doi:10.1017/S1431927614004322
Microsc. Microanal. 20 (Suppl 3), 2014

High Resolution EELS Study of $Ge_{1-y}Sn_y$ and $Ge_{1-x-y}Si_xSn_y$ Alloys

Liying Jiang[1], Toshihiro Aoki[2], John Kouvetakis[3], and José Menéndez[1]

[1] Department of Physics, 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 Chemistry and Biochemistry, Arizona State University, Tempe, AZ 85287, USA

Germanium (Ge) is a promising material for photonic device applications. Direct band gap photoluminescence is seen from elemental Ge due to the small energy separation between the L- and Γ-valleys [1]. This separation is further reduced by alloying Ge with Sn [2]. Unfortunately, the solid solubility of Sn in Ge is very low (<1.1%) limiting the thermal stability of $Ge_{1-y}Sn_y$ alloys with high Sn contents (y>0.011) [3]. Ternary $Ge_{1-x-y}Si_xSn_y$ alloys exhibit an enhanced thermodynamic stability relative to $Ge_{1-y}Sn_y$ analogs due to increased mixing entropy afforded by the incorporation of Si in the $Ge_{1-y}Sn_y$ lattice [4]. The incorporation of Si not only improves the thermal stability but changes the electronic structure [5]. The atomic distribution in these alloys at the sub-nanometer scale is of paramount concern, since even slight deviations from randomness can have a dramatic impact on the electronic structure. In this study $Ge_{1-y}Sn_y$ and $Ge_{1-x-y}Si_xSn_y$ were grown on Ge buffered Si(100) and were extensively characterized by cross-section transmission electron microscopy (XTEM) and "element-selective" electron energy loss spectral (EELS) mapping. The TEM samples were prepared using mechanical thinning and dimple polishing followed by argon-ion-milling. High-angle annular-dark-field (HAADF) images and EELS spectra were collected on JEOL ARM 200F operated at 200kV with spot size 0.13nm in aberration corrected STEM mode. Atom-selective EELS mapping of lattice columns was performed using a GATAN Enfinium™ spectrometer.

Multiple scans throughout each sample revealed well defined ionization peaks corresponding to Ge (L) and Sn (M) core edges at 1217 eV and 483 eV, respectively. The Si L edge at 99eV and K edge at 1836eV are visible in $Ge_{1-x-y}Si_xSn_y$ alloys, but the signal is not sufficiently strong to generate a well-defined Si map. Figure 1 reveals that the $Ge_{1-y}Sn_y$/Ge interface is chemically sharp and the Sn atoms are evenly distributed in the Ge matrix. EELS analysis was performed in a 2x2 nm^2 area of a 60 nm thick specimen indicating full Sn substitutionally and random distribution of the atoms in the Ge host lattice (see Fig. 2). Observation of the $Ge_{1-x-y}Si_xSn_y$/Ge interface in Figure 3 reveals that the Sn distribution is uniform throughout the layer. Figure 4 shows that the Sn atoms occupied Ge lattice columns as evidenced by atomic resolution EELS maps from a sample volume of 4x4 nm^2 in the lateral direction and a thickness of 25-30 nm [5]. Further work to be reported at the meeting will include electronic structure characterization and band gap determination obtained from EELS. The latter results will be compared with optical and theoretical calculations.

References:

[1] G. Grzybowski, *et al*., Phys. Rev. B **84** (20), 205307 (2011).
[2] J. Mathews, *et al.*, Appl. Phys. Lett. **97** (22), 221912 (2010).
[3] F. A. Trumbore, J. Electrochem. Soc. **103** (11), 597 (1956).
[4] J. Xie, *et al.*, Appl. Phys. Lett. **95** (18), 181909 (2009).
[5] L. Jiang *et al*., Chemistry of Materials, **DOI:** 10.1021/cm403801b
[6] The authors acknowedge support from NSF DMR-1309090 and use of facilities in the John M.Cowley center for high resolution electron microscopy at Arizona State University.

