Valence band spectroscopy in V-grooved quantum wires

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Abstract

We present a combined theoretical and experimental study of the anisotropy in the optical absorption of V-shaped quantum wires. By means of realistic band structure calculations for these structures, we show that detailed information on the heavy- and light-hole states can be singled out from the anisotropy spectra *independently of the electron confinement*, thus allowing accurate valence band spectroscopy.

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In semiconductor quantum wires, the anisotropic absorption of light linearly polarized parallel or perpendicular to the wire axis has long been recognized to be the effect of the quasi-one-dimensional (quasi-1D) character of electronic states, combined with heavy- and light-hole (HH and LH) mixing. [1,2] The observation of optical anisotropy has since been considered as an evidence of the 1D character of nanostructured materials. [3] However, even in samples showing the best optical properties, the interpretation of optical spectra is difficult owing to the large broadening of the peaks, which is comparable to the typical inter-subband splittings, and to the lack of realistic band structure calculations, so far limited to wires with model geometries. [2,4–6] Moreover, the confinement-induced blueshift is dominated by the light effective mass of conduction electrons, and information concerning hole states are even more difficult to extract from the spectra.

The aim of this letter is to show, by a combination of optical measurements and band structure calculations, that in realistic structures the details of the optical anisotropy can be traced to HH and LH states and, therefore, allow for a *spectroscopy of hole states*. We shall focus on quasi-1D structures obtained by epitaxial growth on non-planar substrates (V-shaped wires) [7–9], which rank among the best available samples from the point of view of optical properties.

Our samples are arrays of GaAs quantum wires embedded in $(GaAs)_8(AlAs)_4$ superlattices (SL), grown by molecular beam epitaxy (MBE) on V-grooved substrates obtained by holographic lithography and wet etching. In our case, the thickness of the crescent-shape wire which self-organizes at the bottom of the V-groove —as obtained by TEM analysis— is approximately 80 Å. More details on sample parameters are reported in Ref. [10].

In Fig. 1a we show the low-temperature (10 K) photoluminescence excitation (PLE) spectra (thin lines) for a V-shaped wire. The spectra are obtained with light polarized along the wire axis and perpendicular to it (and to the growth direction). We shall focus on the excitonic continuum, i.e., the energy range above 1.575 eV. The broad peaks in PLE spectra are very different from the singularities which one would naively expect from the ideal joint density of states (DOS) of quasi-1D electrons and holes, because of the relatively small

intersubband splittings compared to the inhomogeneous broadening [11] and the effects of Coulomb interaction [12], so that the interpretation is not easy. As we shall demonstrate, however, additional information can be gained by considering the optical anisotropy between the two polarizations (thick line in Fig. 1a), defined as $100 * (I_{\|} - I_{\perp})/I_{\|}$, where $I_{\|}$, $I_{\perp}$ are the PLE intensities for the two polarizations. In this range of energy, the anisotropy lies in the range 10-20%, but goes through a deep minimum around 1.59-1.60 eV, where it is almost suppressed.

We have performed realistic band structure calculations within the envelope function approximation. The HH-LH mixing was modelled by the Luttinger Hamiltonian [13], with proper account of crystallographic directions [14]. As in Ref. [10], the wire potential profile was extracted from a TEM micrograph of the sample. In the present complex geometry, the common approach of first solving the Schrödinger equation for uncoupled HHs and LHs and then using them as a basis set to represent the Luttinger Hamiltonian, results in a very large computation due to the poor convergence with respect to the number of subbands included in the basis. To overcome this difficulty, our approach is to set up a basis set from the eigenstates of only one type of particle, with a fictitious mass to be specified later, confined in the V-shaped wire potential. These are obtained numerically in terms of a plane wave expansion as in Ref. [10]. In this scheme the additional cost in the calculation of matrix elements of the Luttinger Hamiltonian is more than compensated, because only one type of particle needs to be calculated, and, more importantly, the convergence can be improved by properly choosing the fictitious mass. In practice, we find it efficient to choose a value close to the HH mass [15].

