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The effect of extended strain fields on point defect scattering

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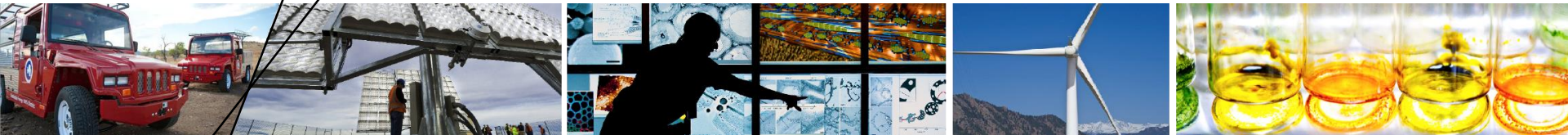
Brenden R. Ortiz, Haowei Peng, Philip Parilla, Stephan Lany, Armando Lopez, and Eric S. Toberer, "The effect of extended strain fields on point defect scattering" in "Nonstoichiometric Compounds VI", ECI Symposium Series, (2016). http://dc.engconfintl.org/nonstoichiometric_vi/37

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Authors

Brenden R. Ortiz, Haowei Peng, Philip Parilla, Stephan Lany, Armando Lopez, and Eric S. Toberer

Extended Strain Fields and Point-Defect Phonon Scattering



Brenden Ortiz, Haowei Peng, Armando Lopez, Phillip Parilla, Stephan Lany, Eric Toberer

Colorado School of Mines, Golden, CO

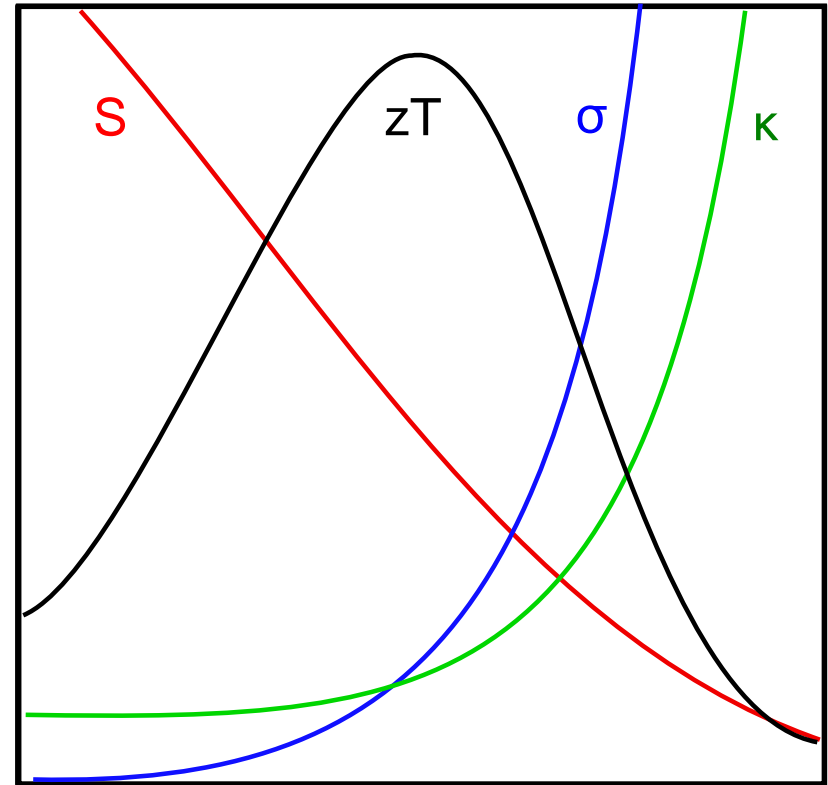
National Renewable Energy Laboratory, Golden, CO

ECI Non-Stoichiometric Compounds (Santa Fe 2016)

Thermoelectrics as an Optimization Problem

$$zT(\kappa, S, \sigma, T) = \frac{S^2 \sigma}{\kappa} T$$

$$zT = zT(\eta, m^*, \tau, E_g, \dots)$$



Carrier Concentration

+

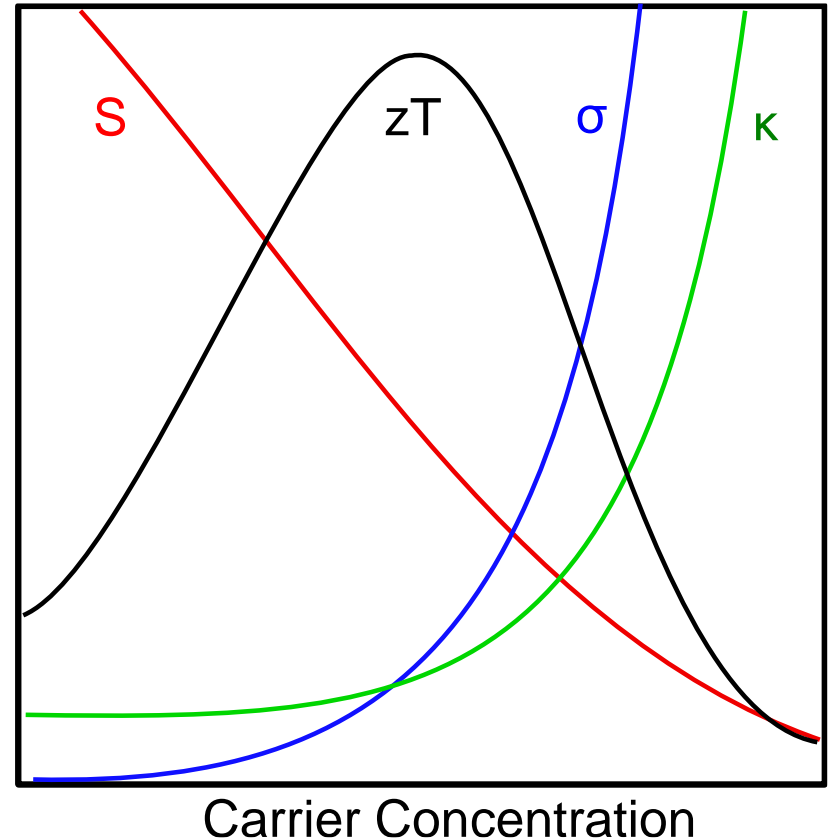
Each transport phenomenon **also** has T dependence

Thermoelectrics as an Optimization Problem

$$zT(\kappa, S, \sigma, T) = \frac{S^2 \sigma}{\kappa} T$$

$$zT = zT(\eta, m^*, \tau, E_g, \dots)$$

The thermal conductivity, κ , is commonly targeted for optimization.

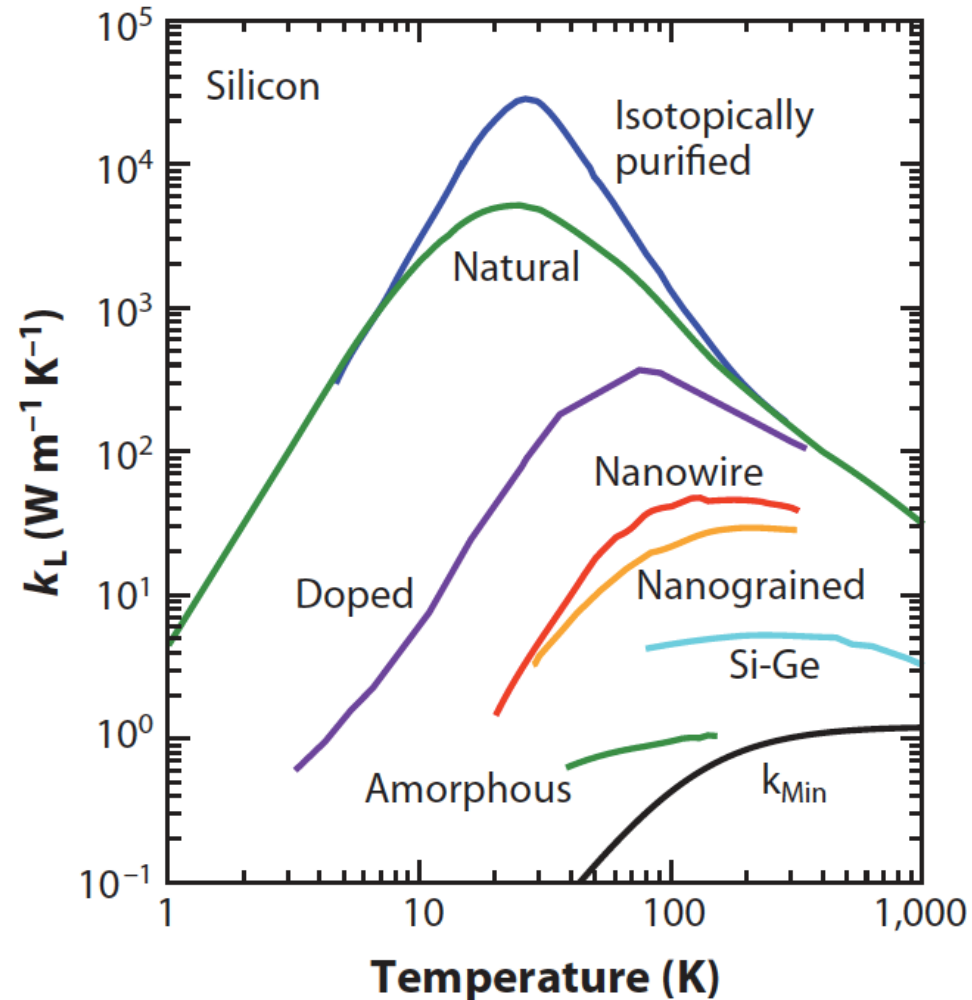


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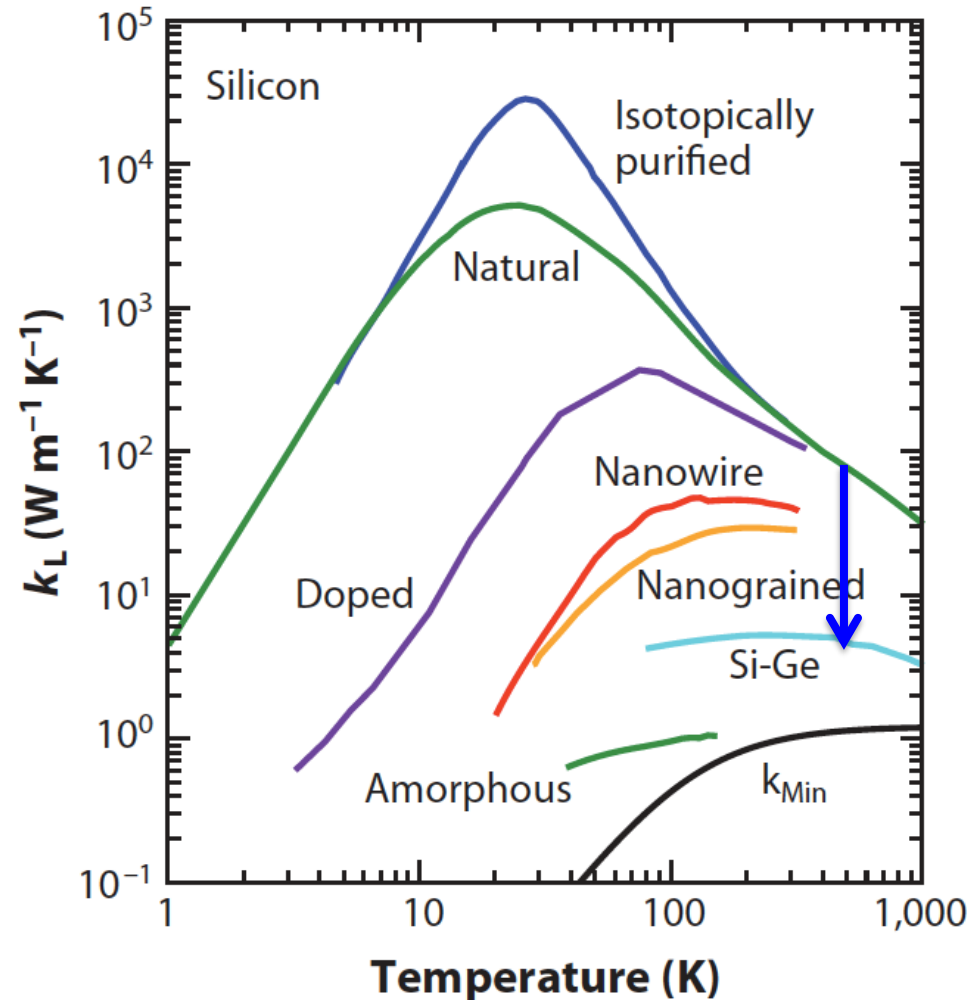
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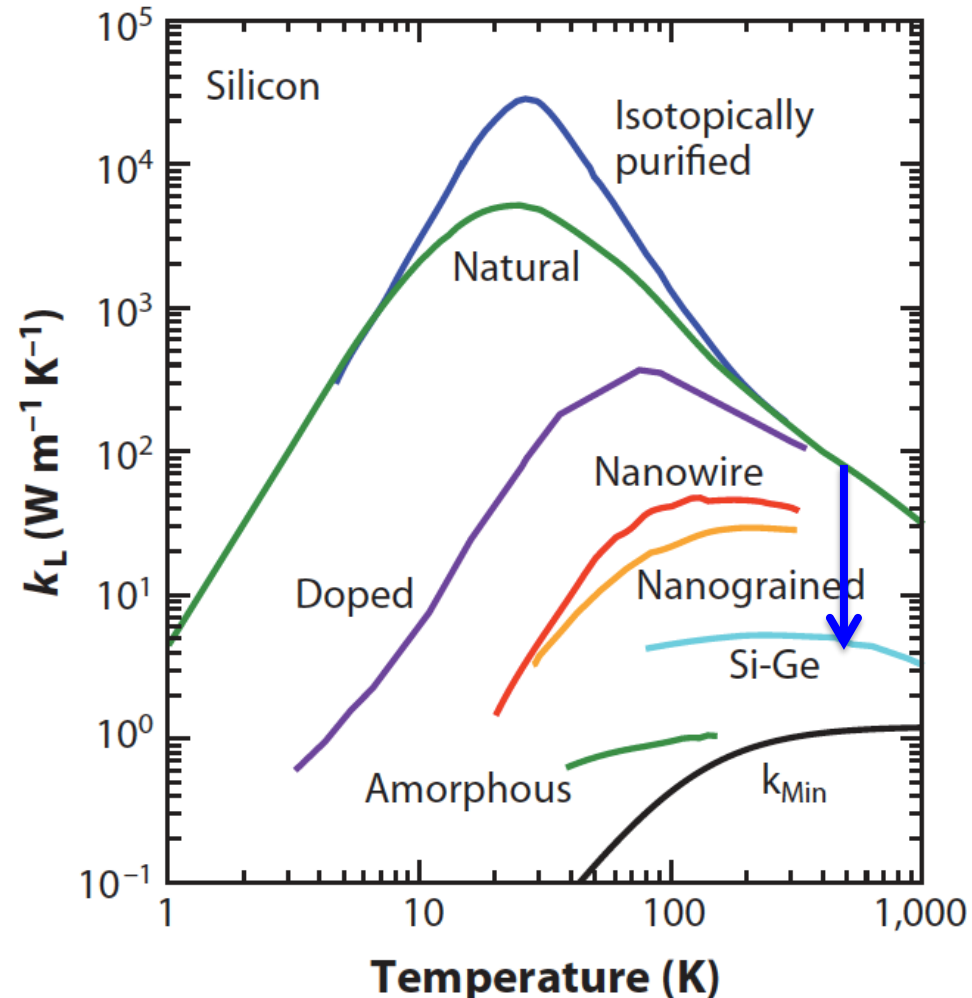
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$$zT(\kappa, S, \sigma, T) = \frac{S^2 \sigma}{\kappa} T$$

