The tin impurity in $Bi_{0.5}Sb_{1.5}Te_3$ alloys

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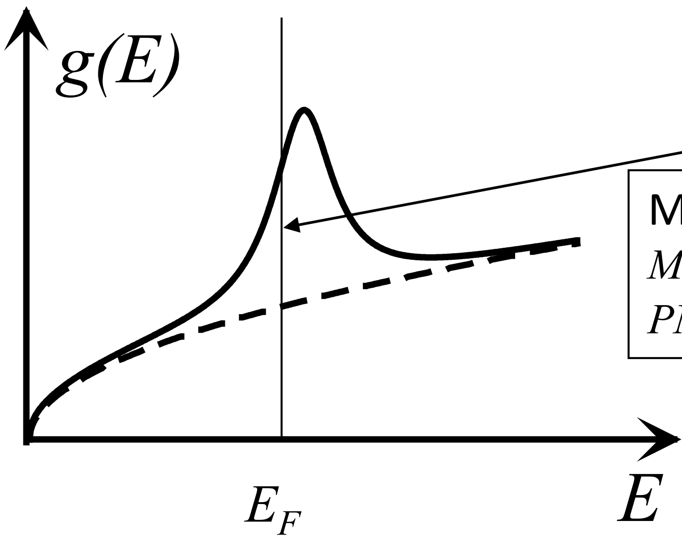
Resonant levels increase thermopower

Mott relation for degenerate statistics

$$S = \frac{\pi^2}{3} \cdot \frac{k_B}{e} \cdot k_B T \cdot \left[\frac{g(E)}{n(E)} + \frac{1}{\mu(E)} \cdot \frac{\partial \mu(E)}{\partial E} \right]_{E=E_F}$$



Mechanism 1
Mahan and Sofo,
*PNAS **93** 7436, 1996*

Mechanism 2: Resonant scattering

*A. Blandin & J. Friedel, Le Journal de Physique et le Radium **20** 160, 1959*

In PbTe: Yu. Ravich, CRC Handbook on Thermoelectrics, D. M. Rowe, Ed. 1995

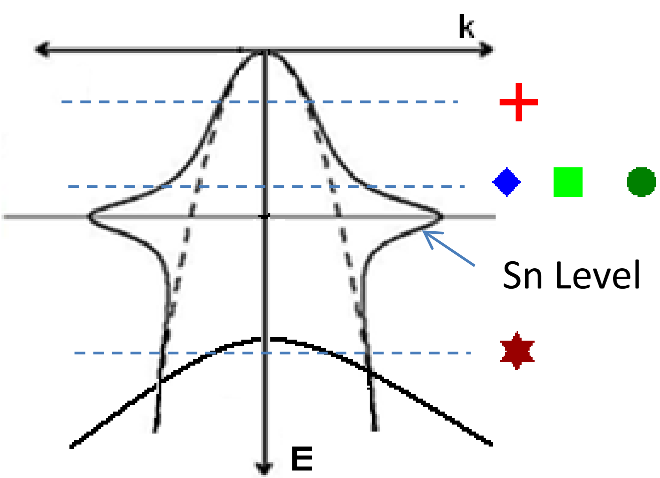
*In Bi_2Te_3: M. K. Zhitinskaya, S. A. Nemov and T. E. Svechnikova, Phys. Solid State **40** 1297, 1998*

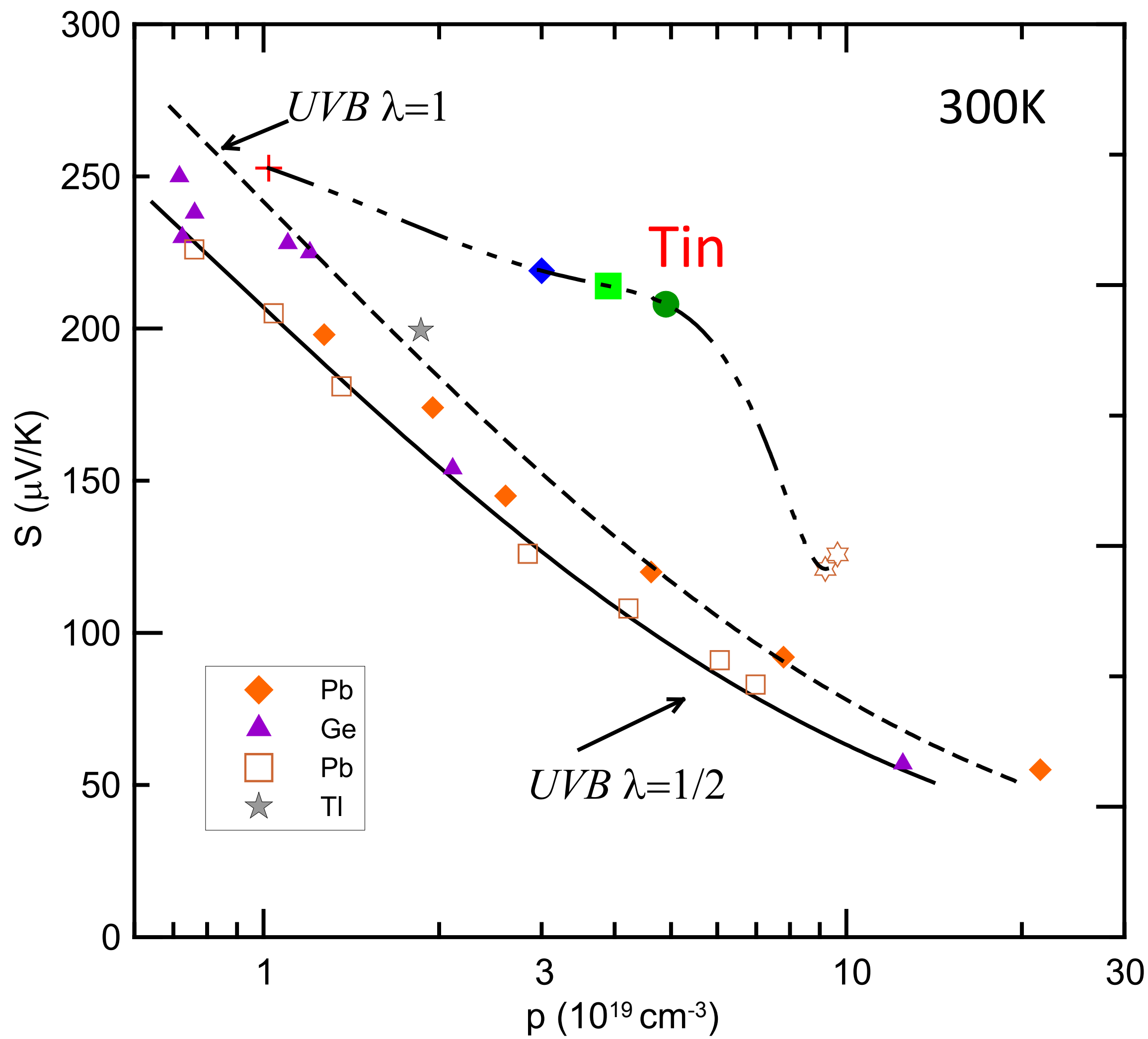
- Works great at cryogenic temperatures
- Will NOT give high *zT* at operating temperatures where acoustic/optic phonon scattering dominates