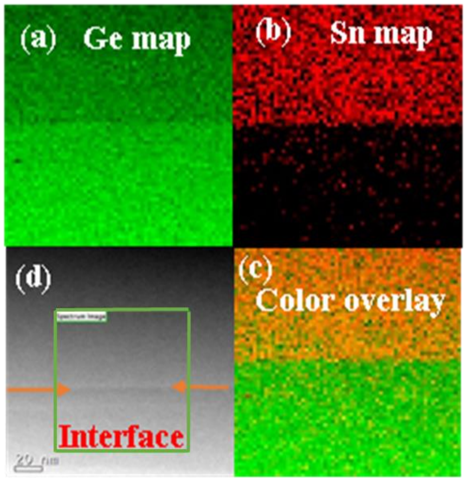

Figure 1.Low magnification EELS elemental mapping of $Ge_{0.96}Sn_{0.04}$/Ge interface. (a) Ge map extracted from EELS spectrum (b) Corresponding map of Sn atoms colored red (c) hybrid map of Sn and Ge near the interface, Sn is uniformly distributed in Ge matrix.(d) HAADF image identifying

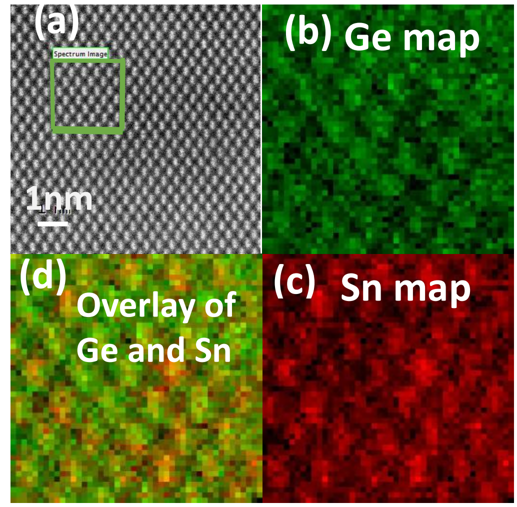

Figure 2.Atomic resolution EELS elemental mapping of $Ge_{0.96}Sn_{0.04}$ film. (a) HAADF high-resolution image dentifying the rectangular region analyzed by EELS. (b) Map of Ge atoms extracted from the EELS (c) Corresponding map of Sn atoms (d) Hybrid map of the Sn and Ge patterns

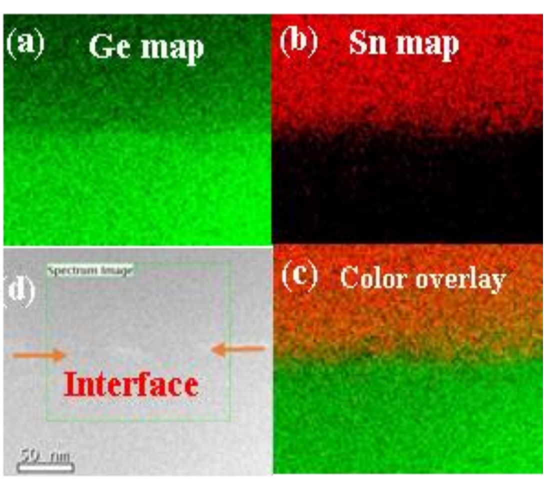

Figure 3. Low magnification EELS elemental mapping of $Ge_{0.915}Si_{0.035}Sn_{0.053}$ /Ge interface. (a) Ge map extracted from EELS spectrum (b) Corresponding map of Sn atoms colored red (c) hybrid map of Sn and Ge near the interface, Sn is uniformly distributed in Ge matrix. (d) HAADF image identifying the EELS analysis area.

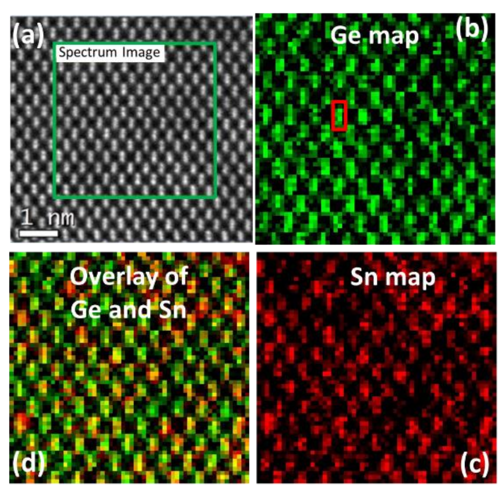

Figure 4. Atomic resolution EELS elemental maps of $Ge_{0.915}Si_{0.035}Sn_{0.053}$ sample. (a) HAADF high-resolution image identifying the rectangular region analyzed by EELS. (b) Map of Ge atoms extracted from the EELS spectrum showing the characteristic dimer rows colored green (c) Corresponding map of Sn atoms colored red (d) Hybrid map of the Sn and Ge patterns

https://doi.org/10.1017/S1431927614004322 Published online by Cambridge University Press