Cross-Plane Seebeck Coefficient Measurement of Misfit Layered Compounds \((\text{SnSe})_n(\text{TiSe}_2)_n\) \((n = 1,3,4,5)\)

Zhen Li,† Sage R. Bauers,‖ Nirakar Poudel,† Danielle Hamann,‖ Xiaoming Wang,⊥ David S. Choi,⑦ Keivan Esfarjani,⊥ Li Shi,‡ David C. Johnson,‖ and Stephen B. Cronin⊥,‡,⑦

†Department of Electrical Engineering, ‡Department of Chemistry, and ⑦Department of Physics and Astronomy University of Southern California, Los Angeles, California 90089, United States
‖Department of Chemistry, University of Oregon, Eugene, Oregon 97403-1253, United States
⊥Department of Physics and Astronomy University of Toledo, Toledo, Ohio 43606-3390, United States,
‡Institute for Advanced Materials, Devices and Nanotechnology, Rutgers University, Piscataway, New Jersey 08854, United States
⑦Department of Mechanical Engineering, The University of Texas at Austin, Austin, Texas 78712-0292, United States
⊥Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904-4746, United States

Supporting Information

ABSTRACT: We report cross-plane thermoelectric measurements of misfit layered compounds \((\text{SnSe})_n(\text{TiSe}_2)_n\) \((n = 1,3,4,5)\), approximately 50 nm thick. Metal resistance thermometers are fabricated on the top and bottom of the \((\text{SnSe})_n(\text{TiSe}_2)_n\) material to measure the temperature difference and heat transport through the material directly. By varying the number of layers in a supercell, \(n\), we vary the interface density while maintaining a constant global stoichiometry. The Seebeck coefficient measured across the \((\text{SnSe})_n(\text{TiSe}_2)_n\) samples was found to depend strongly on the number of layers in the supercell \((n)\). When \(n\) decreases from 5 to 1, the cross-plane Seebeck coefficient decreases from \(-31\) to \(-2.5 \text{ \mu V/K}\), while the cross-plane effective thermal conductivity decreases by a factor of 2, due to increased interfacial phonon scattering. The cross-plane Seebeck coefficients of the \((\text{SnSe})_n(\text{TiSe}_2)_n\) are very different from the in-plane Seebeck coefficients, which are higher in magnitude and less sensitive to the number of layers in a supercell, \(n\). We believe this difference is due to the different carrier types in the n-SnSe and p-TiSe_2 layers and the effect of tunneling on the cross-plane transport.

KEYWORDS: Intergrowth compounds, Seebeck coefficient, cross-plane, 2D material stack

Over the past several decades, various methods and nanoscale materials have presented the promise of enhanced thermoelectric figures of merit. Hicks and Dresselhaus predicted an enhanced figure of merit by making nanostructures of Bi_2Te_3 such as quantum-wells. Slack and Nolas introduced the concept of a phonon glass, electron crystal to improve the performance of thermoelectric materials. These and other approaches have shown significant increases in ZT (2-3 fold) at high temperatures. However, the room-temperature ZT has not improved much during the past few decades. One approach to improving the efficiency of thermoelectric materials is to lower the thermal conductivity. In 2007, Chiritescu and others showed that WSe_2, “disordered layered crystals” (i.e., solids that combine order and disorder in the random stacking of two-dimensional crystalline sheets) have a thermal conductivity that is only a factor of 2 larger than air. The low thermal conductivity was attributed to the rotational disorder between the layers, which results in a very short coherence length between the Se-W-Se dichalcogenide layers. Follow-up studies on multicomponent “nanolamine” materials exhibited similarly ultralow thermal conductivities. In the low lattice thermal conductivity limit, the relative impact of the Seebeck coefficient on the figure of merit increases. It is known that the thin-film layered structure superlattices can help to improve the thermoelectric figure of merit. Further studies show that various misfit layered materials can be synthesized by kinetically trapping desired products via self-assembly from modulated elemental reactant (MER) precursors, such as (PbSe) (WSe_2), (SnSe) (MoSe_2), and (SnSe) (NbSe_2). The interfaces in these materials will affect the transport phenomenon, especially along the cross-plane direction and may provide a system with enhanced thermoelectric properties. However, the cross-plane Seebeck and electrical transport properties of these disordered materials have not been reported due to the difficulty in performing these measurements and lack of appropriate measurement techniques.

