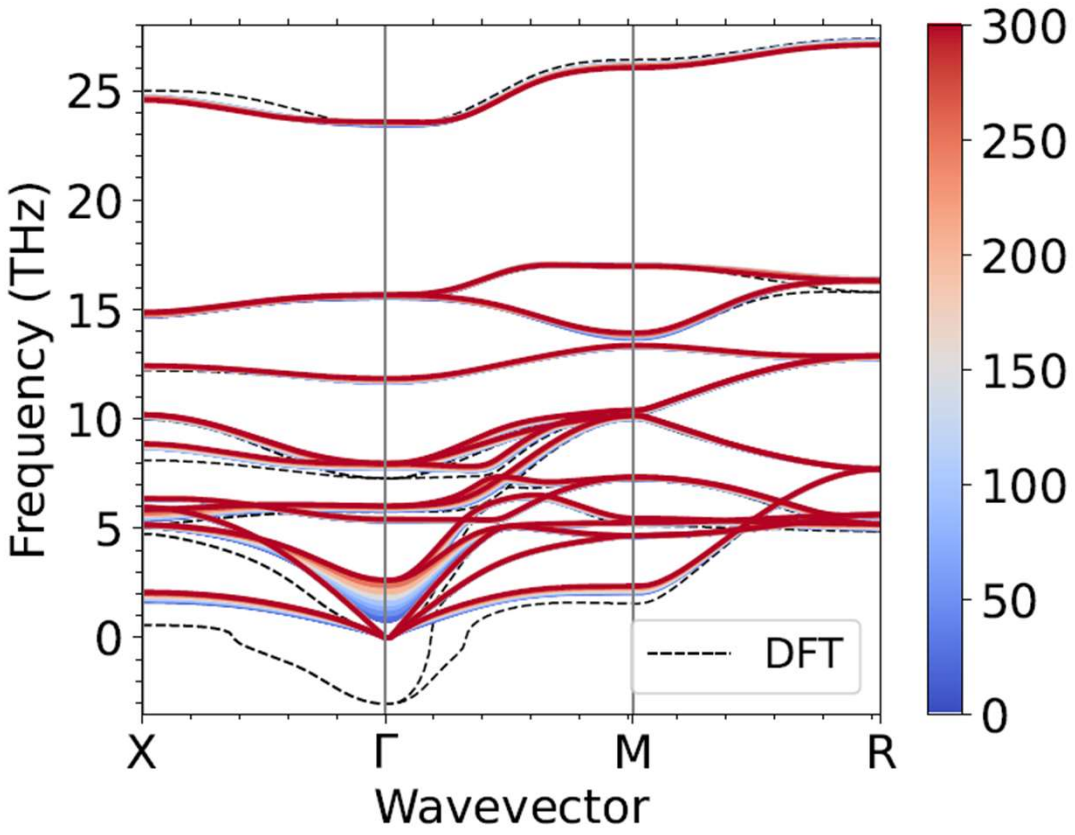
liten
cea tech



Lattice Anharmonicity in Solids: High-Throughput Screening and Machine Learning

Platform for Advanced Scientific Computing Conference – 26/06/2023
Ambroise van Roekeghem

Atomistic calculations at LITEN

Pioneer in ab initio calculations of (bulk) lattice thermal conductivity (ShengBTE)

Specific thermal properties are notably desirable for:

- Thermoelectrics (low thermal conductivity)
- Electronics (high thermal conductivity)

Extensions to multi-scale, defects, interfaces: almaBTE (Stars of Europe prize)

Other interests: anharmonicity beyond quasi-harmonic approximation (QSCAILD), high-throughput screening, machine learning

Recently moved to the battery field (2020)
-> projects in kinetics of degradation, calculation of forces and photoemission in correlated oxides, ionic transport in solids and liquids