Which dominates? Can be proven experimentally by measuring Nernst effect

Starting Point: binary Bi_2Te_3 with Sn

- Lowest Sn% -> compensated
- Middle Sn% have increased Seebeck over Ge and Pb doped
- Highest Sn% lose 'resonance'





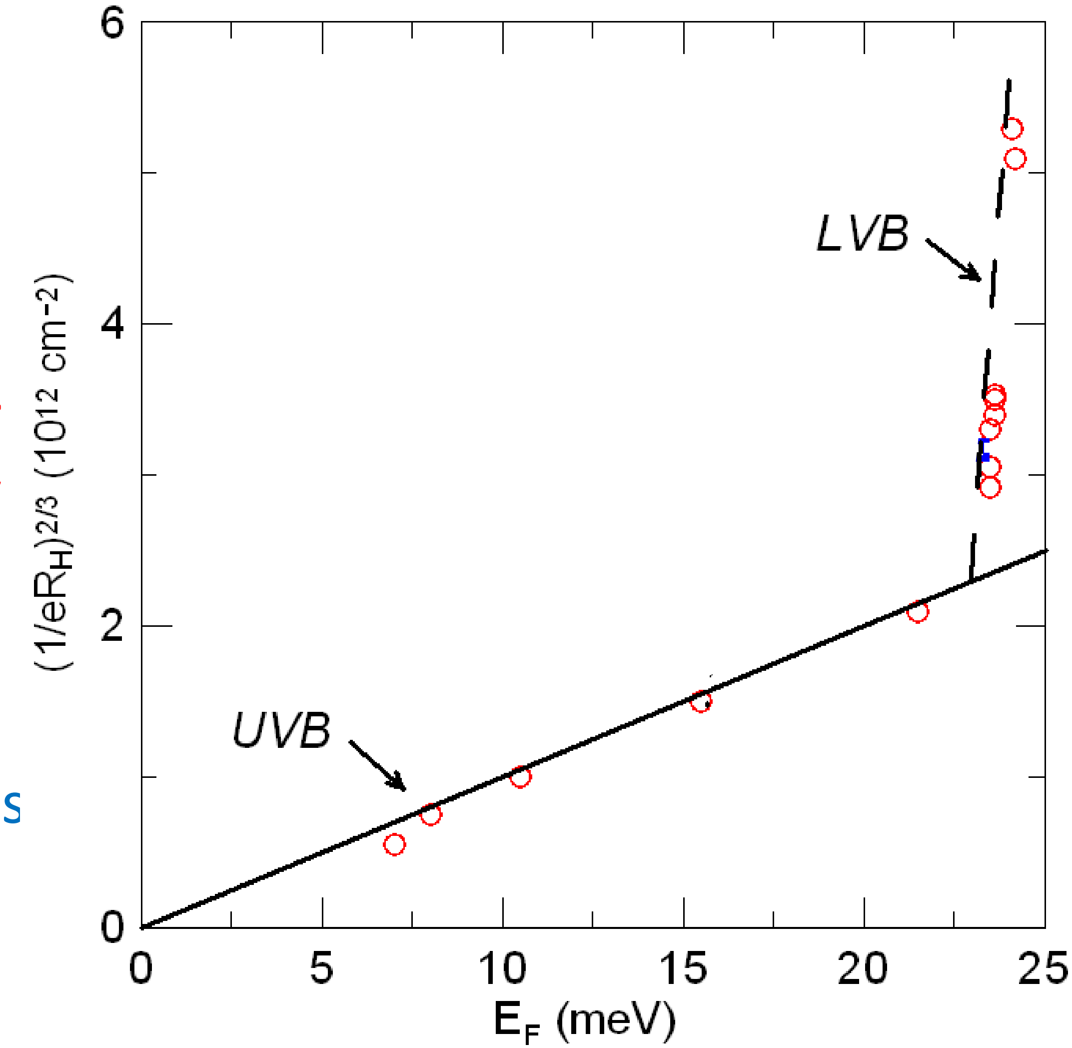
Jaworski, C.M. et al., Phys Rev B **80** 233201 (2009)

