Effects of interfacial atomic segregation on optical properties of InAs/GaSb superlattices

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Largely because of the lack of detailed microscopic information on the interfacial morphology, most electronic structure calculations on superlattices and quantum wells assume abrupt interfaces. Cross-sectional scanning tunneling microscopy STM) measurements have now resolved atomic features of segregated interfaces. We fit a layer-by-layer growth model to the observed STM profiles, extracting surface-to-subsurface atomic exchange energies. These are then used to obtain a detailed simulated model of segregated InAs/GaSb superlattices with atomic resolution. Applying pseudopotential calculations to such structures reveals remarkable electronic consequences of segregation, including a blueshift of interband transitions, lowering of polarization anisotropy, and reduction of the amplitude of heavy-hole wave functions at the inverted interface.

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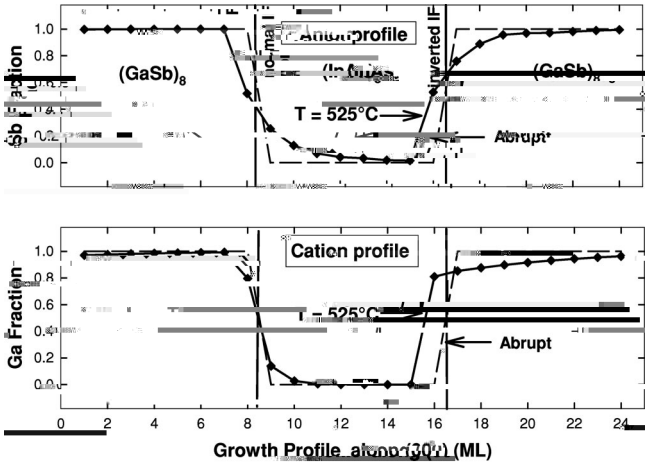
The natural difference in surface energies of the alloy components invariably causes certain atomic species to segregate to the surface during alloy growth. This effect necessarily leads to a structural and chemical asymmetry in quantum wells or superlattices. For example, since In segregates to the surface in preference to Ga, the growth of InAs-on-GaAs can lead to an abrupt interface, whereas the next step of growth of GaAs-on-InAs leads, by necessity, to an intermixed interface as In attempts to float from subsurface layers into the GaAs top layer. This segregation-induced structural asymmetry, noted previously in cation-segregating systems such as AlAs/GaAs and InAs/GaAs,[1,2] leads to significant changes in the optical properties.[3,4] Only recently were such effects explored in anion-segregating systems.[5,6] Of particular interest here is the infrared laser[7] and the light-emitting diode[8] material InAs/GaSb. Since the binary components InAs and GaSb do not share a common element, the two interfaces of an ideal InAs/GaSb quantum well have chemically distinct bonds that do not appear in the respective endpoint components see inset to Fig. 1). Thus, growth of InAs-on-GaSb ''normal interface'') leads to an interfacial Ga-As bond, whereas growth of GaSb-on-InAs ''inverted interface'') leads to an interfacial In-Sb bond. The ensuing lower (C_{2v}) symmetry relative to common-atom quantum wells (D_{2d}) makes the optical properties of systems without a common atom particularly susceptible to segregation effects. Largely because of lack of atomistic information on the structure of segregated interfaces, electronic structure calculations with only few exceptions,[3,4] have thus far assumed ideal, abrupt, interfaces. To address this issue one needs to a) quantify the degree of segregation at the interfaces, and b) establish how does atomic segregation affect the optical properties of the superlattices.

Cross-sectional scanning tunneling microscopy STM) has made a substantial progress in achieving the first task, identifying the interfacial bonding at GaSb/InAs heterojunctions, and measuring the compositional grading in $Ga_xIn_{1-x}Sb$/InAs superlattices caused by Sb segregation at an atomic level.[6] In this paper we tackle task b). We first formulate a layer-by-layer growth model, adjusting the atomic segregation energies of In/Ga and Sb/As so as to fit the detailed segregation profile measured[6] by cross-sectional STM for $Ga_xIn_{1-x}Sb$/InAs. We next apply this model to superlattices $(GaSb)_n/(InAs)_m$ for which optical properties were measured[9]), generating detailed atomistic structures of both ideally abrupt, as well as segregated interfaces, corresponding to a range of growth temperatures. Finally, we apply to these atomistic models of segregated superlattices the pseudopotential electronic structure method, finding the energy levels, wave functions, transition probabilities, and polarization anisotropy. This establishes directly the effect of interfacial segregation on electronic properties. We find the following.

i) Sb segregates primarily at the ''normal interface,'' whereas In segregates primarily at the ''inverted interface.'' However, some secondary segregation occurs too: As segregates somewhat at the inverted interface, but Ga does not segregate at the normal interface.

ii) Segregation reduces significantly the amplitude of the wave function of the first heavy hole hh1) at the inverted interface, hence diminishing significantly the dissimilarity with the normal interface, where hh1 always has a small

tem. The model simulates a layer-by-layer growth starting



and $E_{In/Ga}^{s\to b}$, so at very low growth temperatures ($<375\,^{\circ}$C) only anion segregation will be important, whereas appreciable In segregation is expected at higher temperatures ($>375\,^{\circ}$C).

