Predicting interband transition energies for InAs/GaSb superlattices using the empirical pseudopotential method

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(Received 23 April 2003; published 27 October 2003)

Recent measurements surprisingly show that the lowest valence-to-conduction confined transitions in narrow $(InAs)_8/(GaSb)_n$ and $(InAs)_6/(GaSb)_n$ superlattices *increase* in energy as the barrier thickness n increases. We show that in addition to the mesoscopic geometric quantities (well and barrier sizes), an atomic-scale description of interdiffused interfaces is needed to correctly reproduce the observed spectroscopic trend. The interdiffused interface is modeled via diffusion equations. We compare our atomistic empirical pseudopotential calculation in which only the *bulk binary* data are fit to experiment, with contemporary methods in which agreement with experiment is forced using ideally abrupt interfaces.

DOI: 10.1103/PhysRevB.68.155329 PACS number(s): 73.21.Cd, 71.55.Eq, 71.20.Nr

I. INTRODUCTION: THE NEED FOR BOTH MESOSCOPIC AND ATOMISTIC MODELING OF NANOSTRUCTURE ELECTRONIC PROPERTIES

The use of quantum wells and superlattices in optoelectronics is predicated on designing confined energy levels with given separations. These energies depend both on mesoscopic conditions (e.g., geometric dimensions on a scale of ≈ 100 Å) and on atomistic details (e.g., interfacial segregation and interdiffusion on a scale of ≈ 5 Å). The dependence on atomic-scale properties is evident, for example, by significant changes in interband energies for nominally identical quantum systems grown at two different temperatures. For example, Yang *et al.*[1] found a 30–40 meV increase of a ≈ 300 meV band gap of a $(InAs)_{5.5}/(In_{0.28}Ga_{0.72}Sb)_{10}/(InAs)_{5.5}/(AlSb)_{14}$ structure, when the layer thicknesses were kept constant but the growth temperature of the device was increased from 460 to 500°C. This suggests that interdiffusion changes the band gap. Also, Vurgaftman, Meyer, and Ram-Mohan[2]

consequence is that in C_2

the potential features at the two different interfacial bonds.

Third, whereas in bulk solids the effective potential form factors $V(\mathbf{G})$ are defined only for *bulk* reciprocal-lattice vec-

fying $R_{n\alpha}$. The term β, which scales the kinetic energy in the Schrödinger equation, has been introduced to represent the quasiparticle nonlocal self-energy effects.[14] In fact it can be shown that at the lowest order, the leading effects of the nonlocal many-body potential can be represented by scaling the kinetic energy.[21] This kinetic-energy scaling is needed to simultaneously fit bulk effective masses and band gaps. The crystal potential is written as a superposition of atomic potentials v_α centered around the atomic sites. The potential includes the spin-orbit interaction, thus the wave functions $\psi_i(r)$ are spinors with spin-up and spin-down components.

For the atomic potential v_α we use atomic screened pseudopotentials whose Fourier transform are continuous functions of momentum[22] $\mathbf{q}$. The functions $v_\alpha(\mathbf{q})$ are determined for each atomic species α=Ga, Sb, In, As of the quaternary GaSb/InAs system. To obtain the values of the form factors at the intermediate $\mathbf{G}$ vectors appropriate for a given superstructure we need simply to evaluate the $v_\alpha(\mathbf{q})$ functions at the required $\mathbf{q}=\mathbf{G}$. The parameters entering the expression of the form factors are fitted to the experimentally measured electron and hole effective masses,[23] band gaps (target values at 0 K),[23] spin-orbit splittings,[23] hydrostatic deformation potentials of the band gaps,[23] band offsets,[23] and LDA-predicted single band-edge deformation potentials[24] of the four *binary* systems. The results of the fit are given elsewhere.[25]

