PHYSICAL REVIEW B **67**, 121306(R) (2003)

Electronic versus geometric contrast in cross-sectional STM images of III-V semiconductor heterostructures

S. G. Kim,[1] S. C. Erwin,[2] B. Z. Nosho,[2,*] L. J. Whitman[2]
[1]*Department of Physics and Astronomy, Mississippi State University, Mississippi State, Mississippi 39762*
[2]*Naval Research Laboratory, Washington DC 20375*
(Received 2 December 2002; published 21 March 2003)

We report first-principles calculations of the electronic and geometric structure of the (110) cross-sectional surfaces on InAs/GaSb superlattices, and compare the results with scanning tunneling microscopy images of filled electronic states. In both the predicted and measured images the InAs surfaces appear lower than GaSb, a height difference we show is caused primarily by differences in the electronic structure of the two materials. In contrast, local variations in the apparent height of surface atoms at InSb- or GaAs-like interfaces arise primarily from geometric distortions associated with local differences in bond length. Our calculated energies for atomic intermixing indicate that anion exchanges are exothermic at GaAs interfaces but endothermic at InSb interfaces. This difference may explain why GaAs interfaces are typically more disordered than InSb interfaces in these heterostructures.

DOI: 10.1103/PhysRevB.67.121306 PACS number(s): 68.35.Ct, 68.37.Ef, 68.65.Cd, 73.20.−r

Cross-sectional scanning tunneling microscopy (XSTM) has emerged as a powerful technique to characterize III-V semiconductor heterostructures.[1–11] Precise characterization of these materials is made possible by the fact that a zinc-blende III-V crystal readily cleaves along the {110} faces, producing a nearly defect-free surface that presents a cross-sectional view through a single lattice plane of structures grown on (001) substrates.[1] Tunneling microscopy is particularly useful for III-V (110) surfaces because of the simple surface structure, illustrated in Fig. 1(a). The III atoms relax towards the surface and V atoms away, shifting charge between the atoms and leaving the III dangling bond essentially empty and the V surface orbital filled. Because the STM surface topography in constant-current images approximately corresponds to contours of constant integrated charge density, only the III dangling bonds are seen in empty-state images of III-V {110} surfaces, while V orbitals are seen in filled-state images.[12,13] Therefore, XSTM images provide an apparently straightforward chemical identification of the atoms observed.

Since the first report of atom-selective STM images of GaAs(110),[12] and the observation of a heterostructure using XSTM,[1] a major issue has been delineating between electronic and geometric sources of height contrast. For nominally homogeneous materials where isolated impurity atoms are observed, such as dopants or substitutional defects, electronic origins of contrast have dominated the discussions.[14] For heterostructures, there are three contrast issues to be considered. First, the different III-V materials in a heterostructure usually have a different topographic height in filled-state images. Until the past few years,[6–10] discussion of this difference focused on electronic effects, specifically on the band gaps and band alignments (for filled states, the valence-band maximum), and the associated number of bands contributing to the tunneling.[1–5] The second contrast issue is related to the relative appearance of point defects associated with interdiffusion between the materials. For example, Harper *et al.* originally described As defects in GaSb as appearing lower in height because of the position of the As highest occupied molecular orbital.[15] Finally, there is the local height of interfacial bonds to consider. For systems without a common anion, such as InAs/GaSb, two different types of interfacial bonds are possible (InSb and GaAs bonds, in this case), and it has recently been proposed, based on crystallographic arguments, that the local XSTM height is primarily determined by local bond lengths.[7,8]

