}Ga_{0.47}As/InP$ QWW's with a quantum-well thickness $L_x = 5$ nm and wire widths $L_x = 8-1000$ nm and of the photoluminescence and photoluminescence excitation spectra from $In_xGa_{1-x}As/GaAs$ QWW's with $L_x = 5$ nm and $L_x = 29-1000$ nm. The experiments were carried out on wire arrays with a typical size of 50×50-μm^2 size ($In_xGa_{1-x}As/InP$) and 100×100-μm^2 size ($In_xGa_{1-x}As/GaAs$), respectively. In addition to the wire arrays, mesa structures of the same size were placed on the samples serving as 2D references.

Typical emission spectra from the $In_xGa_{1-x}As/InP$ QWW structures for two directions of the luminescence light polarization (parallel and perpendicular to the wire axis) are shown for different wire widths in Fig. 2. The spectra consist of one emission line corresponding to excitonic recombination. As the wire is decreased down to 16 nm, we observe an increase of the emission energy by about 40 meV. As can be shown by model calculations, this shift is expected for small widths due to lateral quantization effects.[9] The PL spectra from the 2D reference mesa do not show any linear polarization. However, the spectra from wires with widths of 105 nm, for which no energy shift due to lateral quantization has yet been observed, show significant intensity differences for the two different polarization directions. This effect is further increased for the 41-nm-wide wires, but saturates for narrower wires, as can be seen from the PL spectra of the 16-nm-wide structures. Therefore, the polarization-resolved photoluminescence measurements show a strongly different luminescence intensity variation with L_x for polarization with the electric field of the PL signal parallel ($I_\parallel$) and normal ($I_\perp$) to the wire axes.

In Fig. 3 the linear polarization degree $\sigma(I_x) = (I_\parallel - I_\perp)/(I_\parallel + I_\perp)$ of the photoluminescence signal obtained from the deep-etched $In_xGa_{1-x}As/InP$ wire structures is displayed as a function of the inverse wire width. Starting from small values of (100 nm$/L_x$), i.e., from large wire widths, the polarization degree σ increases linearly with $1/L_x$ and then saturates for wire widths smaller