Shubinov-de Haas oscillations -> Hall vs E_F

Tin Content	Oscillation Frequency	Fermi Surface Area
$Bi_{2-x}Sn_xTe_3$	$[\Delta(1/B)]^{-1}$ T	(m^{-2})
x=0.0025	12.7	1.21E+17
x=0.0075	11.4	1.09E+17
x=0.015	13.5	1.29E+17

Carrier density from Hall

Slope of line = effective mass



Fermi Level from SdH

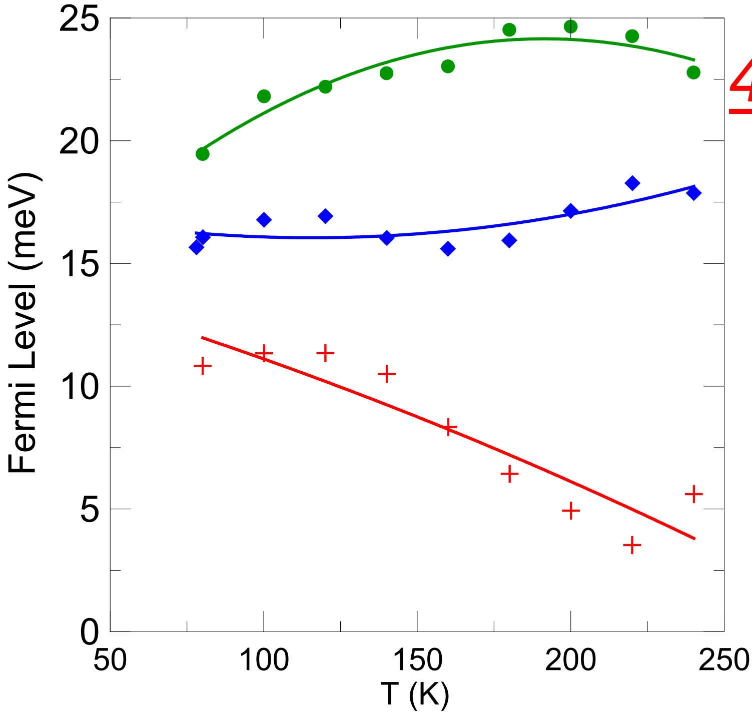
1: Kulbachinskii, Physical Review B, 1994, Vol. 50, 23, 16921.
2: Kohler, H.: Physica Status Solidi (b), 1976, Vol. 74.