Band parameters are taken from GaAs bulk values. The only unknown parameters are the barrier heights for conduction and valence electrons: in fact, the embedding short period SL is modeled by a homogeneous barrier with an effective band offset. The conduction band offset was taken to be 150 meV, a value that was independently proved to reproduce PLE and magnetoluminescence experiments [10]. The criterium for the choice of the valence band offset is a by-product of the present work and will be discussed below.

The calculated absorption spectra and the relative optical anisotropy, obtained from the full band structure within the dipole approximation, are shown in Fig. 1b. A gaussian broadening of $\pm 4.5\,\mathrm{meV}$ has been included. The anisotropy (thick line) shows a minimum, and then reaches a value of $\sim 20\,\%$. The comparison with experiments is quite favourable for energies around the minimum, while the agreement worsens in the high energy range, where the calculated anisotropy drops rapidly and finally changes sign. This discrepancy might be due to our "effective" description of the barriers which affects particularly the higher-lying hole states.

As for the calculated absorption spectra, apart from a redshift of the continuum onset with respect to the experimental curves (which is within the uncertainty in the wire geometry), the main difference lies in the relative intensities of the two broad features at 1.59 eV and 1.61 eV in the experiment. This might be due to the Coulomb interaction, which is neglected in the present calculation, and causes a change in the excitonic continuum. Note, however, that while each single polarization is significantly changed by excitonic effects, the relative anisotropy seems to be much less affected.

The suppression of the optical anisotropy at $\sim 1.59\,\mathrm{meV}$ is due to states which are mainly of LH character: As an indication of the role of LH states, we also show in Fig. 1b the calculated absorption spectra for light linearly polarized parallel to the growth direction (z direction). Although measurements with this polarization are actually difficult because of the small active volume, it is instructive to calculate, since z-polarized light only couples with the LH component of hole states. [2] Figure 1b shows that the minimum in anisotropy actually lies in the range where the z-polarized absorption (and therefore the LH character) has a sudden increase.

Due to the large DOS, the electron and hole states at $k = 0$ (k is the 1D wavevector along the wire axis) are mainly responsible for the features in the spectra. Due to the quasi-1D confinement provided by the wire barriers, and in contrast to the case of quantum wells, these states are of mixed HH and LH character. [2] Figure 2 shows the calculated HH character of $k = 0$ hole states (labeled by the quantum number n_h) vs. the confinement energy. [16] Even

in a strongly confined system as the present V-shaped wire, the ground state is almost a pure HH state (92 %); the transition involving this hole state ($n_h = 1$) and the first electron state ($n_e = 1$) is mainly responsible for the first peak in the calculated spectrum. [17] The excited hole states show an increasingly mixed HH-LH character, so that the classification in HH and LH states, familiar from quantum wells, becomes inappropriate. Simultaneously to the decrease in the HH character, the wavefunctions of excited states become more and more extended along the "wings" of the wire, in analogy to the simpler case of conduction electrons [10], and, consequently, they couple to excited electron subbands, mostly with $n_e = n_h$); starting from the $n_h = 2$ level in Fig. 2, they contribute to the broad features above ~ 1.59 in the calculated spectra. However, this progression is interrupted by the state indicated with an arrow in Fig. 2. This state is of mainly LH character (56 %), and for brevity it will be indicated below as *the* LH state. Although this is a highly excited state ($n_h = 13$), its wavefunction, shown in the insets, is localized in the center of the wire. Therefore, due to the large overlap, this state couples primarily with the first conduction subband ($n_e = 1$), making a contribution to the low energy part of the spectrum. The large LH component, moreover, makes that the intensity for the two polarizations is reversed with respect to the strongly HH ground state, causing the dip in the anisotropy at 1.59 meV.

An immediate consequence of the above reasoning is that, since both the ground HH state and the LH state couple with the lowest electron subband, the difference in energy between the onset of the continuum and the position of the dip in the anisotropy is a direct measure of the energy splitting between the ground HH and the first LH states, *independently of the electron confinement*. This splitting, which in the experiment of Fig. 1a is ~ 16 meV, has been used in our calculation to estimate the effective valence band offset; its value was finally taken as 85 meV. [18]

In conclusion we have shown by realistic band structure calculations that the optical anisotropy of V-shaped quantum wires contains detailed information on the valence subbands, which can be singled out from the conduction electron contribution, allowing for a spectroscopy of the hole states.

