Prediction of barrier localization in modulated nanowires

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(Received 16 April 2004; accepted 23 July 2004)

It is shown that the phenomenon of inversion recently discovered in a one-band model [L. C. Lew Yan Voon and M. Willatzen, J. Appl. Phys. **93**, 9997 (2003)] is much more general and is present in both multiband theories and in the excited states. Predictions of the one-band and of a four-band model are in good agreement for the ground state. A critical radius of around 15 Å (7 Å) is obtained for holes in InGaAs/InP (GaAs/AlAs) modulated nanowires. This phenomenon should be readily observable in both optical spectroscopy and transport. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1792803]

A type of nanostructures, so-called modulated nanowires, has recently been grown.[1–4] Material systems studied so far are GaAs/GaP,[1] Si/SiGe,[2] InAs/InP,[3] and ZnSe/CdSe.[4] Typical dimensions have been radii of 200–400 Å. While Bjork *et al.*[3] have reported atomically sharp modulation, the structures of Gudiksen *et al.*[1] have much more gradual modulation. It has been predicted that these structures might find applications as nano bar codes, waveguides, lasers, and light-emitting diodes, and that nanowire superlattices are an improvement over plain nanowires due to the coupling of the superlattice longitudinal confinement to the nanowire radial confinement.[1] Among the characteristic properties experimentally reported so far, we note photoluminescence from the GaAs layer in the GaAs/GaP structure,[1] and the nonhomic conductance of a modulated nanowire with a single barrier layer.[3]

Due to the large radii of the structures grown so far, there is not yet any experimental evidence of quantum phenomena in these structures. Nevertheless, we recently predicted, on the basis of a parabolic one-band model with an infinite potential barrier outside the nanowire that a drastic change in the localization of the electron states can occur below a certain critical radius.[5] This model was chosen because it leads to analytic solutions but it is also realistic for the conduction states of large-gap materials and for a free-standing (i.e., not embedded) nanowire; we have since independently verified the results numerically.[6,7] For a normal III-V semiconductor system, this would apply to the conduction band but not, in general, to the valence band. Indeed, it is known that the lowest hole state in a cylindrical quantum wire has mixed heavy-hole and light-hole characters.[8,9] Therefore, the existence of a critical radius for holes is not guaranteed. Our goal here is to give a general demonstration of the effect by solving the nonparabolic multiband problem. The theory is directly applicable to the hole states. We also predict an effect of the dependence of the critical radius on the longitudinal structure and of multiple critical radii when the excited states are considered.

The band structure of the modulated nanowires with an [001] wire axis is calculated using a four-band *kp* theory within the axial approximation, following the method of Sercel and Vahala.[8] It involves expressing the *kp* Hamiltonian in terms of cylindrical polar coordinates ρ, ϕ, z and noting that a good quantum number is the projection of the total angular momentum (of the envelope function and Bloch state) along the rod axis (labeled z),

$$F_z = L_z + J_z, \tag{1}$$

where the angular momentum L_z of the envelope function can only take integer values, and the J_z values belong to the $J=\frac{3}{2}$ subspace of heavy holes (HHs) and light holes (LHs). The total wave function is then written as

$$\psi(\mathbf{r}) = \sum_{J_z} f_{J_z}(\rho,z) e^{i(F_z-J_z)\phi} \left| \frac{3}{2} J_z \right\rangle, \tag{2}$$

where $f_{J_z}(\rho,z)$ are the envelope-function components. There is a double degeneracy with respect to the sign of F_z due to inversion symmetry. Two new features of our theory are the use of the Burt-Foreman Hamiltonian (instead of the Luttinger-Kohn one) and the presence of confinement along the wire axis. While the two Hamiltonians are in good agreement for the GaAs/AlAs system, there are substantial differences for InGaAs/InP. The coupled differential equations were solved using the finite-element method.

We represented the superlattice structure by using a finite number of periods; it turns out that only a couple of periods are necessary for convergence. Specifically, we found that, for radii away from the critical radius, the lowest two levels converge already to within 1 meV after only one period of the superlattice, i.e., the energies of the first two states do not change with the superlattice period after only one period. We attribute the rapid convergence to the presence of barrier layers in the modulated structure which reduce the interbilayer coupling of the states even more over the plain nanowire case, and to the heavy mass of the lowest hole state.

