

## THERMOELECTRICITY: AB INITIO APPROACHES

Journées Nationales de la Thermoélectricité - 18/10/2018  
Ambroise van Roekeghem

- Ab initio calculations
- Lattice thermal conductivity
- Electronic properties
- High-throughput screening
- Machine learning

$$ZT = \frac{\sigma S^2 T}{\kappa}$$

# AB INITIO CALCULATIONS

$$\left\{ -\frac{\hbar^2}{2m} \sum_j \nabla_j^2 - \sum_{j,\ell} \frac{Z_\ell e^2}{|r_j - R_\ell|} + \frac{1}{2} \sum_{j \neq j'} \frac{e^2}{|r_j - r'_{j'}|} - E \right\} \Psi = 0$$

*Quantum Mechanics of Many-Electron Systems.*

By P. A. M. DIRAC, St. John's College, Cambridge.

(Communicated by R. H. Fowler, F.R.S.—Received March 12, 1929.)

## § 1. *Introduction.*

The general theory of quantum mechanics is now almost complete, the imperfections that still remain being in connection with the exact fitting in of the theory with relativity ideas. These give rise to difficulties only when high-speed particles are involved, and are therefore of no importance in the consideration of atomic and molecular structure and ordinary chemical reactions, in which it is, indeed, usually sufficiently accurate if one neglects relativity variation of mass with velocity and assumes only Coulomb forces between the various electrons and atomic nuclei. The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble. It therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation.

## DENSITY FUNCTIONAL THEORY

Finding and storing the many-body wave function  $\psi(r_1, \dots, r_N)$  is a problem that grows exponentially with N.

Hohenberg-Kohn theorem (1963) states that the groundstate density  $n(r)$  of interacting electrons in an external potential  $v(r)$  determines this potential uniquely.

This means that  $n(r)$  contains all the information about the Hamiltonian and thus about the ground state properties of the system.

Instead of trying to find the wave function that gives the minimum energy, we look for the density that minimizes the energy.

See Walter Kohn's Nobel (1998) lecture

# DENSITY FUNCTIONAL THEORY

The Kohn-Sham equations (1964)

$$\left( -\frac{1}{2} \nabla^2 + v_{eff}(r) - \epsilon_j \right) \varphi_j(r) = 0 ,$$

$$n(r) = \sum_{j=1}^N | \varphi_j(r) |^2 ,$$

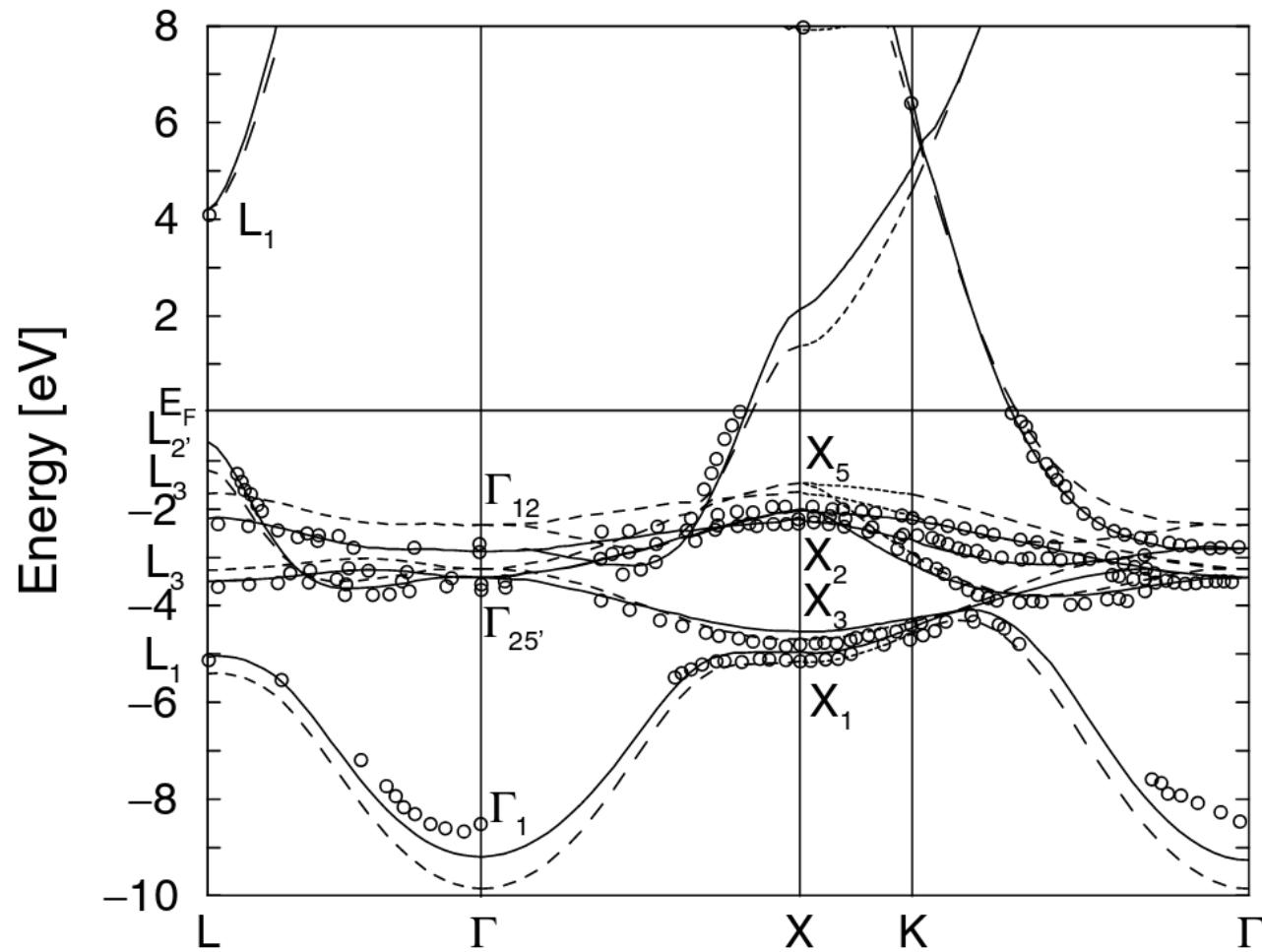
$$v_{eff}(r) = v(r) + \int \frac{n(r')}{| r - r' |} dr' + v_{xc}(r) \quad v_{xc}(r) \equiv \frac{\delta}{\delta \tilde{n}(r)} E_{xc}[\tilde{n}(r)] |_{\tilde{n}(r)=n(r)}$$

allow to find the ground-state density using independent electrons, provided that the exchange-correlation potential is known exactly.

One of the most successful exchange-correlation functionals is based on the local density approximation:  $v_{xc}(r) = v_{xc}(n(r))$

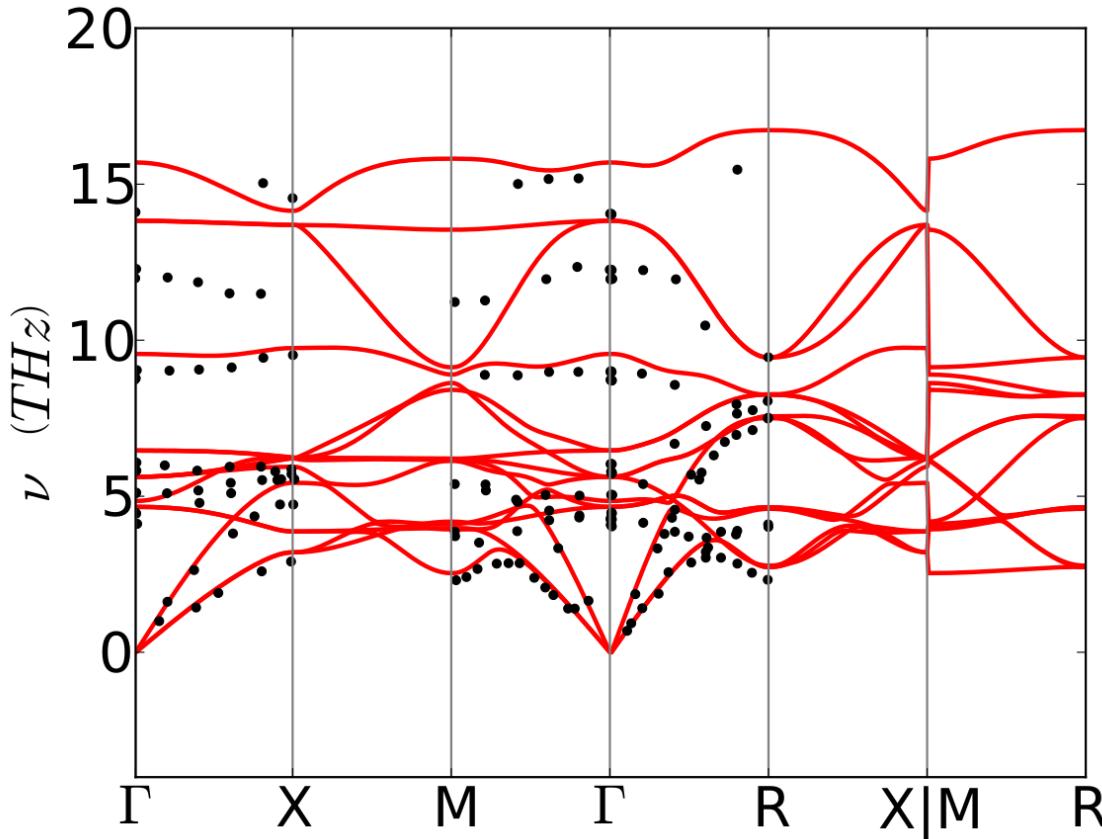
DFT-based programs are now some of the most used tools in computational solid state physics.