REFERENCES

[1] P.C. Sercel and K.J. Vahala, App. Phys. Lett. **57**, 545 (1990).

[2] U. Bockelmann and G. Bastard, Phys. Rev. B **45**, 1688 (1992).

[3] For a review see: R. Cingolani and R. Rinaldi, Rivista Nuovo Cimento **16**, 1 (1993); see also: R. Rinaldi, M. Ferrara, R. Cingolani, U. Marti, D. Martin, F. Morier-Gemoud, P. Ruterana, and F. K. Reinhart, Phys. Rev. B **50**, 11795 (1994).

[4] D.S. Citrin and Y.-C. Chang, Phys. Rev. B **43**, 11703 (1991).

[5] H. Ando, S. Nojima, and H. Kanbe, Phys. Rev. B **43**, 11703 (1992).

[6] G. Goldoni and A. Fasolino, Phys. Rev. B **52**, 14118 (1995).

[7] E. Kapon, D.M. Hwang, and R. Bhat, Phys. Rev. Lett. **63**, 430 (1989).

[8] M. Gailhanou, T. Baumbach, U. Marti, P.L. Silva, F.K. Reinhart, and M. Ilegems, Appl. Phys. Lett. **62**, 1623 (1993).

[9] S. Tiwari, G.D. Pettit, K.R. Milkove, F. Legoues, R.J. Davis, and J.M. Woodall, Appl. Phys. Lett. **64**, 3536 (1994).

[10] R. Rinaldi, R. Cingolani, M. Lepore, M. Ferrara, I.M. Catalano, F. Rossi, L. Rota, E. Molinari, P. Lugli, U. Marti, D. Martin, F. Morier-Gemoud, P. Ruterana, and F. K. Reinhart, Phys. Rev. Lett. **73**, 2899 (1994).

[11] E. Molinari, F. Rossi, L. Rota, P. Lugli, R. Rinaldi, M. Lepore, and R. Cingolani, in *Proc. 22nd Internat. Conf. on the Physics of Semiconductors*, edited by D.J. Lockwood, World Scientific, Singapore (1994), p. 1707.

[12] F. Rossi and E. Molinari, Phys. Rev. Lett. **76**, 3642 (1996); Phys. Rev. B **53**, 16462 (1996).

[13] J. M. Luttinger, Phys. Rev. **102**, 1030 (1956).

[14] J.-B. Xia, Phys. Rev. B **43**, 9856 (1991).

[15] The choice of such fictitious mass is purely dictated by computational convenience: once convergence is achieved, it does not affect the final results in any way.

[16] The angular momentum quantization axis is chosen along the direction of strongest confinement, i.e. the [001] growth direction.