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Alloys as a source of point defect scattering

Mass contrast simple, but role of **strain field** not obvious in **complex crystals**



Thermoelectrics as an Optimization Problem

1

18

IA																VIII A					
1 1,008* H Wasserstoff	2 II A												13 III A	14 IV A	15 VA	16 VIA	17 VII A	2 4,003 He Helium			
3 6,94* Li Lithium	4 9,012 Be Beryllium											5 10,81* B Bor	6 12,01* C Kohlenstoff	7 14,01* N Stickstoff	8 16,00* O Sauerstoff	9 19,00 F Fluor	10 20,18 Ne Neon				
11 22,99 Na Natrium	12 24,31* Mg Magnesium	3 III B	4 IV B	5 V B	6 VI B	7 VII B	8 VIII B	9 VIII B	10 VIII B	11 IB	12 IIB	13 26,98 Al Aluminium	14 28,09* Si Silicium	15 30,97 P Phosphor	16 32,06* S Schwefel	17 35,45* Cl Chlor	18 39,95 Ar Argon				
19 39,10 K Kalium	20 40,08 Ca Calcium	21 44,96 Sc Scandium	22 47,87 Ti Titan	23 50,94 V Vanadium	24 52,00 Cr Chrom	25 54,94 Mn Mangan	26 55,85 Fe Eisen	27 58,93 Co Cobalt	28 58,69 Ni Nickel	29 63,55 Cu Kupfer	30 65,38* Zn Zink	31 69,72 Ga Gallium	32 72,63 Ge Germanium	33 74,92 As Arsen	34 78,96 Se Selen	35 79,90* Br Brom	36 83,80 Kr Krypton				
37 85,47 Rb Rubidium	38 87,62 Sr Strontium	39 88,91 Y Yttrium	40 91,22 Zr Zirkonium	41 92,91 Nb Niob	42 95,96* Mo Molybdän	43 [98] Tc Technetium	44 101,1 Ru Ruthenium	45 102,9 Rh Rhodium	46 106,4 Pd Palladium	47 107,9 Ag Silber	48 112,4 Cd Cadmium	49 114,8 In Indium	50 118,7 Sn Zinn	51 121,8 Sb Antimon	52 127,6 Te Tellur	53 126,9 I Iod	54 131,3 Xe Xenon				
55 132,9 Cs Caesium	56 137,3 Ba Barium	57-71 La Lanthan	72 178,5 Hf Hafnium	73 180,9 Ta Tantal	74 183,8 W Wolfram	75 186,2 Re Rhenium	76 190,2 Os Osmium	77 192,2 Ir Iridium	78 195,1 Pt Platin	79 197,0 Au Gold	80 200,6 Hg Quecksilber	81 204,4* Tl Thallium	82 207,2 Pb Blei	83 209,0 Bi Bismut	84 [209] Po Polonium	85 [210] At Astat	86 [222] Rn Radon				
87 [223] Fr Francium	88 [226] Ra Radium	89-103 Ac Actinium	104 [267] Rf Rutherfordium	105 [268] Db Dubnium	106 [269] Sg Seaborgium	107 [270] Bh Bohrium	108 [269] Hs Hassium	109 [278] Mt Meitnerium	110 [281] Ds Darmstadtium	111 [281] Rg Roentgenium	112 [285] Cn Copernicium	113 [286] Uut Ununtrium	114 [289] Fl Flerovium	115 [288] Uup Ununpentium	116 [293] Lv Livermorium	117 [294] Uus Ununseptium	118 [294] Uuo Ununoctium				

Chemical degrees of freedom can slow down experimental searches

*H: [1,00784, 1,00811]
 Li: [6,938, 6,997]
 B: [10,806, 10,821]
 C: [12,0096, 12,0116]
 N: [14,00643, 14,00728]
 O: [15,99903, 15,99977]
 Mg: [24,304, 24,307]
 Si: [26,084, 26,086]
 S: [32,059, 32,076]
 Cl: [35,446, 35,457]
 Br: [79,901, 79,907]
 Tl: [204,382, 204,385]
 Zn: 65,38(2)
 Se: 78,96(3)
 Mo: 95,96(2)

57 138,9 La Lanthan	58 140,1 Ce Cer	59 140,9 Pr Praseodym	60 144,2 Nd Neodym	61 [145] Pm Promethium	62 150,4 Sm Samarium	63 152,0 Eu Europium	64 157,3 Gd Gadolinium	65 158,9 Tb Terbium	66 162,5 Dy Dysprosium	67 164,9 Ho Holmium	68 167,3 Er Erbium	69 168,9 Tm Thulium	70 173,1 Yb Ytterbium	71 175,0 Lu Lutetium
89 [227] Ac Actinium	90 232,0 Th Thorium	91 231,0 Pa Protactinium	92 238,0 U Uran	93 [237] Np Neptunium	94 [244] Pu Plutonium	95 [243] Am Americium	96 [247] Cm Curium	97 [247] Bk Berkelium	98 [251] Cf Californium	99 [252] Es Einsteinium	100 [257] Fm Fermium	101 [258] Md Mendelevium	102 [259] No Nobelium	103 [262] Lr Lawrencium

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Mission Statement

Can we find a way to accelerate the discovery of effective alloys using simple, intuitive computational models?

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Experiment / Modeling

Ab Initio Computation

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Model System

Classical Alloy Models

Experimental Results

Ab Initio Computation

Toy Models for Strain

Computational Methods

Computational Results

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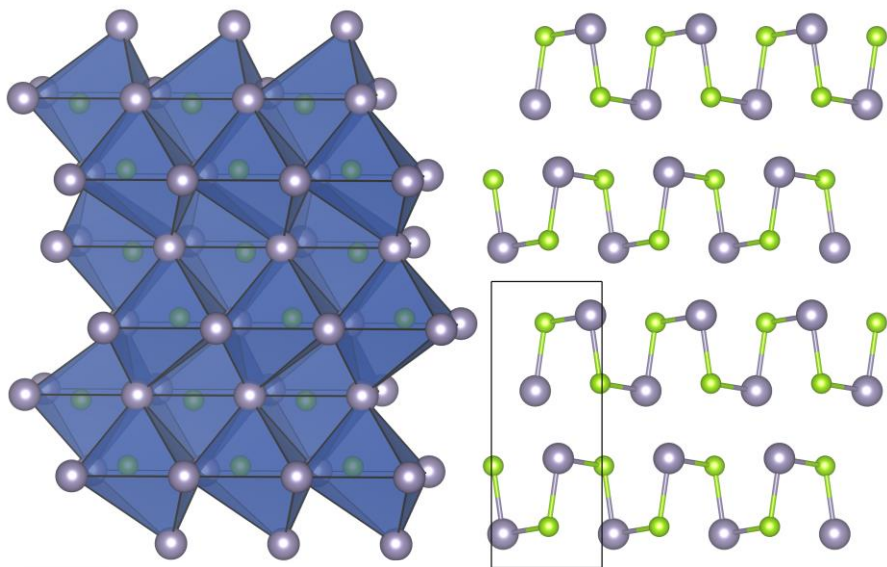
Computational Methods

Computational Results

Effective Proxy?

Experiment – Model System

SnSe (Pnma) + one of Sr, Ba, S, Se, Te
(e.g. $\text{Sn}_{1-x}\text{Ba}_x\text{Se}$)

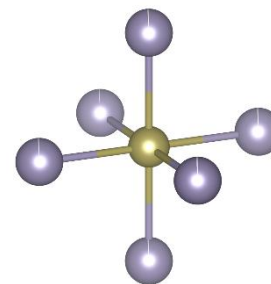


Distorted-rock salt structure

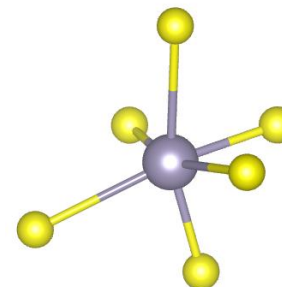
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Rock-salt and non rock-salt endpoints

BaSe, SrSe, SnTe: Rock-salt

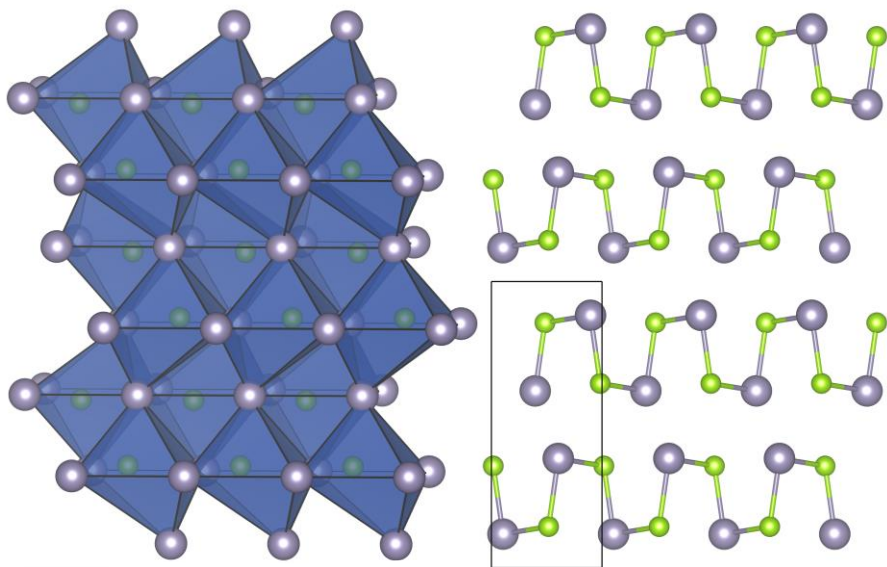


SnS, GeSe: Layered Pnma (distorted-RS)



Experiment – Model System

SnSe (Pnma) + one of Sr, Ba, S, Se, Te
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Synthesis (e.g. $\text{SnSe} + \text{SnSe}_2 + \text{Ba}$)

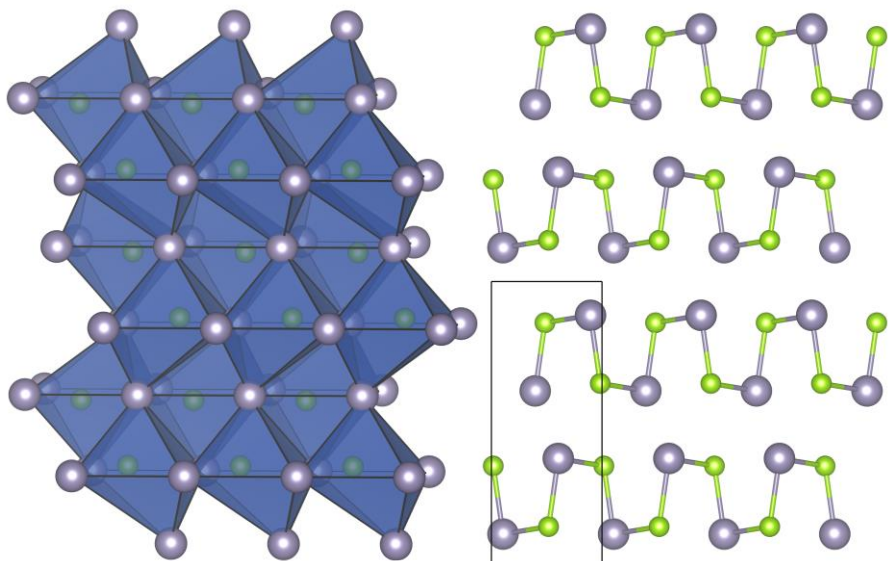
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Ball-milling and inductive hot-pressing