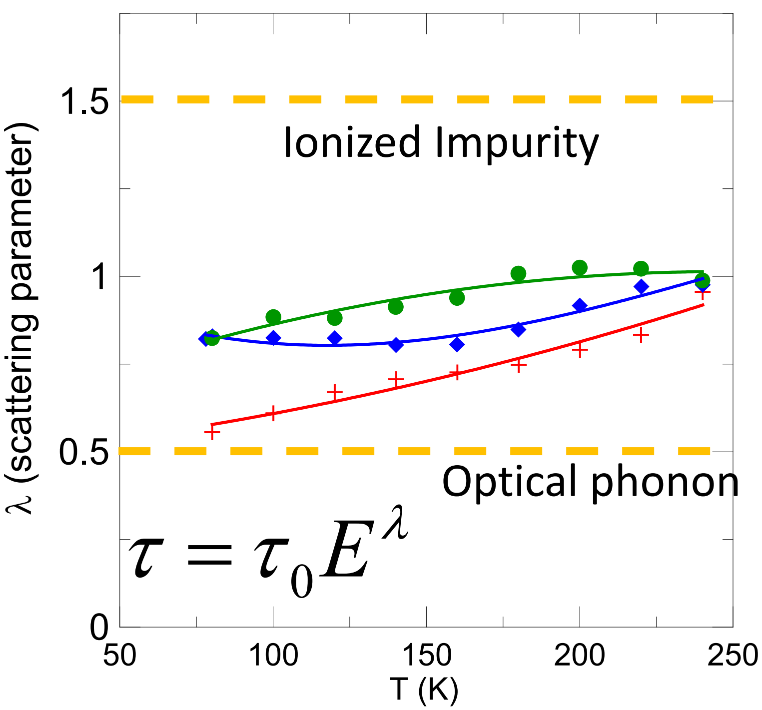
4-parameter fit to transport properties

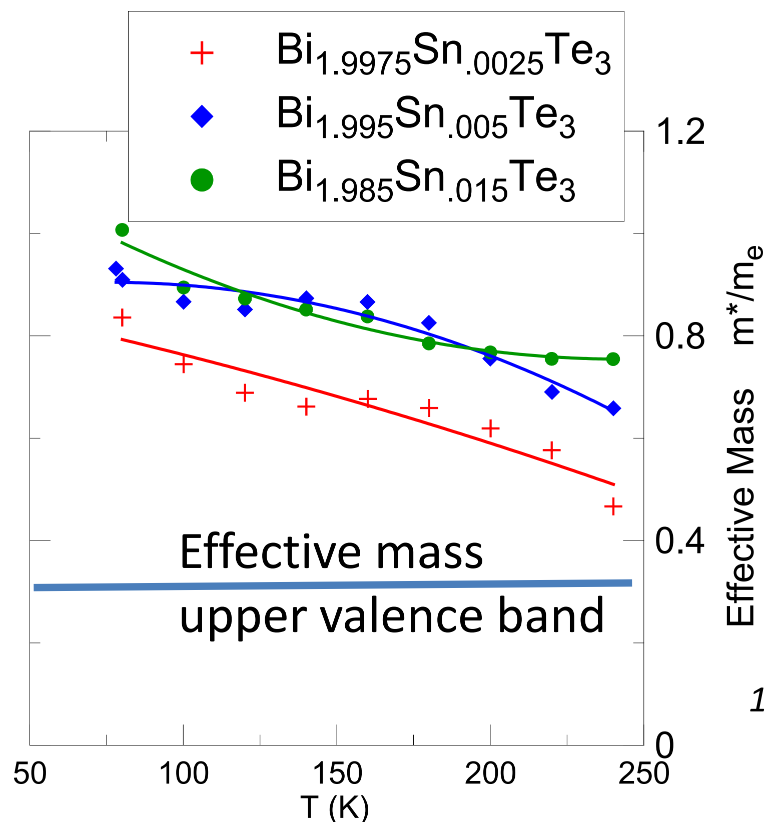
It's this | Not that

$$S = \frac{\pi^2}{3} \cdot \frac{k_B}{e} \cdot k_B T \cdot \left[\frac{g(E)}{n(E)} + \frac{1}{\mu(E)} \cdot \frac{\partial \mu(E)}{\partial E} \right]_{E=E_F}$$

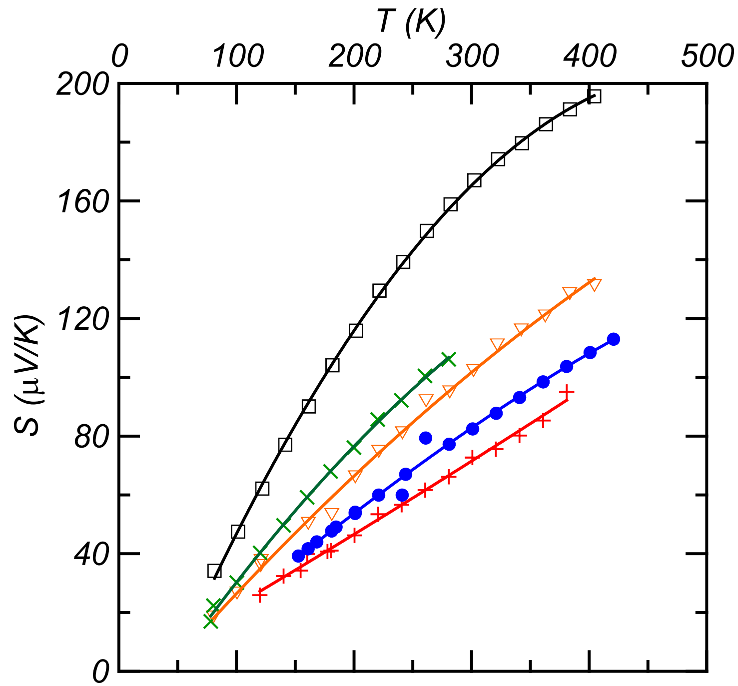
Not resonant scattering, need λ~4[1]







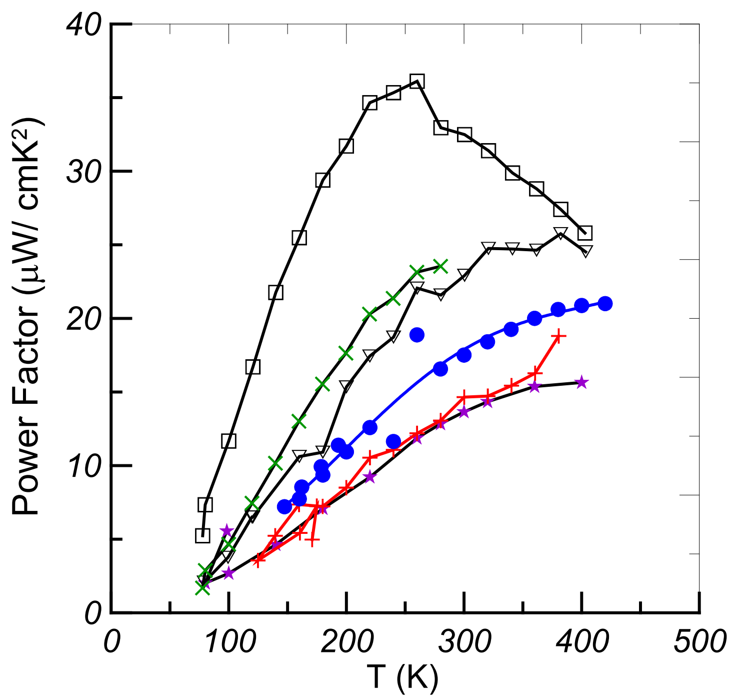
1. *physica status solidi (RRL)* -, Vol. 1, No. 6. (2007), pp. 256-258.