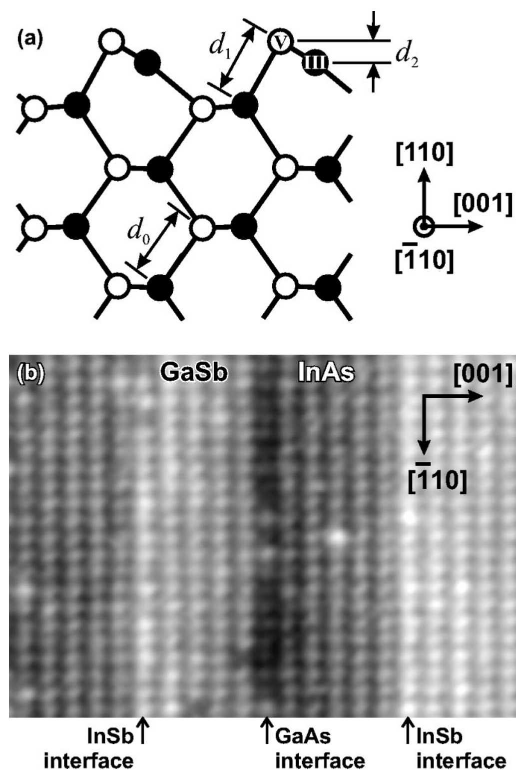

FIG. 1. (a) Relaxed geometry of a III-V (110) surface. d_0 denotes the bond length in the bulk, d_1 the out-of-plane III-V bond length indicated, and d_2 the height difference between III and V surface atoms. (b) Constant-current, filled-state XSTM image of an InAs/GaAs superlattice.

 ©2003 The American Physical Society

Report Documentation Page

Form Approved
OMB No. 0704-0188

Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
DEC 2002		**00-00-2002 to 00-00-2002**

4. TITLE AND SUBTITLE	
Electronic versus geometric contrast in cross-sectional STM images of III-V semiconductor heterostructures	5a. CONTRACT NUMBER
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Naval Research Laboratory,4555 Overlook Avenue SW,Washington,DC,20375**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT

Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
unclassified	**unclassified**	**unclassified**	**Same as Report (SAR)**	**4**	

In this work, we use first-principles methods to describe the electronic and geometric structure of the (110) surfaces of InAs/GaSb superlattices. We find that the apparent surface height difference between the two materials is primarily an electronic structure effect, but the local height differences observed for InSb and GaAs interfacial bonds are mostly geometric in nature. In addition, the calculations reveal that atomic intermixing lowers the energy of GaAs interfaces, favoring disorder.

The XSTM measurements were performed in ultrahigh vacuum using InAs/GaSb superlattice samples grown by solid-source molecular-beam epitaxy, as described in detail previously.[8] All images presented here are of (110) surfaces recorded with filled states at contant current (2.2–2.5 V, 150–200 pA). Figure 1(b) shows a typical filled-state XSTM image for a (110) surface. In our theoretical work, we focus on three aspects of this representative image. First, the large-scale topography shows an alternating pattern of brighter and darker bands (higher and lower apparent heights) corresponding to GaSb and InAs layers, respectively. Second, a higher Sb row is evident at InSb interfaces in this figure, whereas a lower As row is evident at the GaAs interface. Third, the degree of atomic disorder at the two interfaces is qualitatively different: InSb interfaces typically appear atomically abrupt, in contrast to GaAs interfaces which often exhibit significant disorder.

Our calculations are based on density-functional theory, using ultrasoft pseudopotentials as implemented in the VASP code.[16,17] Exchange-correlation effects were treated within the local-density approximation. The plane-wave cutoff was 150 eV, and the sampling of the quasi-one-dimensional Brillouin zone was equivalent to using 64 k-points in the full zone of the primitive fcc cell.

We used supercell geometries to represent the InAs/GaSb heterostructures. Because of the importance of interfacial strain in this material system, we constructed supercells with starting geometries that minimized, as much as possible, any artificial strain at the interfaces. Our procedure consisted of the following three steps.

(1) Bulk calculations were performed to obtain the optimized lattice parameters for four different types of zincblende III-V semiconductor crystals: InAs, GaSb, InSb, and GaAs.

(2) For each of these four different homogeneous materials, we constructed slab supercells representing the unreconstructed (110) surfaces. This was done by periodically replicating the unit cells obtained in the previous step four times along the (110) direction, resulting in slabs containing eight atomic layers. We separated adjacent slabs by a vacuum region corresponding to five atomic layers, which we confirmed was sufficient to make the interaction between slabs negligible. All of the atoms within each slab were then relaxed within the constraint of the fixed in-plane lattice constant determined from the previous step.

(3) Next we periodically replicated the relaxed slabs six times along the (001) direction, and joined two such extended slabs together to form various III-V heterostructures with (001) interfaces and exposed (110) surfaces. For each interfacial bond case (InSb and GaAs), the two slabs were joined at a distance chosen as to allow every interfacial bond to have a bond length corresponding to the bulk lattice constant computed in step (1). The resulting supercells each contained a total of 96 atoms, with four different atomic species, corresponding to a (001) superlattice period of 24 atomic layers. Finally, the positions of all atoms were completely relaxed within the constraints of fixed superlattice period and fixed lattice constant along $[1\bar{1}0]$.