Received: December 29, 2016
Revised: February 8, 2017
Published: February 8, 2017
In this paper, we report the first cross-plane thermoelectric measurement of rotationally disordered misfit layered compounds \((\text{SnSe})_n(\text{TiSe}_2)_n\) \((n = 1,3,4,5)\). Instead of using "indirect" means, such as the Harman transient method\(^8,13,14\) or the time-domain thermoreflectance (TDTR) method\(^5,15,16\), a direct, steady state method is used to measure the cross-plane Seebeck coefficient and thermal conductivity of the material. Here, we fabricate metal resistance temperature detectors (RTD) on the top and bottom of the compounds in order to measure the temperature difference created by the heater\(^17,18\).

The extent of disorder of the compounds studied here, \((\text{SnSe})_n(\text{TiSe}_2)_n\) \((n = 1,3,4,5)\) differ from the previously studied \((\text{PbSe})_n(\text{WSe}_2)_m\) and \((\text{SnSe})_n(\text{MoSe}_2)_m\) compounds.\(^5,19\) Both WSe\(_2\) and MoSe\(_2\) have the transition metal in trigonal prismatic coordination, and the stable structure is a 2H polytype. When grown using the MER approach, a random stacking sequence results because there are two different possible orientations that are very close in energy, and this disorder contributes significantly to reducing thermal conductivity. TiSe\(_2\), however, has the Ti in octahedral coordination and the stable structure is a 1T polytype. When grown using the MER approach, an ordered 1T polytype forms.\(^20\) Hence the disorder at the SnSe–TiSe\(_2\) interfaces is expected to be a more important factor in reducing thermal conductivity than in WSe\(_2\) and MoSe\(_2\) containing intergrowths, which also have disorder between the dichalcogenide planes. The cross-plane Seebeck coefficient and thermal conductivity measured in these materials depend strongly on the number of layers in the supercell \((n)\). By varying \(n\), we vary the interface density while maintaining a constant stoichiometry.

In the work presented here, \((\text{SnSe})_n(\text{TiSe}_2)_n\) cross-plane thermoelectric devices are fabricated on a Si wafer with a 300 nm oxide. First, the bottom metal resistance temperature detector (RTD) is patterned by using electron beam lithography followed by metal deposition, as illustrated in Figure 1. After that, another layer of PMMA is spin-coated on the substrate, and an 18 μm × 18 μm window is opened on top of the bottom RTD. A 50 nm thick \((\text{SnSe})_n(\text{TiSe}_2)_n\) film is then deposited on the substrate, by physical vapor deposition (see Methods section).\(^21\) After the lift-off process, the designed precursor film is annealed into the layered structure in an inert N\(_2\) \(([\text{O}_2] < 1 \text{ ppm})\) environment by heating at 350 °C for 30 min. In addition to the actual devices, various control samples are prepared in the same round of deposition for transmission electron microscopy (TEM) and X-ray diffraction studies. The top metal RTD is patterned in the same way as the bottom metal RTD. The samples are then capped with a 50 nm insulating film of Al\(_2\)O\(_3\) deposited by atomic layer deposition.
(ALD) at 200 °C using trimethylaluminum (TMA) and water as precursors (Cambridge Nanotech ALD, Savannah 200). Lastly, a serpentine metal heater with 5 nm Ti and 35 nm Pd is patterned on top of the Al2O3 layer. The heater also contains four probes in order to measure the heating power precisely. The device fabrication process and an optical image of a device after fabrication are shown in Figure 1.

High-angle annular dark field (HAADF) scanning transmission electron microscopy of the film is shown in Figure 2a, for n = 1 and n = 3 samples. According to these TEM images, no regular stacking orientations are observed between the different layers, highlighting the interlayer rotational disorder in the films. However, for n = 3 blocks of like-constituents have regular registration and a similar structure is expected for all other films with n > 1. Also, a few layering defects are observed. It is not known whether defects are an artifact of self-assembly of the laminate structures or of beam damage produced during the preparation of the lamella used for imaging. According to the zoom-out scanning transmission electron microscopy (STEM) images (see Figure S1), the interface density of the materials may increase a little due to the layer defects. However, the overall linear relation between the layer number and interface density will be not changed. Figure 2b shows X-ray diffraction data taken from the (SnSe)n(TiSe2)m, n = 3 film. The first 14 (00l) peaks are indexed corresponding to a unit cell of 3.561 nm. The number is slightly larger than the sum by adding the thickness of the three layer SnSe and TiSe2 together. The cross-plane distance between bulk SnSe and TiSe2 is 0.57 and 0.60 nm, respectively, but the interfaces between constituents could reasonably be assumed to increase the thickness of the laminate relative to these values. The measured supercell thickness for the n = 3 structure is slightly less than three times the value of the previously published n = 1 laminate,17 because no additional SnSe-TiSe2 interfaces are formed with increasing n. Twelve Kiessig fringes are observed between Bragg reflections indicating 14 total repeats of the superstructure and a film thickness of 49.8 nm. The Kiessig fringes persist to high Q indicating (by the Parratt relationship)22 that the nanolaminate stack consists of parallel interfaces that are approaching atomically smooth. We have also performed Raman spectroscopy on the (SnSe)n(TiSe2)m sample (see Figure S2), which do not show any Raman peaks belonging to either bulk SnSe23 or TiSe224 indicating that the phonon modes in the misfit compound are different. Atomic force microscopy (AFM) images are taken of the films before the ALD deposition to confirm their thickness when incorporated into the RTD device (Figure S3).