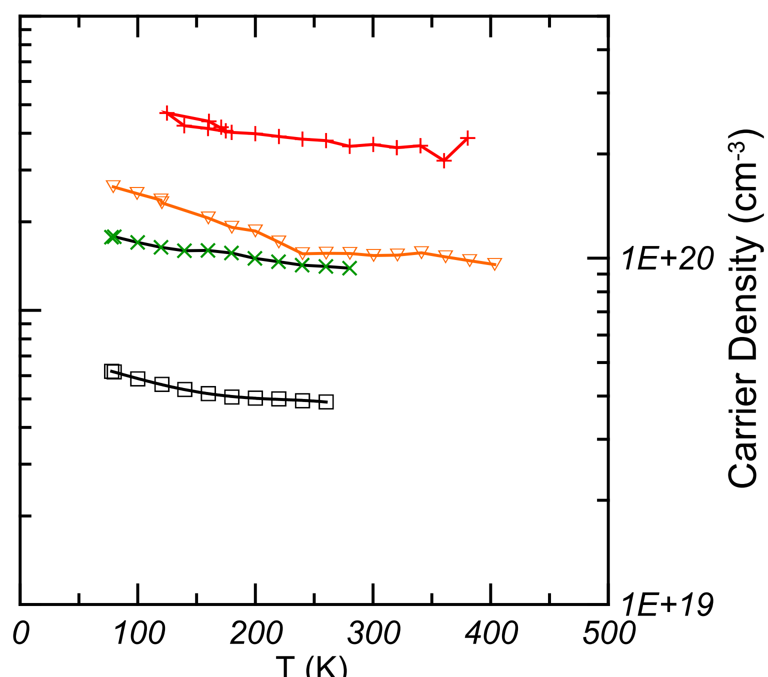


$(Bi_{.25}Sb_{.75})_{2-x}Sn_xTe_{3+\delta}$

Single Crystal

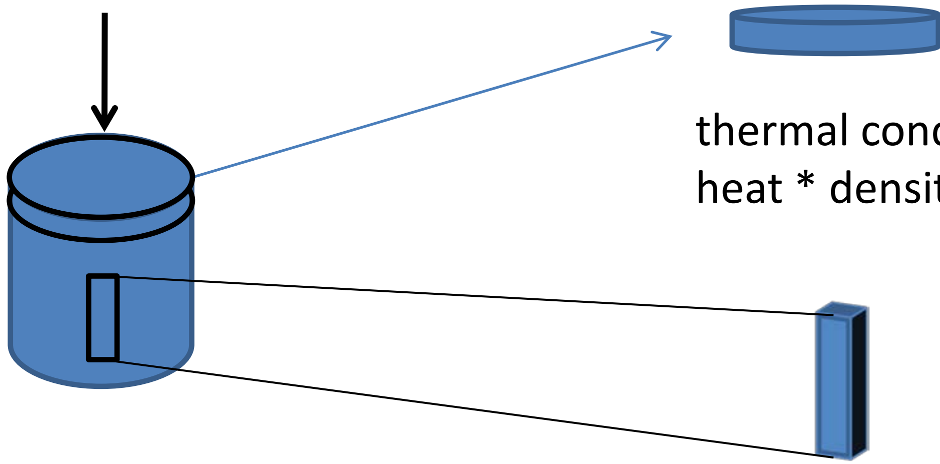
Carrier concentration too high





Powder Metallurgy: Measurement

All properties measured in same direction along pressing

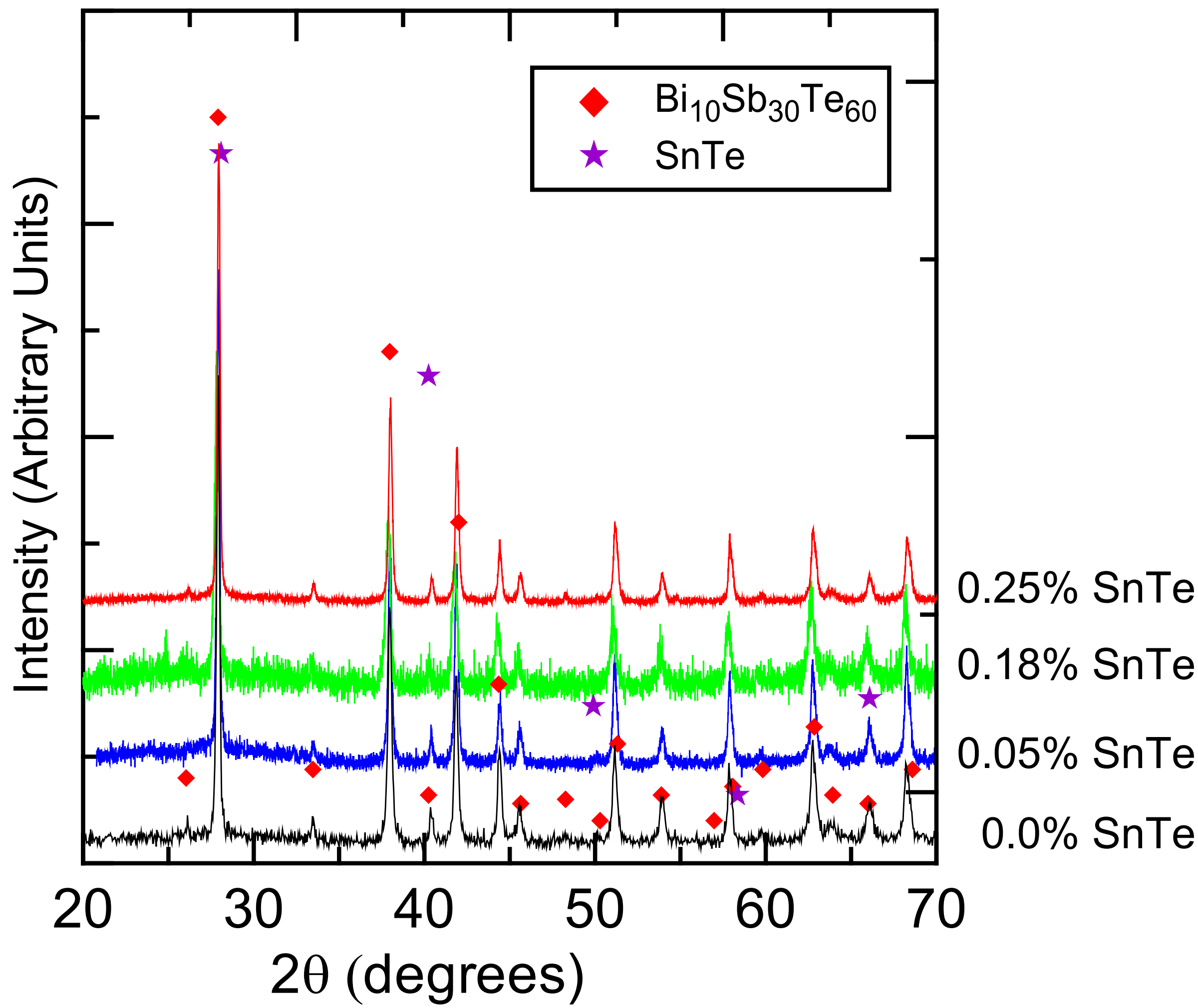


thermal conductivity = specific heat * density * flash diffusivity

- Thermopower
- electrical resistivity
- low temperature thermal conductivity
- Hall Coefficient
- Nernst Coefficient