## DENSITY FUNCTIONAL THEORY



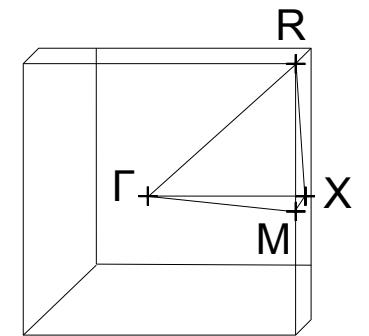
Marini et al, Phys. Rev. Lett. 88, 016403 (2001)

## PHONONS



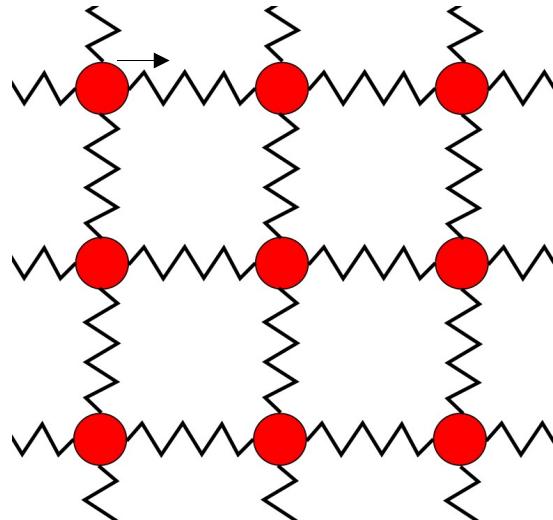
Lattice vibrations

→ Wavelength and energy



# COMPUTING PHONONS

Small displacements



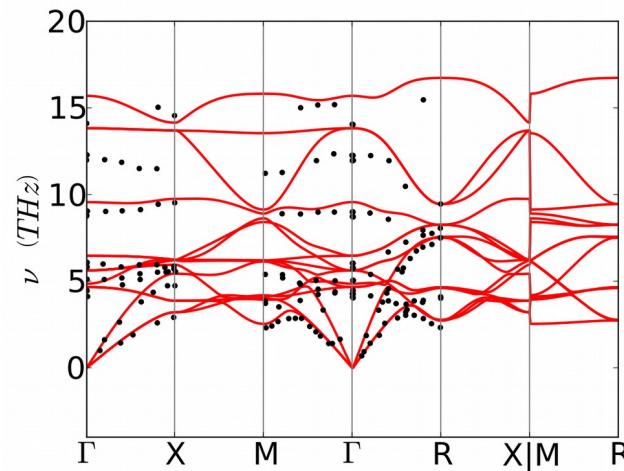
$$F_i^\alpha = \sum_{j\beta} \Phi_{ij}^{\alpha\beta} u_j^\beta + \frac{1}{2} \sum_{jk\beta\gamma} \Psi_{ijk}^{\alpha\beta\gamma} u_j^\beta u_k^\gamma$$

→ 2<sup>nd</sup> and 3<sup>rd</sup> order  
force constants

→ Phonon spectrum  
And thermal conductivity

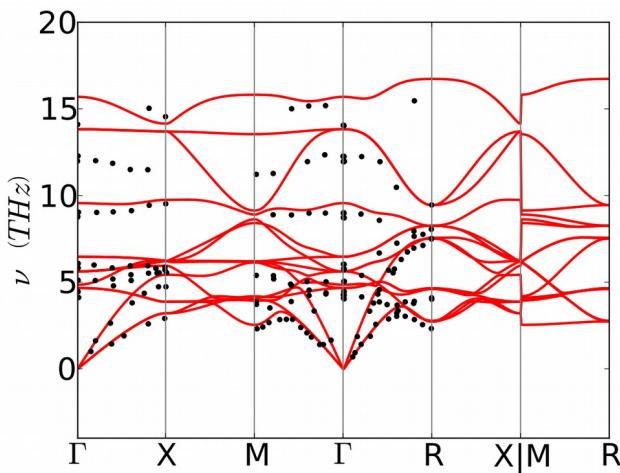
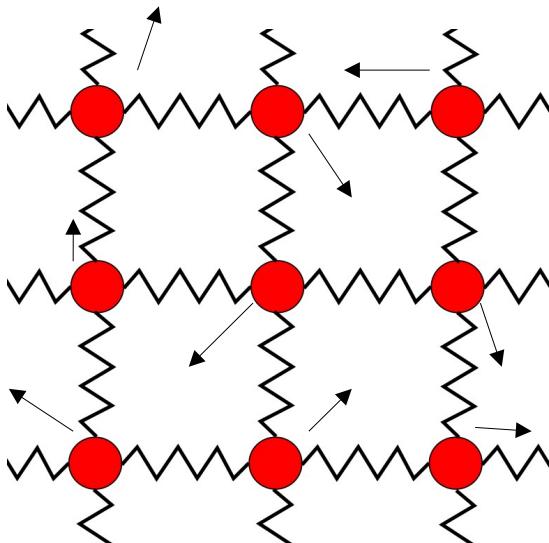
$$D_{ij}(k) = \sum_R \frac{\Phi_{Ri,0j}}{\sqrt{M_i M_j}} e^{ikR}$$

$$\sum_j D_{ij}(k) e_j^n(k) = \omega_{nk}^2 e_i^n(k)$$



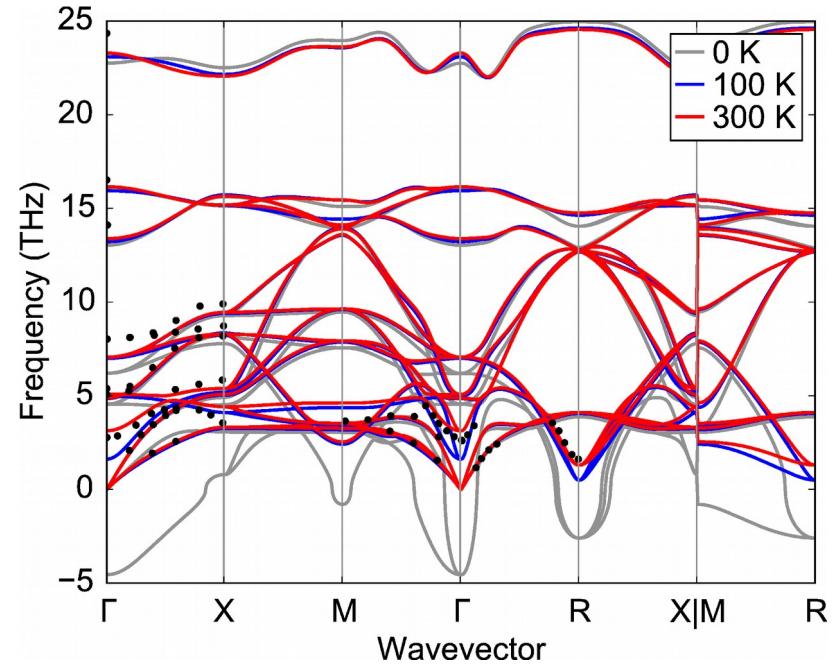
# COMPUTING PHONONS

Quantum statistics, finite T



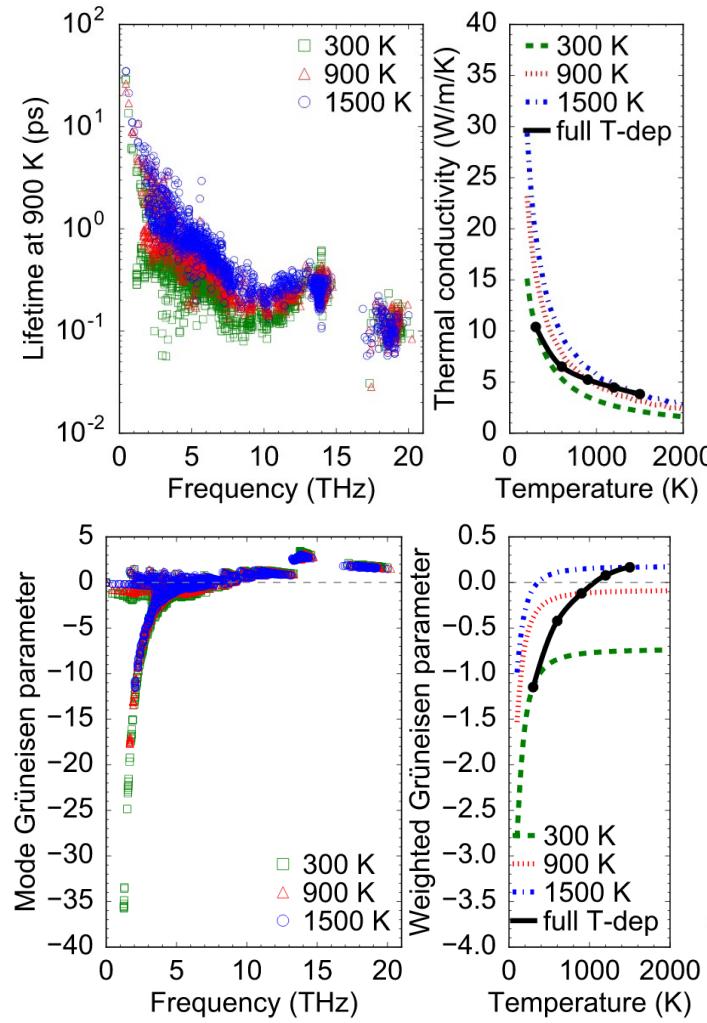
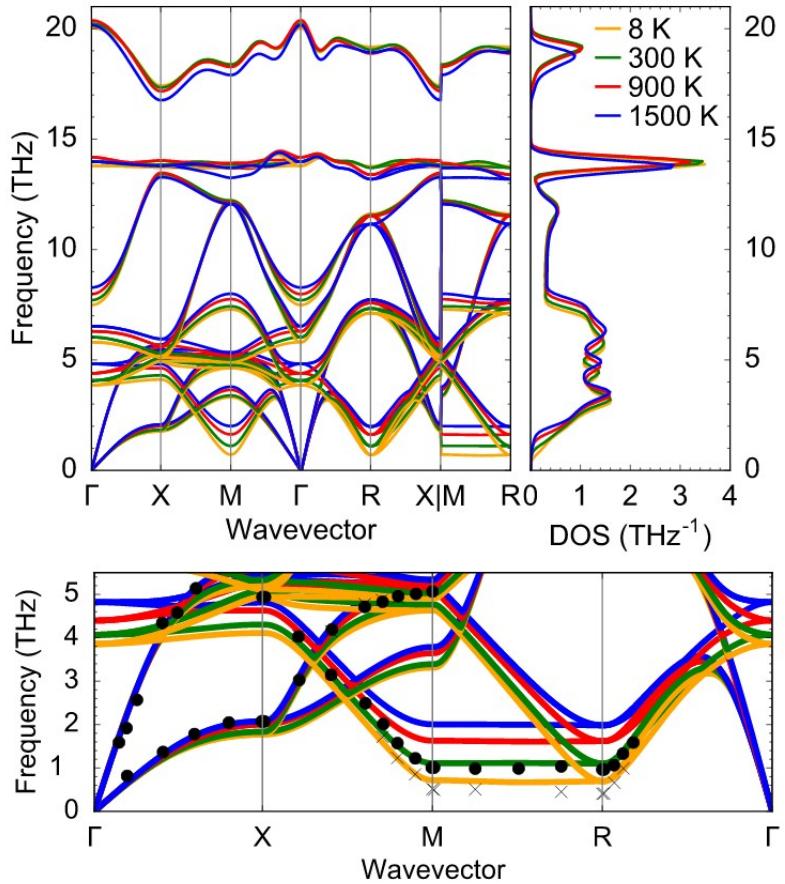
$$\rho_h(\{u_{i\alpha}\}) \propto \exp(-\frac{1}{2}u^T \Sigma^{-1} u)$$

$$\Sigma(i\alpha, j\beta) = \frac{\hbar}{2\sqrt{M_i M_j}} \sum_m \omega_m^{-1} [1 + 2n_B(\omega_m)] \epsilon_{mi\alpha} \epsilon_{mj\beta}^*$$