TABLE I. Lattice constants a_0 and bond lengths d_i of the relevant III-V materials, in angstrom. See Fig. 1(a) for definitions of the different bond lengths.

	a_0	a_0	d_0	d_1	d_2
InAs	6.01	6.06	2.60	2.62	0.77
GaSb	6.04	6.10	2.62	2.65	0.76
InSb	6.43	6.47	2.78	2.80	0.86
GaAs	5.60	5.65	2.42	2.44	0.70

[a]Lattice constant from this work.
[b]Experimental lattice constant.

The equilibrium lattice constants calculated in step (1) are listed, along with their experimental values, in the first two columns of Table I. The agreement is very good, with all errors less than 1%. The relaxed (110) surfaces of the four homogeneous materials, as obtained from step (2), show the surface buckling obtained in many previous studies.[13] The calculated buckling, illustrated in Fig. 1(a) and tabulated in the last three columns of the table, is in good quantitative agreement with experiment; in particular, the height difference between III and V atoms for InAs is 0.77 Å, in excellent agreement with the value of 0.78 Å determined by low-energy electron diffraction.[18] After relaxing the various III-V heterostructures described in step (3), we simulated XSTM images using the method of Tersoff and Hamann.[19] To simulate filled-state images, we integrated the local density of states (LDOS) from 1 eV below the Fermi level up to the Fermi level; the surface of constant integrated LDOS then corresponds to the ideal STM topography.

Our results for InAs/GaSb heterostructures with InSb interfacial bonds are shown in Fig. 2. The geometry of the fully relaxed (110) surface is displayed in Fig. 2(b). As in the case of homogeneous structures, the surface atoms buckle, causing the group-V atoms (As and Sb) to move outward and the group-III atoms (In and Ga) to move inward. At the interface, Sb atoms relax still further outward so as to partially relieve compressive strain in the InSb bonds. The resulting simulated XSTM image closely resembles the measured image, as indicated by the inset of Fig. 2(a) and the calculated XSTM profile across the row maxima shown in Fig. 2(c). Away from the interface, the topographic maxima (from the integrated LDOS) are 0.15 Å higher on the GaSb than on the InAs, in good agreement with the height difference of about 0.2 Å typically observed in XSTM images. Interestingly, the difference in height between the actual Sb and As atoms associated with the topography is much smaller, 0.06 Å, demonstrating that the XSTM height difference is primarily caused by the surface electronic structure. Based on the calculated structure, the opposite appears true for the local to-

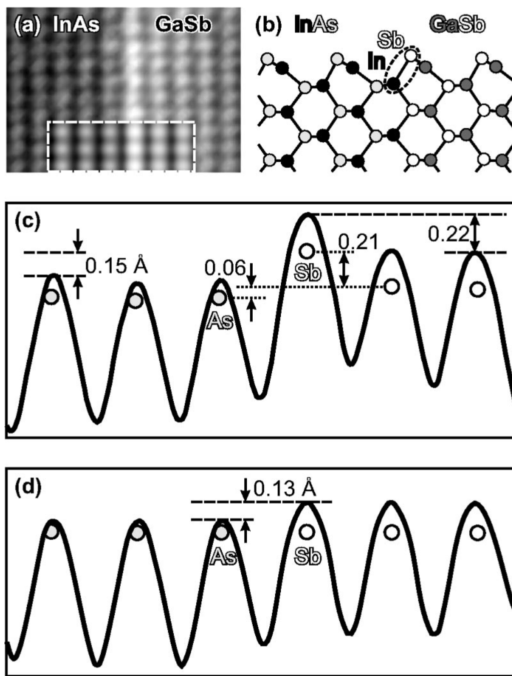

FIG. 2. (a) An XSTM image of an InAs-GaSb interface with InSb interfacial bonds. Inset: Simulated XSTM image of this structure. Both gray scales span about 1 Å. (b) Side view of the fully relaxed surface geometry for this interface. (c) Line profile for the simulated image along the [001] direction (across the row maxima). Circles denote the relaxed positions of the surface As and Sb atoms. (d) Line profile for a simulated image of two homogeneous structures joined together at their ideal lattice positions and allowed to relax the electronic but not the geometric structure.

pographic height difference observed at the InSb interface. The Sb row forming InSb bonds is geometrically higher by about 0.2 Å than the Sb atoms on the GaSb surface, essentially the same height difference that occurs in the integrated LDOS, demonstrating that the observed height difference in this case is associated with the geometric structure.