Despite the general inapplicability of a one-band model to the hole states, we first use the one-band model[5] in order

TABLE I. Luttinger parameters and computed effective masses along [001].

	γ_1	γ_2	γ_3	$m_{HH}=(\gamma_1-2\gamma_2)^{-1}$	$m_{LH}=(\gamma_1+2\gamma_2)^{-1}$
GaAs[a]	6.85	2.10	2.90	0.377	0.090
AlAs[a]	3.45	0.68	1.29	0.478	0.208
InAs[b]	20.4	8.3	9.1		
InP[b]	4.95	1.65	2.35	0.606	0.121
$In_{0.53}Ga_{0.47}As$	14.03	5.39		0.308	0.040

[a]Reference 15.
[b]Reference 11.

to estimate the critical radius, assuming the latter to exist. It was shown[5] that the critical radius R_c is given by

$$R_c^2 = \frac{(m_B - m_W)}{m_B m_W V_B} \alpha_{lm}^2, \tag{3}$$

where V_B is the original barrier height, m_B and m_W are the effective masses in the (conventional) barrier and well layers, and α_{lm} is the lth zero of the Bessel function of the first kind J_m. Equation (3) predicts that the critical radius increases with decreasing effective masses, decreasing band offset, increasing difference in effective masses between the barrier and well materials, and for higher states. We studied two popular unstrained systems that have been used for making plain nanowires: GaAs/AlAs and $In_{0.53}Ga_{0.47}As$/InP. Band parameters for all the materials are given in Table I. For the valence-band offsets, we used 0.53 eV for GaAs/AlAs (Ref. 10) and 0.39 eV for $In_{0.53}Ga_{0.47}As$/InP.[11] The InGaAs/InP is expected to lead to a larger HH critical radius than GaAs/AlAs due to the larger difference in Luttinger parameters between $In_{0.53}Ga_{0.47}As$ and InP than between GaAs and AlAs, and also to the smaller valence-band offset for the former. The above equation gives a critical radius for the lowest HH (LH) state of about 10 Å (31 Å) for $In_{0.53}Ga_{0.47}As$/InP and of about 5 Å (16 Å) for GaAs/AlAs.

We next carried out the four-band calculations for modulated nanowires with layer thicknesses of 50 Å and a circular cross section with radii ranging from 5 Å to 15 Å. A critical radius is identified when the envelope function shifts from a well layer (here $In_{0.53}Ga_{0.47}As$ and GaAs) to a barrier layer (here InP and AlAs). The sum squared of the envelope functions for one structure with $R=15$ Å and one with $R=10$ Å are shown in Fig. 1 (first row) for $In_{0.53}Ga_{0.47}As$/InP. In the longitudinal direction, there are two well layers and three barrier layers, each of width 50 Å. A change in the localization of the envelope function is clearly seen. For $In_{0.53}Ga_{0.47}As$/InP(GaAs/AlAs), the actual critical radius for the lowest state is found to be 14–15 Å (7–8 Å). This is in good agreement with the one-band calculation. While the corresponding diameter is rather small for the lowest state of GaAs/AlAs, it is of the order of 30 Å for $In_{0.53}Ga_{0.47}As$/InP, a value within the limit of applicability of the theory and a structure that is realizable in the laboratory. Also, our earlier prediction of a critical radius of about 20 Å for electrons in GaAs/AlAs modulated nanowires[5] means that just below that radius, the ground electron state would be inverted but not the ground valence state. Thus, the fundamental absorption should be strongly suppressed below the critical radius. One would also expect the transport properties to be significantly affected. For example, below and above the critical radius, the states are mostly localized leading to little current flow under an applied voltage across the wire. However, at the critical radius, the barrier height is effectively zero leading to a strong transmission (but not complete due to the effective-mass mismatch). Thus, the current-voltage curve should display a large peak at the critical radius.

A one-band calculation would be accurate if there is little heavy hole–light hole mixing in the ground state. The four components (squared) of the envelope function are also given in Fig. 1. Note that the color codings for the components in the panels are independent of each other. Both above and below the inversion, the predominant component is for $J_z=+1/2$ but there is some band mixing.