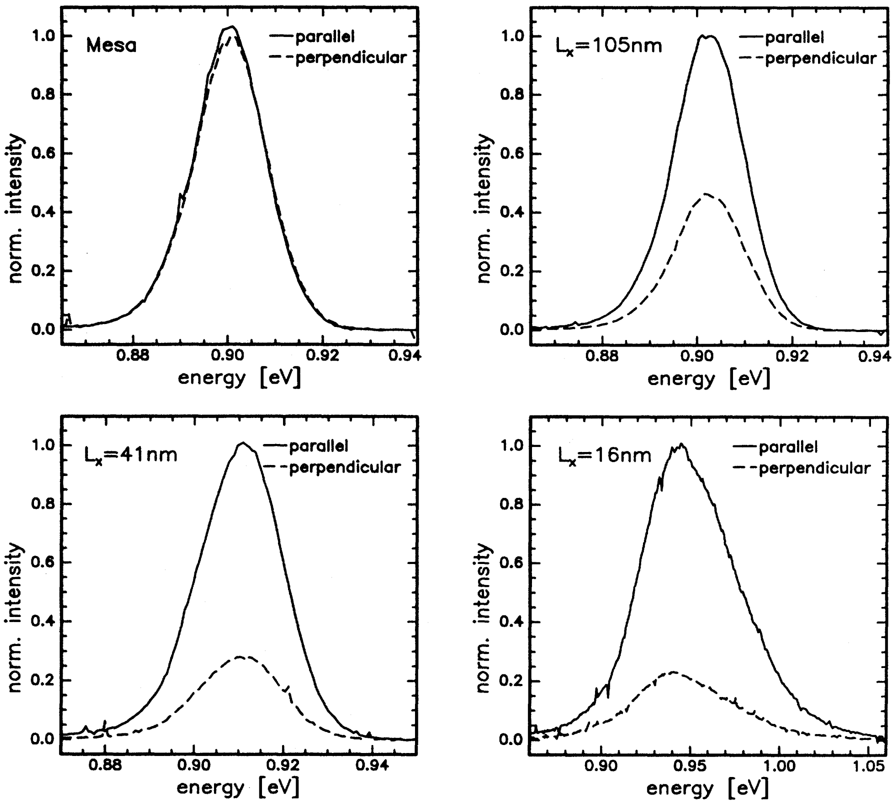

FIG. 2. Photoluminescence (PL) spectra from deep-etched $In_xGa_{1-x}As/InP$ wires for different wire widths. The PL emission was detected at $T=2$ K for polarizations of the luminescence light parallel and perpendicular to the wire orientation.

than about 50 nm. The polarization degree σ reaches maximum values of about 60%. Experiments on samples with different distances ΔL between the wires show that σ does not depend significantly on ΔL until $\Delta L \gg L_x$.

Figure 4 displays the corresponding dependence of the photoluminescence polarization degree σ on the inverse wire width for the barrier-modulated $In_xGa_{1-x}As/GaAs$ wires. The qualitative behavior is the same as for the deep-etched QWW's, i.e., a strong increase of σ with decreasing wire width for wide wires and a saturation for widths below 50 nm are observed. A significant difference exists in the maximum value of σ, which reaches about 20% for the barrier-modulated wires, compared to about 60% obtained for the deep-etched QWW's. As will be shown below, the smaller maximum value of σ is a consequence of the small etch depths in

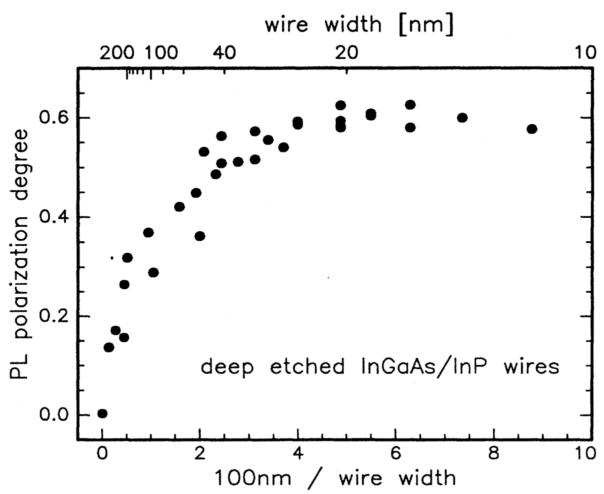

FIG. 3. Wire width dependence of the polarization degree of the photoluminescence emission obtained from deep-etched $In_xGa_{1-x}As/InP$ wires.

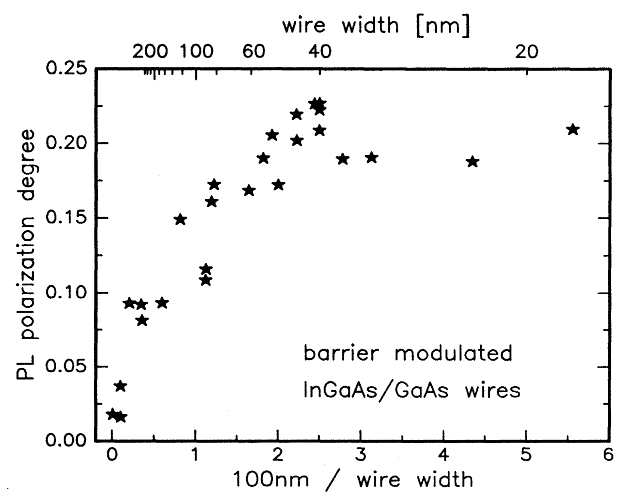

FIG. 4. Wire width dependence of the polarization degree of the photoluminescence emission obtained from barrier-modulated $In_xGa_{1-x}As/GaAs$ wires.