In order to more explicitly delineate the relative contributions of geometric and electronic structure relaxation on the apparent STM topography, we calculated the electronically-relaxed structure of an "ideal" InAs/GaSb heterostructure, with all interfacial atoms frozen at ideal positions. In this ideal geometry all atoms in the top layer have exactly the same height, and thus differences in heights across the computed topography originate from purely electronic effects. As shown in Fig. 2(d), the electronic structure alone creates a difference in height of 0.13 Å between the InAs and GaSb surfaces, close to the 0.15 Å difference calculated with full relaxation, further supporting our conclusion that electronic structure underlies the measured height difference between the two materials.

Our analogous results for InAs/GaSb heterostructures with GaAs interfacial bonds are summarized in Fig. 3, with the calculated topography again in qualitatively good agreement with that observed experimentally. At this interface the structural relaxation is quite different from the InSb case. As shown in Table I, GaAs bonds have the shortest surface bond length d_1 and thus As atoms near the interface relax even

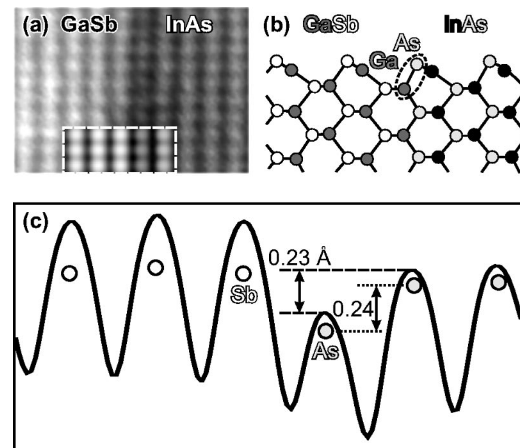

FIG. 3. (a) An XSTM image of an InAs/GaSb interface with GaAs interfacial bonds. Inset: Simulated image of this structure. (b) Side view of the fully relaxed surface geometry for this interface. (c) Line profile for the simulated image along the [001] direction.

further inward than Sb relaxes outward at InSb interfaces. The resulting geometric height of the As row at the GaAs interface is 0.24 Å lower than the As atoms far from the interface. However, as we saw at the InSb interface, the computed topographic height difference is almost identical, 0.23 Å, indicating that the local depression of the GaAs interface is almost completely geometric in origin.

Finally, we address why interfacial roughness appears to depend on the interfacial bond type, with GaAs interfaces generally observed to be more disordered. Experimentally, most defects occur close to the interfaces, suggesting that they arise from simple Ga-In or As-Sb exchanges across the interface, rather than from bulk defects such as vacancies or cation-anion antisites. An example of an apparent Sb atom observed in an As site at a GaAs interface is highlighted in Fig. 4(a). To confirm the structural assignment of such features, we theoretically modeled such a defect by replacing one of the surface As atoms at a GaAs interface with an Sb

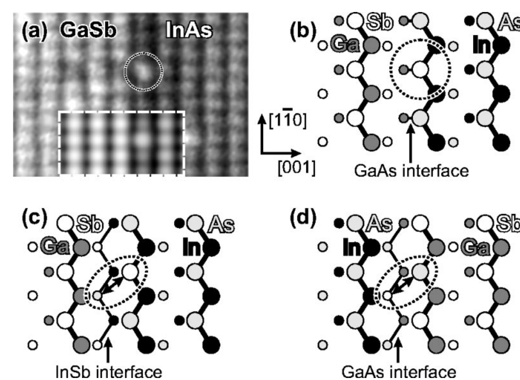

FIG. 4. (a) An XSTM image of InAs/GaSb with a GaAs interface. An apparent Sb atom in an As site at the interface is circled. Inset: Simulated XSTM image for such a defect. (b) Top view of the Sb-in-As-site defect model structure, with the top-layer atoms shown larger. (c) Model used to calculate the energetics of an As-Sb exchange across an InSb interface. (d) Model for an As-Sb exchange across a GaAs interface.

atom, as illustrated in Fig. 4(b). The simulated XSTM image for this structure, shown in the inset of Fig. 4(a), closely resembles the experimental result.

To investigate the energetics of anion interfacial defects, we consider the simplest defects that both preserve the global stoichiometry and satisfy local chemical bonding requirements, As-for-Sb exchanges. Such exchanges represent a simple mechanism for interfacial disorder at a nominally abrupt interface. We studied the energetics of exchanging adjacent As and Sb atoms both at an InSb interface and at a GaAs interface, as shown in Figs. 4(c) and 4(d), respectively. The structures were fully relaxed before and after the exchange and the change in the total energy then computed. At the InSb interface, the exchange raised the total energy by 7 meV. Surprisingly, at the GaAs interface, the same process actually *lowered* the total energy by 22 meV; i.e., the formation of such defect pairs is *exothermic*. This result implies that abrupt GaAs interfaces are thermodynamically unstable. Therefore, although kinetic barriers may suppress anion exchanges, one should generally expect GaAs interfaces to be more disorderd than InSb interfaces (as widely observed).