Quantum rods have been reported to display a level crossing of the lowest two states near an aspect ratio of 1:1.[12] We have also recently obtained a similar behavior, whereby the ground state went from "bell shaped" to one with a node in the z direction.[13] We found that this does not occur for the modulated structure for an aspect ratio below 1:10. This would suggest that the band-structure contribution to the linearly-polarized emission reported previously[1] would be less likely with the modulated nanowires, an important consideration for optoelectronic devices. This would also support the calculations of Wang *et al.*[14] that the polarization properties can be accounted for using electromagnetic theory. The work of Sercel and Vahala on quantum wires also showed that the ground state always has the quantum number $F_z=\pm 1/2$; we have found this to be true for the modulated nanowires too.

Equation (3) predicts that the critical radius increases with the energy of the state [the energy varies approximately linearly with α_{lm}, refer to Eq. (2) of Ref. 5]. The multiband calculation shows that the critical radii for excited states are indeed larger than for the ground state but not by as much as predicted by the one-band model. For example, for $In_{0.53}Ga_{0.47}As$/InP, the first excited state is inverted for a radius of 18 Å, an increase of only 20% over the ground-state value compared to an increase of 60% as suggested by Eq. (3).

Finally, the one-band model predicts that the critical radius is independent of the actual structure in the longitudinal direction as long as a modulation exists.[5] However, since the current problem is mathematically nonseparable (for the two spatial coordinates), one would expect a dependence of the critical radius on the structure in the longitudinal direction. We investigated this by performing calculations for modulated nanowires with various layer widths. We did find a