In order to perform the thermoelectric measurement, the top and bottom metal RTDs are first calibrated in a temperature-controlled stage. Figure 3 shows the calibration data and temperature difference measured under electrical heating of the (SnSe)n(TiSe2)m device with n = 5. First, the resistance of the RTD is measured at various temperatures between 300 and 330 K. Several thermal cycles are performed in order to anneal out any strain in the samples due to thermal expansion, until the resistance is stable. We then normalize the resistance with respect to the room temperature resistance (i.e., R/Ro, Ro is resistance at 300 K) and plot this as a function of the temperature increase ΔT (T − T0, To = 300 K) (see Figure 3b). The relation between resistance and temperature is established by a linear fit of this data. The resistance of the RTDs are then measured as a function of heater current (see Figure 3c). From the ΔR−ΔT relation (Figure 3b), we obtain the temperature change as a function of heater power, which is fit to a linear function, as shown in Figure 3d. Similar calibration and temperature difference measurements are performed for the top RTD and for the other devices with n = 1,3,4. (see Supporting Information). The top and bottom RTDs are also used to measure the thermovoltage.

The thermoelectric voltage drop across the sample was measured with both positive and negative heating voltages. These two sets of measurements give almost the same result, indicating that the voltage difference is in fact a thermoelectric effect, rather than a potential drop induced by the heater voltage. Also, electrical measurements between the Pd heater and the top RTD show leakage currents less than 50 pA for applied bias voltages up to 5 V (>0Ω), which is shown in Figure S7 of the Supporting Information. Figure 4 shows the thermoelectric voltage plotted as a function of the temperature difference across the (SnSe)n(TiSe2)m film for the four devices with n = 1,3,4,5. All data sets were fit to linear functions with the slope corresponding to the Seebeck coefficient, as indicated in these figures. Here, the cross-plane Seebeck coefficient increases from −2.5 to −31 μV/K as n varies from 1 to 5.

In our sample configuration, the distance between the silicon heat sink and the metal heater is only 400 nm and the heating area is 21 μm × 20 μm. For lateral heat transport, the cross-sectional area of the metal electrodes is 40 nm × 10 μm, which is only about 0.1% of the cross-plane transport area. Here, we can compare the vertical and lateral thermal resistivities to prove the thermal transport is dominated by the vertical component. The vertical transport traverses 50 nm Al2O3, 300 nm SiO2, and 50 nm (SnSe)n(TiSe2)m films. The total vertical thermal resistance can be calculated using the thermal conductivities of these films and their corresponding thicknesses: κALD ≈ 1.6 W/K·m,25,26 tALD = 50 nm, κSiO2 ≈ 1.25 W/K·m,27,28 tSiO2 = 300 nm, and the thermal resistance of the (SnSe)n(TiSe2)m film is about 500 K/W (as calculated below), we have...
Figure 3. Temperature calibration of the bottom (a–d) and top (e–h) metal RTDs of (SnSe)$_n$(TiSe$_2$)$_n$, $n = 5$ device. (a,e) The resistance is the RTD measured at different temperatures. The resistance stabilizes after a few thermal cycles. (b,f) Normalized resistance ($R/R_o$, $R_o$ is the resistance at 300 K) plotted as a function of $\Delta T$ ($T - T_o$, $T_o = 300$ K). By linear fitting of this data, the relation between resistance and temperature is acquired. (c,g) The resistance change of the RTDs under various heating currents. (d,h) Temperature change of the metal RTDs plotted as a function of heating power.
On the other hand, considering the lateral thermal resistance of the Ti/Pd heater, the electrical conductivity, $\sigma_{\text{metal}}$, measured from the heater is $1.2 \times 10^6$ S/m. On the basis of Wiedmann-Franz law