No issue with anisotropy error in zT

Cold Press + Sintered $Bi_{10}Sb_{30}Te_{60}$ +$SnTe_x$



x= 0.05%, 0.10%, 0.18%, 0.25%

Inorg. Mater., vol: 22 pg: 515, (1986)
Inorg. Chem., vol: 36 pg: 260, (1997)

Idea: increase S by maximizing ionized impurity scattering

Seebeck, in general: $$S = \frac{k_B}{q}\frac{1}{\sigma}\int_0^{\infty}\sigma(E)\frac{(E-E_F)}{k_B T}\left(\frac{\partial f}{\partial E}\right)dE$$

Non-degenerate statistics: The Pisarenko relation

$$\tau = \tau_0 E^{\lambda}$$

$$S = \frac{k_B}{q}\left[\lambda + \frac{5}{2} + \ln\left(\frac{\frac{2}{h^3}\left(2\pi m_d^* k_B T\right)}{n}\right)\right]$$

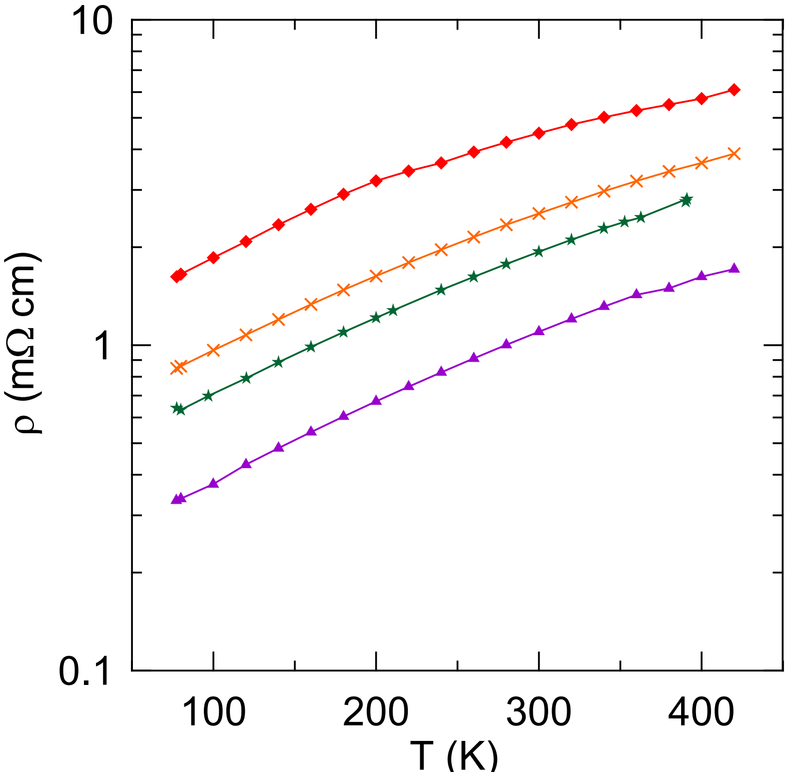
Works in bismuth[1], but will it increase power factor in BiSbTe?

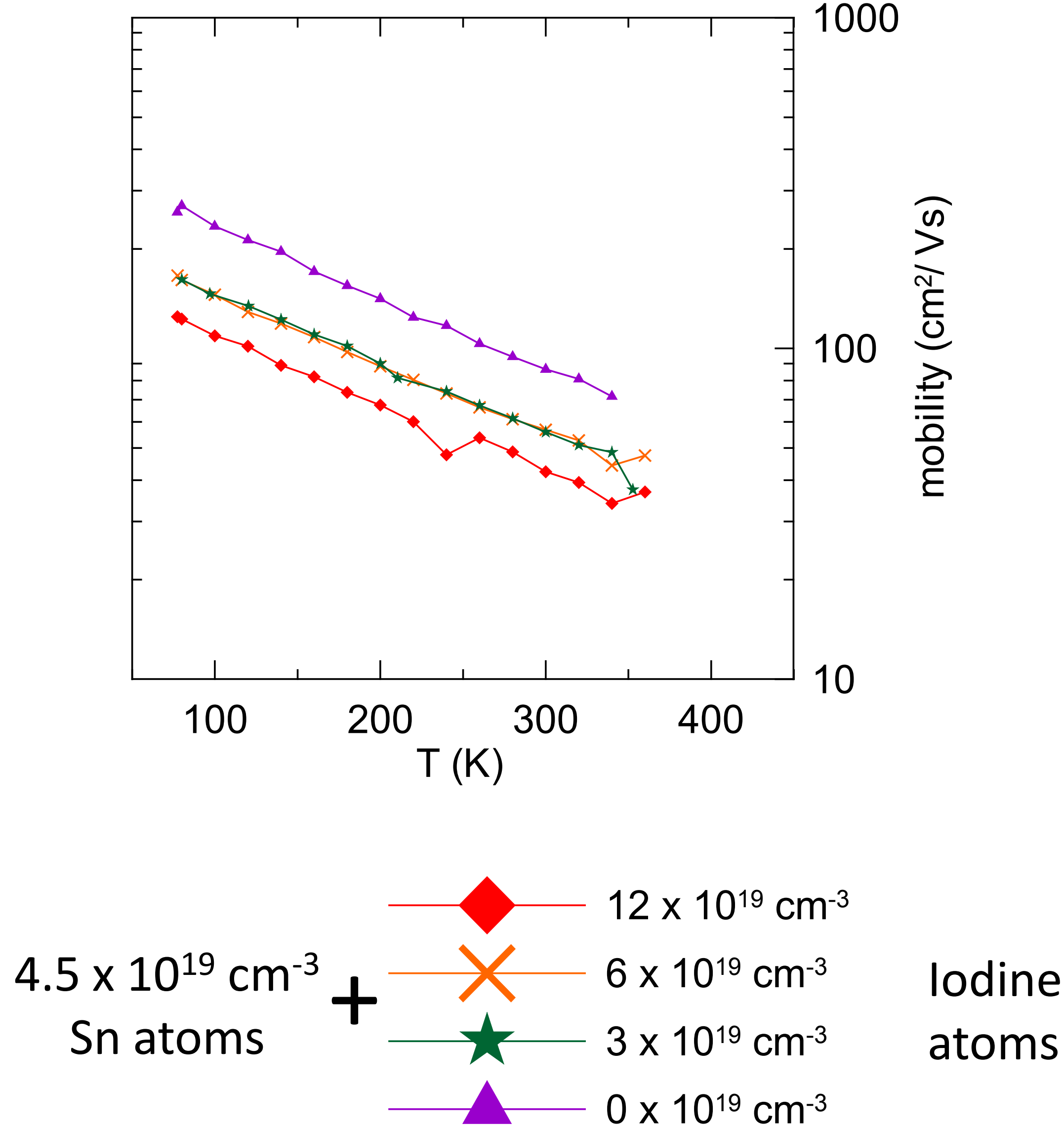
-Tradeoff between increased thermopower and decreased mobility