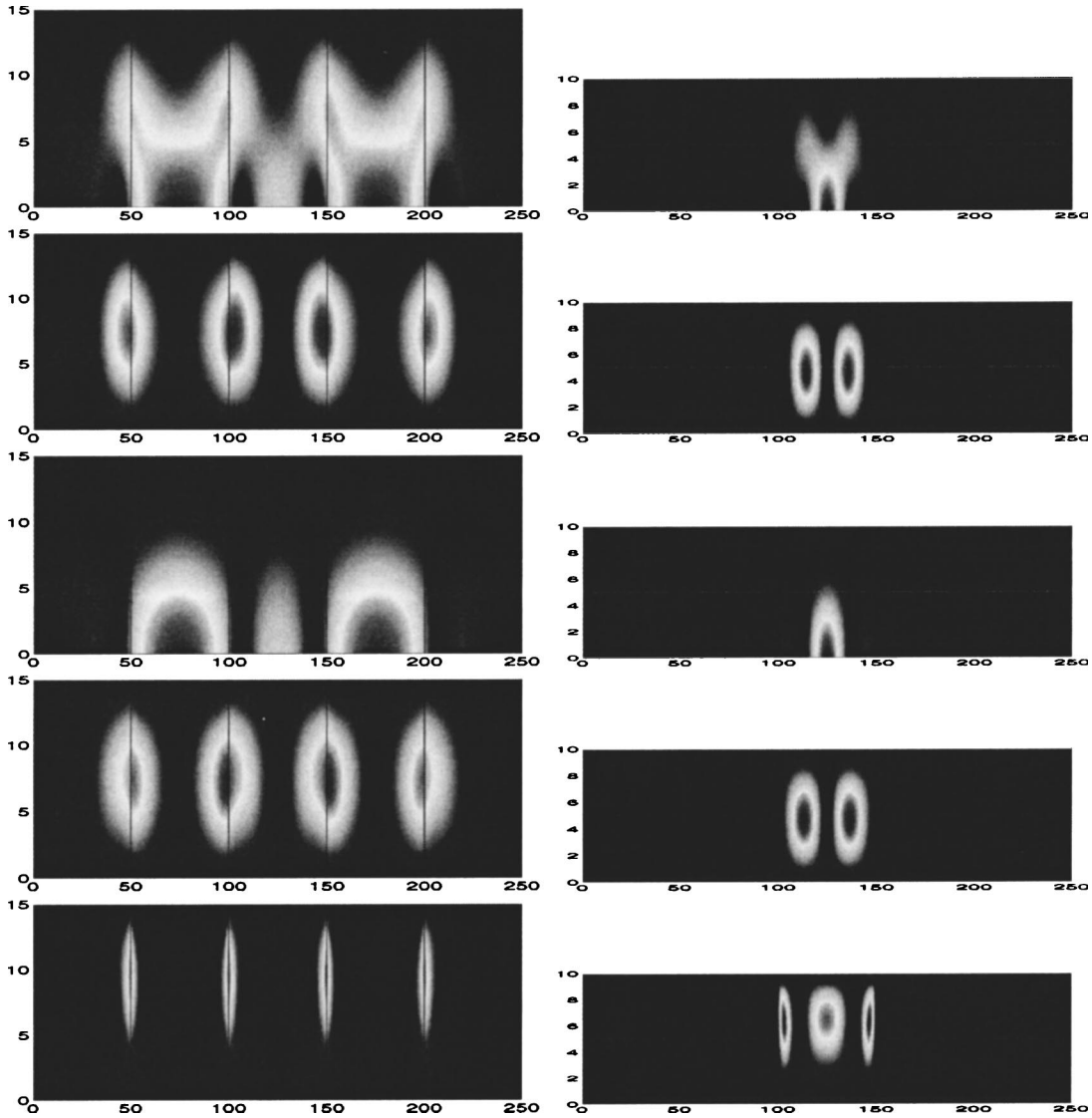

FIG. 1. Lowest valence state above inversion (left, with $R=15$ Å) and below inversion (right, with $R=10$ Å). The y axis is the radial direction, the x axis is the z direction with 50 Å wide layers of (from left to right) InP, $In_{0.53}Ga_{0.47}As$, InP, $In_{0.53}Ga_{0.47}As$, and InP. The top plots are for the sum squared of the envelope-function components, while the remaining rows are for each of the four components in the following order: $J_z=+3/2,+1/2,-1/2,-3/2$.

small dependence of the critical radius on the layer widths. For example, for InGaAs/InP, decreasing both the layer thicknesses from 50 Å to 20 Å reduced the critical radius from 14–15 Å to near 12 Å.

In summary, the existence of critical radii for the inversion of hole states in modulated nanostructures is demonstrated using a multiband calculation. It is proposed to observe the inversion via interband absorption or transport experiments in GaAs/AlAs modulated nanowires of radii near 20 Å.

ACKNOWLEDGMENTS

This work was supported by an NSF CAREER award (NSF Grant No. 9984059) and a Balslev award (Denmark).

[1]M. S. Gudiksen, L. J. Lauhon, J. Wang, D. C. Smith, and C. M. Lieber, Nature (London) **415**, 617 (2002).
[2]Y. Wu, R. Fan, and P. Yang, Nano Lett. **2**, 83 (2002).
[3]M. T. Björk *et al.*, Nano Lett. **2**, 87 (2002).
[4]R. Solanki, J. Huo, J. L. Freeouf, and B. Miner, Appl. Phys. Lett. **81**, 3864 (2002).
[5]L. C. Lew Yan Voon and M. Willatzen, J. Appl. Phys. **93**, 9997 (2003).
[6]M. Willatzen, R. V. N. Melnik, C. Galeriu, and L. C. Lew Yan Voon, Math. Comput. Simul. **65**, 385 (2004).
[7]C. Galeriu, L. C. Lew Yan Voon, R. N. Melnik, and M. Willatzen, Comput. Phys. Commun. **157**, 147 (2004).
[8]P. C. Sercel and K. J. Vahala, Phys. Rev. B **B 42**, 3690 (1990).
[9]P. C. Sercel and K. J. Vahala, Appl. Phys. Lett. **57**, 545 (1990).
[10]L. C. Lew Yan Voon, L. R. Ram-Mohan, H. Luo, and J. K. Furdyna, Phys. Rev. B **47**, 6585 (1993).
[11]S. A. Stoklitsky, Q. X. Zhao, P. O. Holtz, B. Monemar, and T. Lundström, J. Appl. Phys. **77**, 5256 (1995).
[12]J. Hu, L.-S. Li, W. Yang, L. Manna, L.-W. Wang, and A. P. Alivisatos, Science **292**, 2060 (2001).
[13]L. C. Lew Yan Voon, B. Lassen, R. Melnik, and M. Willatzen, Nano Lett. **4**, 289 (2004).
[14]J. Wang, M. S. Gudiksen, X. Duan, Y. Cui, and C. M. Lieber, Science **293**, 1455 (2001).
[15]*In Numerical Data and Functional Relationships in Science and Technology*, Landolt-Börnstein, New Series, Group III, Vol. 22, Pt. A, edited by O. Madelung (Springer, Berlin, 1982).