Thermoelectric Properties of CoAsSb: An Experimental and Theoretical Study

Xiaoyan Tan,1 Kasey P. Devlin,2 Xiaoyu Deng,3 Chang-Jong Kang,3 Mark Croft,3 Corey E. Frank,1 Chongin Pak,4 Saul Lapidus,5 Susan M. Kauzlarich,2 Valentin Taufour,6 Gabriel Kotliar*,3, Martha Greenblatt*,1

1 Department of Chemistry and Chemical Biology, Rutgers, The State University of New Jersey, Piscataway, New Jersey, 08854, United States
2 Department of Chemistry, University of California, Davis, California, 95616, United States
3 Department of Physics and Astronomy, Rutgers, The State University of New Jersey, Piscataway, New Jersey, 08854, United States
4 Department of Chemistry and Biochemistry, Florida State University, Tallahassee, Florida, 32306, United States
5 Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois, 60439, United States
6 Department of Physics, University of California, Davis, California, 95616, United States

Corresponding authors’ e-mail: greenbla@chem.rutgers.edu and kotliar@physics.rutgers.edu

Figure S1. Powder X-ray pattern compared with calculated arsenopyrite and marcasite structure types...
Figure S2. Scanning electron microscope image of CoAsSb particles...
Figure S3. X-ray diffraction pattern of samples before and after TGA measurement.
Figure S4. The fitting of magnetization as a function of field of CoAsSb measured at 1.8 and 50 K.
Figure S5. The inverse magnetic susceptibility as a function of temperature of CoAsSb.
Figure S6. Electronic and lattice thermal conductivities of CoAsSb as a function of temperature.
Figure S7. Seebeck coefficient (a) and thermal conductivity (b) of CoAsSb as a function of temperature below room temperature.
Figure S8. Electronic band structure and density of states plots for CoAsSb with DFT relaxation calculation, mBJ exchange potential method.
Figure S9. The calculated electronic thermal conductivity of CoAsSb as a function of temperature with the BoltzTrap package.
Figure S10. The Fermi level of density of states of CoAsSb0.883 with LDA rigid band approximation method based on CoAsSb.
Figure S11. Comparison of transport properties of CoAsSb, and CoAsSb0.883 calculated with LDA rigid band approximation.
Figure S12. Transport properties as a function of chemical potential near the Fermi level of CoAsSb at 300 K.
Figure S1. Powder X-ray pattern compared with calculated arsenopyrite and marcasite structure types.

Table S1. Summary of Rietveld refinement of CoAsSb synchrotron powder X-ray pattern.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Unit cell parameters</th>
<th>x, y, z</th>
<th>s.o.f</th>
<th>Rf</th>
<th>R_{bagg}</th>
<th>Chi²</th>
</tr>
</thead>
<tbody>
<tr>
<td>CoAsSb</td>
<td>a = 6.23704(6) Å</td>
<td>Co: 0.2837(2), 0.0061(3), 0.2827(2)</td>
<td>Co: 1</td>
<td>2.77</td>
<td>4.55</td>
<td>2.07</td>
</tr>
<tr>
<td>P2_1/c</td>
<td>b = 6.13276(4) Å</td>
<td>As: 0.3473(2), 0.3610(2), 0.1657(2)</td>
<td>As: 1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>c = 6.21963(6) Å</td>
<td>Sb: 0.1519(1), 0.6357(1), 0.3604(2)</td>
<td>Sb: 0.883</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>β = 116.6396°(4)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CoAs</td>
<td>a = b = 3.57813(4) Å</td>
<td>Co: 0, 0, 0</td>
<td>Co: 1</td>
<td>3.90</td>
<td>7.11</td>
<td></td>
</tr>
<tr>
<td>P6_3/mmc</td>
<td>c = 5.17478(8) Å</td>
<td>As: 0.3333, 0.6667, 0.25</td>
<td>As: 1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure S2. Scanning electron microscope image of CoAsSb particles.
Figure S3. X-ray diffraction pattern of samples before and after TGA measurement.

Figure S4. Magnetization as a function of field of CoAsSb measured at 1.8 (solid red squares with heavy solid line) and 50 K (open blue circles). Subtracting the estimated linear portion of the 1.8 K magnetization yields an estimate for the impurity related component, $m_{nl}$ (solid red squares with light solid line). For reference the field dependence of scaled Brillouin functions at 1.8 K for $S=1/2$, 1 and 3/2 are shown in the figure. Here the impurity content, per formula unit, would be approximately 0.1 % and 0.3% for the $S=1$ and 3/2 cases, respectively.
Figure S5. The inverse magnetic susceptibility as a function of temperature of CoAsSb.

Figure S6. Electronic and lattice thermal conductivities of CoAsSb as a function of temperature.
Figure S7. Experimental Seebeck coefficient (a) and thermal conductivity (b) of CoAsSb as a function of temperature, below room temperature.

Figure S8. Electronic band structure and density of states plots for CoAsSb obtained by mBJ exchange potential method with DFT relaxed structure.

Figure S9. The calculated electronic thermal conductivity of CoAsSb as a function of temperature with the BoltzTrap package.
Figure S10. The Fermi level of density of states of CoAsSb$_{0.883}$ with LDA rigid band approximation method based on CoAsSb.

Figure S11. Comparison of transport properties of CoAsSb, and CoAsSb$_{0.883}$ with LDA rigid band approximation.

Figure S12. Transport properties as a function of chemical potential near the Fermi level of CoAsSb at 300 K.