Comp. Phys. Comm. 2014

Comp. Phys. Comm. 2017

QSCAILD
Comp. Phys. Comm. 2021

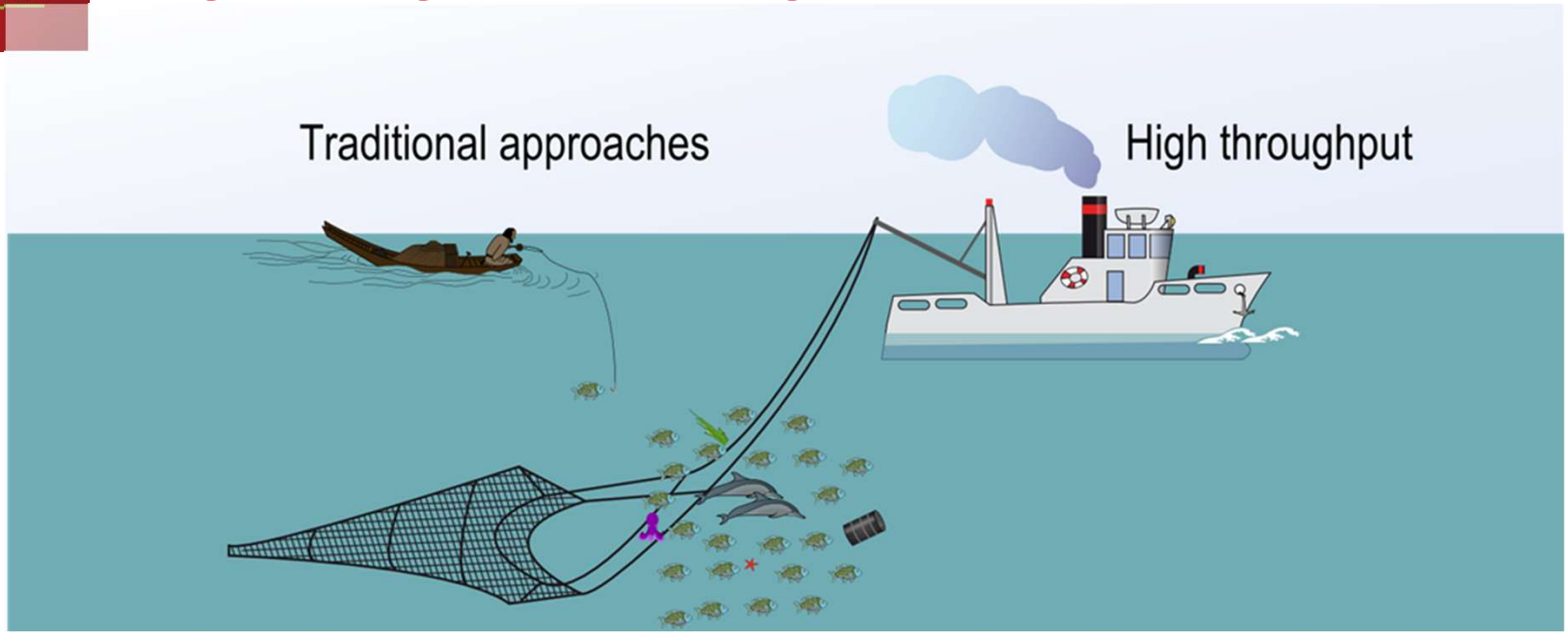
Outline

Stability at finite temperature

Thermal conductivity

Dielectric properties

High-throughput screening



MATERIALS PROJECT

Energy & Environmental Science

RSC Publishing

PAPER

View Article Online

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

Yabi Wu,[a] Predrag Lazic,[a] Geoffroy Hautier,†[a] Kristin Persson[b] and Gerbrand Ceder*[a]

Cite this: *Energy Environ. Sci.*, 2013, 6, 157

AFLOW

Automatic - FLOW for Materials Discovery

nature materials

REVIEW ARTICLE

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

The high-throughput highway to computational materials design

Stefano Curtarolo[1,2]*, Gus L. W. Hart[2,3], Marco Buongiorno Nardelli[2,4,5], Natalio Mingo[2,6], Stefano Sanvito[2,7] and Ohad Levy[1,2,8]