Exp. data at 300 K: Cowley, Phys. Rev. 134, A981 (1964)  
and Stirling, J. Phys. C: Solid State Physics 5, 2711 (1972)

# FINITE-TEMPERATURE PHONON CALCULATIONS



$$\kappa \propto T^{-0.6}$$

$$\gamma_i = -\frac{V}{\omega_i} \frac{\partial \omega_i}{\partial V}$$

$$\gamma = \frac{\sum \gamma_i c_{vi}}{\sum c_{vi}}$$

$$\alpha_V = \gamma \frac{C_v \rho}{K_T}$$

$$\gamma_m = -\frac{1}{6\omega_m^2} \sum_{ijk\alpha\beta\gamma} \frac{\epsilon_{mi\alpha}^* \epsilon_{mj\beta}}{\sqrt{M_i M_j}} r_k^\gamma \Psi_{ijk}^{\alpha\beta\gamma} e^{i\mathbf{q}\cdot\mathbf{r}_j}$$

van Roekeghem, Carrete and Mingo, Physical Review B 94, 020303(R) (2016)

# BOLTZMANN'S TRANSPORT EQUATION

$$J^\alpha = - \sum_\beta \kappa^{\alpha\beta} (\nabla T)^\beta$$

$$\mathbf{J} = \sum_p \int f_\lambda \hbar \omega_\lambda \mathbf{v}_\lambda \frac{d\mathbf{q}}{(2\pi)^3}$$

$$\frac{df_\lambda}{dt} = \left. \frac{\partial f_\lambda}{\partial t} \right|_{\text{diffusion}} + \left. \frac{\partial f_\lambda}{\partial t} \right|_{\text{scattering}} = 0$$

$$\left. \frac{\partial f_\lambda}{\partial t} \right|_{\text{diffusion}} = -\nabla T \cdot \mathbf{v}_\lambda \frac{\partial f_\lambda}{\partial T}$$

$$f = f_0 - \frac{C}{\hbar\omega} v_g \tau_{eff} \frac{dT}{dx} \quad \quad \kappa = \int C v_g^2 \tau_{eff}$$

$$1/\tau_\lambda^0 \equiv \sum_{\lambda' \lambda''}^+ \Gamma_{\lambda \lambda' \lambda''}^+ + \sum_{\lambda' \lambda''}^- \frac{1}{2} \Gamma_{\lambda \lambda' \lambda''}^- + \sum_{\lambda'} \Gamma_{\lambda \lambda'}$$

$$\Gamma_{\lambda \lambda' \lambda''}^+ = \frac{\hbar \pi}{4} \frac{f'_0 - f''_0}{\omega_\lambda \omega_{\lambda'} \omega_{\lambda''}} |V_{\lambda \lambda' \lambda''}^+|^2 \delta(\omega_\lambda + \omega_{\lambda'} - \omega_{\lambda''})$$

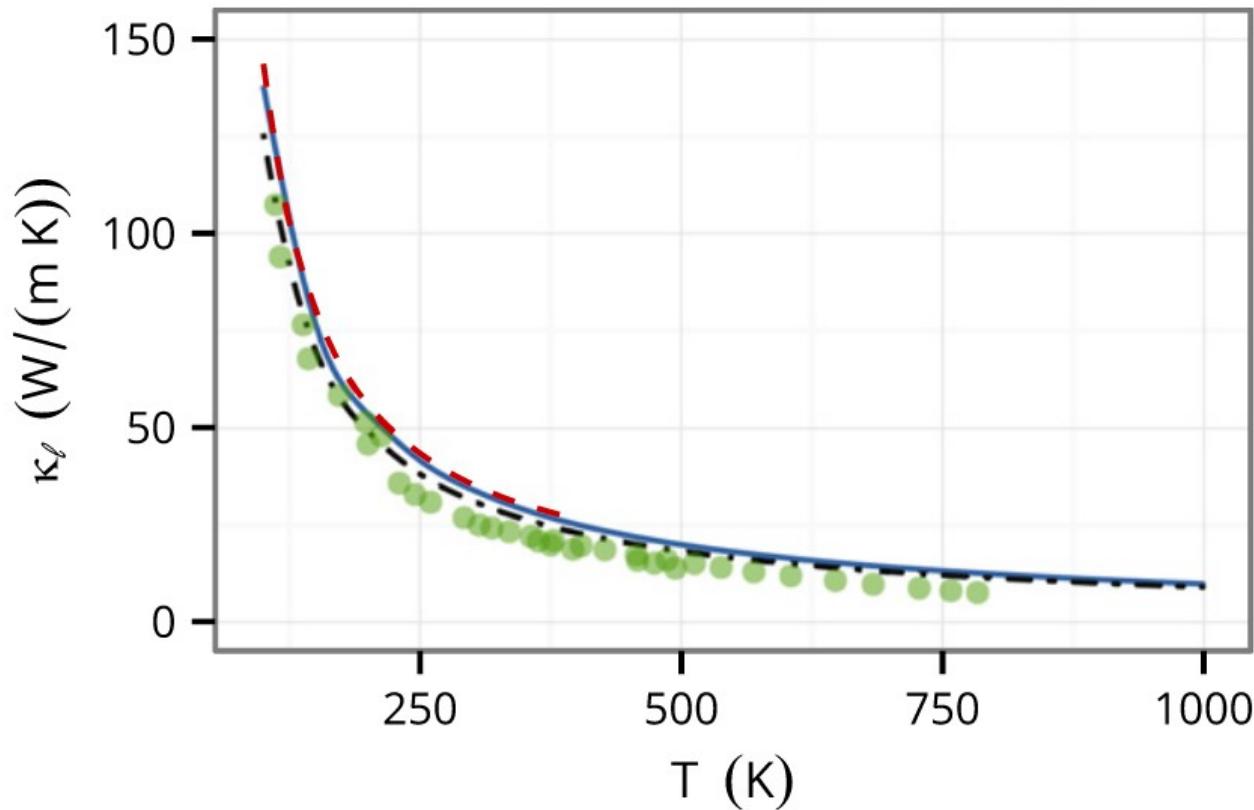
$$\Gamma_{\lambda \lambda' \lambda''}^- = \frac{\hbar \pi}{4} \frac{f'_0 + f''_0 + 1}{\omega_\lambda \omega_{\lambda'} \omega_{\lambda''}} |V_{\lambda \lambda' \lambda''}^-|^2 \delta(\omega_\lambda - \omega_{\lambda'} - \omega_{\lambda''})$$

$$\Gamma_{\lambda \lambda'} = \frac{\pi \omega^2}{2} \sum_{i \in \text{u.c.}} g(i) |\mathbf{e}_\lambda^*(i) \cdot \mathbf{e}_{\lambda'}(i)|^2 \delta(\omega_\lambda - \omega_{\lambda'})$$

$$V_{\lambda \lambda' \lambda''}^\pm = \sum_{i \in \text{u.c.}} \sum_{j,k} \sum_{\alpha \beta \gamma} \Phi_{ijk}^{\alpha \beta \gamma} \frac{e_\lambda^\alpha(i) e_{p', \pm \mathbf{q}'}^\beta(j) e_{p'', -\mathbf{q}''}^\gamma(k)}{\sqrt{M_i M_j M_k}}$$