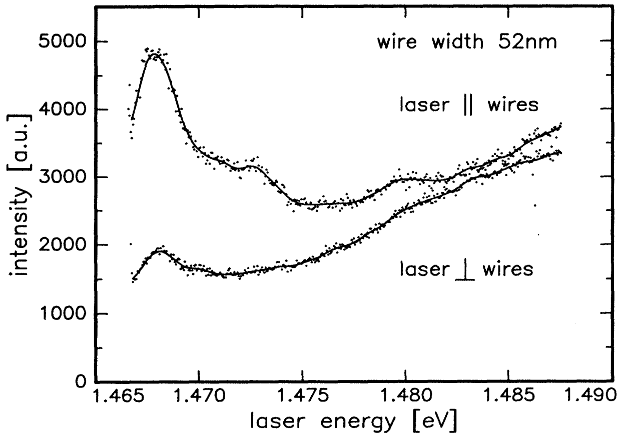

FIG. 5. Photoluminescence excitation (PLE) spectra of 52-nm-wide barrier-modulated $In_xGa_{1-x}As/GaAs$ wire structures taken with laser polarization parallel and perpendicular to the wire orientation.

the barrier-modulated structures.

In addition, we have investigated linear polarization effects in the photoluminescence excitation spectra from barrier-modulated $In_xGa_{1-x}As/GaAs$ QWW structures. The integral $(I_\parallel + I_\perp)$ exciton luminescence signal was registered from the sample for different polarization directions of the exciting laser beam. Figure 5 displays photoluminescence excitation (PLE) spectra obtained from 52-nm-wide $In_xGa_{1-x}As/GaAs$ wires for polarization directions of the exciting laser parallel and perpendicular to the wire axis. The three features observed in the spectra at laser energies of 1.468, 1.473, and 1.480 eV result from transitions between the first three lateral wire subbands of electrons and heavy holes.[13] As can be seen from the figure, the PLE signal is significantly larger for a laser excitation parallel to the wire axis than for a perpendicular excitation. Thus the absorption properties of these wire structures exhibit significant polarization effects, too. Close to the QWW band gap the degree of polarization of the PLE spectra is comparable to that observed in the emission spectra ($\sigma \approx 0.4$ at 1.468 eV). For higher energies the polarization degree of the PLE decreases strongly (e.g., $\sigma \approx 0.05$ at 1.485 eV). This behavior is not expected from the present model. The change of the polarization degree of the PLE with energy may be due to band-structure effects, which are expected to modify the polarization in narrow wires. Furthermore, at higher energies, excitation of the QWW's via defects in the GaAs barrier layers may contribute to the observed reduction of the degree of polarization.

IV. DISCUSSION

There are at least two reasons for the appearance of a linear polarization in the absorption and luminescence spectra of the QWW's. The first one is due to the mixing of $j=\frac{1}{2}$ and $\frac{3}{2}$ hole states in the one-dimensional valence subbands.[4] The second one is connected to the electric-field modulation in the vacuum/semiconductor QWW nanostructures.[6,8,12]

The theory of interband transitions in quasi-1D GaAs-based structures with mixed valence bands was recently developed by Bockelmann and Bastard.[4] Their calculations are based on the effective-mass approximation (EMA). They show that the mixing of the heavy- and light-hole states in the valence subbands results in different values of the dipole matrix elements for optical transitions between the valence-band and conduction-band states for different linear photon polarizations with respect to the wire orientation. However, this effect is important only when the lateral size L_x of the QWW's approaches the thickness L_z of the QW. In particular, for GaAs wires it is expected not to exceed a few percent for $L_x/L_z > 3$, but can reach 30–40% for the case of $L_x = L_z$.[4] Because much wider structures exhibit a strong linear polarization in our experimental data, this mechanism cannot explain the origin of the observed polarization anisotropy.