$$\frac{R_{\text{vertical}}}{\kappa} = L T, \quad L = 2.44 \times 10^{-8} \text{ W} \cdot \Omega \cdot \text{K}^{-2}$$

the thermal conductivity of the heater can be calculated, $\kappa_{\text{metal}} = 8.8$ W/K-m. Given the width of the metal connection line is 1 $\mu$m, the transport length is the same as the vertical transport length, which is around 100 nm, then the lateral thermal resistance of the metal is

$$R_{\text{electrode}} = \frac{L_{\text{electrode}}}{A_{\text{electrode}} \rho_{\text{metal}}} = \frac{L_{\text{electrode}}}{W_{\text{electrode}} t_{\text{metal}} \kappa_{\text{metal}}}$$

$$= \frac{100 \text{ nm}}{1 \mu \text{m}} \approx 3.3 \times 10^5 \text{ K/W}$$

which is much larger than the vertical thermal resistance. Thus, while the metal electrodes have a large thermal conductivity, the thermal transport across the film can be considered as pseudo-1D transport. Further COMSOL simulation based on our device configuration can be found in the Supporting Information. The value of the film’s thermal conductivity provided by the COMSOL simulation is 12% smaller than the

Figure 4. Seebeck coefficient measured from (SnSe)$_n$(TiSe$_2$)$_n$ compounds, $n = 1, 3, 4, 5$. Here, both positive and negative voltages are applied to the heater to confirm that the voltage difference measured from the top and bottom RTD is due to the heating and not purely electrical effects.

Figure 5. Cross-plane (a) Seebeck coefficient and (b) thermal conductivity with different layer configuration $n$. The Seebeck coefficient is increased from $-2.5$ to $-31$ $\mu$V/K as $n$ varies from 1 to 5, and the thermal conductivity is increased from 0.17 to 0.34 W/K-m.
calculation result from the 1D mode. The difference is within the experimental error range, and shows that the pseudo 1D transport is a good approximation of our device.

Given the temperature difference $\Delta T$ and heating power $Q$ in Figure 3d, we can calculate the thermal resistance of the film using the relation $R = \frac{\Delta T}{Q} \approx 500 \text{ K/W}$, where $\kappa$ is the thermal conductivity, $t$ is the thickness, and $A$ is the area of film. The effective thermal conductivity can be obtained by $\kappa = \frac{Q}{\Delta T}$ where $A$ is $420 \mu m^2$ and $t$ varies from 47 to 60 nm for the different samples according to the AFM measurement (Figure S3). The calculated thermal conductivity also changes with the layer configuration, $n$. The 11th sample has the lowest thermal conductivity of 0.17 W/K·m, while the thermal conductivity of the 55th sample is 0.34 W/K·m. Figure 5 shows the Seebeck coefficient and thermal conductivity plotted as a function of layer configuration, $n$. The 11th configuration, $n$, is labeled by $n=1$. In the first case labeled by $n=1$, previous calculations on phosphorene layers have shown that tunneling can be important and would cause a lowering of the Seebeck coefficient compared to the thermionic limit where no tunneling takes place and more heat is carried over the barrier. Tunneling can be suppressed for thicker layers such as $n > 3$ case, and therefore one expects the $n=3$ structure to have a larger Seebeck than the $n=1$ (left figure) case, although tunneling is still present there but to a lesser degree. Overall, the LDOS in TiSe$_2$ is much larger than in SnSe around the Fermi level.

