Atom Probe Tomography of Low-Dimensional Materials: III-As Nanowire Heterostructures and Doped Layered Chalcogenides

L. J. Lauhon[+]

Department of Materials Science and Engineering, Northwestern University, Evanston, IL 60208, United States.

We will describe the application of atom probe tomography (APT) to the analysis of facet driven composition fluctuations in GaAs-AlGaAs nanowire core-shell heterostructures and the distribution of Ag dopant atoms in $(PbSe)_5(Bi_2Se_3)_3$. AlGaAs is a ternary semiconductor whose composition can be tuned smoothly from GaAs to AlAs in molecular beam epitaxial growth. However, transmission electron microscopy and APT of GaAs-AlGaAs core-shell heterostructures have revealed facet dependent segregation.[1] Furthermore, APT analysis has linked quantum dot like emission spectra to composition fluctuations that exceed those expected for a random alloy,[1] despite the absence of a miscibility gap at typical growth temperatures. When the shell growth temperature is reduced from 560 °C to below 400 °C, the *non*-randomness of the alloy distribution in the AlGaAs shell is greatly reduced.[2] These observations will be explained in terms of a facet dependent segregation that is kinetically suppressed at reduced growth temperatures.

van der Waals heterostructures in layered or two-dimensional (2D) materials represent an entirely new class of ultrathin heterostructure. Doping of the constituent 2D materials provides a route to tuning electronic properties and forming new types of heterojunctions between semiconductors, metals, and superconductors. A nanoscale perspective on the dopant distribution can provide important insights into electronic structure and physical behaviors. APT analysis of Ag doped $(PbSe)_5(Bi_2Se_3)_3$ [3] shows that Ag dopes both Bi_2Se_3 and PbSe layers in $(PbSe)_5(Bi_2Se_3)_3$, and correlations in the position of Ag atoms suggest a pairing across neighboring Bi_2Se_3 and PbSe layers. Density functional theory (DFT) calculations confirm the favorability of substitutional doping for both Pb and Bi and provide insights into the observed spatial correlations in dopant locations. This work demonstrates the feasibility of APT analysis of 2-D materials.

[+] Author for correspondence: lauhon@northwestern.edu

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[2] Bernhard Loitsch, Nari Jeon, Markus Döblinger, Julia Winnerl, Eric Parzinger, Sonja Matich, Ursula Wurstbauer, Hubert Riedl, Gerhard Abstreiter, Jonathan J Finley, Lincoln J Lauhon, and Gregor Koblmüller. "Suppression of alloy fluctuations in GaAs-AlGaAs core-shell nanowires". *Applied Physics Letters* **109**, 093105 (2016).

[3] Xiaochen Ren, Arunima K Singh, Lei Fang, Mercouri G Kanatzidis, Francesca Tavazza, Albert V Davydov, and Lincoln J Lauhon. Atom Probe Tomography Analysis of Ag Doping in 2D Layered Material $(PbSe)_5(Bi_2Se_3)_3$. *Nano Lett ASAP*. DOI: 10.1021/acs.nanolett.6b02104.