On the other hand, the transition matrix element of luminescence and absorption depends not only on the electron-hole wave functions, but also on the electromagnetic-field eigenmodes. The nanostructures of interest are regular arrays of $In_xGa_{1-x}As/GaAs$ or $In_xGa_{1-x}As/InP$ quantum wires with height h, lateral width L_x, and separated by a distance $\Delta L \gg h, L_x$. Let us note first of all that the distribution of the electric field in such structures drastically depends on the angle between the wire axes and the direction of the electric field $\mathbf{E}_\infty$ in the beam far from the interface. This is a consequence of the large difference between the vacuum and the semiconductor dielectric constants. For example, if $\mathbf{E}_\infty$ is parallel to the wires, the electric field inside the structure is homogeneous. If $\mathbf{E}_\infty$ is perpendicular to the wires, the electric field is very inhomogeneous.

In a dipole approximation the luminescence and absorption transition matrix elements are proportional to the scalar product of the local electric field $\mathbf{E}$ and the interband dipole momentum $\mathbf{d}$ averaged over the electron-hole wave function. Both the absorption and emission, which are proportional to the square of the same matrix element, acquire the same polarization dependence, due to the local electric-field dependence on the $\mathbf{E}_\infty$ orientation. In order to calculate the S/V QWW absorption and emission, one first has to find the local electric-field distribution inside the QWW structure. The problem is greatly simplified due to the fact that the characteristic QWW sizes h and L_x are small compared to the electromagnetic wavelength λ, i.e., $L_x, h \ll \lambda$. For the barrier-modulated wires a photon energy of about 1.5 eV leads to a wavelength in the material of about 250 nm. Although the experimental range of L_x extends up to 1000 nm, the polarization effects are observed mainly in case of narrow wires with width less than 100 nm. When using the static approximation one neglects the retardation effects. We would like to stress that these effects become small for the most interesting region of the wire widths investigated. Hence the static approximation being very crude for wide wires is more adequate for narrow wires, where the polarization effects are most significant. Basically this means that one can use the electrostatic approximation when calculating the electromagnetic eigenmodes inside the QWW's. For QWW arrays the problem reduces to the 2D Poisson equation, which can be easily solved numerically.

In Fig. 6 examples of the electric-field distribution $\mathbf{E}_\perp$ for the case of $\mathbf{E}_\infty$ perpendicular to the quantum wire axis are shown for different wire width to height ratios. For the calculations a dielectric constant $\epsilon=12$ was assumed inside the wires. The dotted lines display the profile of the wires used for the calculations. It corresponds approximately to the etch profile of the present structures obtained from scanning electron micrographs. The electric-field vectors are characterized by arrows according to their lengths and directions. As can easily be seen from Fig. 6, the most dramatic changes in the electric-field distribution occur in the vicinity of the wire surface. The strength of the changes increases with decreasing wire width to height ratio. For small width to height ratios there is a strong reduction of the absolute value of the electric field inside the wires. Additionally, the electric-field distribution is strongly inhomogeneous in this case. Due to this inhomogeneity the location of the active layer inside the wire plays an important role for the polarization properties, especially for small width to height ratios. For increasing width to height ratios this inhomogeneity inside the wires disappears gradually.