Here, we believe that the different carrier types in the p-SnSe and n-TiSe$_2$ layers nearly cancel each other’s contribution to the Seebeck coefficient for the samples with small $n$. In Figure 6, we display the local density of states (LDOS) of two heterostructures: On the left, (SnSe/TiSe$_2$)$_3$ is sandwiched between two gold substrates formed of four (left electrode) and five (right electrode) layers each. On the right, (SnSe)$_3$/ (TiSe$_2$)$_3$ is placed between the same gold structures. The plots show the LDOS contours in the (layer number, energy) plane. Whereas white indicates absence of states (i.e., bandgap), increasing LDOS values can be seen to vary from blue to red. The horizontal red dashed line shows the position of the Fermi level at zero bias across the whole heterostructure. We can see the general trend that while TiSe$_2$ possesses a small pseudogap right below the Fermi energy, the SnSe layers have their bandgap right above the Fermi energy except on the first layer attached to gold which seems to cause an Ohmic contact. Therefore, there is n-type and p-type doping in the TiSe$_2$ and SnSe layers, respectively. Similar results of p-n-p-n structure due to charge exchange and “self-doping” are also observed from misfit layered materials in other studies. In addition, in the first case labeled by $n=1$ (left), previous calculations on phosphorene layers have shown that tunneling can be important and would cause a lowering of the Seebeck coefficient compared to the thermionic limit where no tunneling takes place and more heat is carried over the barrier. Tunneling can be suppressed for thicker layers such as $n > 3$ case, and therefore one expects the $n=3$ (right figure) structure to have a larger Seebeck than the $n=1$ (left figure) case, although tunneling is still present there but to a lesser degree. Overall, the LDOS in TiSe$_2$ is much larger than in SnSe around the Fermi level.
the Fermi level, and therefore most of the thermoelectric current is carried by electrons above the Fermi level rather than holes or states below it. As a result, the overall Seebeck of the device is negative. Because of the tunneling effect, the Seebeck coefficients measured from the film increase when the layer configuration is changed from 11 to 55.

In summary, we have demonstrated a novel device configuration for measuring the cross-plane thermal and thermoelectric transport through a thin film of layered materials, such as \((\text{SnSe})_n(\text{TiSe}_2)_m\) \((n = 1,3,4,5)\). In contrast to transient methods, such as the Harman method or the TDTR method, our method directly measures the steady-state temperature difference between the top and bottom surface of the thin film. Here, due to increased interfacial phonon scattering, the cross-plane thermal resistivity increases by a factor of 2, when \(n\) is changed from 5 to 1. In addition, the cross-plane Seebeck coefficient increases from \(-2.5\) to \(-31 \mu V/K\) as \(n\) varies from 1 to 5, which is considerably smaller than the in-plane Seebeck coefficients \((-50\) to \(-90 \mu V/K, see Figure S9 of the Supporting Information\). We believe that the reduced value of the cross-plane Seebeck coefficient is the result of tunneling transport, which has been predicted to have substantially lower values than diffusive (in-plane) transport. The massive proliferation of graphene and other 2D materials, such as TMDCs, provides new opportunities to engineer these layered materials to achieve a better thermoelectric performance. The device structure used in this experiment can serve as a general approach to characterize the cross-plane transport properties of these layered materials.

## METHODS

**Preparation of \((\text{SnSe})_n(\text{TiSe}_2)_m\) Films.** The nanolaminate films for this study were formed by the self-assembly of designed precursors. Precursors were prepared by physical vapor deposition of elemental sources in a purpose-built high vacuum chamber (pressure \(< 5 \times 10^{-7}\) Torr). Elemental Sn and Ti were evaporated with 6 kV electron guns and Se with a Knudsen effusion cell. Layers of each element were sequentially deposited to form the precisely layered precursors. The distance between sources and substrates were on the order of 1 m and evaporation rates were monitored and maintained below 1 Å s\(^{-1}\) at a position approximately halfway between the sources and substrate. Films were deposited both onto unpatterned substrates for structural characterization as well as substrates patterned for cross-plane transport measurements. The deposition parameters were calibrated beginning from the previously reported \(((\text{SnSe})_{1.2})(\text{TiSe}_2)_4\) compound and by increasing the integer number of layers of each constituent. After the deposition, the films were crystallized into the layered structures in an inert \(\text{N}_2\) \(([\text{O}_2] < 1 \text{ ppm})\) environment by gently heating to 350 °C for 30 min.

**TEM and X-ray Diffraction Characterization of \((\text{SnSe})_n(\text{TiSe}_2)_m\) Films.** X-ray diffraction data were collected at the Advanced Photon Source (APS) on beamline 33BM-C with an X-ray energy of 16 keV in an Ar sample environment. Data were collected under secular conditions in a locked-coupled out-of-plane geometry. Because of the texture of the nanolamine thin films, peaks in this geometry uniquely correspond to the supercell and can thus be indexed as 00l reflections. Two scan conditions were used with the region at low angle having a smaller step-size to capture the Kiessig oscillations and the region at high angle using a larger step size to better facilitate data collection throughput. HAADF-STEM data were collected on an aberration corrected FEI Titan 80-300 TEM/STEM at 300 kV with a 120 mm camera length. Lamella for STEM characterization were prepared with a dual-beam FIB and thinned to approximately 10 nm.

**Raman Spectroscopy.** Raman and photoluminescence spectra are taken using a Renishaw InVia spectrometer with a 532 nm laser \((10 \mu W)\) focused through a 100× objective lens. Raman spectra were collected at room temperature under ambient conditions.

