1923
doi:10.1017/S1431927615010399 Paper No. 0960
Microsc. Microanal. 21 (Suppl 3), 2015

Atomic Scale Structure and Bonding Configurations in Monocrystalline $Al_{1-x}B_xPSi_3$ Alloys Grown Lattice Matched on Si(001) Platforms

P. Sims[1], T. Aoki[2], J. Menéndez[3], and J. Kouvetakis[1]

[1.] Department of Chemistry & Biochemistry, Arizona State University, Tempe, AZ 85287 USA
[2.] LeRoy Eyring Center for Solid State Science, Arizona State University, Tempe, AZ 85287 USA
[3.] Department of Physics & Astronomy, Arizona State University, Tempe, AZ 85287 USA

Atomic-resolution imaging and element-selective mapping of composition and structure in crystalline solids using aberration corrected microscopy with probe sizes comparable to atomic spacing is a powerful technique which has to date been successfully used to study known systems with classical structures such as hexagonal BN [1] and perovskites [2]. In this study we have used this method to directly investigate atomic scale structure, and bonding configurations in newly synthesized Al_{1-x} B_xPSi_3/Si(001) alloys comprised of earth abundant elements. These materials are grown on Si buffered Si(001) *via* reaction of $Al(BH_4)_3$ and $P(SiH_3)_3$ using low pressure gas source molecular beam epitaxy. The reactions deliver tetrahedral $AlPSi_3$ and $BPSi_3$ building blocks, which are expected to incorporate intact into the crystal, creating a Si-like framework containing isolated Al-P and B-P donor-acceptor pairs. The addition of B into $AlPSi_3$ facilitates lattice matching of the material with the Si template yielding fully relaxed structures devoid of mismatch induced defects as required for application in photovoltaic devices.

In this study the bonding patterns of these materials are directly resolved and the constituent atoms are explicitly identified thereby yielding the alloy composition/structure at the Angstrom level. The work shows that the new materials are likely constructed *via* directed molecular assembly of chemical building blocks sharing the same chemical composition and basic tetrahedral motifs as the solid phases. Another unique functionality is the enhanced light absorption that is significantly extended relative to Si making these Si-based lattice-matched systems likely candidates as top junction materials in tandem solar cells designs for next generation Si-based PV technologies with efficiencies exceeding that of Si.

XTEM (JEOL-4000EX) and probe-corrected STEM/EELS experiments were performed using a Nion UltraSTEM 100 equipped with HERMES^{TM} and Gatan Enfinium^{TM} EELS spectrometer. Figures 1 (a) and (b) are XTEM/STEM images of the entire epilayer and interface region, demonstrating high quality epitaxy of monocrystalline layers devoid of interfacial dislocations and threading defects. The diffraction pattern reveals a Si-like structure with a lattice parameter nearly equal to that of Si, depending on the amount of B uptake, which reaches up to 6 at.% relative to Al. EELS indicates that the films are single phase alloys with uniform elemental distributions and not a mixture of Si and zinc-blende $Al_{1-x}B_xP$. Atomic resolution mapping identified distinct randomly oriented motifs for the Al-P pairs as expected for single phase-alloy with a disordered structure. The Si component is uniformly arranged throughout individual crystal columns consistent with a diamond lattice (Figure 2(d)). Raman spectroscopy provides evidence that the B atoms are incorporated as isolated B-P units with dilated bond distances relative to zinc-blend BP (Figure 3(a)). The single phase character of the samples is corroborated by triple axis XRD analysis of 004, 135 and 224 reflections (Figure 3 (b)). Measurements of the dielectric function using spectroscopic ellipsometry revealed higher absorption relative to bulk Si in the visible range [3].

References:

[1] K. Suenaga *et al*, J. Electron Microsc. **61** (2012) p. 285.
[2] D. Muller *et al*, Science, **319** (2008) p. 1073.
[3] This work was supported by NSF and NREL. The authors acknowledge the use of facilities in the John M. Cowley center for High Resolution Electron Microscopy at Arizona State University.
[4] P. Sims *et al*, Chem. Mater., submitted

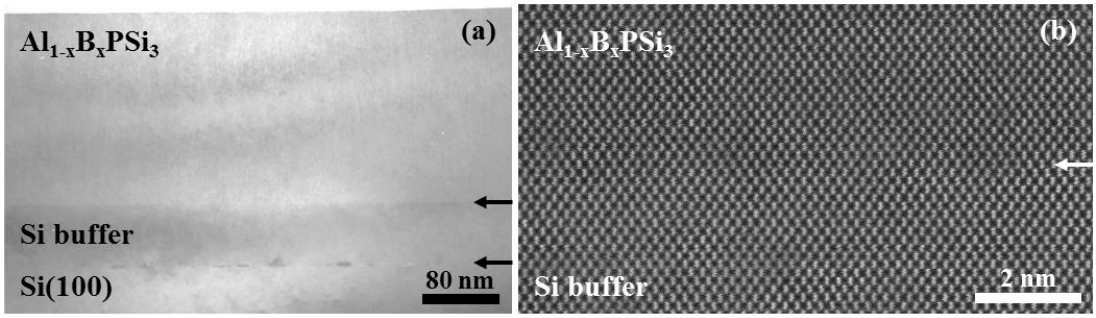

Figure 1. TEM/STEM images of $Al_{1-x}B_xPSi_3$/Si(001); (a) XTEM of full epilayer grown lattice matched on Si(001) wafer. The arrows denote the location of the interfaces. (b) STEM HAADF image showing full alignment of the (111) lattice fringes. The close chemical and structural relationship between the two materials allows near perfect crystal integration devoid of interfacial defects.