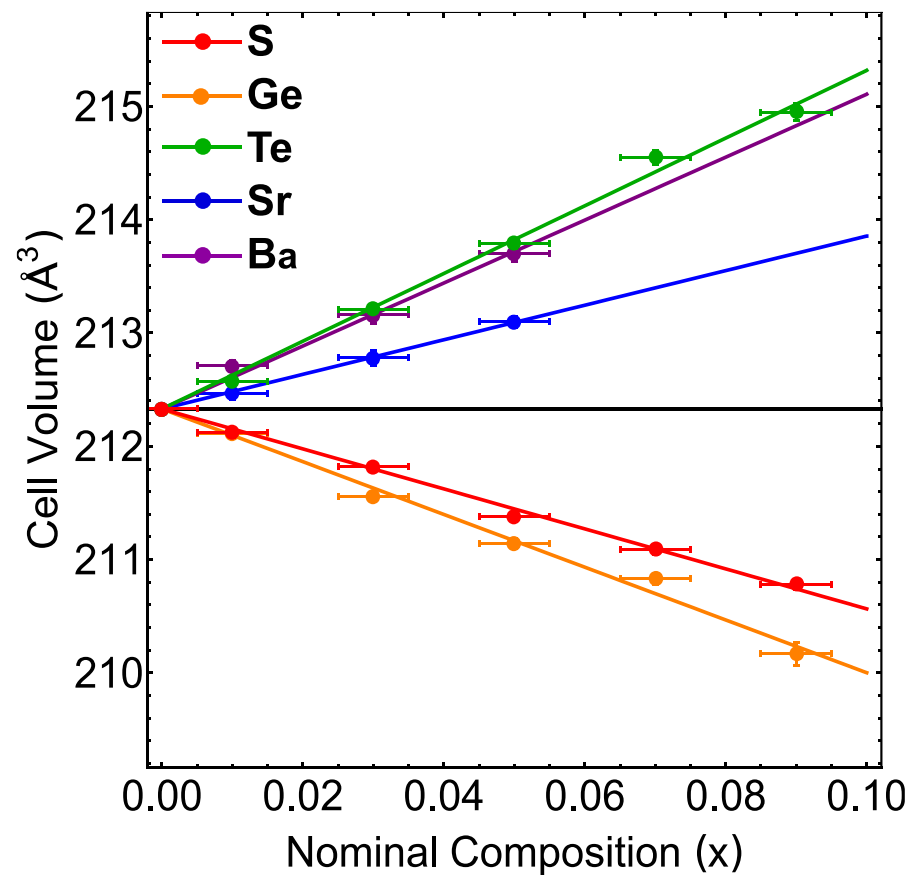


Experiment – Model System

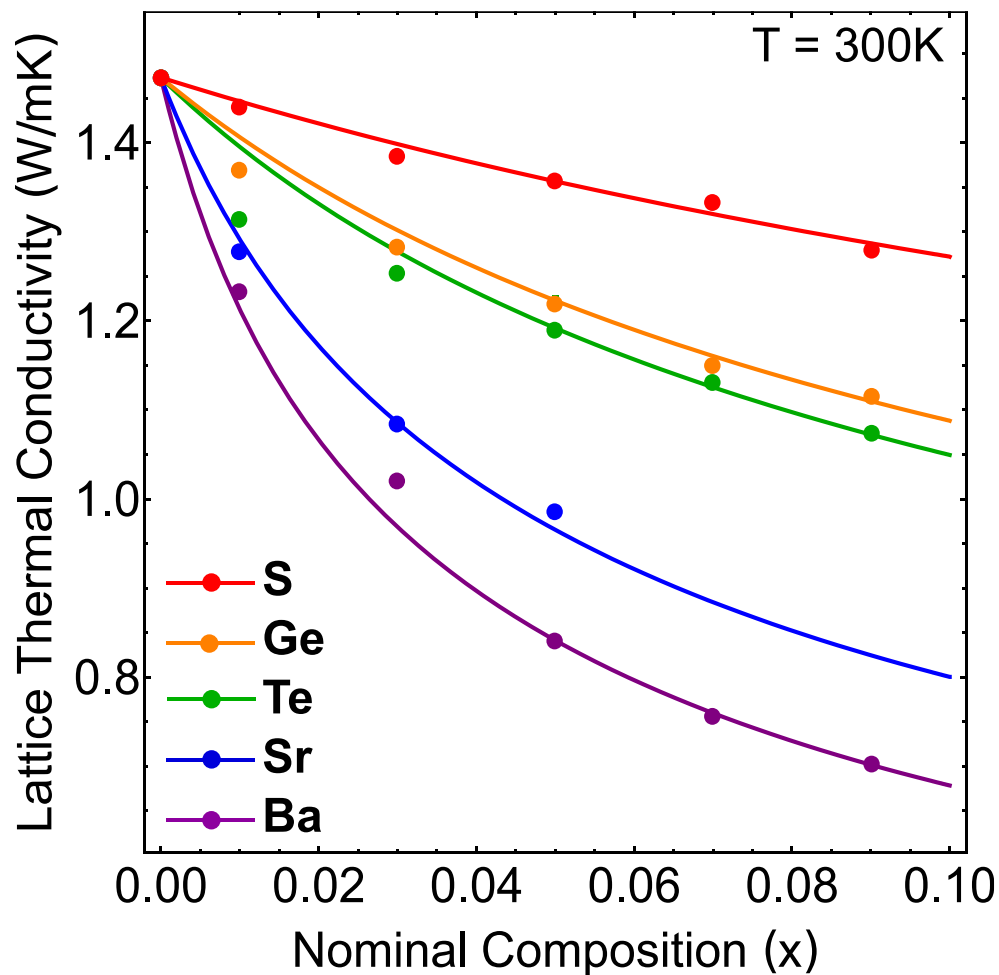
SnSe (Pnma) + one of Sr, Ba, S, Se, Te
(e.g. $\text{Sn}_{1-x}\text{Ba}_x\text{Se}$)



X-ray Diffraction, Rietveld Refinement
+
Vegard's Law



Experiment – Model System



Note large spread in the thermal conductivity w/alloying species.

Depressions range from slight (e.g. Sulfur) to severe (e.g. Ba)

Need to model to understand role of chemistry on scattering.

Abeles Model for Alloy Scattering¹

[1] B. Abeles, Phys. Rev., 1963, 131, 1906–1911.

Experiment – Transport and Modeling

Abeles model for composition dependent thermal conductivity

Thermal Conductivity of Alloy (κ_{alloy}) ...

$$\kappa_{\text{alloy}} = \kappa_0 \left(\frac{\tan^{-1}(u)}{u} \right)$$

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Disorder Scaling Parameter (u) ...

$$u = \left(\frac{\pi^2 \Theta \Omega}{h v_s^2} \kappa_0 \Gamma_{\text{tot}} \right)^{1/2}$$

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Net Scattering Factor (Γ_{tot}) ...

$$\Gamma'_{\text{tot}} = \Gamma'_m + \Gamma'_s$$

Experiment – Transport and Modeling

$$\Gamma'_{\text{tot}} = \Gamma'_m + \Gamma'_s \quad \left\{ \begin{array}{l} \Gamma'_m = x(1-x) \frac{a}{a+b} \left(\frac{\Delta M_{\text{Sn},X}}{M_{\text{Sn},X}} \right)^2 \\ \Gamma'_s = x(1-x) \varepsilon \frac{a}{a+b} \left(\frac{\Delta r_{\text{Sn},X}}{r_{\text{Sn},X}} \right)^2 \end{array} \right.$$

Experiment – Transport and Modeling

Mass Contrast

$$\Gamma'_{\text{tot}} = \Gamma'_m + \Gamma'_s \quad \left\{ \begin{array}{l} \Gamma'_m = x(1-x) \frac{a}{a+b} \left(\frac{\Delta M_{\text{Sn,X}}}{M_{\text{Sn,X}}} \right)^2 \\ \Gamma'_s = x(1-x) \varepsilon \frac{a}{a+b} \left(\frac{\Delta r_{\text{Sn,X}}}{r_{\text{Sn,X}}} \right)^2 \end{array} \right.$$

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Radii Contrast

Experiment – Transport and Modeling

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Free parameter

Radii Contrast

Experiment – Transport and Modeling

Mass Contrast

$$\Gamma'_{\text{tot}} = \Gamma'_m + \Gamma'_s \left\{ \begin{array}{l} \Gamma'_m = x(1-x) \frac{a}{a+b} \left(\frac{\Delta M_{\text{Sn},X}}{M_{\text{Sn},X}} \right)^2 \\ \Gamma'_s = x(1-x) \varepsilon \frac{a}{a+b} \left(\frac{\Delta r_{\text{Sn},X}}{r_{\text{Sn},X}} \right)^2 \end{array} \right.$$

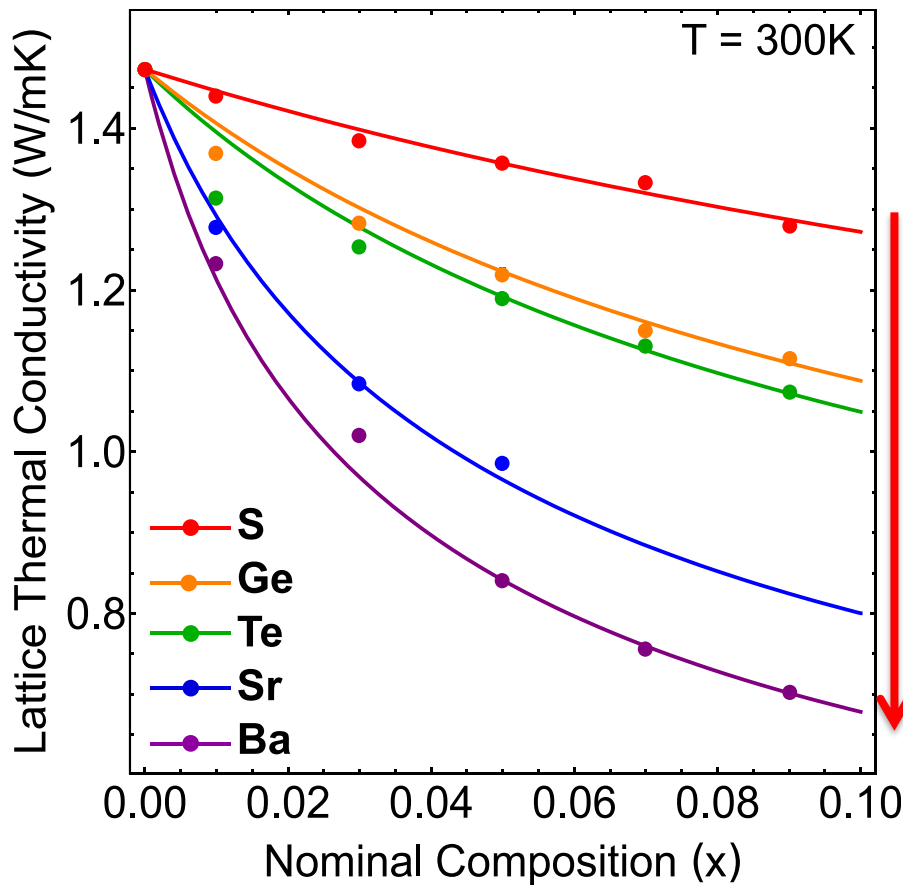
Free parameter

Radii Contrast

$$\mathbf{K}_{\text{alloy}} \left(\mathbf{K}_0, \nu_s, x, \varepsilon, r_\alpha \dots, m_\alpha \dots \right)$$

Experimental Summary

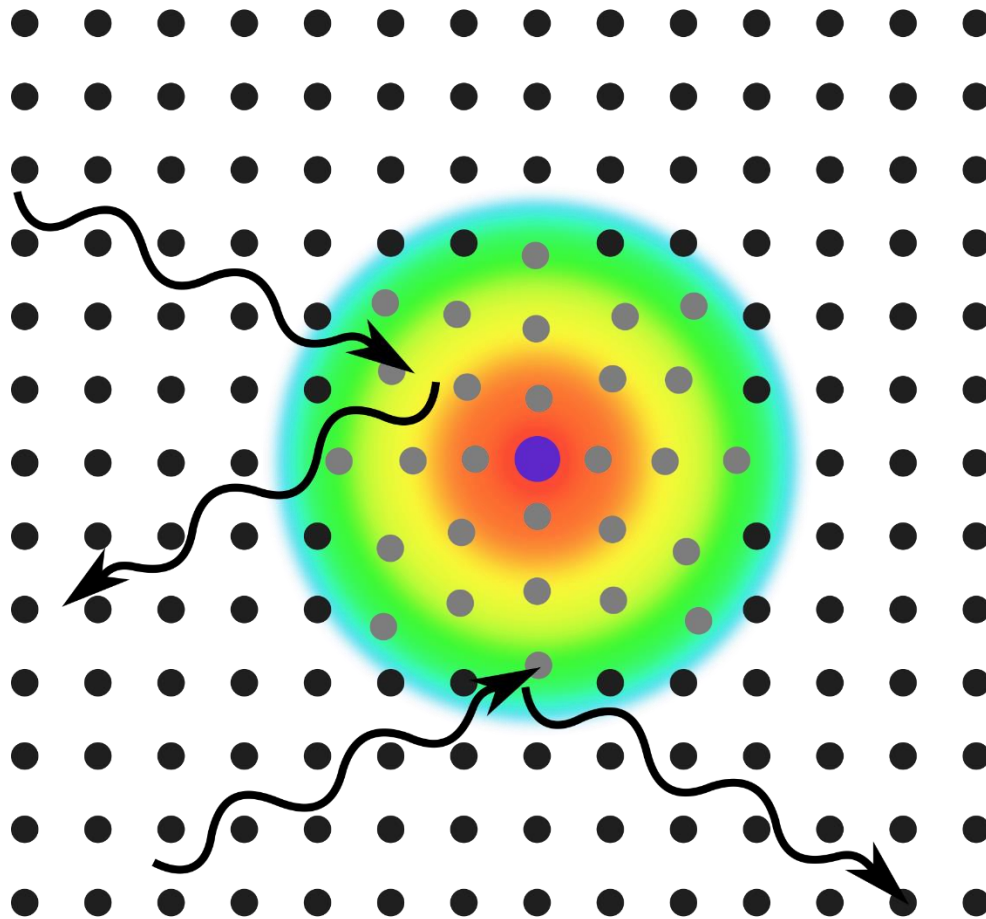
$$K_{\text{alloy}}(K_0, \nu_s, x, \mathcal{E}, r_\alpha \dots, m_\alpha \dots)$$



Increasing Strain Contribution
+
Decreasing Thermal Conductivity

Alloy	Γ_s/Γ_m	Γ'_s
S	0	0
Ge	1.73	0.0063
Te	1.99	0.0168
Sr	17.8	0.0297
Ba	85.0	0.0491

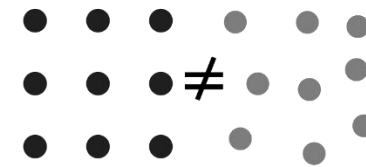
Computational Toy Models



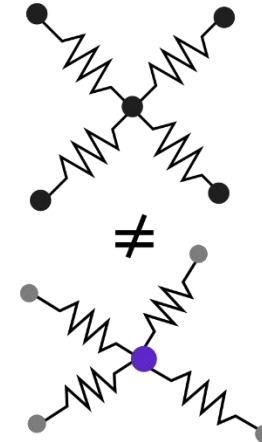
Mass Contrast:

$$m_{\bullet} \neq m_{\bullet}$$

Registry Loss:



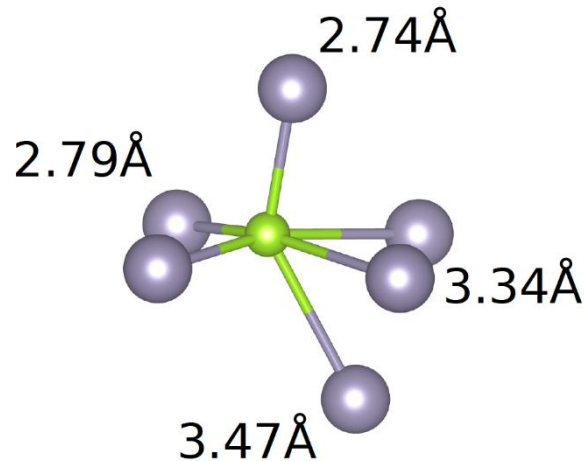
Bond Strain:



Computational Methods

Approach 1: Pair-Distribution Function (Supercell)

