New semiconductor alloys, $GaSb_xN_{1-x}$ for photoelectrochemical water splitting: computations and experiments

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ABSTRACT

Applicability of the $Ga(Sb_x)N_{1-x}$ alloys for practical realization of photoelectrochemical water splitting is investigated using first-principles density functional theory incorporating the local density approximation and generalized gradient approximation plus the Hubbard U parameter formalism. Prior results with calculations revealed that a relatively small concentration of Sb impurities is sufficient to achieve a significant narrowing of the band gap, enabling absorption of visible light.[1] Theoretical results predict that $Ga(Sb_x)N_{1-x}$ alloys with 2 eV band gaps straddle the potential window at moderate to low pH values, thus indicating that dilute $Ga(Sb_x)N_{1-x}$ alloys could be potential candidates for splitting water under visible light irradiation. See Figure 1. Theoretical computations with Sb composition beyond 7% change the electronic band gap from direct to indirect.

Experimental synthesis is carried out using metal organic chemical vapor deposition using trimethyl gallium (TMGa) and Trimethyl Antimony (TMSb) and ammonia. Crystalline $GaSb_xN_{1-x}$ films were obtained at x values ranging from 0-5%. The synthesis was carried out on different planar substrates and GaN nanowires. Optical measurements confirm that severe band gap reduction occurs with incorporation of antimony in to GaN as predicted by the theoretical calculations. X-Ray Diffraction (XRD) results confirm the lattice expansion at small concentrations of antimony. This presentation will highlight our results with both synthesis and photoelectrochemical characterization of GaSbxN1-x alloys.

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References:

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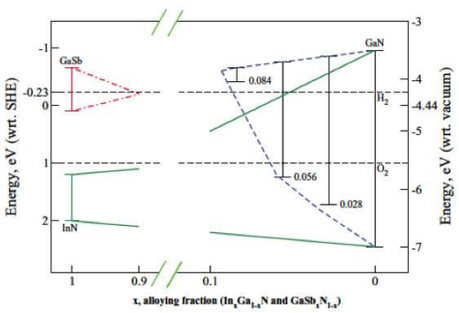

Figure 1. FIG. 4: A qualitative comparison of the band edges of $In_x(Ga_{1-x})N$ and $Ga(Sb_x)N_{1-x}$ alloys is presented. The almost linear band edge variation in $In_x(Ga_{1-x})N$ alloys is indicated by the solid green line. The projected band edge variation of $Ga(Sb_x)N_{1-x}$ is plotted using the reported band gap values for dilute nitrogen alloys, indicated by the dash-dot line. The band-edge and band gap positions calculated in this work are indicated by the vertical lines and the variation is plotted as indicated by the dashed lines emerging from GaN band edges. It is evident that $Ga(Sb_x)N_{1-x}$ alloy with 2 eV band gap straddles the H_2/O_2 redox potential window and that very low Sb incorporation is su_cient to reduce the band gap of $Ga(Sb_x)N_{1-x}$ alloys from UV to the visible region. (Reference 1).