**Temperature Calibration.** To calibrate the top and bottom RTDs, the sample is loaded into a cryostat (XE-102 OPT, Cryo Industries). The temperature is varied from 300 to 330 K while a lock-in amplifier (Stanford Research Systems, SR830) is used to measure the resistance change of the metal RTDs. The lock-in amplifier is used to provide a constant alternating current \((ac)\) source by putting a large \((1 \text{ MΩ})\) resistance in series with the metal RTD \((R < 10 \Omega)\). The voltage difference of the RTD is measured using the lock-in amplifier. The frequency of the lock-in is set to 1 kHz. To prevent electric heating during this calibration, a very small current \((1 \mu A)\) is used for the four probe measurement. In order to measure the resistance change of the RTD under different heating powers, a Keithley 2401 SourceMeter is used as a power source for the heating. A four-point measurement is performed under various direct current \((dc)\) heating currents while the voltage drop across the serpentine heater is measured to determine the total heating power.

**Seebeck Coefficient Measurement.** The thermal voltage is measured using a HP 34401A multimeter with a SR560 low-noise voltage amplifier in order to improve the signal-to-noise ratio. Both positive and negative heating voltages are applied to the heater in order to prove that the voltage measured is thermoelectric in nature. All the electric measurements are performed under vacuum.

**First-Principles Calculation.** First-principles electronic structure calculations are done using density functional theory (DFT) as implemented in the SIESTA code,\(^{31}\) which is an efficient real-space DFT package with numerical orbital basis set. We utilize norm-conserving pseudopotentials generated by the Troullier-Martins scheme.\(^{32}\) Exchange-correlation effects are treated by the generalized gradient approximation within the Perdew–Burke–Ernzerhof (PBE) formulation.\(^{33}\) Double zeta plus polarization (DZP) basis set are constructed for all the atoms. A cutoff of 300 Ry for the real-space grid integration is adopted. Brillouin zone integrations are carried out by using Monkhorst–Pack\(^{34}\) mesh of \(4 \times 4 \times 1\) \(k\)-points for the self-consistent calculation while the \(k\) mesh was increased to \(20 \times 20 \times 1\) for the DOS calculations. The heterostructure is constructed based on the experimental lattice parameters, 0.58 nm for SnSe, 0.60 nm for TiSe\(_2\), 0.59 nm for SnSe–TiSe\(_2\) interface based on the STEM data. A parameter of 0.277 nm is used for Au–SnSe and Au–TiSe\(_2\) interfaces.\(^{35}\) For the in-plane configuration, the lattice parameters of gold are fixed while those of SnSe and TiSe\(_2\) are strained to adapt to gold.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b05402.

Raman spectra and AFM measurements of the \((\text{SnSe})_n(\text{TiSe}_2)_m\) \((n = 1,3,4,5)\) flakes, temperature
calibration of metal RTDs of all the devices, COMSOL simulation of the heating transport, and the in-plane Seebeck coefficient measured from the same material (PDF)

**AUTHOR INFORMATION**

**Corresponding Author**
*(S.B.C.)* Phone: (213) 740-8787. E-mail: scronin@usc.edu.

**ORCID**
Zhen Li: 0000-0003-0886-2994
Danielle Hamann: 0000-0002-9262-1060

**Notes**
The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This research was supported by the Department of Energy (DOE) Award Nos. DE-FG02-07ER46376 (Z.L.), DE-FG02-07ER46377 (L.S.), and NSF Award No. 1402906. (N.P. and K.E.). Authors S.R.B., D.H., and D.C.J. acknowledge support from the National Science Foundation under Grant DMR-1266217. Use of the Advanced Photon Source, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science by Argonne National Laboratory, was supported by the U.S. DOE under Contract No. DE-AC02-06CH11357. The authors thank Jenia Karapeeva for assistance in collection of the synchrotron X-ray diffraction data at beamline 33-BM-C. We also acknowledge support through the Collaborative Access Team (CAT): Pooled Resources for Electron Microscopy Informatics, Education and Research (PREMIER) Network Program and the Chemical Imaging Initiative at Pacific Northwest National Laboratory (PNNL) and the Environmental Molecular Sciences Laboratory, a national scientific user facility sponsored by DOE’s Office of Biological and Environmental Research at PNNL. PNNL is a multiprogram national laboratory operated by Battelle for DOE under Contract No. DE-AC05-76RL01830.

**REFERENCES**