NOMAD

NREL

NATIONAL RENEWABLE ENERGY LABORATORY

High Performance Computing Center

Materials Database

ARTICLES

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

nature chemistry

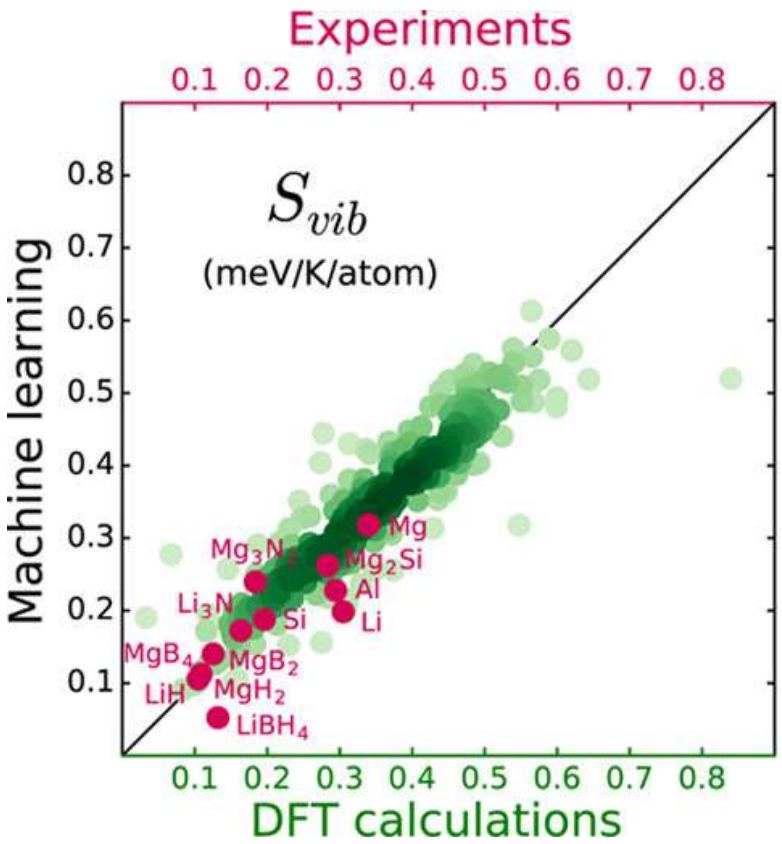
Prediction and accelerated laboratory discovery of previously unknown 18-electron ABX compounds

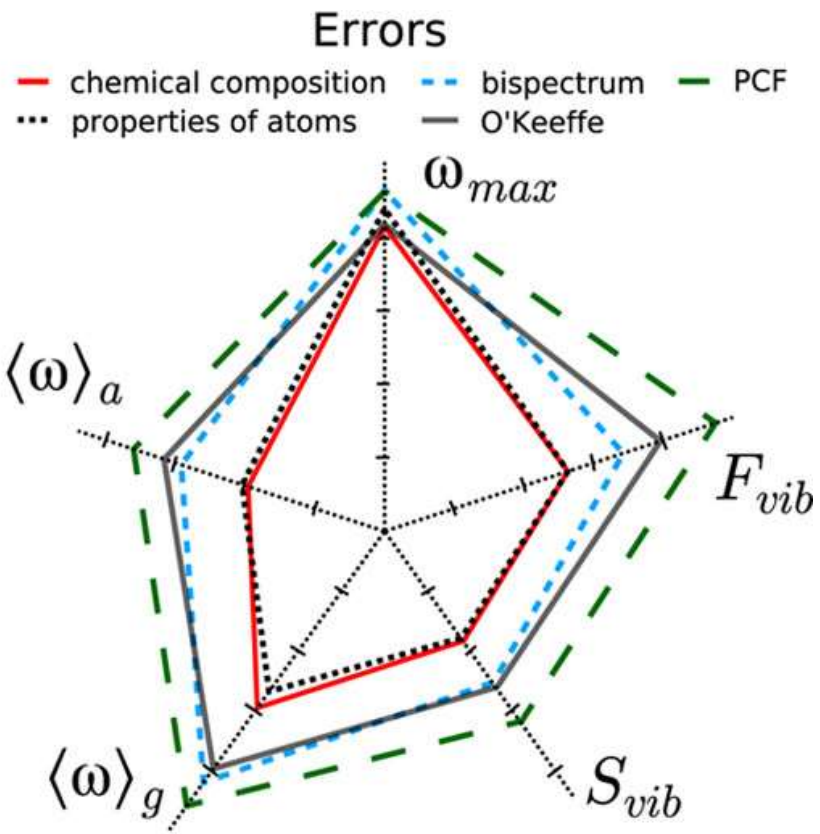
Romain Gautier[1], Xiuwen Zhang[2], Linhua Hu[1], Liping Yu[2], Yuyuan Lin[1], Tor O. L. Sunde[1], Danbee Chon[1], Kenneth R. Poeppelmeier[1]* and Alex Zunger[2]*