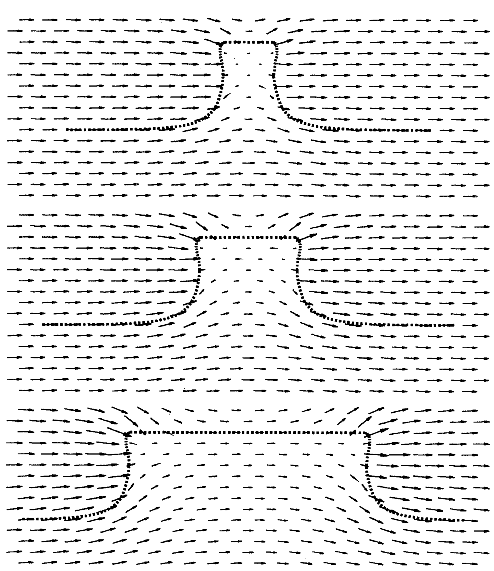

FIG. 6. Electric-field configuration of a light wave with a far-field polarization perpendicular to the wire axis in the vicinity of the wires for three different height-to-width ratios. The dotted lines show the assumed profiles of the different wires. The electric-field distribution is characterized by the arrows according to their length (absolute value of the electric field) and direction.

Bearing all this in mind, one can write the following equations for the absorption coefficients α and $\alpha_\parallel$ for the electric field oriented perpendicular and parallel to the wire axis, respectively:

$$\alpha_\perp \propto \frac{\langle c|\mathbf{E}_\perp \cdot d|v\rangle^2}{|\mathbf{E}_{\perp,\infty}|^2}, \tag{1}$$

$$\alpha_\parallel \propto \frac{\langle c|\mathbf{E}_\parallel \cdot \mathbf{d}|v\rangle^2}{|\mathbf{E}_{\parallel,\infty}|^2}. \tag{2}$$

Here $\mathbf{E}_\parallel$ and $\mathbf{E}_\perp$ are local electric fields for two polarizations (obviously $\mathbf{E}_\parallel=\mathbf{E}_\infty$ if $\mathbf{E}_\infty$ is parallel to the wires), and the averaging has to be performed over the electron-hole wave function corresponding to the state that provides the dominant impact to the luminescence or absorption at the measured frequency. For example, in the case of QWW exciton luminescence, the electron-hole function is that of the QWW-confined exciton and is mainly located in the wire region.

Now for the linear polarization degree of the absorption and of the PL, we can write

$$\sigma=\frac{\alpha_\parallel-\alpha_\perp}{\alpha_\parallel+\alpha_\perp}=\frac{\langle c|\mathbf{E}_\parallel\cdot\mathbf{d}|v\rangle^2-\langle c|\mathbf{E}_\perp\cdot\mathbf{d}|v\rangle^2}{\langle c|\mathbf{E}_\parallel\cdot\mathbf{d}|v\rangle^2+\langle c|\mathbf{E}_\perp\cdot\mathbf{d}|v\rangle^2}. \tag{3}$$

If the electric field is homogeneous inside the electron-hole localization region, one can remove the electric-field amplitude from the averaging integral. Then the linear polarization degree is governed only by the anisotropy of the interband dipole matrix elements.[4] This is not the case for S/V nanostructures. In the following, we will neglect the anisotropy of the interband dipole matrix element and use the following simple equation for the linear polarization degree:

$$\sigma \approx \frac{\langle E_\parallel\rangle^2-\langle E_\perp\rangle^2}{\langle E_\parallel\rangle^2+\langle E_\perp\rangle^2}, \tag{4}$$

where $\langle E_\parallel\rangle$ and $\langle E_\perp\rangle$ are the corresponding electric fields (averaged over the electron-hole wave functions), calculated with $E_{\parallel,\infty}=E_{\perp,\infty}$ far from the sample surface.

The results of our calculations of the linear polarization degree σ are shown in Fig. 7. Here the dependence of the photoluminescence polarization degree on the wire height to width ratio is shown for different locations z of the QW layer in the wires. The value of z is positive for deep-etched wires and negative for barrier-modulated wires, where the optically active QW layer is situated below the etched surface. Additionally, the experimental results for the deep-etched $In_xGa_{1-x}As/InP$ wires (filled circles) and the barrier-modulated $In_xGa_{1-x}As/GaAs$ wires (filled stars) are displayed, which differ in the location z of the optically active QW layer with respect to the wire height (etch depth) h. In the case of the barrier-modulated wires the etch depth h is equal to the GaAs cap layer thickness of 20 nm, and the bottom of the 5 nm-thick optically active $In_xGa_{1-x}As$ layer is situated 5 nm below the etched surface [i.e., $z=-5$ nm, see also Fig. 1(b)]. This corresponds to a ratio $z/h=-0.25$. The geometric conditions of the deep-etched $In_xGa_{1-x}As/InP$ wires are roughly described by a z/h

