doi:10.1017/S1431927614003924
Microsc. Microanal. 20 (Suppl 3), 2014

Observing the Interplay Between Composition and Phonon Transport in $Bi_2Te_{3-x}Se_x$ Alloys using ADF STEM

J. H. Dycus[1], A. Oni[1], X. Sang[1], T. Chan[1], C. Koch[1], J. M. LeBeau[1]

[1.] Department of Materials Science and Engineering/ North Carolina State University, Raleigh, NC 27695, USA.

Bismuth telluride-selenide alloys are some of the most commonly used thermoelectrics at room temperature. In these materials, phonon scattering plays a pivotal role in controlling the thermoelectric properties where decoupling phonons and charge carriers induces a net heat flux that results in an electric potential. Understanding the heat flow requires knowledge of phonon scattering behavior as a function of alloying, at defects, and within the microstructure. In $Bi_2Te_{3-x}Se_x$ alloys, the structure consists of stacked 'quintuple' layers in the sequence Te(1)-Bi-Te(2)-Bi-Te(1) [1]. Se can substitute into either the Te(1) or Te(2) sites. The position of alloying impurities in the crystal lattice greatly influences thermoelectric properties as the local bonding environment can change at different crystallographic locations. Additionally, impurities will change the bond strengths in the structure, further affecting phonon transport. Determining impurity site preference and influence on bonding thus provides a critical piece of the puzzle needed to completely understand the relationship between alloy content and properties.

In this talk, we will discuss an atomic scale structural and chemical study of $Bi_2Te_{3-x}Se_x$ alloys using aberration corrected scanning transmission electron microscopy (STEM). Further, we employ RevSTEM, a new technique that corrects for drift and distortion in electron microscopy, that provides precise and accurate intensity measurements enabled by image averaging [2]. From HAADF, see Figure 1 and Figure 2(a), we will show that Se appears randomly distributed across the Te sites as the intensity of Te(1) and Te(2) columns are equivalent. By employing state-of-the-art atomic resolution energy dispersive x-ray spectroscopy (EDS), Se resides primarily in the Te(2) crystal sites as also shown in Figure 1. Using this information from EDS, we will show that a typical atomic number HAADF interpretation suggests that Te(2) atom columns should appear darker (Se=34, Te =52), but this is not observed.

Through a combination of images acquired at different ADF collection angles and the results from EDS, we will demonstrate that the observed intensities are due to phonon scattering resulting from Se incorporation. As shown in Figure 2, LAADF STEM imaging shows a significant drop in intensity at the Te(2) for the alloy, which is not observed in pure Bi_2Te_3. We will show that, through a comparison of experiment and simulations, the results can be explained by reduction of the average thermal displacements in the Te(2) layers. These results also help to explain the prior literature suggesting that the average bond strength of Te(2) layer changes upon incorporation of Se [1,3].

References:

[1] J. R. Wiese and L. Muldawer, Journal of Physics and Chemistry of Solids **15** (1960), p. 13.
[2] X. Sang and J. M. LeBeau, Ultramicroscopy **138** (2014), p. 28. [3] G. R. Miller *et al*, Journal of Applied Physics **34** (2004), p. 1398.

[4] The authors acknowledge the use of the Analytical Instrumentation Facility (AIF) at North Carolina State University, which is supported by the State of North Carolina and the National Science Foundation.

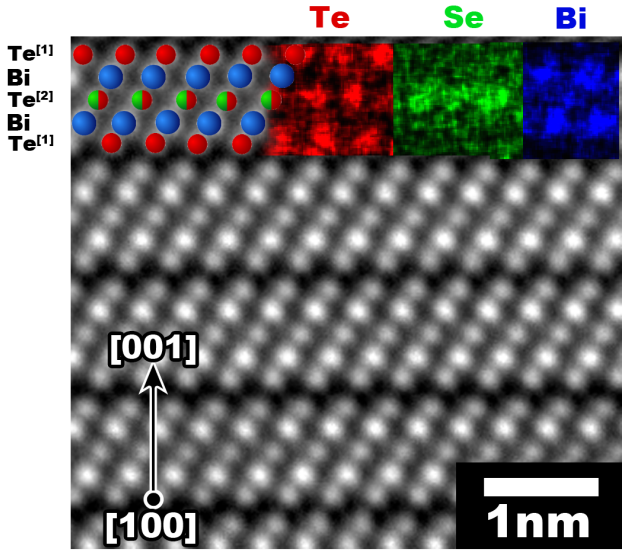

Figure 1. HAADF STEM image of $Bi_2Te_{2.7}Se_{0.3}$ viewed along [100]. The corresponding Te, Se, and Bi atomic resolution EDS maps are shown at the top. Note the Se signal resides in the Te(2) position.

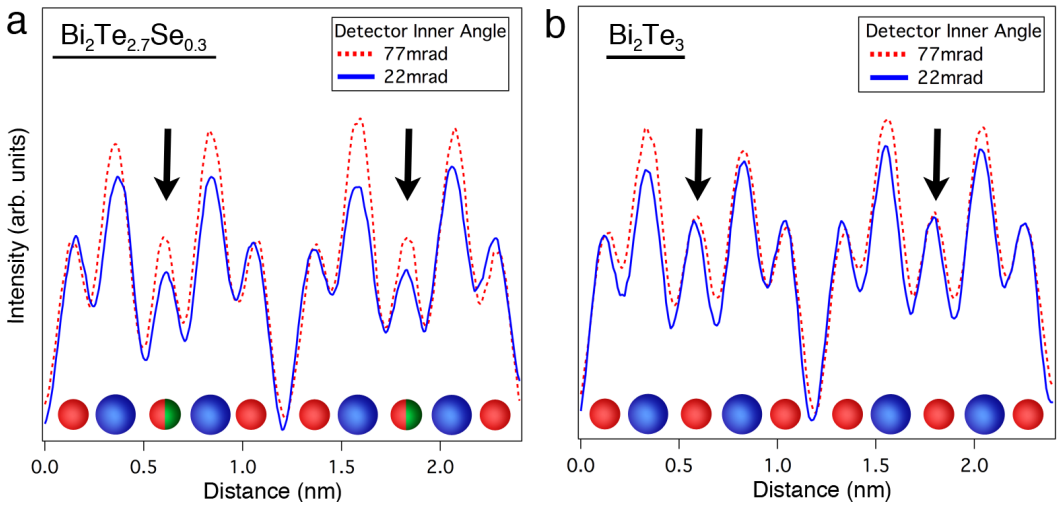

Figure 2. Line profiles from [100] $Bi_2Te_{2.7}Se_{0.3}$ (a) and Bi_2Te_3 (b) extracted from images at the indicated ADF collection angles. Atomic positions of Bi (blue), Te (red), and Se (green) placed below the corresponding intensity from ADF. Arrows indicate the atomic columns that change intensity at different collection angles for $Bi_2Te_{2.7}Se_{0.3}$ (a) but not Bi_2Te_3 (b). Note that the intensities have been normalized to the Te(1) columns to emphasize the changes at Te(2).

https://doi.org/10.1017/S1431927614003924 Published online by Cambridge University Press