$$g(i) = \sum_s f_s(i) [1 - M_s(i) / \overline{M}(i)]^2$$

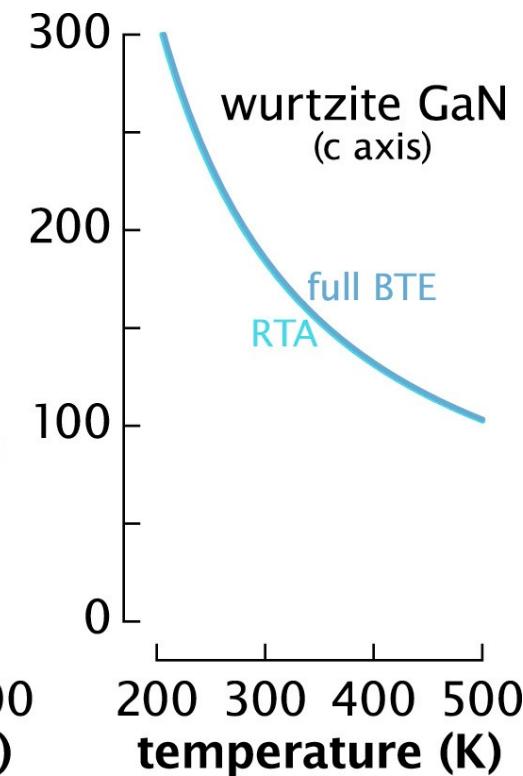
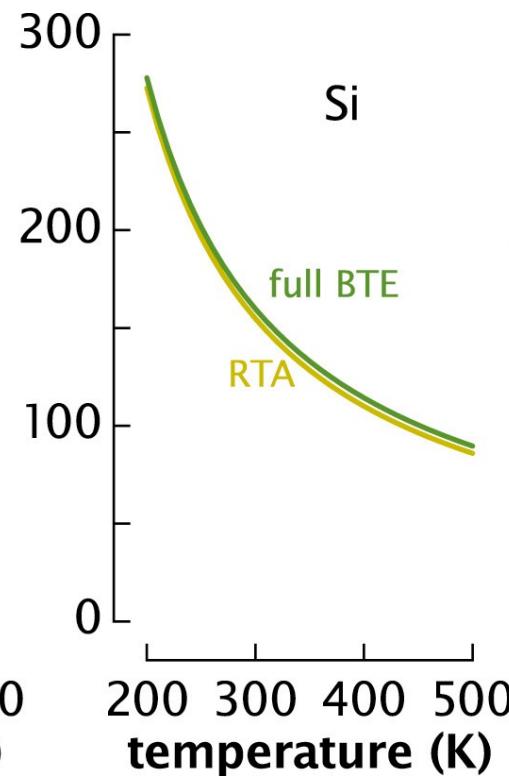
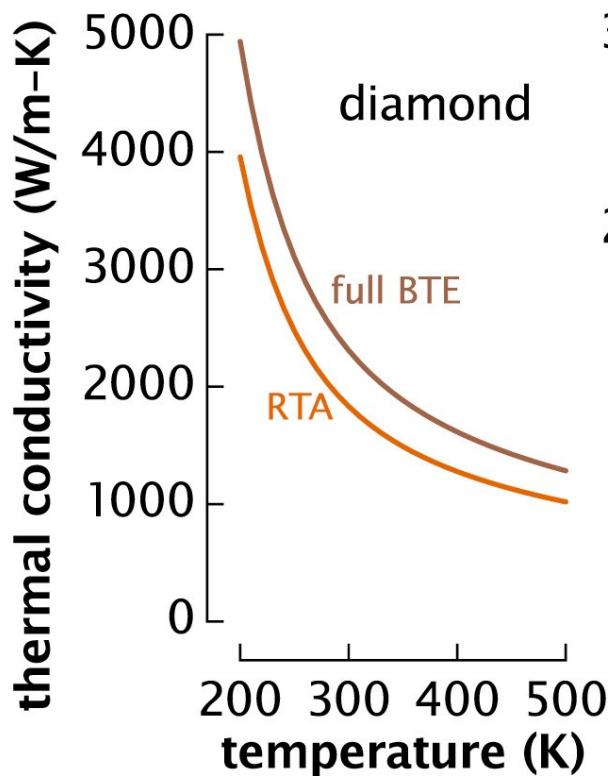
# BOLTZMANN'S TRANSPORT EQUATION



ShengBTE: Computer Physics Communications 185, 1747 (2014)

# BOLTZMANN'S TRANSPORT EQUATION

$$\tau_\lambda = \tau_\lambda^0 + \tau_\lambda^0 \Delta_\lambda$$



ShengBTE: Computer Physics Communications 185, 1747 (2014)

**homogeneous materials**

Single crystals  
Alloys

Steady state  
and transient  
heat flow

**with defects**

Vacancies  
Substitutionals  
Interstitials  
Dislocations

**nanostructures**

Superlattices  
Thin films

**multiscale structures**

1D multilayers  
2D and 3D  
Interfaces

J. Carrete *et al.*, “almaBTE : A solver of the space–time dependent Boltzmann transport equation for phonons in structured materials”, *Computer Physics Communications*, vol. 220, pp. 351–362, Nov. 2017. [www.almabte.eu](http://www.almabte.eu)

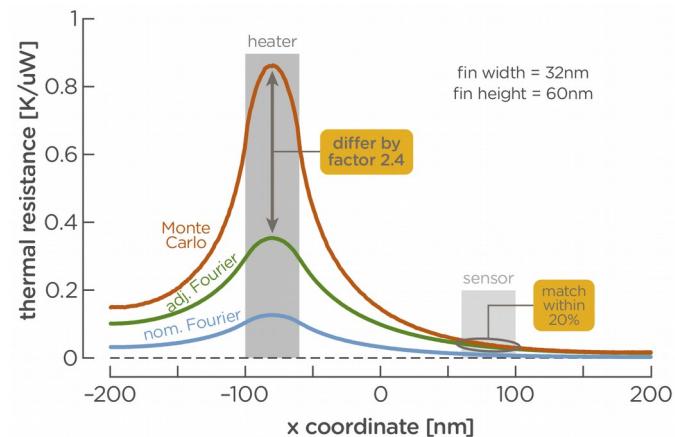
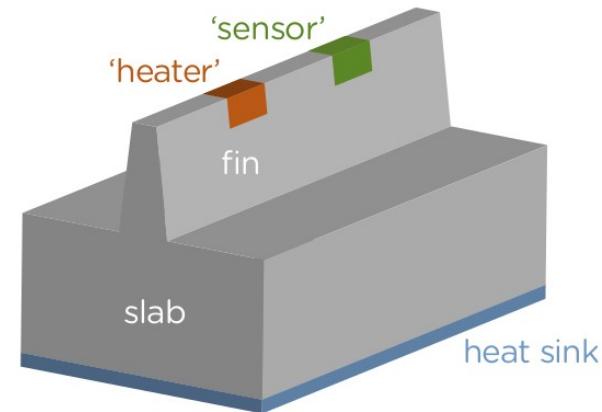
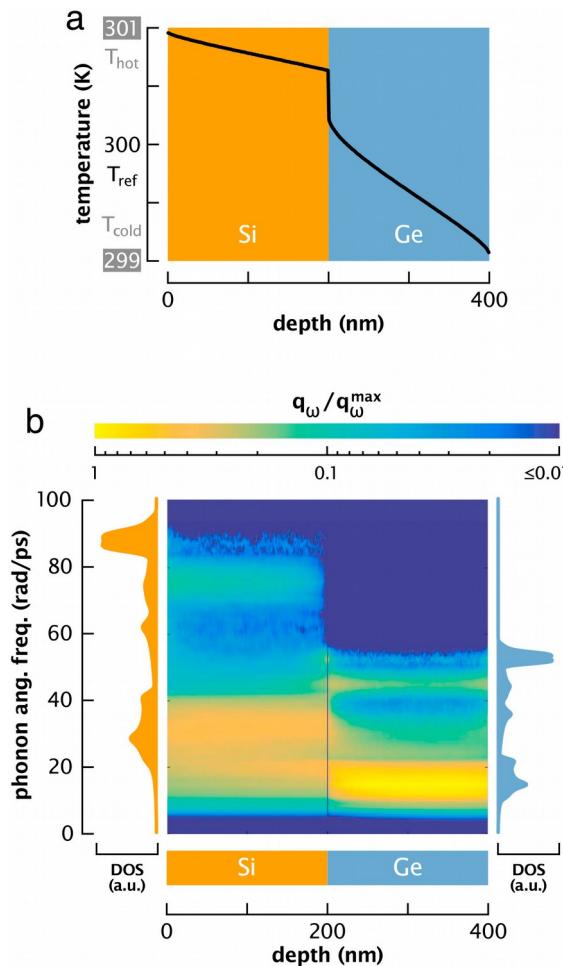
6 partners from academia to industry

13 citations in less than one year

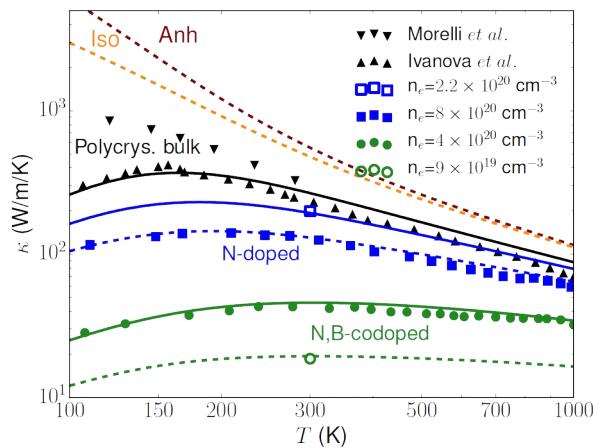
88 members on the online forum

31 related publications from our group since 2016

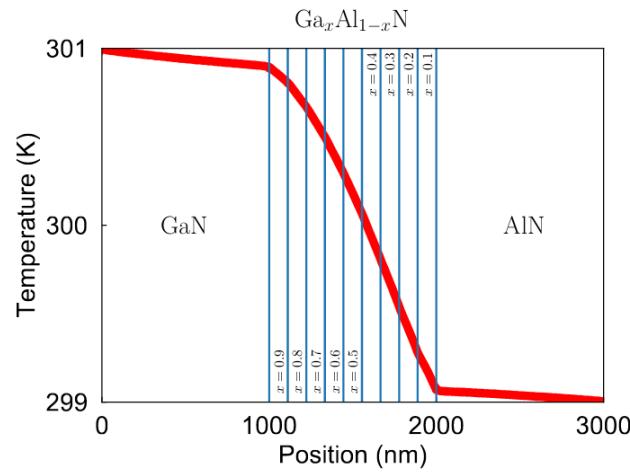
(including 1 Adv. Mat., 1 Phys. Rev. X, 1 Phys. Rev. Lett., 9 Phys. Rev. B, 1 Phys. Rev. Mat.)