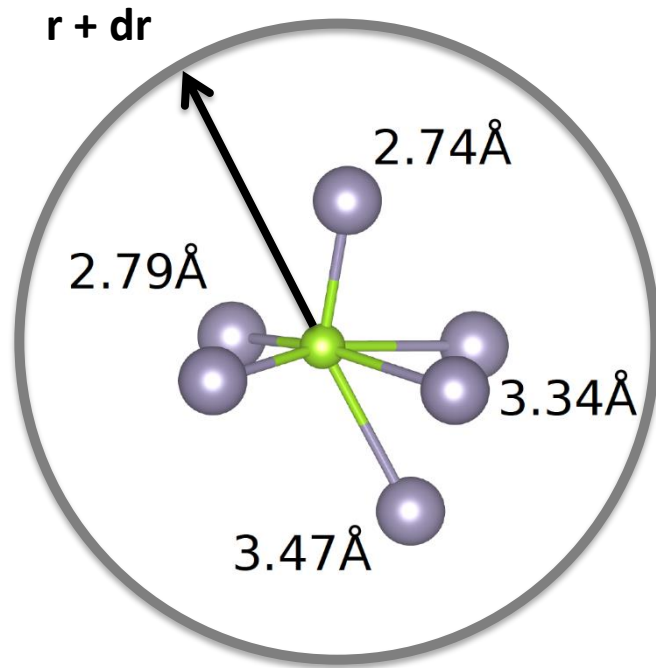
32-atom special quasi-random structures (SQS) to mimic 25% alloy PDF's



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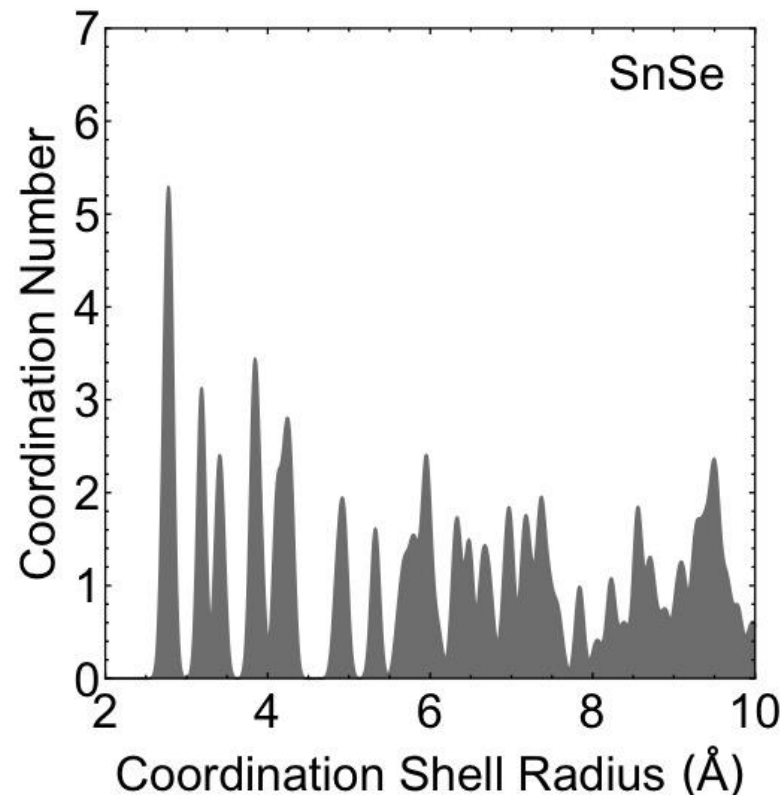
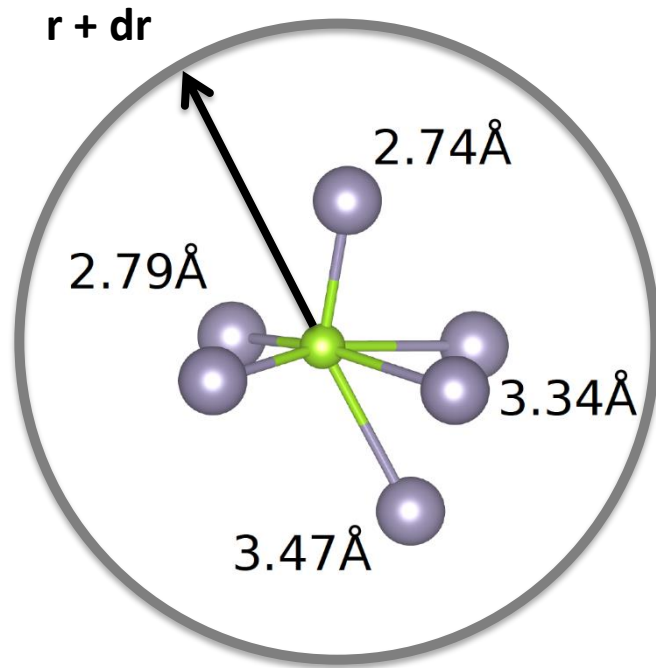
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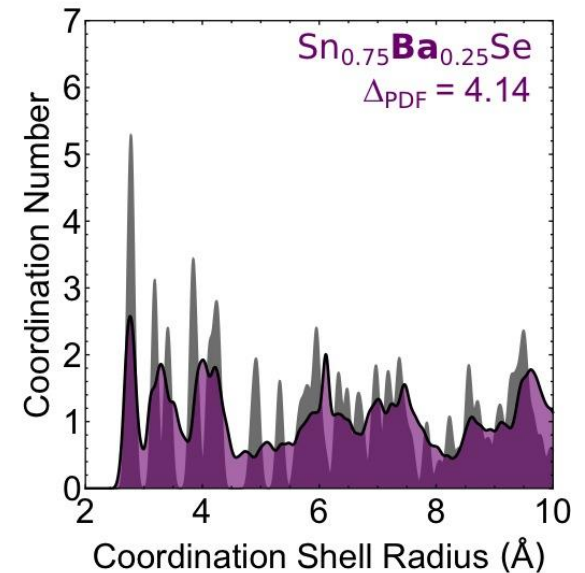
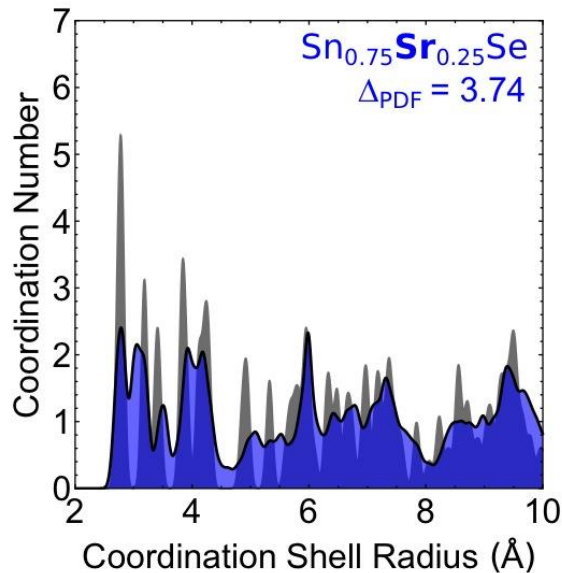
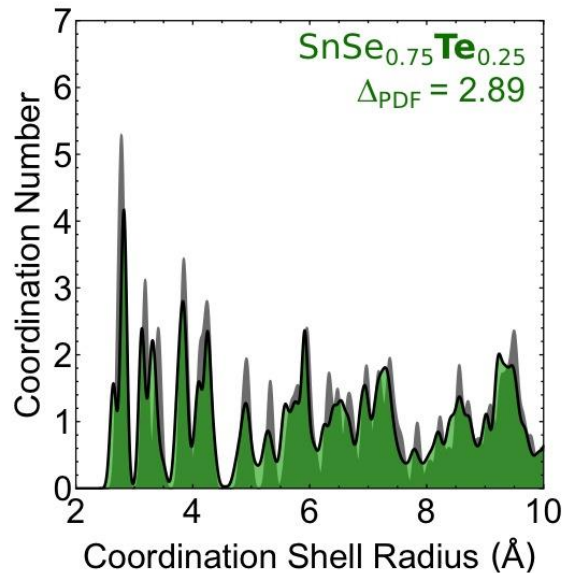
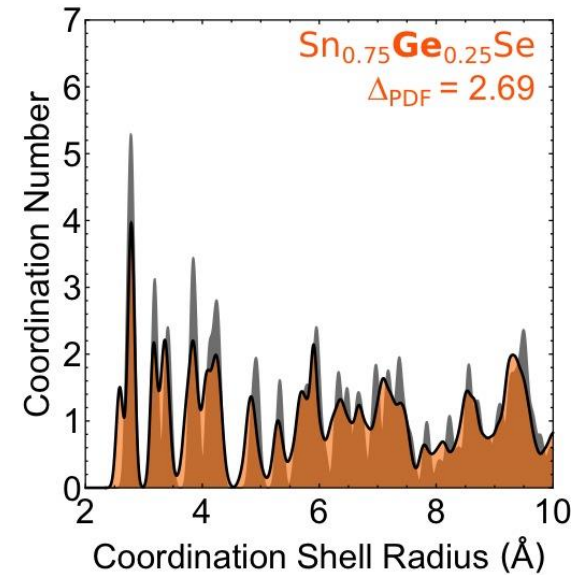
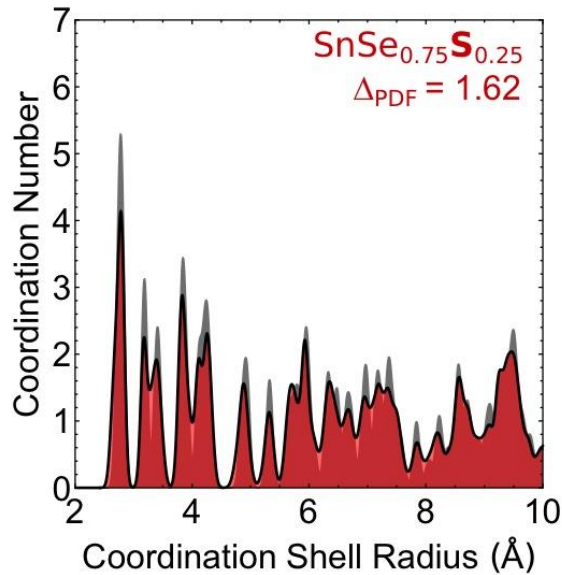
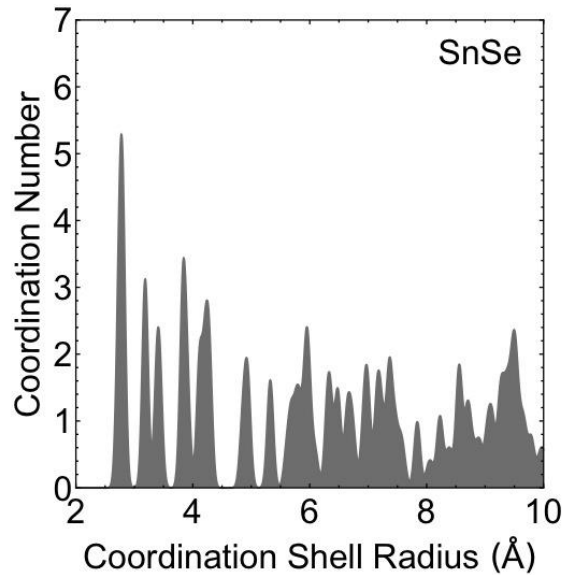
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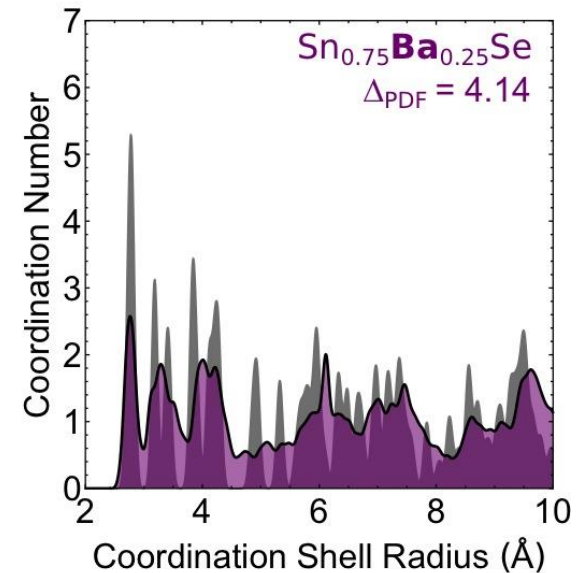
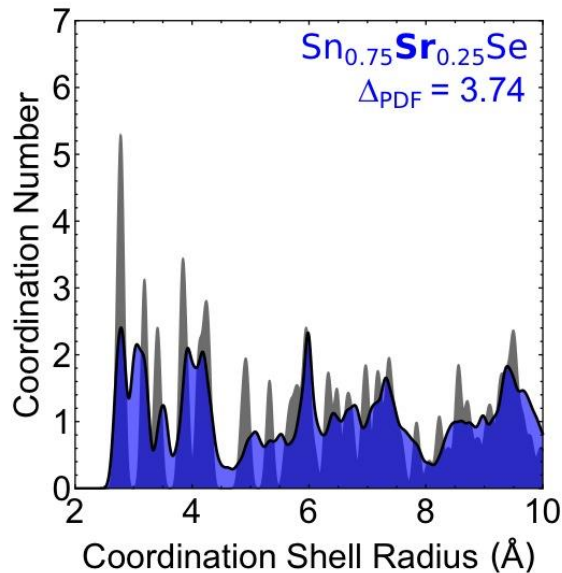
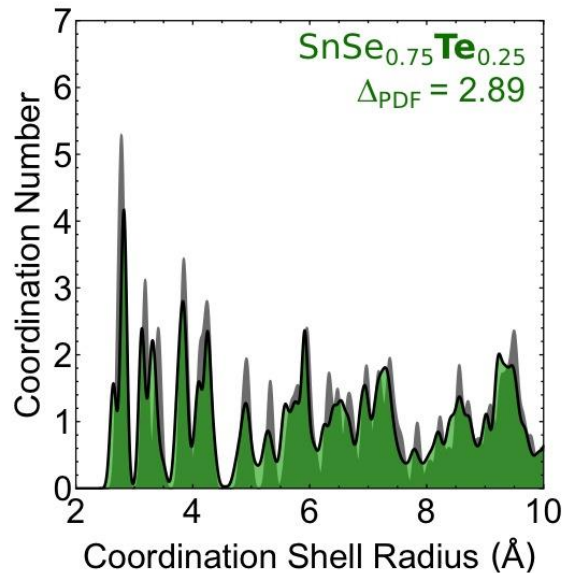