In summary, we have used first-principles electronic-structure methods to clarify the interpretation of XSTM images of (110) surfaces on cleaved InAs/GaSb heterostructures, focusing on the differences between interfaces with InSb versus GaAs bonds. We find that the apparent height differences between the InAs and GaSb surfaces are largely associated with the electronic structure, whereas the local height differences at the InSb and GaAs interfaces are caused by geometric relaxation from the partial relief of local bond strain. Finally, our calculations of the energies associated with interfacial exchange of anions reveal that As-for-Sb exchanges are exothermic at GaAs interfaces, but endothermic at InSb interfaces. This result may explain why GaAs interfaces are generally more disordered than InSb interfaces in InAs/GaSb heterostructures.

The authors would like to thank B.R. Bennett and M.J. Yang for fabricating the InAs/GaSb superlattice samples. This work was supported by the Office of Naval Research and the Defense Advanced Research Projects Agency.

*Present address: HRL Laboratories, LLC, 3011 Malibu Canyon Rd., Malibu, CA 90265.

[1] O. Albrektsen, D.J. Arent, H.P. Meier, and H.W.M. Salemink, Appl. Phys. Lett. **57**, 31 (1990).
[2] S. Gwo, K.-J. Chao, C.K. Shih, K. Sadra, and B.G. Streetman, Phys. Rev. Lett. **71**, 1883 (1993).
[3] R.M. Feenstra, D.A. Collins, D.Z.Y. Ting, M.W. Wang, and T.C. McGill, Phys. Rev. Lett. **72**, 2749 (1994).
[4] A.R. Smith, K.-J. Chao, C.K. Shih, Y.C. Shih, K.A. Anselm, and B.G. Streetman, J. Vac. Sci. Technol. B **13**, 1824 (1995).
[5] J. Harper, M. Weimer, D. Zhang, C.-H. Lin, and S.S. Pei, J. Vac. Sci. Technol. B **16**, 1389 (1998).
[6] R.M. Feenstra, Physica B **273-274**, 796 (1999).
[7] J. Steinshnider, M. Weimer, R. Kaspi, and G.W. Turner, Phys. Rev. Lett. **85**, 2953 (2000).
[8] B.Z. Nosho, W. Barvosa-Carter, M.J. Yang, B.R. Bennett, and L.J. Whitman, Surf. Sci. **465**, 361 (2000).
[9] D.M. Bruls, J.W.A.M. Vugs, P.M. Koenraad, H.W.M. Salemink, J.H. Wolter, M. Hopkinson, M.S. Skolnick, F. Long, and S.P.A. Gill, Appl. Phys. Lett. **81**, 1708 (2002).
[10] S.L. Zuo, Y.G. Hong, E.T. Yu, and J.F. Klem, J. Appl. Phys. **92**, 3761 (2002).
[11] B.Z. Nosho, B.R. Bennett, L.J. Whitman, and M. Goldenberg, Appl. Phys. Lett. **81**, 4452 (2002).
[12] R.M. Feenstra, J.A. Stroscio, J. Tersoff, and A. P. Fein, Phys. Rev. Lett. **58**, 1192 (1987).
[13] B. Engels, P. Richard, K. Schroeder, S. Blügel, P. Ebert, and K. Urban, Phys. Rev. B **58**, 7799 (1998), and references therein.
[14] P. Ebert, Surf. Sci. Rep. **33**, 121 (1999), and references therein.
[15] J. Harper, M. Weimer, D. Zhang, C.-H. Lin, and S.S. Pei, J. Vac. Sci. Technol. B **16**, 1389 (1998).
[16] G. Kresse and J. Hafner, Phys. Rev. B **47**, 558 (1993).
[17] G. Kresse and J. Furthmüller, Phys. Rev. B **54**, 11 169 (1996).
[18] C. Mailhiot, C.B. Duke, and D.J. Chadi, Phys. Rev. B **31**, 2213 (1985).
[19] J. Tersoff and D.R. Hamann, Phys. Rev. Lett. **50**, 1998 (1983).