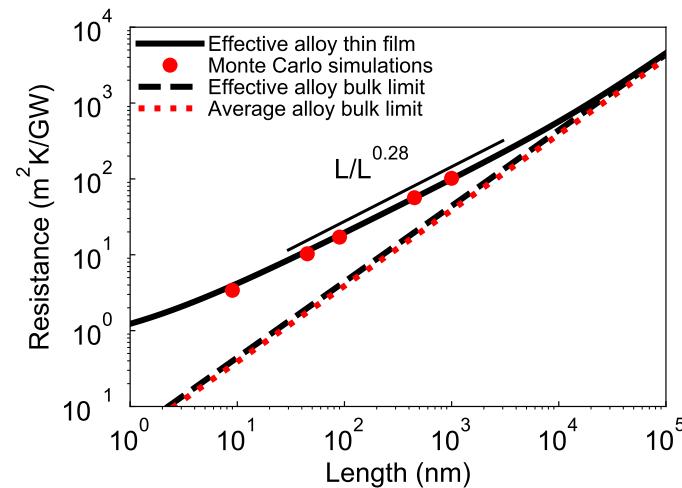
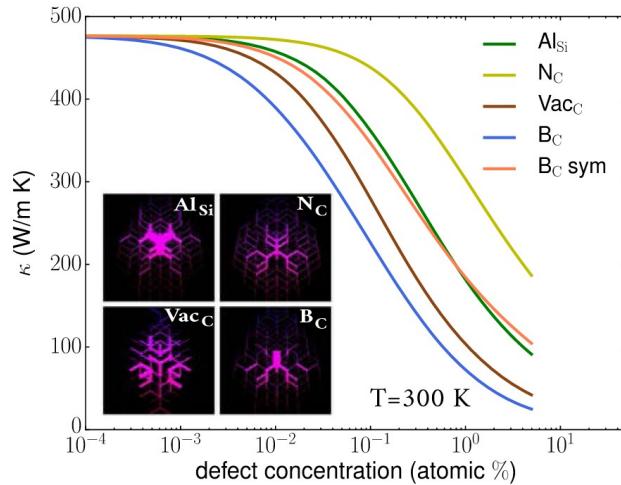
Vermeersch et al, unpublished



Katre et al, Phys. Rev. Lett. 119, 075902 (2017)



van Roekeghem et al, arXiv:1809.11046 (2018)



# ONSAGER'S COEFFICIENTS AND THE BTE

Linearized Boltzmann's transport equation in the relaxation time approximation

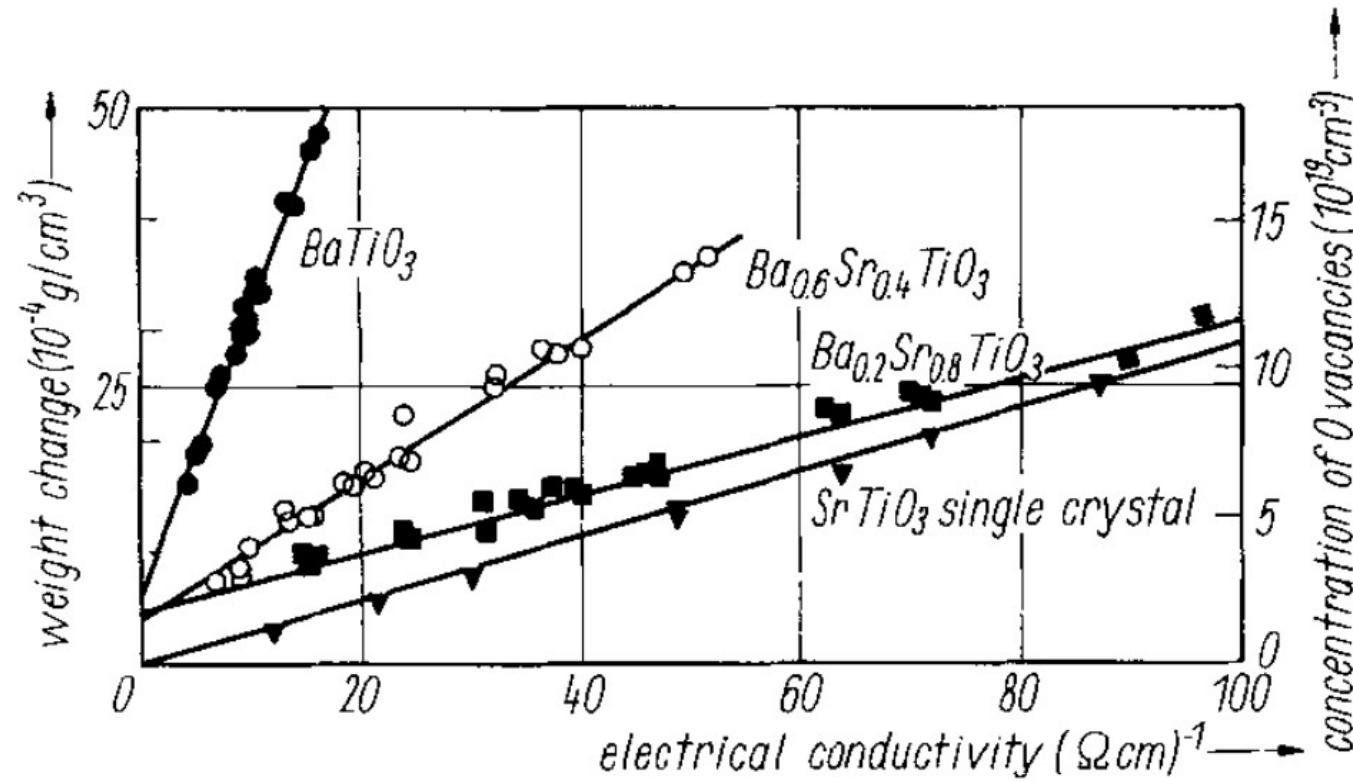
$$\begin{aligned} j_e &= \mathcal{L}^{(0)} \mathbf{E} + \frac{\mathcal{L}^{(1)}}{qT} (-\nabla T) & \sigma &= \mathcal{L}^{(0)} & S &= \frac{1}{qT} \frac{\mathcal{L}^{(1)}}{\mathcal{L}^{(0)}} \\ j_Q &= \frac{\mathcal{L}^{(1)}}{q} \mathbf{E} + \frac{\mathcal{L}^{(2)}}{q^2 T} (-\nabla T) & \Pi &= \frac{\mathcal{L}^{(1)}}{q \mathcal{L}^{(0)}} \end{aligned}$$

$$\begin{aligned} \sigma(\varepsilon, T) &= \int \sum_h \mathbf{v}_{b,\mathbf{k}} \otimes \mathbf{v}_{b,\mathbf{k}} \tau_{b,\mathbf{k}} \delta(\varepsilon - \varepsilon_{b,\mathbf{k}}) \frac{d\mathbf{k}}{8\pi^3} \\ \mathcal{L}^{(\alpha)}(\mu; T) &= q^2 \int \sigma(\varepsilon, T) (\varepsilon - \mu)^\alpha \left( -\frac{\partial f^{(0)}(\varepsilon; \mu, T)}{\partial \varepsilon} \right) d\varepsilon \end{aligned}$$

(See Electrons and Phonons by Ziman, Ashcroft-Mermin, Boltztrap papers)

# ELECTRON SCATTERING

- Electron-phonon scattering
- Impurity scattering
- Electron-electron scattering

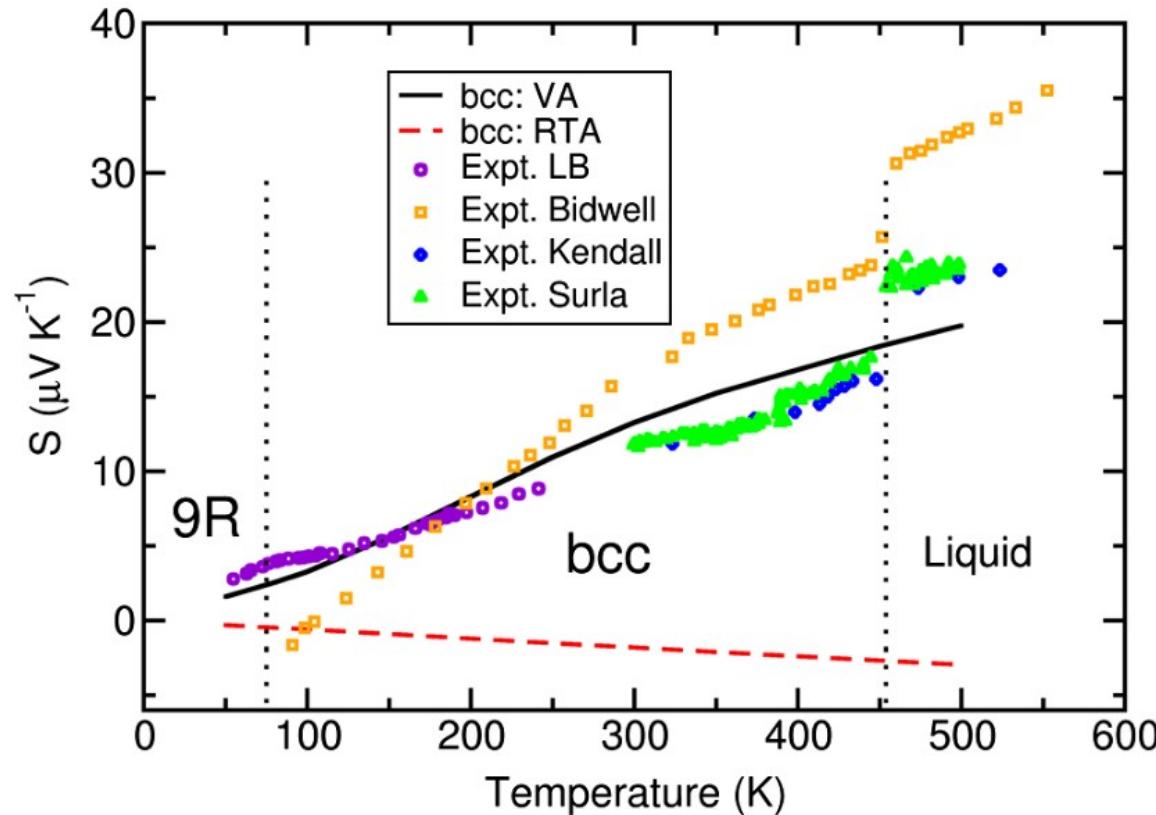


Gerthsen et al, Phys. Stat. Sol. (a) 13, 127 (1972)

# COMPUTING THE SEEBECK COEFFICIENT

Constant relaxation time approximation

→ S becomes independent of  $\tau$  and  $\sigma/\tau$  becomes constant



Xu and Verstraete, Phys. Rev. Lett. 112, 196603 (2014)