Computational Methods – Pair Distribution Fn



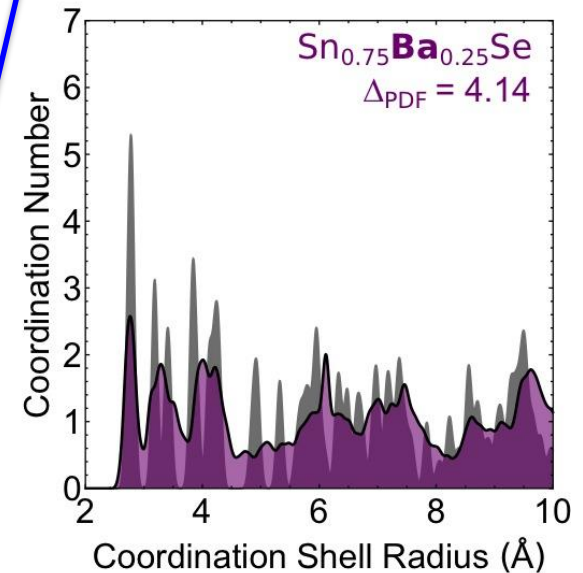
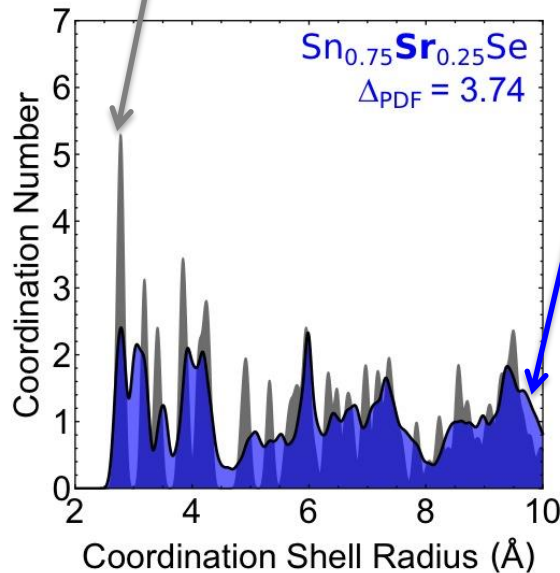
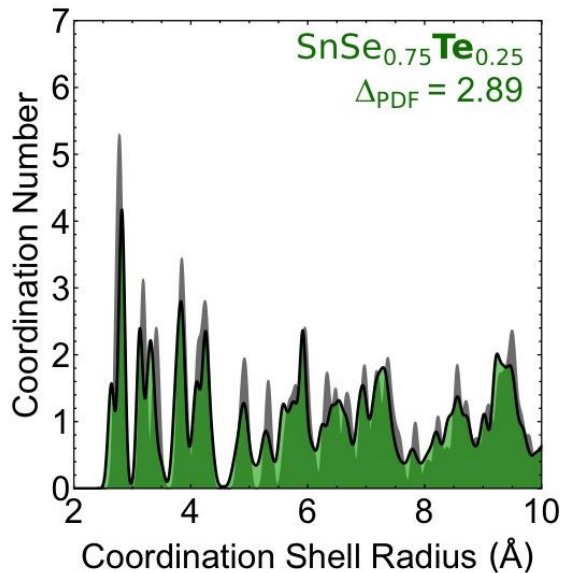
Computational Methods – Pair Distribution Fn

$$\Delta_{\text{PDF}} = \int_0^{r_{\text{max}}} |\text{PDF}_{\text{SnSe}}(r) - \text{PDF}_{\text{Alloy}}(r)| dr$$



Computational Methods – Pair Distribution Fn

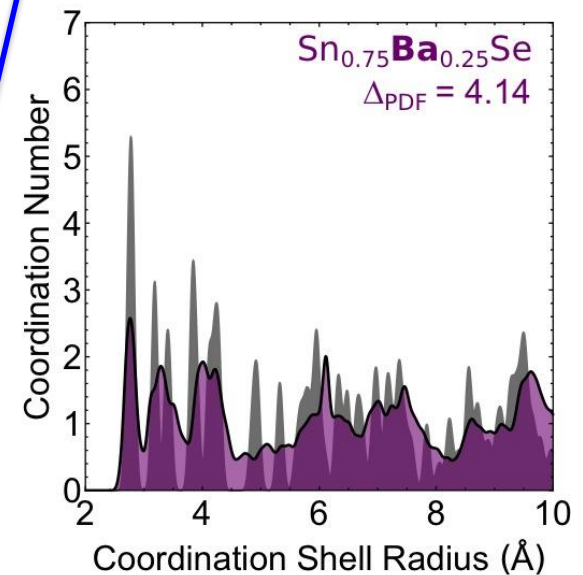
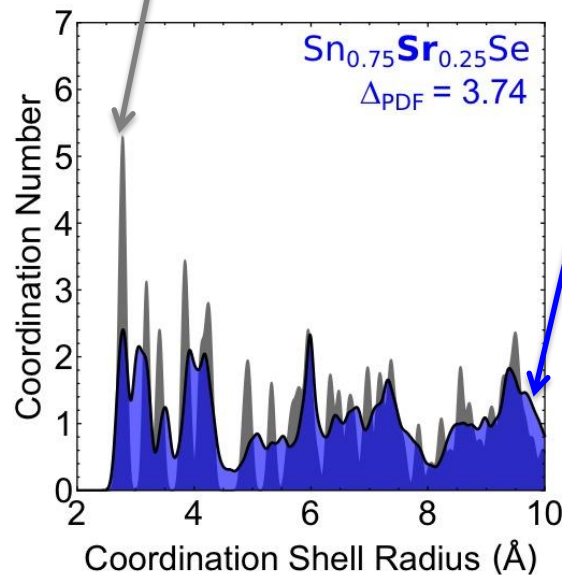
$$\Delta_{\text{PDF}} = \int_0^{r_{\text{max}}} |\text{PDF}_{\text{SnSe}}(r) - \text{PDF}_{\text{Alloy}}(r)| dr$$



Computational Methods – Pair Distribution Fn

$$\Delta_{\text{PDF}} = \int_0^{r_{\text{max}}} |\text{PDF}_{\text{SnSe}}(r) - \text{PDF}_{\text{Alloy}}(r)| dr$$

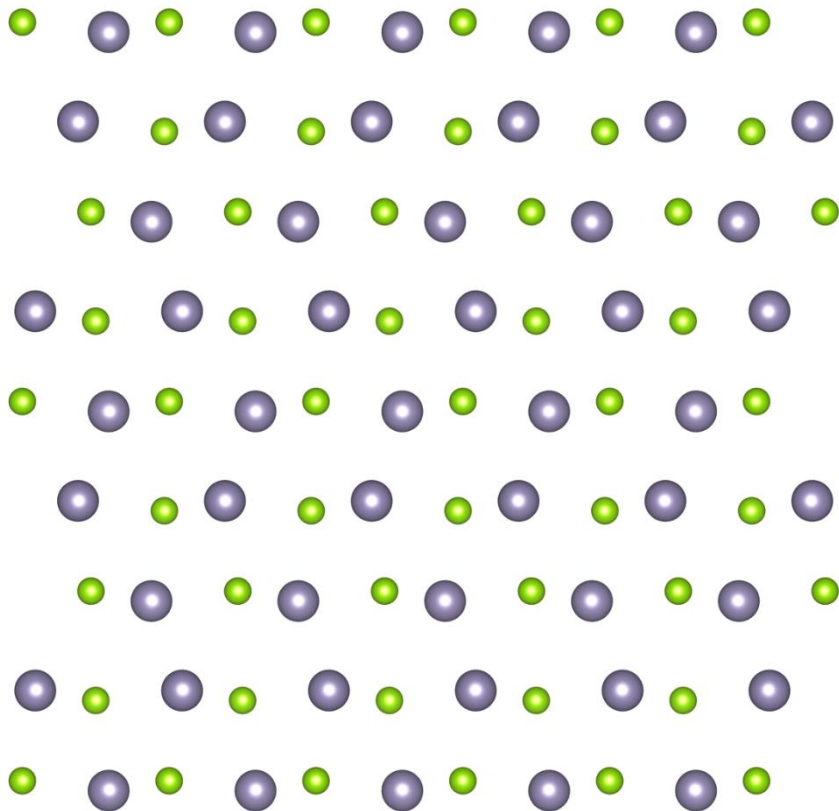
Alloy	Δ_{PDF} (Å)
SnSe	0
S	1.62
Ge	2.69
Te	2.89
Sr	3.74
Ba	4.14



Computational Methods

Approach 2: Single Atom Distortion (Supercell)

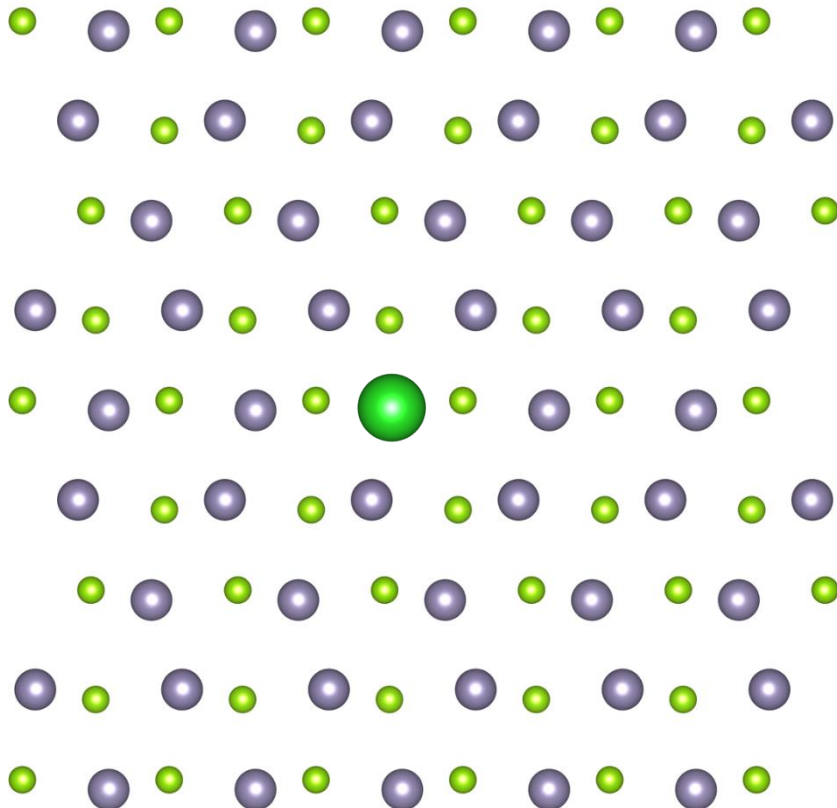
256-atom supercell with singular atom replaced by alloying species



Computational Methods

Approach 2: Single Atom Distortion (Supercell)

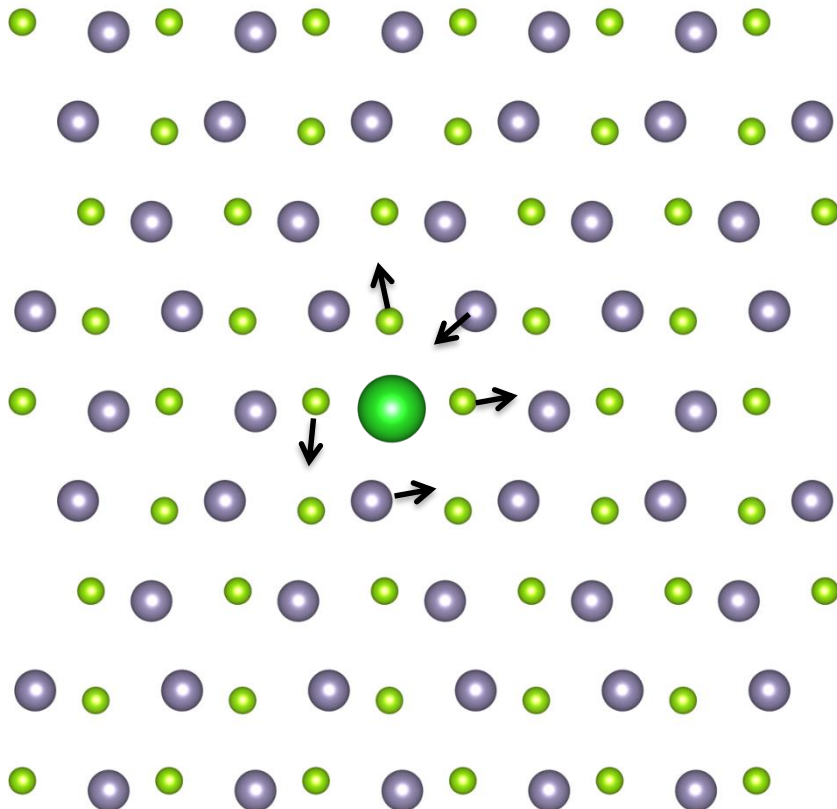
256-atom supercell with singular atom replaced by alloying species



Computational Methods

Approach 2: Single Atom Distortion (Supercell)

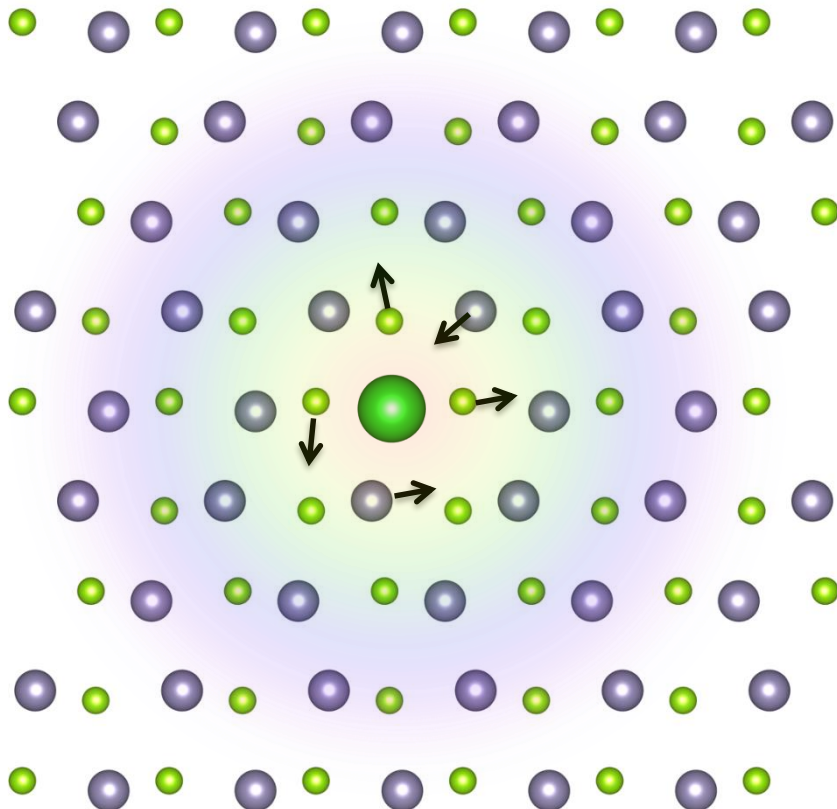
256-atom supercell with singular atom replaced by alloying species