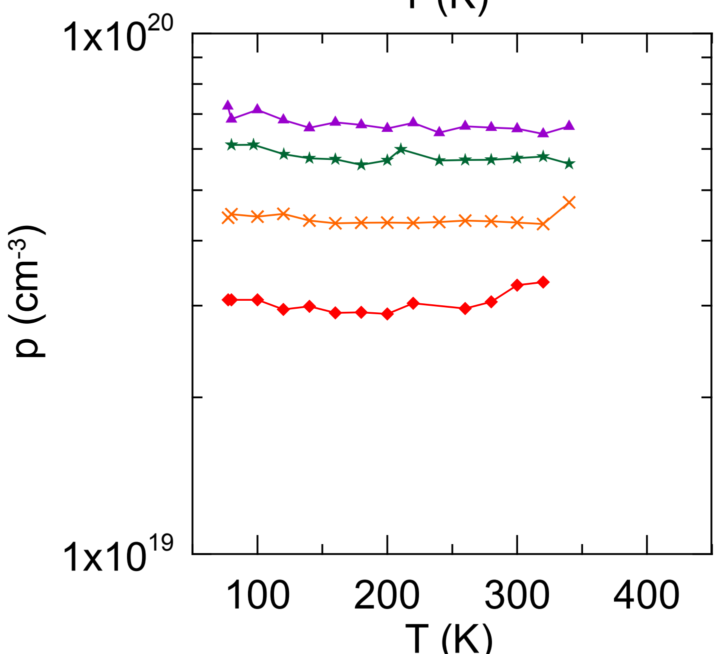
Add both tin and iodine to alloys

1. Heremans J.P. et al., Phys. Rev. B **14**, 5156 (1976)

SPS: $Bi_{.5}Sb_{1.5}Te_3$ + Tin + Iodine







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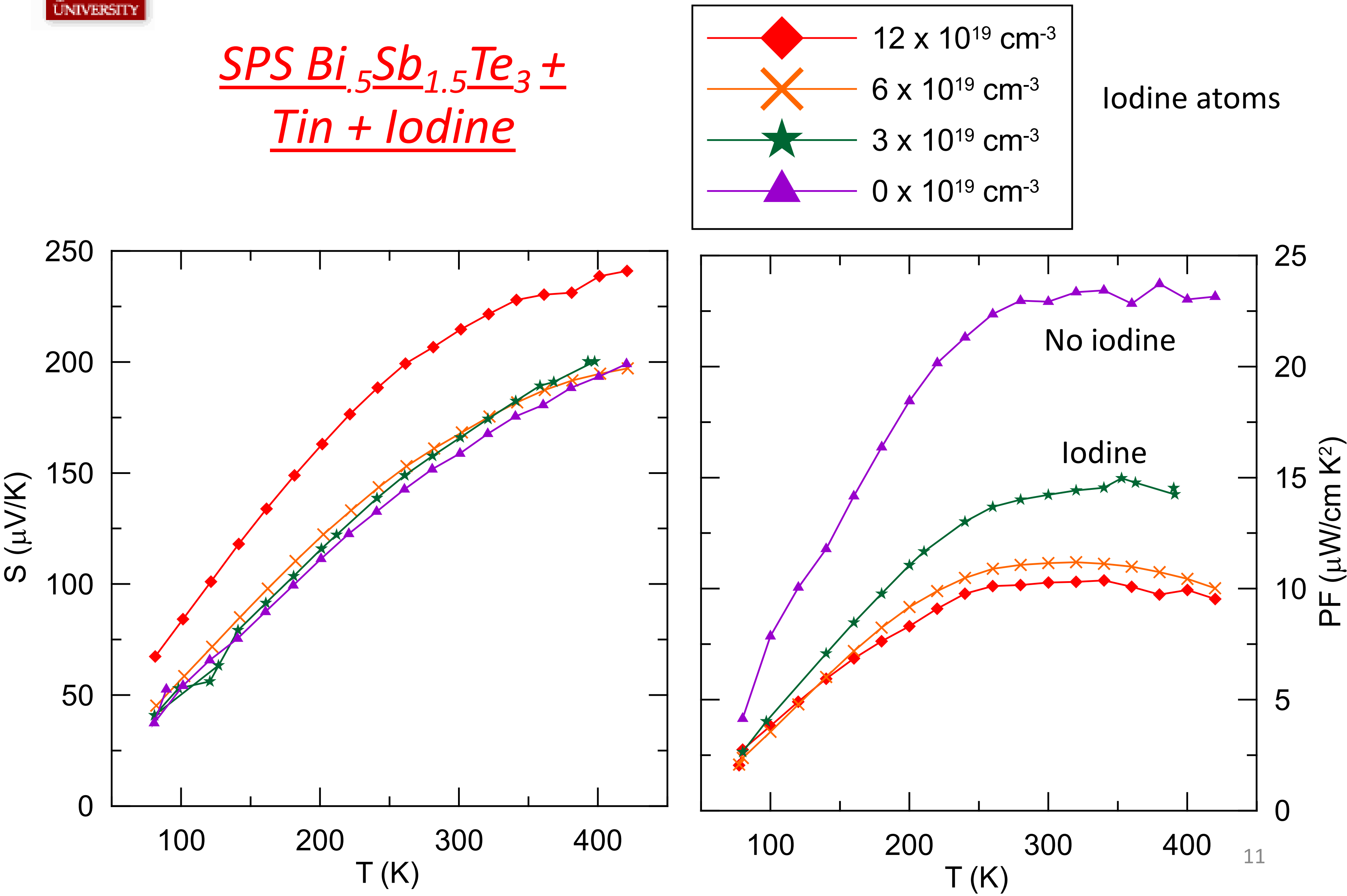