Having obtained the segregation parameters for the InAs/GaSb system, we next model the atomistic structure of the superlattices used for optical studies.[10] We consider 001) $(InAs)_8/(GaSb)_{16}$ and $(InAs)_8/(GaSb)_8$ superlattices lattice matched to a GaSb substrate. While we have modeled the profile along the [001] growth direction e.g., Fig. 1) no experimental information is available on the atomistic arrangement in the perpendicular substrate 001) plane. We thus assume random arrangements in these planes, consistent with the planar composition profile dictated by the growth model. To achieve this we use a surface unit cell containing 16 atoms in the 001) plane, which are distributed randomly. Once we determine the superlattice configuration consistent with the solution of the growth model at a given growth temperature T_g, we permit local atomic displacements by a valence force field approach.[11]

Figure 2 shows the anion and cation segregation profiles obtained for a $(InAs)_8/(GaSb)_{16}$ superlattice at an high growth temperature. We see the following.

i) Segregation leads to the penetration of Sb and In deeply into the InAs and GaSb layers, respectively. The penetration length increases with growth temperatures. At $T_g = 525\,^{\circ}$C, Sb penetrates 5–6 ML into InAs, while In penetration length is much larger because of the larger $\Delta_{In/Ga}$), being about 11 ML.

ii) Sb segregation occurs primarily at the normal interfaces InAs-on-GaSb) where in the abrupt geometry a Ga-As bond exists, while In segregation occurs at the inverted interface GaSb-on-InAs), where in the abrupt geometry an In-Sb bond exixts. Our profiles at low (400 °C) growth temperatures closely agree with the STM images of the anion sublattice of Ref. 6 where it is seen that the anion intermixing is much larger at the normal interfaces than at the inverted interfaces[6] see inset in Fig. 1).

iii) There is also a substantial anion intermixing at the inverted interface and a smaller) cation intermixing at the normal interface. This is related to the difference $\Delta_{In/Ga}$ and $\Delta_{Sb/As}$ between the barrier energies [Eq. 1). If Δ is small, the segregation of the species with a higher energy barrier i.e., As) becomes noticeable at high T_g. We find $\Delta_{Sb/As} = 70$ meV, while $\Delta_{In/Ga}$ is much larger, 200 meV. This explains why at high T_g the anion profile at the inverted interface is more broadened for As segregation) than the analogous cation profile at the normal interface no Ga segregation). We will see below, that segregation at the *inverted interface* leads to a narrowing of the InAs well.

Using the calculated segregation profiles we generate detailed atomistic models for different growth temperatures. The electronic and optical properties of such structures are then obtained using the pseudopotential method.[12] The atomic pseudopotentials are obtained fitting the observed band gaps, effective masses, and deformation potentials of the constituent bulk binaries: GaSb, InAs, InSb, and GaAs.[13] Spin-orbit coupling is included as in Ref. 14.

Figures 3 a) and 3 b) shows the square of the (xy-averaged) wave functions of the hh1 hole state of the $(InAs)_8/(GaSb)_{16}$ superlattice. We see that the hh1 wave function is strongly affected by segregation: whereas in the

abrupt geometry 3a), the hh1 amplitude on the normal interface is much smaller than that on the inverted interface, in the segregated geometry 3b) the amplitude of hh1 on the inverted interface is reduced substantially and becomes similar to the amplitude on the normal interface. Segregation affects to a lesser degree the lh1 and $e1$ wave functions not shown). Another interesting result is related to the behavior of the potential [Figs. 3 b) and 3 d)]: segregation affects mostly the inverted interface where there is a shift of the interface itself and the InAs well becomes 1 ML narrower.

The interband transition energies and dipole oscillator strengths at the Brillouin-zone center have been calculated as a function of the growth temperature for the $(InAs)_8/(GaSb)_{16}$ superlattice Fig. 4). We see the following.

1) A segregation-induced *increase* blueshift) of the interband transition energies with growth temperature until $T_g = 425\,^{\circ}C$. The blueshift is due to the narrowing of the InAs well for electrons) and the broadening of the GaSb well for holes) with In segregation. The electron state becomes more confined with increasing T_g, whereas hole states become less confined, but their energies change at a smaller rate, so interband energies increase with T_g)formal