Computational Methods

Approach 2: Single Atom Distortion (Supercell)

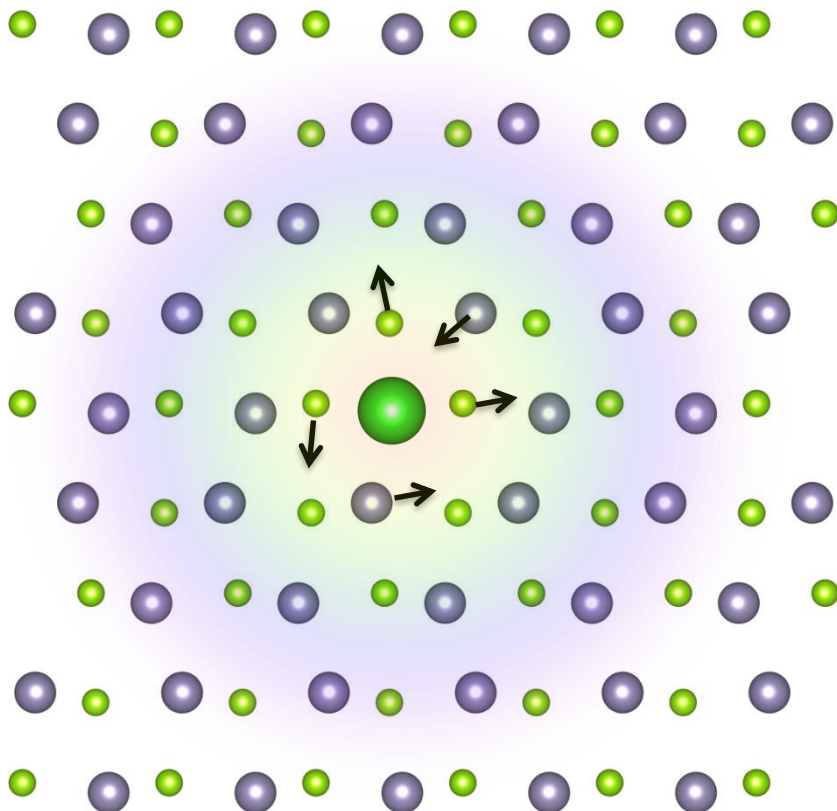
256-atom supercell with singular atom replaced by alloying species



Computational Methods

Approach 2: Single Atom Distortion (Supercell)

256-atom supercell with singular atom replaced by alloying species



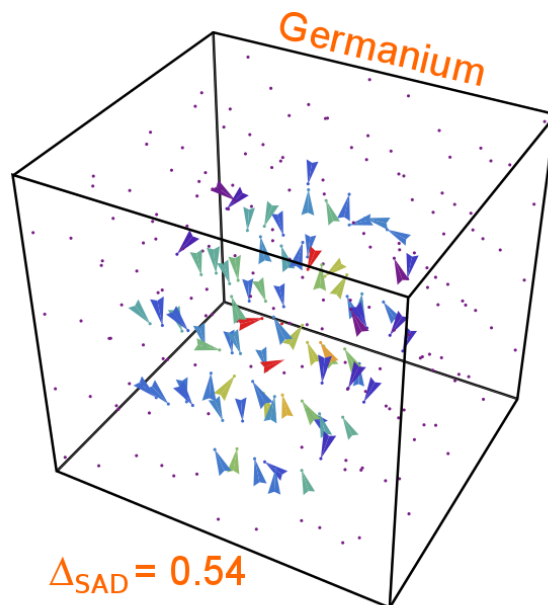
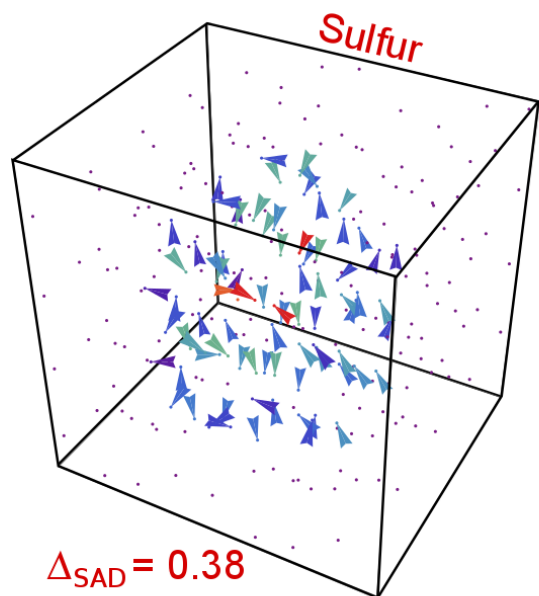
Change in local coordination around species...

How far from source atom does it extend?

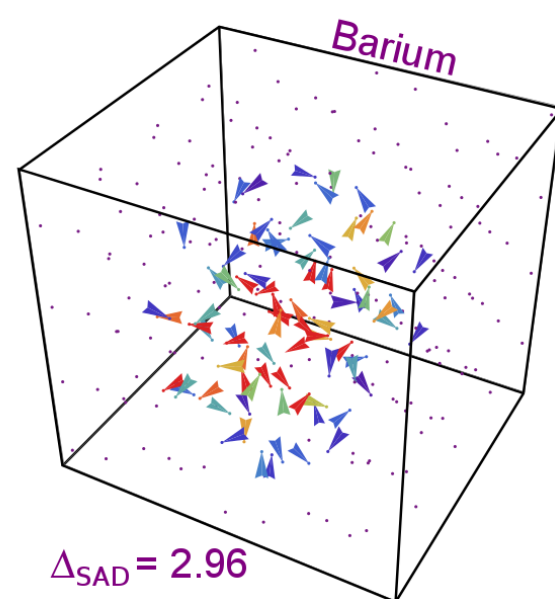
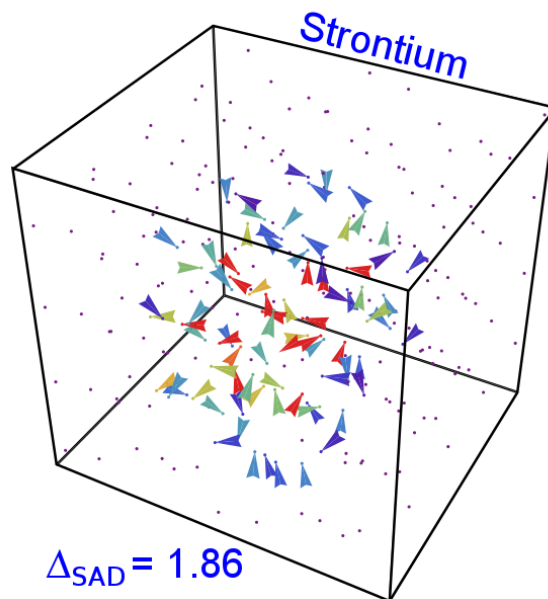
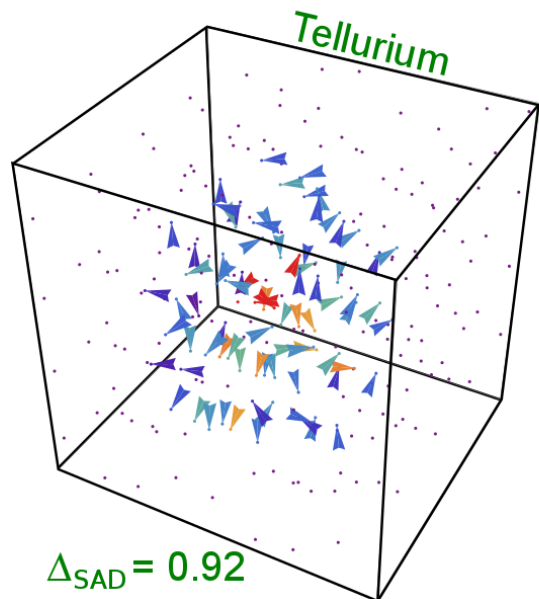
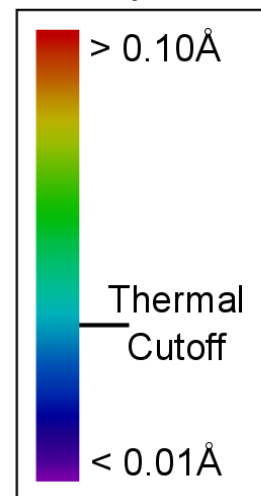
How large of a distortion?

Relation with chemistry?

Computational Methods – Single Atom Distortion



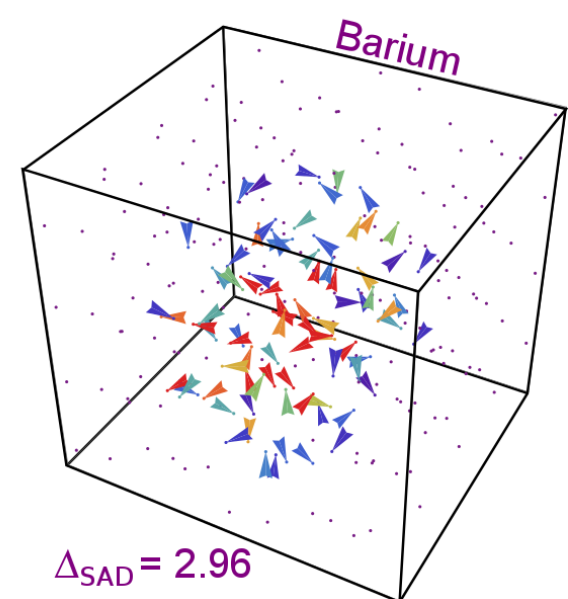
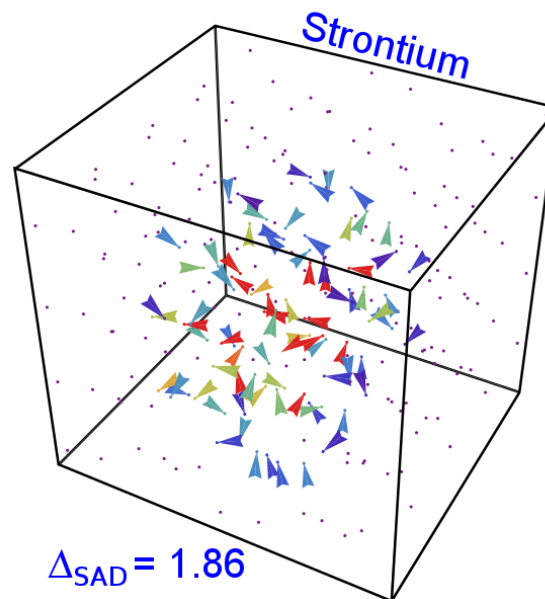
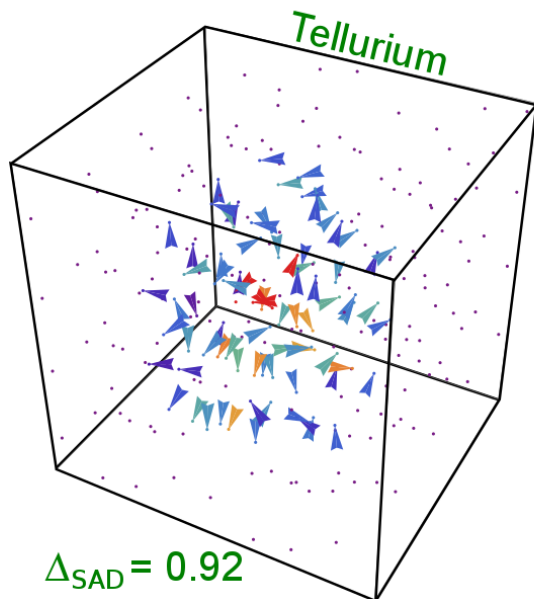
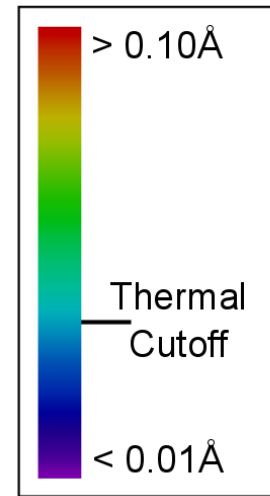
Atomic Displacement



Computational Methods – Single Atom Distortion

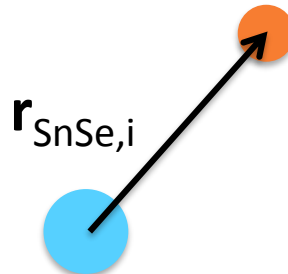
$$\Delta_{\text{SAD}} = \sum_{i=1}^N |\vec{r}_{\text{Alloy},i} - \vec{r}_{\text{SnSe},i}|$$

Atomic Displacement

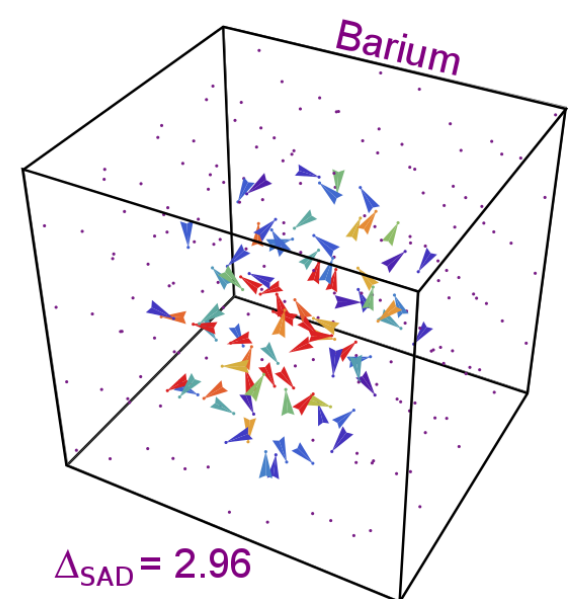
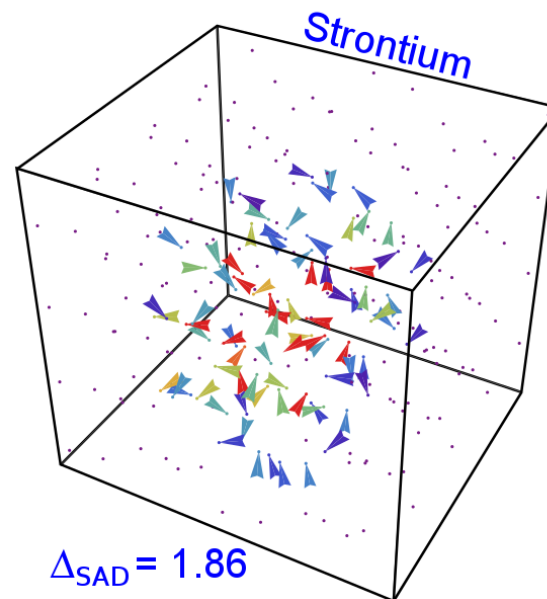
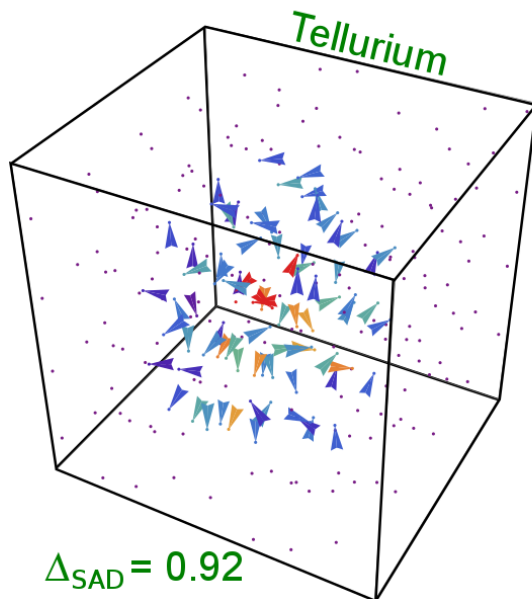
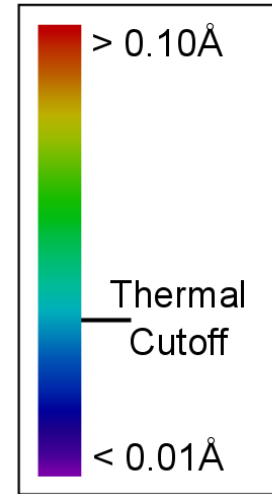