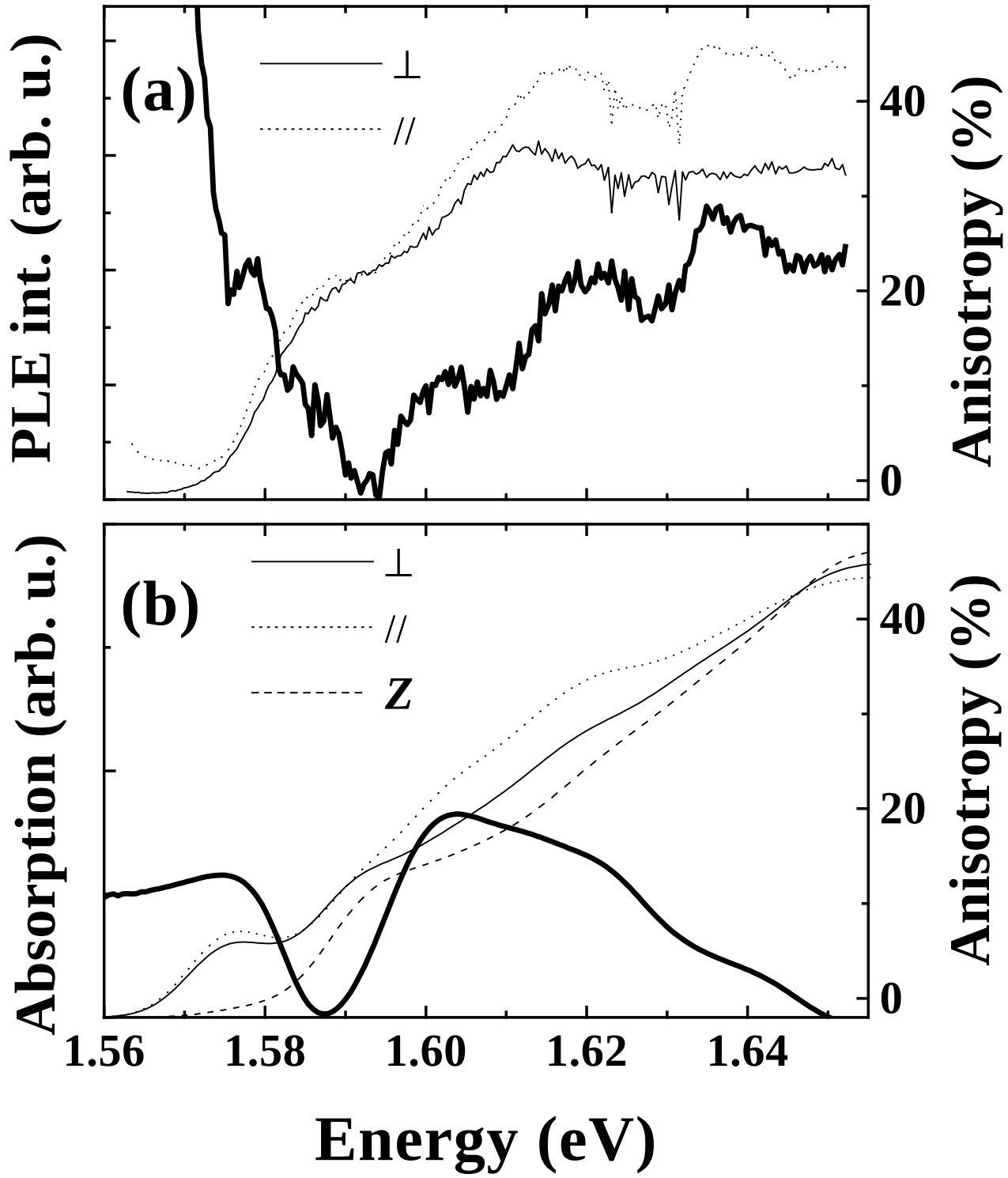
[17] To help with the interpretation of the theoretical spectra we record here the electron confinement energy of the three lowest subbands $n_e = 1, 2, 3$: 43.3 meV, 57.3 meV, and 65.3 meV. The band gap is 1.519 eV.

[18] A variation of the valence band offset of ± 15 meV shifts the HH-LH splitting of ± 1.3 meV.

FIGURES

FIG. 1. a) Low-temperature PLE spectra measured with incoming light linearly polarized perpendicular to the wire axis (thin solid line), parallel to the wire axis (dotted line), and relative anisotropy (thick solid line). b) Calculated absorption spectra with light linearly polarized perpendicular to the wire axis (thin solid line), parallel to the wire axis (dotted line), parallel to the z direction (dashed line), and relative anisotropy (thick solid line).

FIG. 2. Percentage of HH character for the lowest hole subbands at $k = 0$ vs subband energy. The arrow indicates the first state which is mainly (56 %) of LH character. Insets: total charge density, and HH- and LH-projected charge densities for the state indicated with the arrow ($n_h = 13$). Darker regions correspond to larger values of the charge density; thin lines represent the potential profile used in the calculation. In-plane axis are in nm (note the different scale in the two directions). For clarity, in the three panels grey levels are set to different scales.



G. Goldoni et al. - Fig. 1

Goldoni et al. - Fig. 2