# NANOGRAIN LIMIT

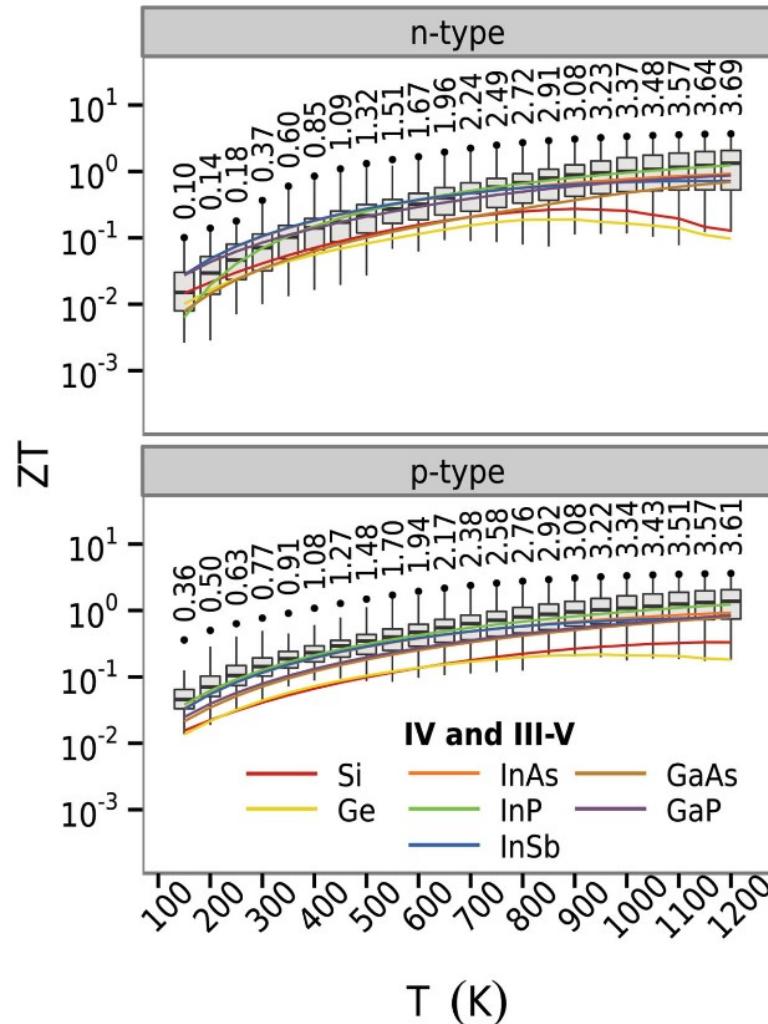
$$ZT = \frac{\sigma S^2 T}{\kappa}$$

$$\sigma = \lambda \frac{e^2}{k_B T} M_0^{(\text{FD})}$$

$$S = \frac{\lambda}{\sigma} \frac{e}{k_B T^2} M_1^{(\text{FD})}$$

$$\kappa_e = \lambda \frac{1}{k_B T^2} M_2^{(\text{FD})} - \sigma S^2 T$$

$$\kappa_I = \lambda \frac{1}{k_B T^2} M_2^{(\text{BE})}$$



Carrete et al, Advanced Functional Materials 24, 7427 (2014)

$$\sigma = \mathcal{L}^{(0)} \quad S = \frac{1}{qT} \frac{\mathcal{L}^{(1)}}{\mathcal{L}^{(0)}}$$

+ Sommerfeld development

→ Mott's formula:  $S = -\frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\partial \ln \sigma}{\partial \mu} = \frac{\pi^2}{3} \frac{k_B^2 T}{e\rho} \frac{\partial \rho}{\partial \mu}$

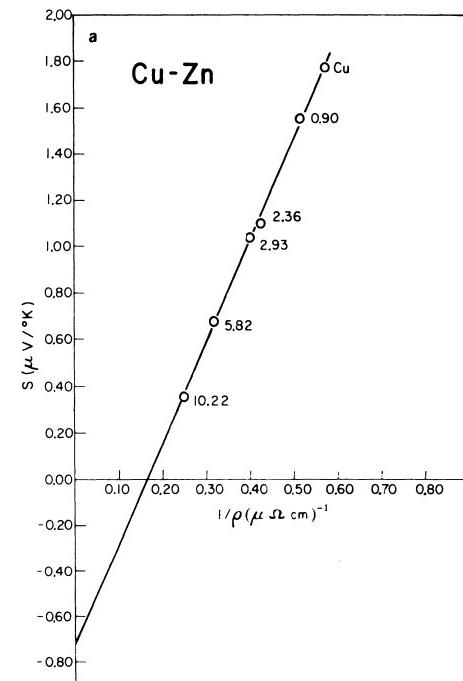
Multicomponent alloys and Matthiessen's rule:

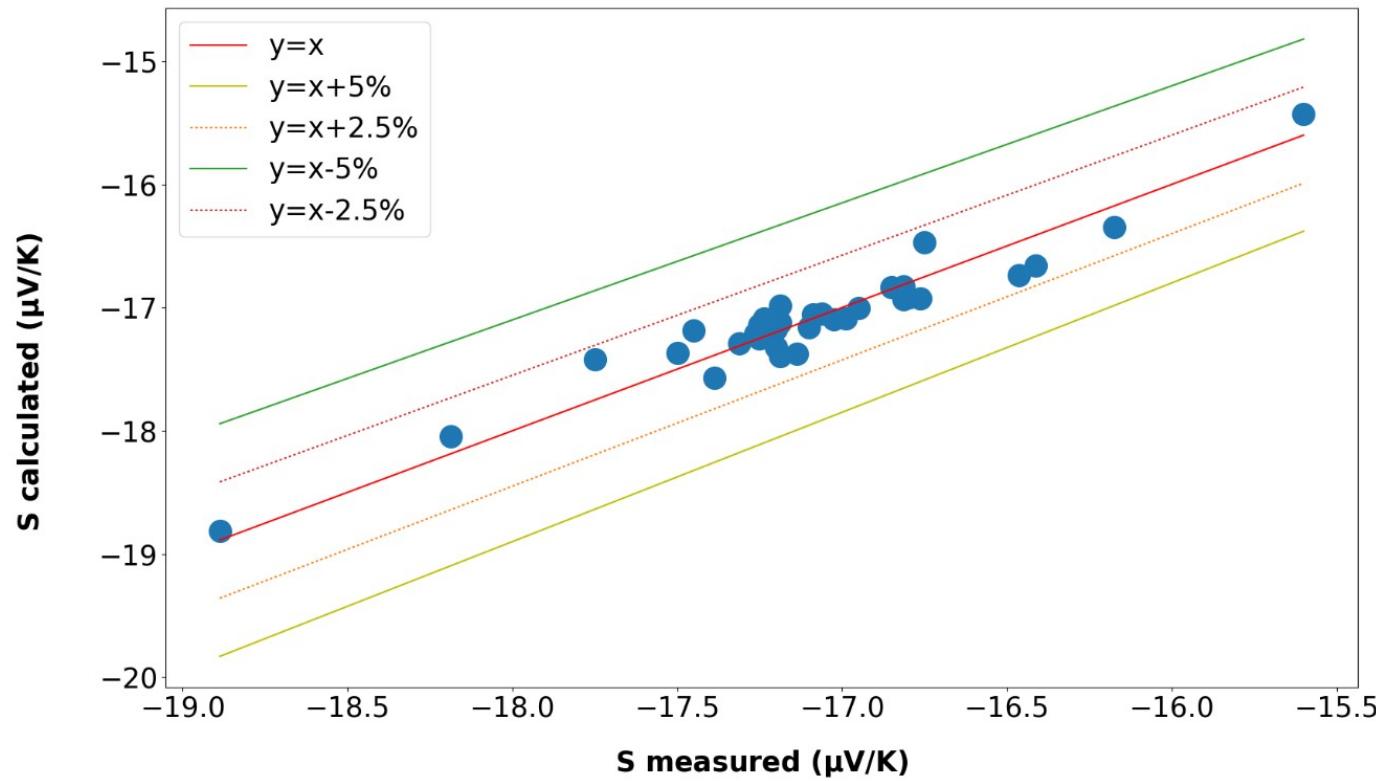
$$\rho = \rho_0 + \sum_j \rho_j = \rho_0 + \sum_j x_j r_j$$

$$S = \frac{\rho_0}{\rho} S_0 + \sum_j \frac{\rho_j}{\rho} S_j$$

→ Gorter-Nordheim formula and plots

See Thermoelectric Power of Metals,  
Blatt, Schroeder, Foiles and Greig

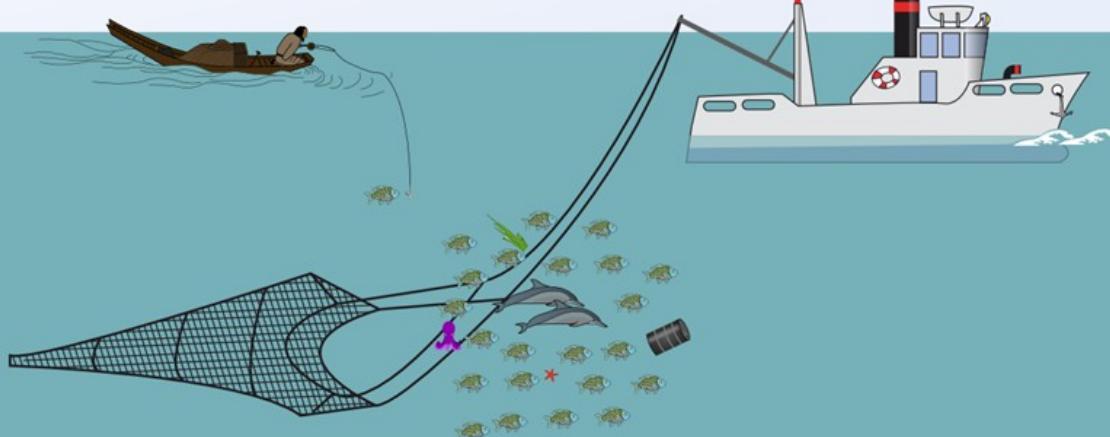




PhD Maximilien Saint-Cricq

# HIGH-THROUGHPUT SCREENING

Traditional approaches



High throughput



Energy &  
Environmental Science

## MATERIALS PROJECT

RSC Publishing

PAPER

[View Article Online](#)  
[View Journal](#) | [View Issue](#)

First principles high throughput screening of oxynitrides  
for water-splitting photocatalysts

Cite this: *Energy Environ. Sci.*, 2013, 6,

157

Yabi Wu,<sup>a</sup> Predrag Lazić,<sup>a</sup> Geoffroy Hautier,<sup>t,b</sup> Kristin Persson<sup>b</sup> and Gerbrand Ceder<sup>a\*</sup>



**AFLOW**  
Automatic - FLOW for Materials Discovery

**REVIEW ARTICLE**

PUBLISHED ONLINE: 20 FEBRUARY 2013 | DOI: 10.1038/NMAT3568

The high-throughput highway to computational  
materials design

Stefano Curtarolo<sup>1,2\*</sup>, Gus L. W. Hart<sup>2,3</sup>, Marco Buongiorno Nardelli<sup>2,4,5</sup>, Natalio Mingo<sup>2,6</sup>,  
Stefano Sanvito<sup>2,7</sup> and Ohad Levy<sup>1,2,8</sup>