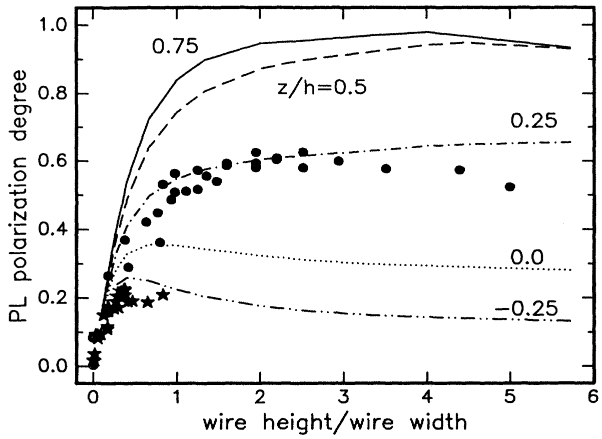

FIG. 7. Polarization degree as a function of the wire height-to-width ratio calculated for five locations z of the QW layer in the quantum wire with respect to the wire height h. Negative values z/h correspond to barrier-modulated QWW's. The filled circles and stars represent the experimental data for deep-etched and barrier-modulated QWW's, respectively.

ratio of 0.25. The etch depth of the $In_xGa_{1-x}As/InP$ wires is approximately the same for all wire widths investigated ($h \approx 20$ nm). The polarization degree increases for an increase of the height to width ratio h/L_x and approaches a maximum for a QW location in the middle of the quantum wire [deep-etched QWW's, Fig. 1(a)]. The comparison of the measured and calculated variations of σ for emission with respect to the wire height to wire width ratio demonstrates a good agreement for both types of QWW's. Due to the simplifications made in the calculation, an absolute agreement cannot be expected, but the qualitative and to a far extent also the quantative behavior are well described by our model. The results shown in Fig. 7 were calculated by averaging over the electron-hole wave functions in the simple infinite barrier approximation, without accounting for the Coulomb correlation. An averaging over the finite barriers EMA wave functions[11] changes the calculated polarization degree only on the order of 3%.

In order to check the sensitivity of the calculated polarization degree to the averaging procedure, a simple averaging of the electric field over the QWW volume has been performed, too. Only for barrier-modulated QWW's, σ depends on the form of the weighting function. This is due to a larger inhomogeneity of the electric field near the bottom of the unetched barrier wire (see Fig. 6). We would like to add that if the wire width is greater than the exciton radius, averaging over the spatial distribution of excitons should be performed, depending on the exciting photon energy, relaxation rates, and diffusion processes of the electron-hole system. The latter may introduce a slight variation of the calculated polarization degree.

V. CONCLUSIONS

We have shown that the strong linear polarization of the photoluminescence along the axis of the etched quantum wires may be understood by taking into account the modification of the photon eigenmodes in the vicinity of the quantum-well wires. The modification appears to have a stronger impact on the linear polarization effects than the explicit form of the electron-hole wave function. The polarization degree of the photoluminescence excitation spectra at the band edge is also described by our model. However, we observe a significant reduction of the polarization degree for higher excitation energies, which is not described by our model. This effect may be due to band structure-induced changes of the polarization. The profile of the quantum-well wires and the etch depth cause strong changes in the polarization behavior, that cannot be accounted for by the electron-hole system renormalization. The present experiments clearly demonstrate the drastic difference between the local and mean electric fields in the near-surface region of the wire arrays. This difference should be taken into account for a correct description of the optical properties of semiconductor-vacuum nanostructures.

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