To obtain the correct behavior of the band-edge energies under hydrostatic or biaxial strain deformations we have built the response to the strain directly into the screened atomic pseudopotentials v_α, adding an explicit strain dependent term $\delta v_\alpha(\epsilon)$. This term plays a crucial role in describing the variation of the valence-band edge and, separately, the conduction-band edge under arbitrary strains. This allows us to describe the modification of the valence- and conduction-band offsets when the systems are subjected to hydrostatic or biaxial deformation conditions such as in the case of epitaxial growth on a lattice-mismatched substrate. We fitted not only the experimental hydrostatic deformation potentials of the band gap, but also the *ab initio* calculated hydrostatic deformation potentials of the valence-band maximum.[24] Even though the binary GaSb and InAs systems are nearly lattice-matched (the lattice mismatch is relatively small, 0.6%), the interface Ga-As and In-Sb bonds are strongly deformed (their lattice mismatch with InAs and GaSb is about 6−7%) when the InAs/GaSb superlattices are grown on a GaSb (or InAs) substrate. Our scheme takes into account automatically the change in the valence-and conduction-band offsets of each constituent, including the interface bonds, due to changes in the biaxial constraints or local bonding deformations without the need to readjust any parameter. As a consequence, the heavy-hole wave function we calculate for the InAs/GaSb (001) superlattice has a much larger amplitude on the In-Sb interface bond than on the Ga-As bonds (see Ref. 25) in agreement with the results of *ab initio* calculations[15] as we also show in Fig. 2. Figure 2 shows a direct comparison between the heavy-hole charge density of a $(\mathrm{GaSb})_5/(\mathrm{InAs})_5$ superlattice integrated over the Brillouin zone (i.e., calculated and summed over the special *k* points) obtained from an *ab initio* calculation (in this case it is the self-consistent charge density) [Fig. 2(b)] and from the current atomistic empirical pseudopotential [Fig. 2(a)]. From this comparison we see that our empirical pseudopotential is able to reproduce the charge redistribution along the superlattice growth direction and at the two differ-

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AC, BC, AD, BD are the four binary compounds, in our case GaSb, GaAs, InSb, and InAs, whose properties have been directly fitted to extract the atomic pseudopotential parameters. This procedure leads to a potential for the InAs mono-

ness of the GaSb barrier is small, the electron wave functions overlap and extend along the growth direction.

The calculations have been performed both for superlattices with abrupt interfaces as well as for superlattices with interfacial disorder due to atomic segregation during growth. Some degree of interfacial segregation is always present in any real sample.[8] The effect of segregation has been modeled through a kinetic model of molecular-beam epitaxy growth. The details of our method for describing segregation are reported elsewhere.[12] We found that the band gaps of superlattices with segregated interfaces are always larger than the gaps calculated for the same nominal structures but assuming perfectly abrupt interfaces. We report our results for the $(InAs)_8/(GaSb)_n$ and $(InAs)_6/(GaSb)_n$ superlattices with abrupt interfaces in Fig. 5 comparing them with the results of other calculations, all using abrupt interfaces. In Fig. 6 we show our results for segregated superlattices obtained using the growth model with a growth temperature 380 °C and a deposition rate 0.5 ML/s, comparing them with the experimental data. growth.

and Tilton,[16] and two fourteen-band $\mathbf{k}\cdot\mathbf{p}$ calculations.[7] The calculations give the following values for the blueshifts of $n=8$: our EPM gives 95 meV, Dente and Tilton's EPM gives 49 meV, standard EFA gives 19 meV, EFA plus interface terms give 47 meV. The EPM theory of Ref. 20 while taking into account the effects of strain, when applied to the $(\mathrm{InAs})_{10}/(\mathrm{GaSb})_n$ superlattices (not measured yet) not only does not predict any blueshift of the band gap but finds a decreasing of the gap with increasing GaSb layer thickness n. We note the following.

(i) The two EPM calculations differ if the same (abrupt) geometry is assumed. The reason is the incomplete treatment of the interfaces by Dente and Tilton [factors (i)–(iv) outlined in Sec. II].

(ii) The standard $\mathbf{k}\cdot\mathbf{p}$ method hardly gives any blueshift. Only when interfacial potential terms are added,[9] fit to agree with the experimental data themselves, does one get the observed blueshift. However, the theory is not predictive since it requires an adjustable parameter to reproduce the data themselves.

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