**ARTICLES**

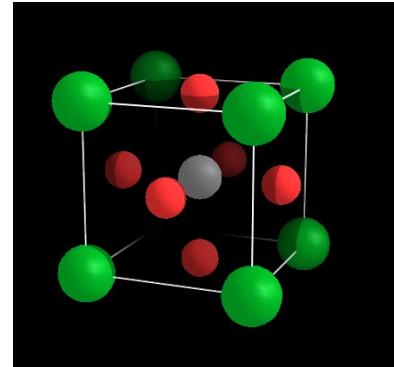
PUBLISHED ONLINE: 24 MARCH 2015 | DOI: 10.1038/NCHEM.2207

Prediction and accelerated laboratory discovery of  
previously unknown 18-electron ABX compounds

Romain Gautier<sup>1\*</sup>, Xiuwen Zhang<sup>2\*</sup>, Linhua Hu<sup>2</sup>, Liping Yu<sup>2</sup>, Yuyuan Lin<sup>1</sup>, Tor O. L. Sunde<sup>1</sup>,  
Danhee Choi<sup>1</sup>, Kenneth R. Poeppelmeier<sup>1\*</sup> and Alex Zunger<sup>2\*</sup>

# HIGH-THROUGHPUT SCREENING

$\text{ABX}_3$  with X=O or F



H

Li Be

Na Mg

K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Br Kr

Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd In Sn Sb Te I Xn

Cs Ba La Hf Ta W Re Os Ir Pt Au Hg Tl Pb Bi Po At Rn

He

B C N O F Ne

Al Si P S Cl Ar

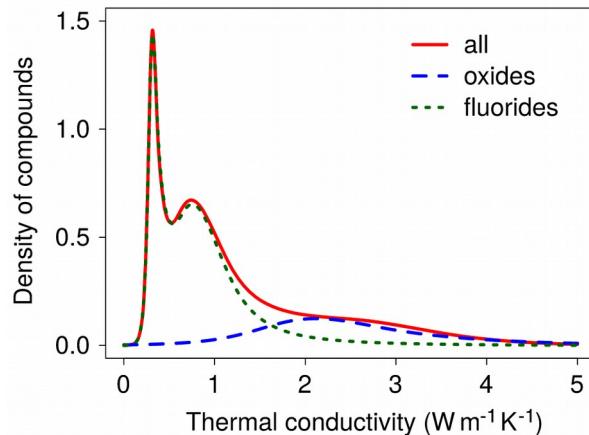
- 8000 possible combinations
- 400 non-magnetic semi-conductors
- 90 found mechanically stable at 1000 K
  
- 35 already synthesized perovskites
- 17 mentioned only as non-perovskites
- 38 potentially new compounds
- 2 with negative thermal expansion at 300 K

# HIGH-THROUGHPUT SCREENING

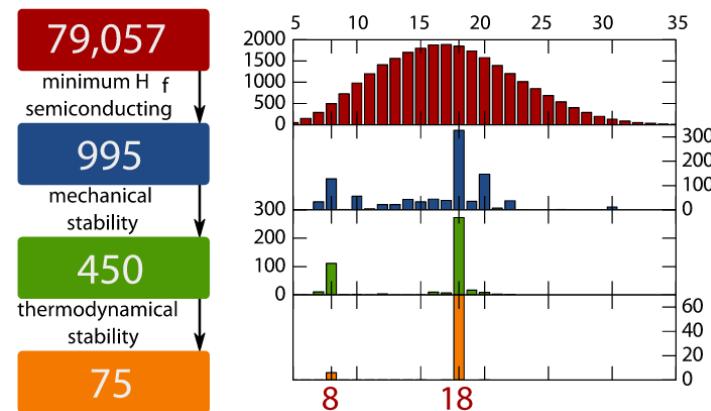
	$\kappa_{1000}$	$\kappa_{300}$	References		$\kappa_{1000}$	$\kappa_{300}$	References		$\kappa_{1000}$	$\kappa_{300}$	References
<b>CaSiO<sub>3</sub></b>	4.89		[21]	<b>CdYF<sub>3</sub></b>	1.29	3.51		<b>TlOsF<sub>3</sub></b>	0.62	0.95	
<b>RbTaO<sub>3</sub></b>	3.61		[22]	<b>RbCaF<sub>3</sub></b>	1.15	2.46 (3.2)	[23–25]	<b>InZnF<sub>3</sub></b>	0.61	1.86	
<b>NaTaO<sub>3</sub></b>	3.45		[26]	<b>HgInF<sub>3</sub></b>	1.15	3.85		<b>CsCdF<sub>3</sub></b>	0.59	1.73	[27]
<b>CuCF<sub>3</sub></b>	3.32	8.79	[28]	<b>AlFeF<sub>3</sub></b>	1.14			<b>AlMgF<sub>3</sub></b>	0.56		
<b>SrSiO<sub>3</sub></b>	3.23	10.10	[29]	<b>PbHfO<sub>3</sub></b>	1.12		[30]	<b>AuZnF<sub>3</sub></b>	0.53		
<b>NaNbO<sub>3</sub></b>	3.05	(1.5)	[31–33]	<b>AgMgF<sub>3</sub></b>	1.11		[34]	<b>InOsF<sub>3</sub></b>	0.52		
<b>BaHfO<sub>3</sub></b>	3.04 (4.5)	8.26 (10.4)	[35]	<b>ZnScF<sub>3</sub></b>	1.10	3.66		<b>RbSrF<sub>3</sub></b>	0.51		[36]
<b>KNbO<sub>3</sub></b>	2.94	(10)	[31, 33]	<b>RbFeF<sub>3</sub></b>	1.09	4.62	[37]	<b>CsSrF<sub>3</sub></b>	0.50	1.13	[36]
<b>TlTaO<sub>3</sub></b>	2.86		[38]	<b>TlMgF<sub>3</sub></b>	1.06	3.42	[39]	<b>BeYF<sub>3</sub></b>	0.48	2.34	
<b>AgTaO<sub>3</sub></b>	2.77		[40, 41]	<b>KCaF<sub>3</sub></b>	1.06		[42]	<b>BeScF<sub>3</sub></b>	0.48	1.59	
<b>KMgF<sub>3</sub></b>	2.74	8.25 (10)	[25, 43]	<b>HgScF<sub>3</sub></b>	1.01	5.42		<b>TlCdF<sub>3</sub></b>	0.44		[27]
<b>GaTaO<sub>3</sub></b>	2.63		[44–46]	<b>CsCaF<sub>3</sub></b>	0.98	3.03	[47] <sup>a</sup>	<b>RbHgF<sub>3</sub></b>	0.43		[48]
<b>BaTiO<sub>3</sub></b>	2.51	4.99 (4-5)	[33, 49]	<b>AuMgF<sub>3</sub></b>	0.96			<b>PdYF<sub>3</sub></b>	0.43	0.99	
<b>PbTiO<sub>3</sub></b>	2.42	(5)	[33]	<b>InMgF<sub>3</sub></b>	0.96	3.53		<b>AlZnF<sub>3</sub></b>	0.39		
<b>SrTiO<sub>3</sub></b>	2.36 (4)	6.44 (10.5)	[51–53]	<b>RbZnF<sub>3</sub></b>	0.91	2.64	[54]	<b>KHgF<sub>3</sub></b>	0.37		[48]
<b>SrHfO<sub>3</sub></b>	2.20 (2.7)	(5.2)	[53, 55]	<b>ZnInF<sub>3</sub></b>	0.88	1.89		<b>RbSnF<sub>3</sub></b>	0.37	0.82	[56]
<b>BaZrO<sub>3</sub></b>	2.13 (2.9)	5.61 (5.2)	[57]	<b>BaSiO<sub>3</sub></b>	0.87		[58]	<b>ZnBiF<sub>3</sub></b>	0.37	1.29	
<b>XeScF<sub>3</sub></b>	1.87	4.40		<b>TlCaF<sub>3</sub></b>	0.86			<b>CsHgF<sub>3</sub></b>	0.37	1.00	[48]
<b>HgYF<sub>3</sub></b>	1.84	5.37		<b>CdScF<sub>3</sub></b>	0.85	2.37		<b>KSnF<sub>3</sub></b>	0.35		[56]
<b>AgNbO<sub>3</sub></b>	1.79		[59, 60]	<b>XeBiF<sub>3</sub></b>	0.82	2.13		<b>CdBiF<sub>3</sub></b>	0.33	0.98	
<b>TlNbO<sub>3</sub></b>	1.75		[38]	<b>AgZnF<sub>3</sub></b>	0.80		[34]	<b>RbPbF<sub>3</sub></b>	0.32		[61]
<b>KFeF<sub>3</sub></b>	1.72	6.37 (3.0)	[62, 63]	<b>PdScF<sub>3</sub></b>	0.79	1.63		<b>BeAlF<sub>3</sub></b>	0.30	1.70	
<b>SnSiO<sub>3</sub></b>	1.66	4.22	[64, 65]	<b>KCdF<sub>3</sub></b>	0.75		[66, 67] <sup>b</sup>	<b>KPbF<sub>3</sub></b>	0.30		[68]
<b>PbSiO<sub>3</sub></b>	1.66	3.69	[69, 70]	<b>BaLiF<sub>3</sub></b>	0.73	2.21	[71, 72]	<b>CsBaF<sub>3</sub></b>	0.29		
<b>AuNbO<sub>3</sub></b>	1.56		[73]	<b>HgBiF<sub>3</sub></b>	0.72	2.37		<b>InCdF<sub>3</sub></b>	0.29		
<b>CaSeO<sub>3</sub></b>	1.42		[74]	<b>ZnAlF<sub>3</sub></b>	0.72	1.92		<b>BaCuF<sub>3</sub></b>	0.28		
<b>NaBeF<sub>3</sub></b>	1.40	2.53	[75, 76]	<b>GaZnF<sub>3</sub></b>	0.69			<b>TlSnF<sub>3</sub></b>	0.27	0.63	[77]
<b>RbMgF<sub>3</sub></b>	1.37	4.54	[78]	<b>RbCdF<sub>3</sub></b>	0.68	1.46	[27]	<b>TlHgF<sub>3</sub></b>	0.26		[79]
<b>GaMgF<sub>3</sub></b>	1.34	2.11		<b>GaRuF<sub>3</sub></b>	0.67			<b>CdSbF<sub>3</sub></b>	0.26		
<b>KZnF<sub>3</sub></b>	1.33	4.15 (5.5)	[25, 80]	<b>CsZnF<sub>3</sub></b>	0.67	1.12	[81]	<b>TlPbF<sub>3</sub></b>	0.22		[82]
<b>ZnYF<sub>3</sub></b>	1.32	3.72		<b>TlZnF<sub>3</sub></b>	0.64	1.96	[83]				

van Roekeghem et al., Phys. Rev. X 6, 041061 (2016)