Computational Methods – Single Atom Distortion

$$\Delta_{\text{SAD}} = \sum_{i=1}^N |\vec{r}_{\text{Alloy},i} - \vec{r}_{\text{SnSe},i}|$$

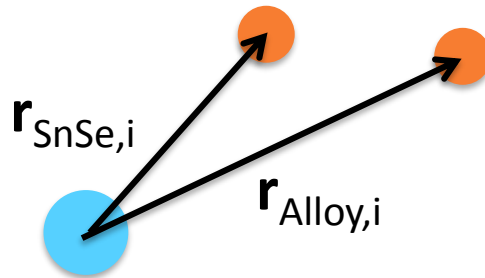


Atomic Displacement

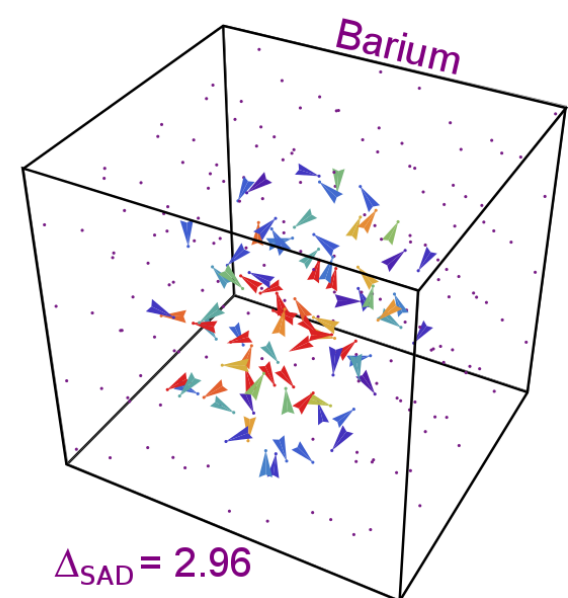
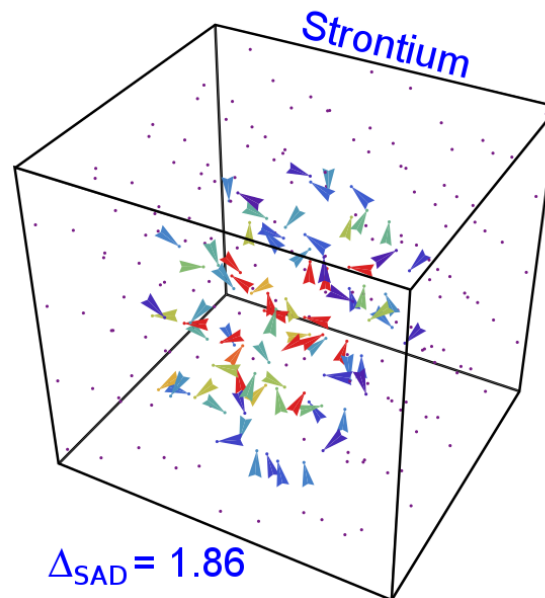
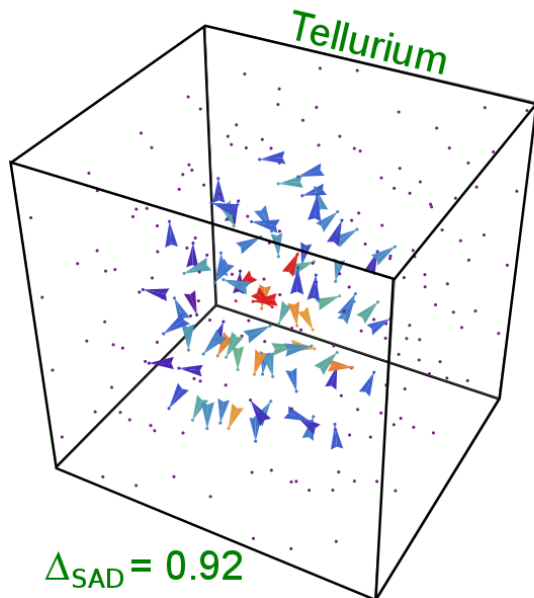
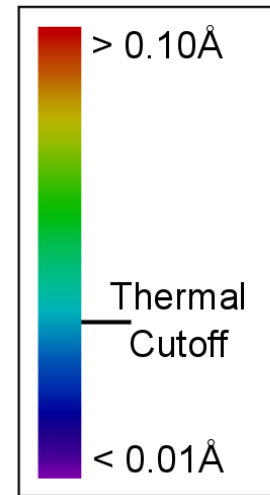


Computational Methods – Single Atom Distortion

$$\Delta_{\text{SAD}} = \sum_{i=1}^N |\vec{r}_{\text{Alloy},i} - \vec{r}_{\text{SnSe},i}|$$

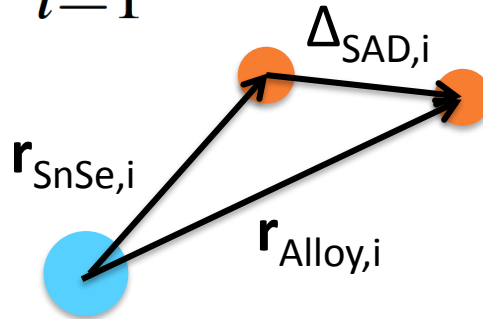


Atomic Displacement

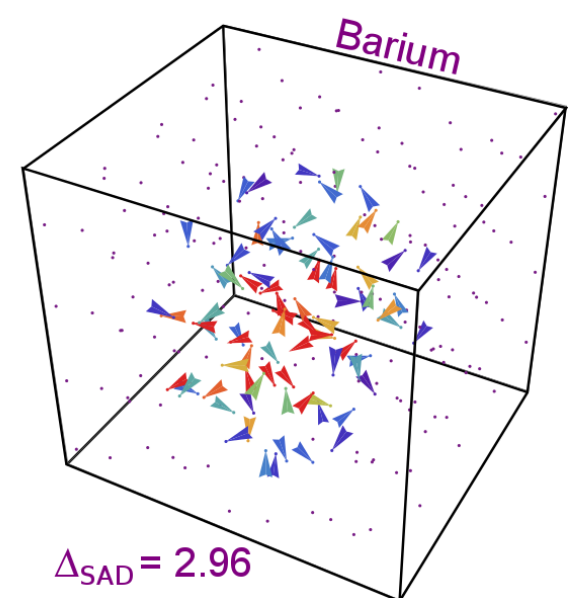
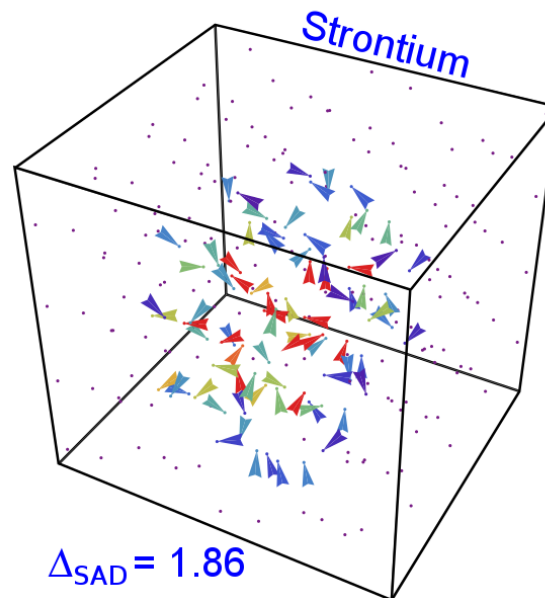
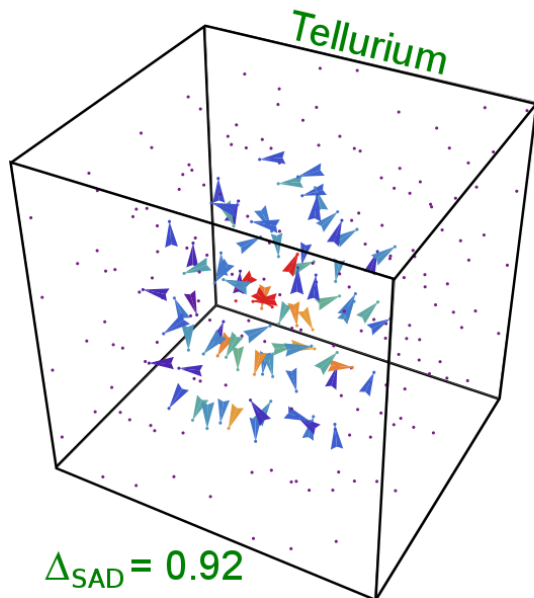
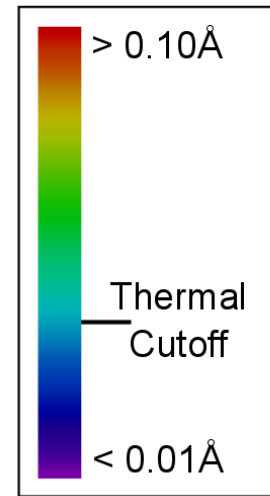


Computational Methods – Single Atom Distortion

$$\Delta_{\text{SAD}} = \sum_{i=1}^N |\vec{r}_{\text{Alloy},i} - \vec{r}_{\text{SnSe},i}|$$

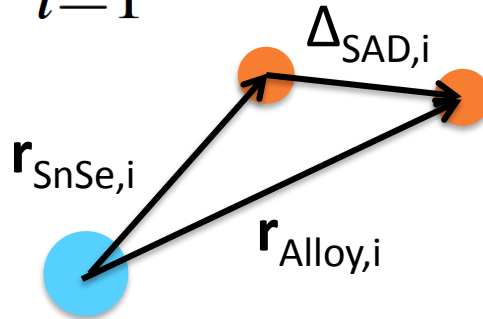


Atomic Displacement

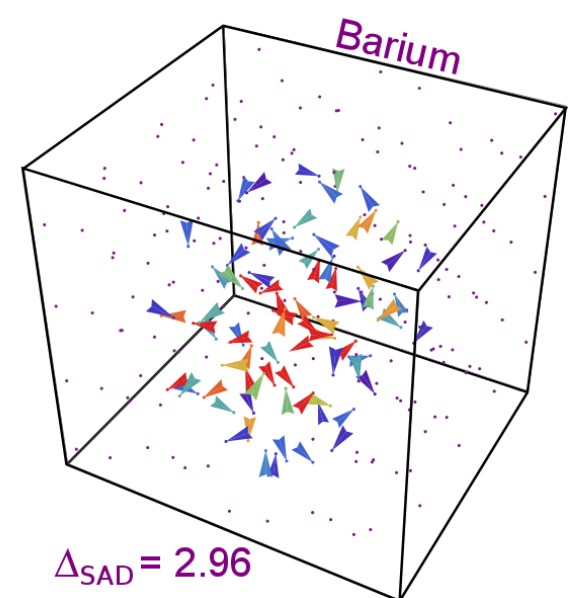
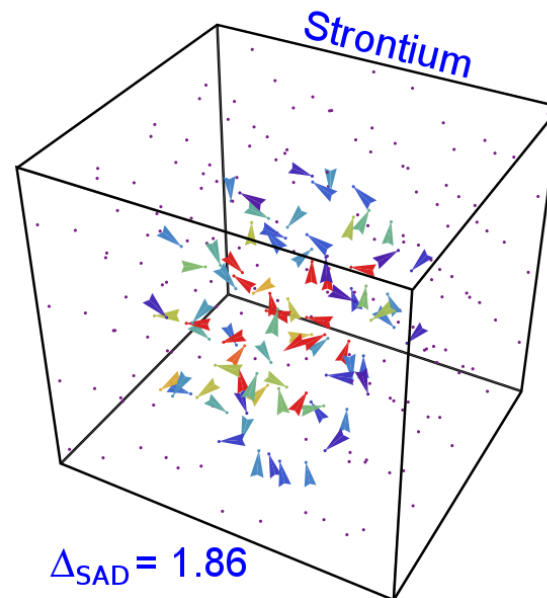
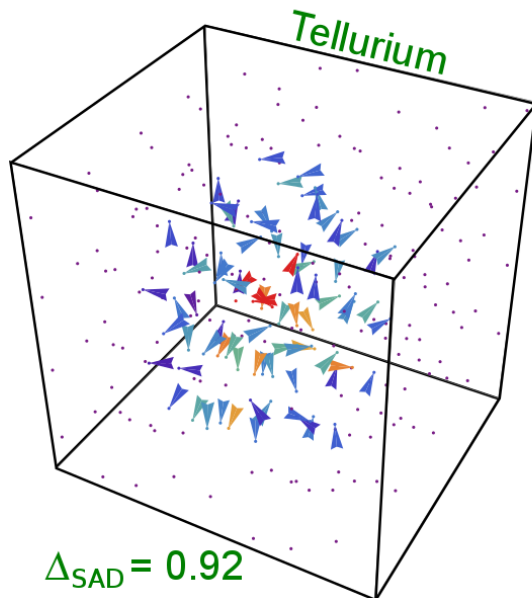


Computational Methods – Single Atom Distortion

$$\Delta_{\text{SAD}} = \sum_{i=1}^N |\vec{r}_{\text{Alloy},i} - \vec{r}_{\text{SnSe},i}|$$



Alloy	Δ_{SAD} (Å)
SnSe	0
S	0.38
Ge	0.54
Te	0.92
Sr	1.86
Ba	2.96



Computational Summary

Approach 3: Bulk Modulus (Supercell)

Standard calculation of total energy in DFT (LDA) as a function of cell volume...