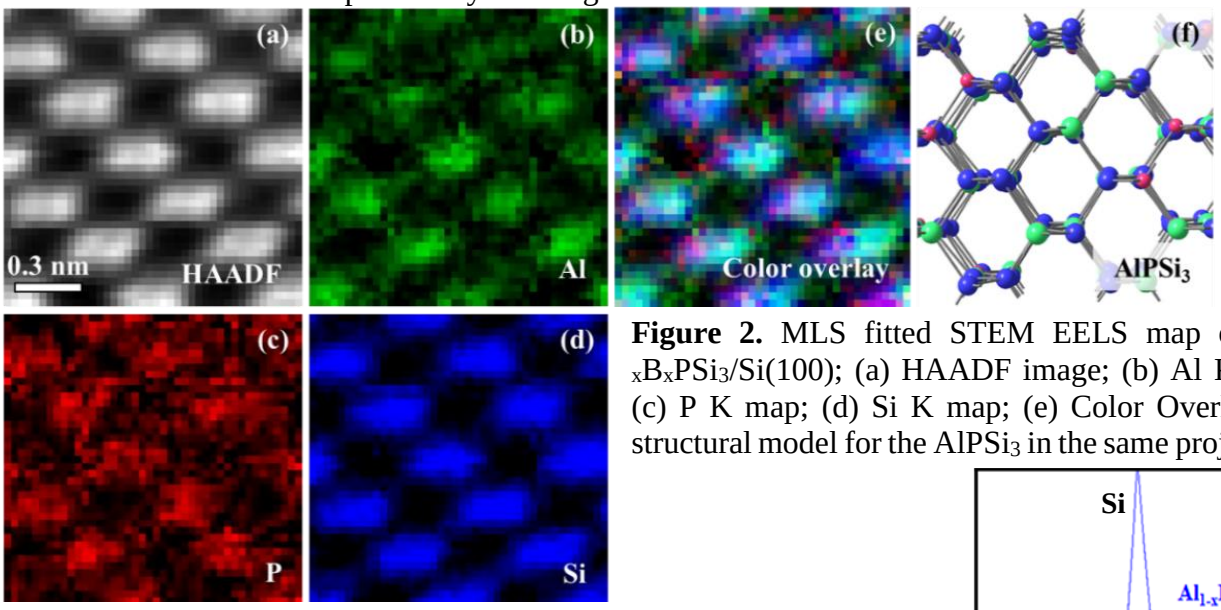

Figure 2. MLS fitted STEM EELS map of $Al_{1-x}B_xPSi_3$/Si(100); (a) HAADF image; (b) Al K map; (c) P K map; (d) Si K map; (e) Color Overlay; (f) structural model for the $AlPSi_3$ in the same projection.

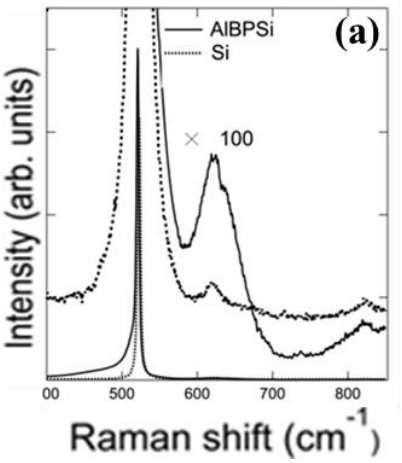

Figure 3. (a) Raman spectra of $Al_{1-x}B_xPSi_3$ and Si. The peak at 631 cm^{-1} is assigned to vibrations form isolated BP pairs in the structure [4].

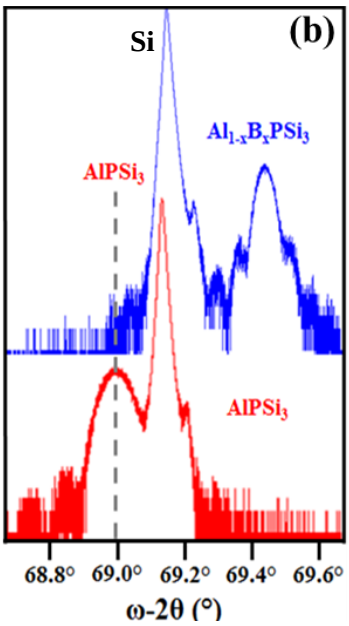

Figure 3. (b) XRD ω-2θ plots of $Al_{1-x}B_xPSi_3$ (blue) and $AlPSi_3$ (red) showing 004 reflections of the diamond lattice

https://doi.org/10.1017/S1431927615010399 Published online by Cambridge University Press