# HIGH-THROUGHPUT SCREENING

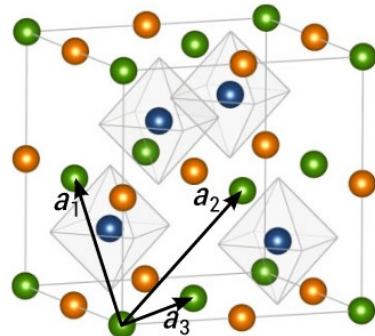


van Roekeghem et al, Phys. Rev. X 6, 041061 (2016)



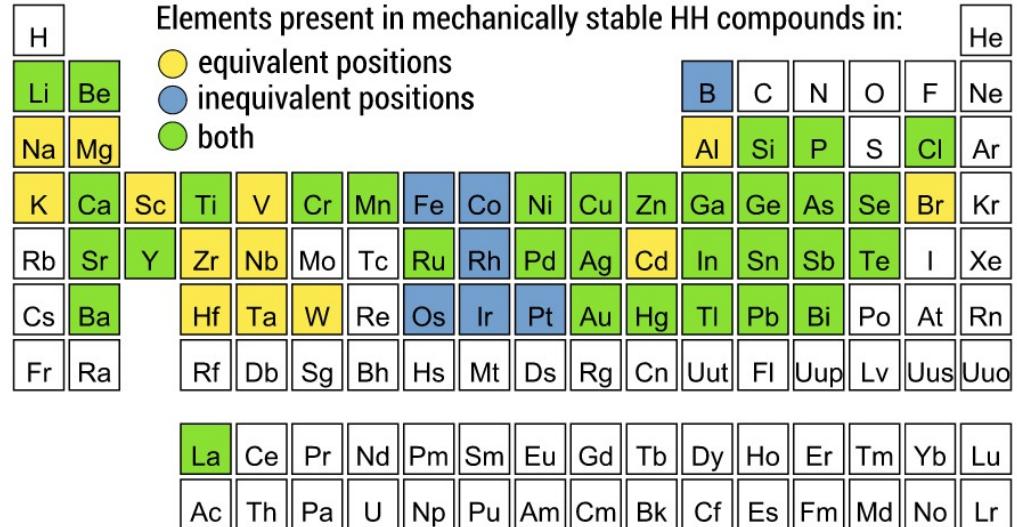
Carrete et al, Phys. Rev. X 4, 011019 (2014)

(a)

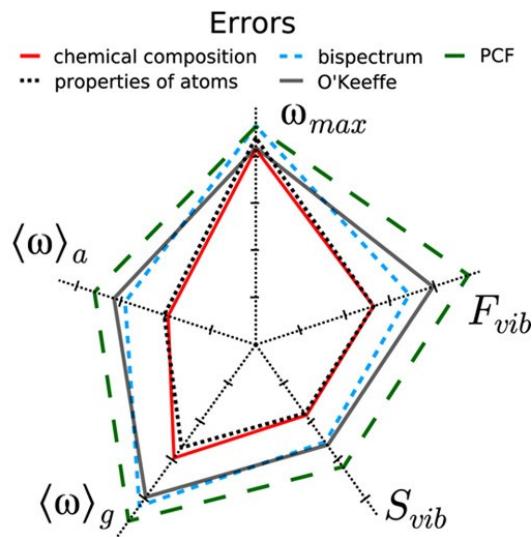
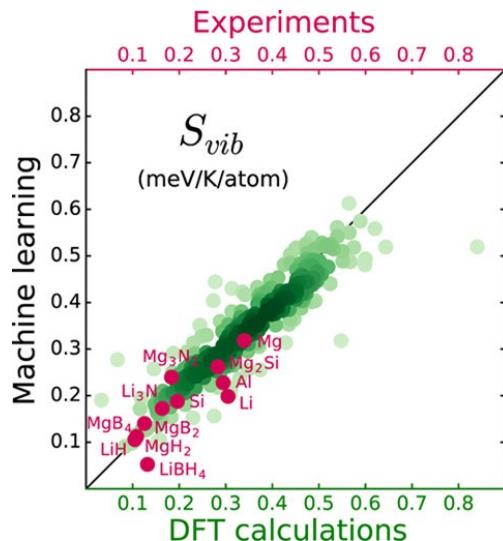


● A | Equivalent positions  
 ● B | positions  
 ● X

(b)



# MACHINE LEARNING



Legrain et al., Chemistry of Materials 29, 6220 (2017)

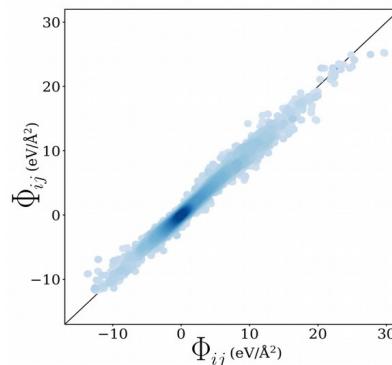
XYZ	probability	space group
ErNiBi	0.954	216 <sup>b</sup>
TmPtBi	0.947	216 <sup>b</sup>
ErPdBi	0.931	216 <sup>c</sup>
MnRuSb	0.931	
TbPtBi	0.913	216 <sup>b</sup>
TbPdBi	0.906	216 <sup>b</sup>
TmPdBi	0.899	216 <sup>b</sup>
EuPdBi	0.890	
MnFeSb	0.885	227 <sup>d</sup>
LuPtBi	0.882	216 <sup>b</sup>
YPtBi	0.864	216 <sup>b</sup>
EuPtBi	0.861	
TiRhSb	0.861	216 <sup>e</sup>
ScPdBi	0.854	216 <sup>b</sup>
MnTeRh	0.846	
HfCoBi	0.844	
LuPdBi	0.831	216 <sup>b</sup>

Legrain et al., J. Phys. Chem. B 122, 625 (2018)

$$\mathbf{D}_{i,j}^{(2)\alpha} = \sum_m e^{-\left|\frac{\mathbf{r}_{ij} + \mathbf{R}_m}{a_\alpha}\right|^2} (\mathbf{r}_{ij} + \mathbf{R}_m) \otimes (\mathbf{r}_{ji} - \mathbf{R}_m)$$

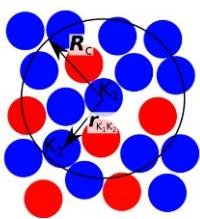
$$\mathbf{D}_{s,s';i,j}^{(2)\alpha} \equiv (\delta_{s_i,s} \delta_{s_j,s'} + \delta_{s_i,s'} \delta_{s_j,s}) \mathbf{D}_{i,j}^{(2)\alpha}$$

$$a_\alpha = \{1, 2, 3, \dots, 30\} \text{ \AA}$$



Legrain et al., arXiv:1803.09827, Journal of Chemical Information and Modeling in press

# MACHINE LEARNING



$$r_{K_i K_j} \Rightarrow f(r_{K_i K_j})$$

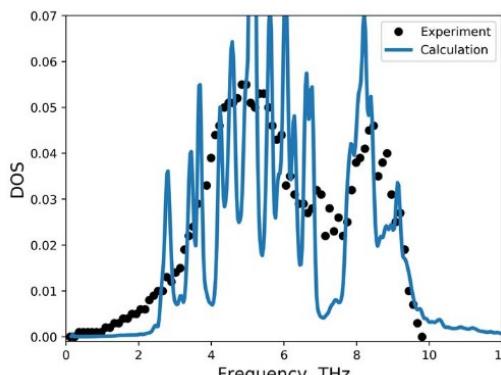
$$f(r_{K_i K_j}) = e^{-\sigma(r_{K_i K_j} - \eta)^2}$$

$$Z_{K_i} \Rightarrow c_{K_i}$$

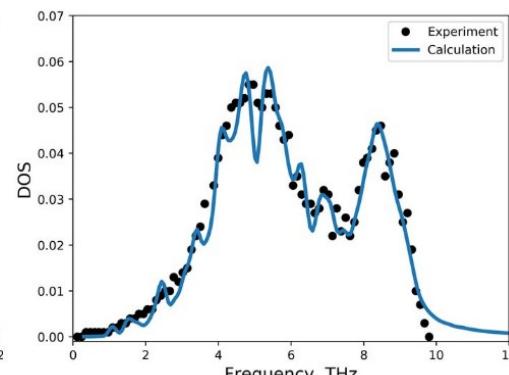
$$E_{tot} = \sum_{i=1}^N E_{K_i}$$

$$J = \sum_i (E_{tot} - E_{DFT})^2 + \lambda \sum_i (F - F_{DFT})^2$$

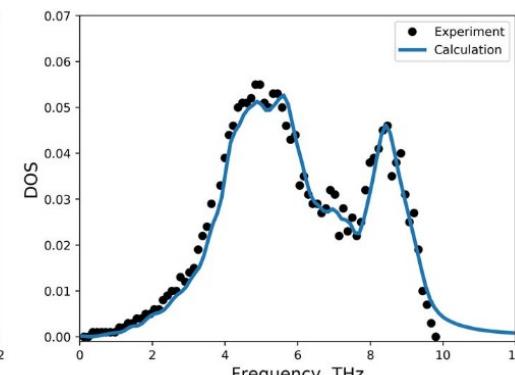
$$F = -\frac{dE_{tot}}{dx}$$



108 atoms



2048 atoms



10976 atoms

## Accuracy

- Energy:  $\sim 1$  meV/atom
- Forces:  $0.003 - 0.05$  eV/ $\text{\AA}$

## Features

- Output: Energy, Forces, Stress tensor
- Efficient training utilizes both energies and forces
- Training data may contain any number of atoms and species

Bochkarev et al, in preparation

## CONCLUSIONS

- Ab initio calculations of the lattice thermal conductivity are reliable.
- For electronic properties, it is still very difficult.
- High-throughput computational screening is increasingly developing.
- Machine learning can help to speed up the process.

# Thank you!

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