Fitting of the Murnaghan equation of state to $E(\Omega)$

$$E(\Omega) = E_0 + \frac{B_0\Omega}{B'_0} \left(\frac{(\Omega_0/\Omega)^{B'_0}}{B'_0 - 1} + 1 \right) - \frac{B_0\Omega_0}{B'_0 - 1}$$

Alloy	B_0 (GPa)
SnSe	42.2
S	40.8
Ge	39.9
Te	38.1
Sr	34.3
Ba	31.5

Hybridization of Experimental and Computation

Do experiment and computation agree?

Alloy	B_0 (GPa)	Δ_{PDF} (Å)	Δ_{SAD} (Å)
SnSe	42.2	0	0
S	40.8	1.62	0.38
Ge	39.9	2.69	0.54
Te	38.1	2.89	0.92
Sr	34.3	3.74	1.86
Ba	31.5	4.14	2.96

Alloy	Γ_s/Γ_m	Γ'_s
S	0	0
Ge	1.73	0.0063
Te	1.99	0.0168
Sr	17.8	0.0297
Ba	85.0	0.0491

Hybridization of Experimental and Computation

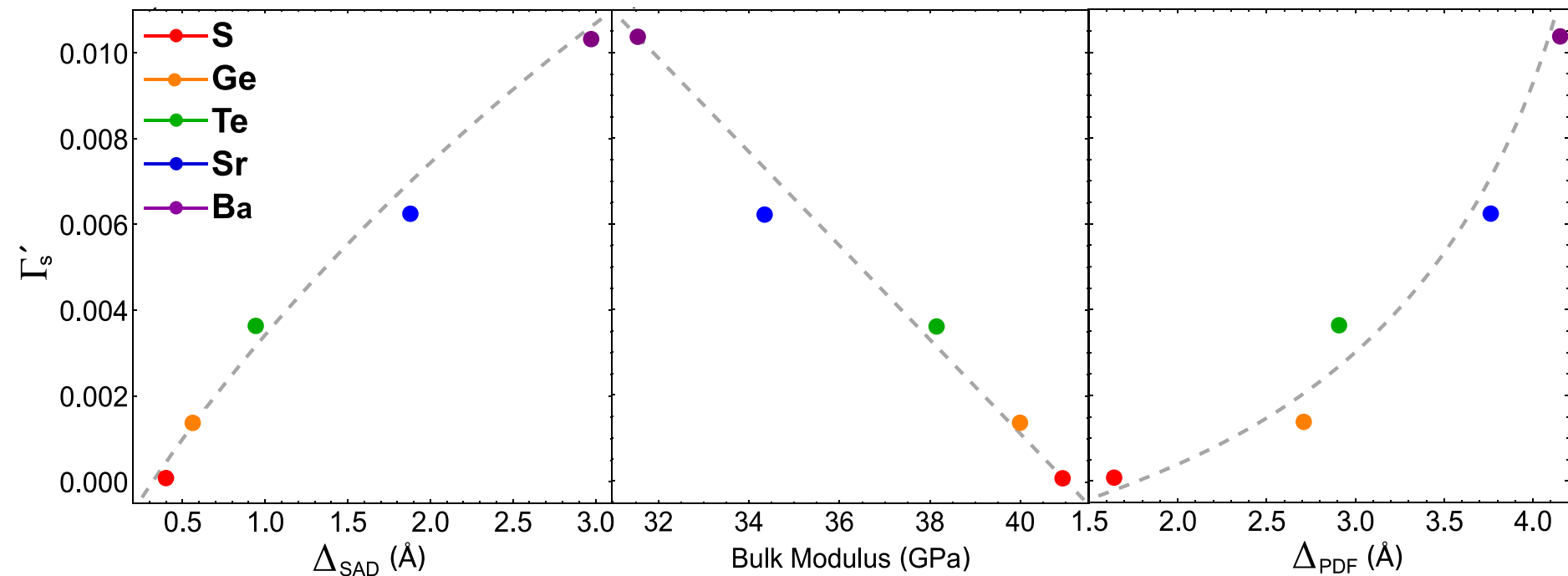
Do experiment and computation agree?

Alloy	B_0 (GPa)	Δ_{PDF} (Å)	Δ_{SAD} (Å)
SnSe	42.2	0	0
S	40.8	1.62	0.38
Ge	39.9	2.69	0.54
Te	38.1	2.89	0.92
Sr	34.3	3.74	1.86
Ba	31.5	4.14	2.96

Alloy	Γ_s/Γ_m	Γ'_s
S	0	0
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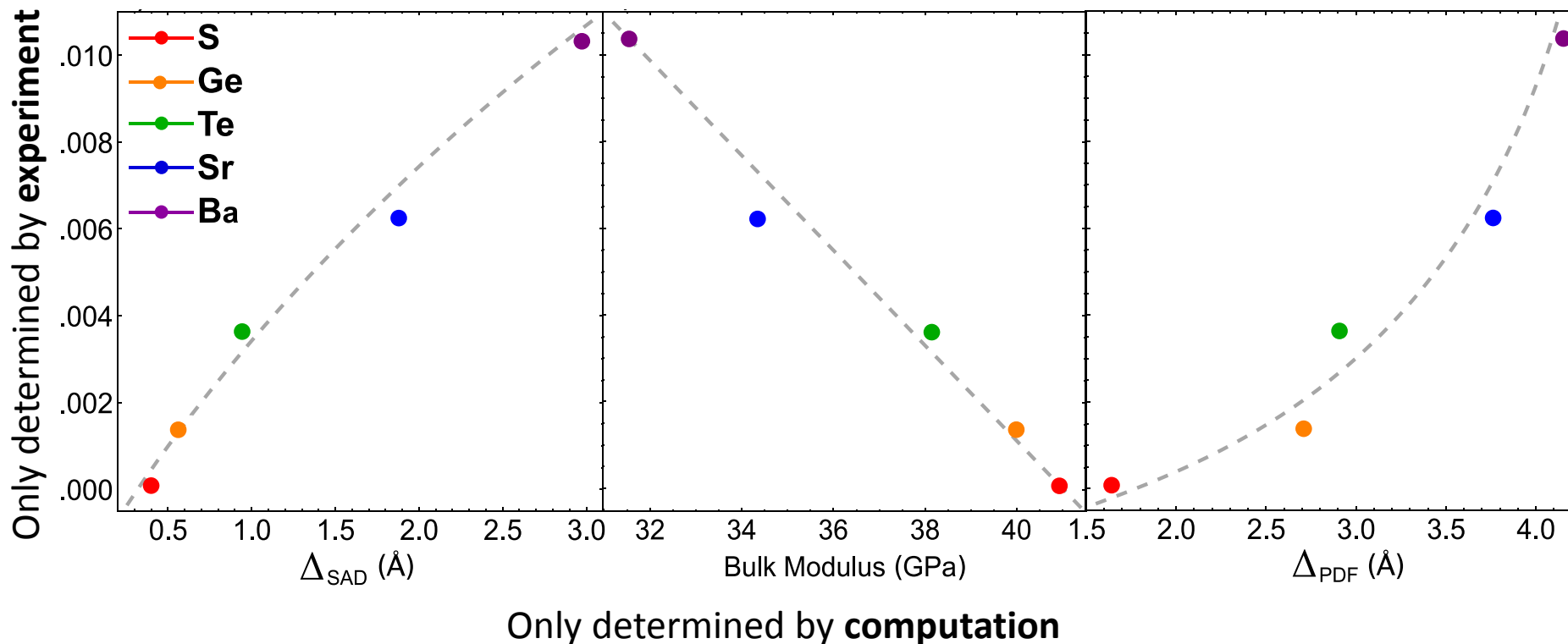
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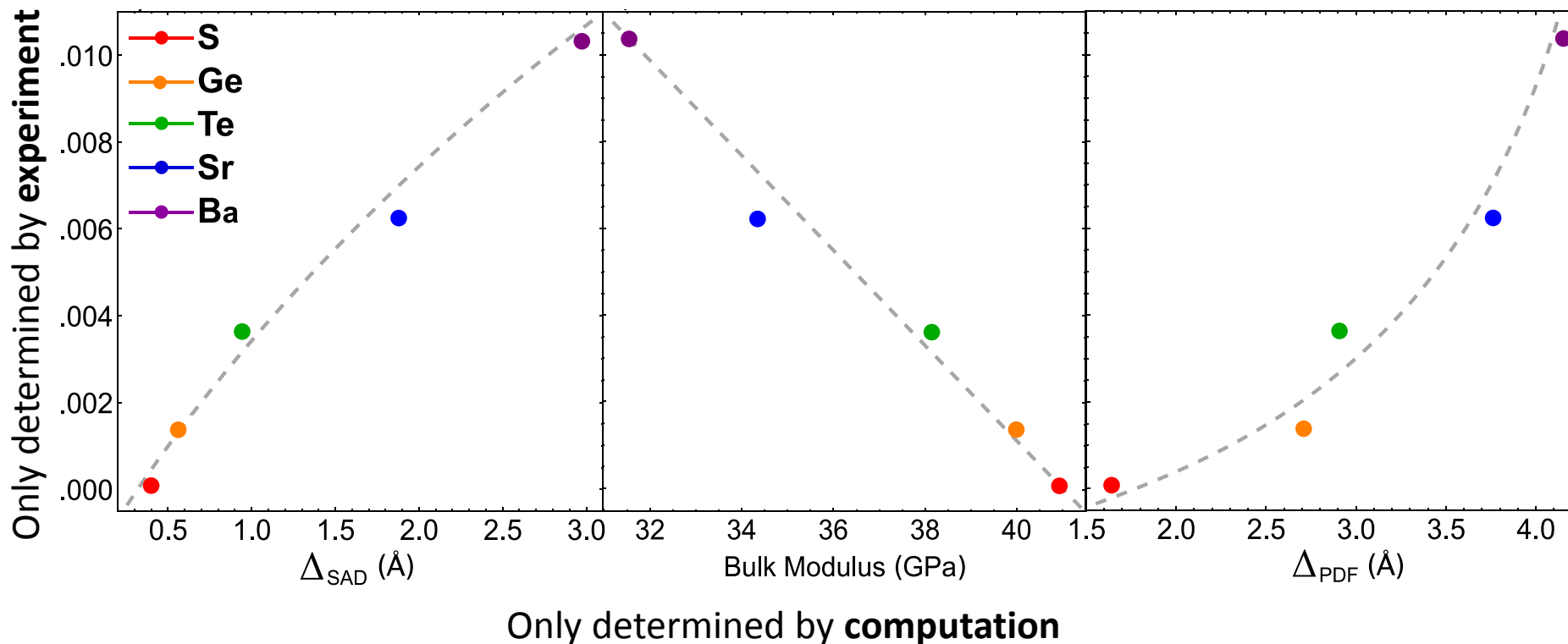
Hybridization of Experimental and Computation

Do experiment and computation agree?



Hybridization of Experimental and Computation

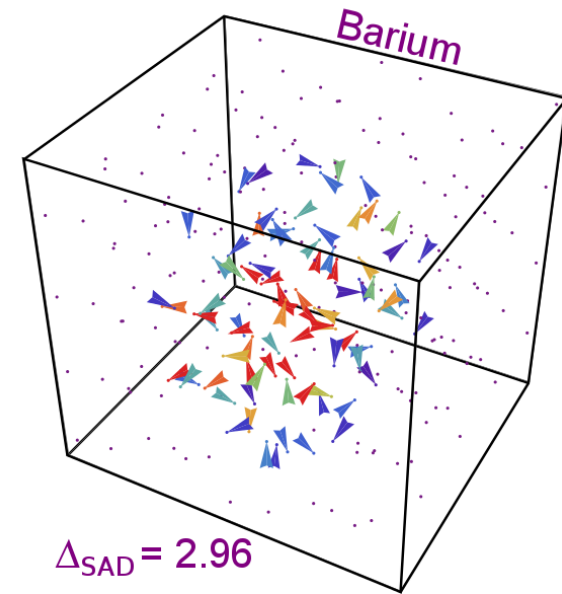
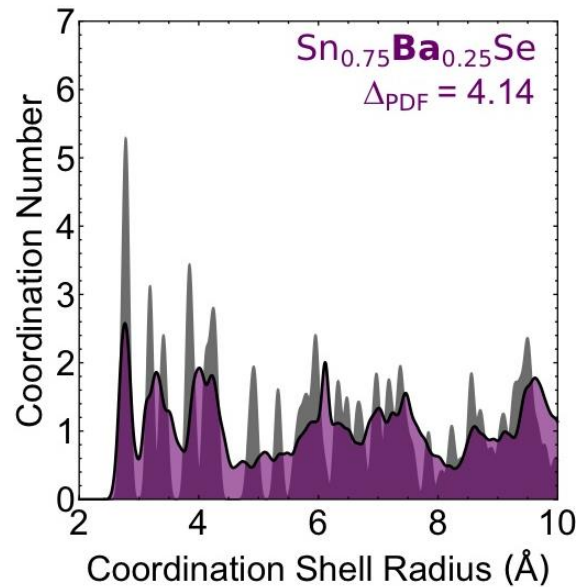
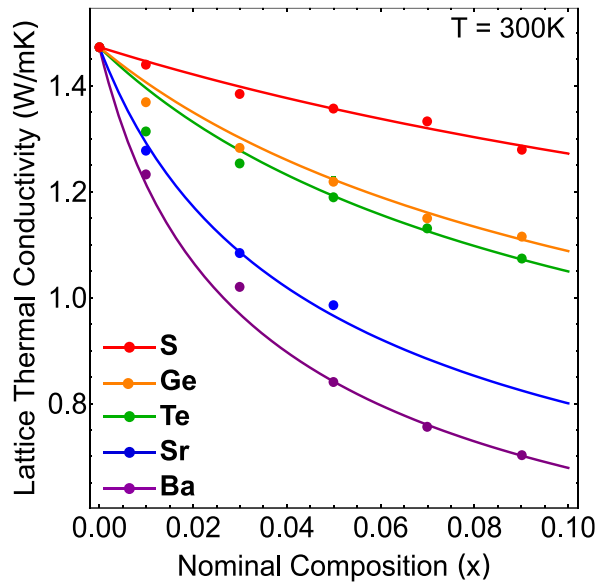
Do experiment and computation agree?



Yes!

Computation successfully ranks relative changes in strain and transport by proxy.

Conclusion



Presented inexpensive, conceptually transparent methods to visualize alloying in SnSe.

Strain effects can be observed far from host lattice site.

Computational ranking successful as proxy for experiment.

Possible down-selection of effective alloying agents.