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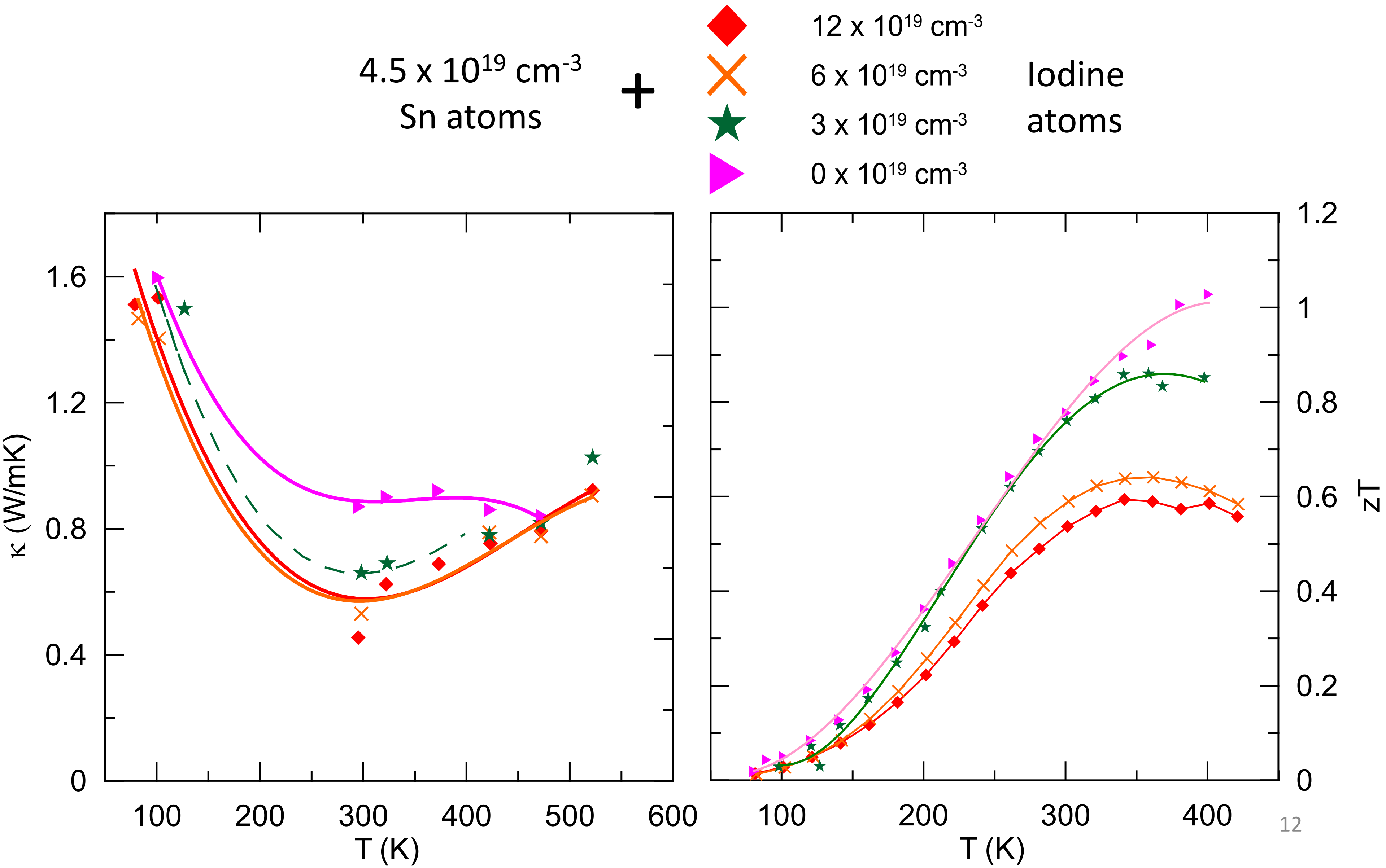

Summation

- Dilute amounts of Sn lead to high zT
- Influence of porosity needs to be further investigated
- Ionized impurity scattering decreases mobility more than increases thermopower
  - Bad for power factor