AiiDA

Predicting vibrational entropies and free energies

Towards screening of chemical reactions at finite temperature





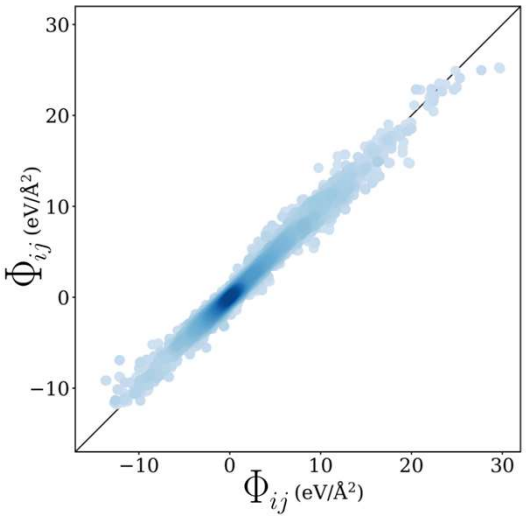
Legrain et al., Chemistry of Materials 29, 6220 (2017)

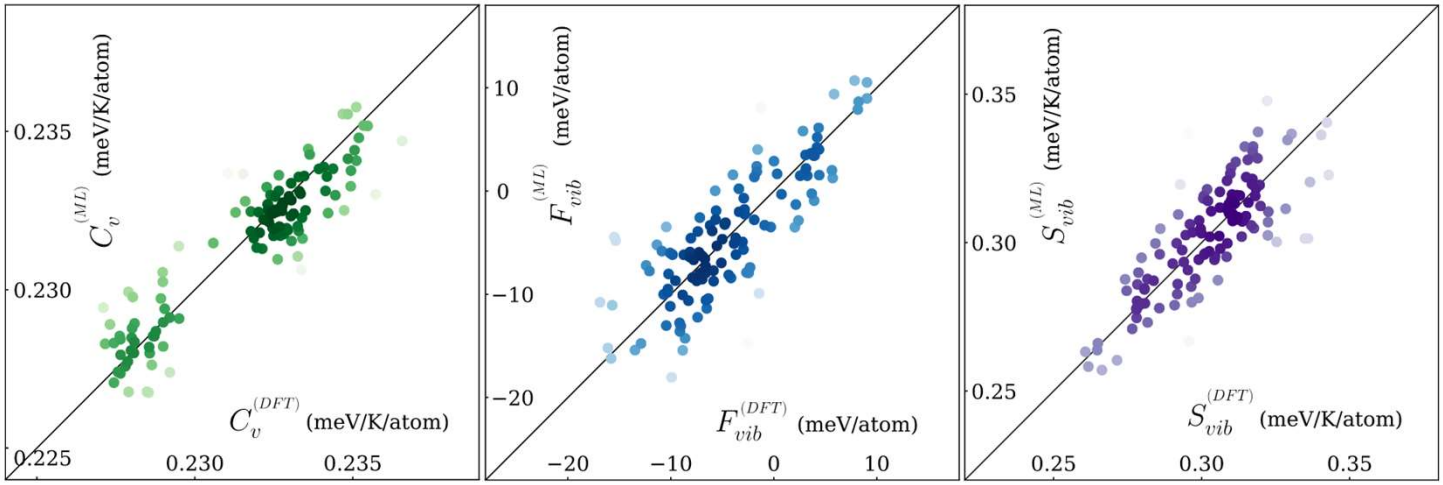
The case of polymorphs

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

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

$$a_\alpha = \{1, 2, 3, ..., 30\} \text{ Å}$$





Legrain et al., Journal of Chemical Information and Modeling 58, 2460 (2018)

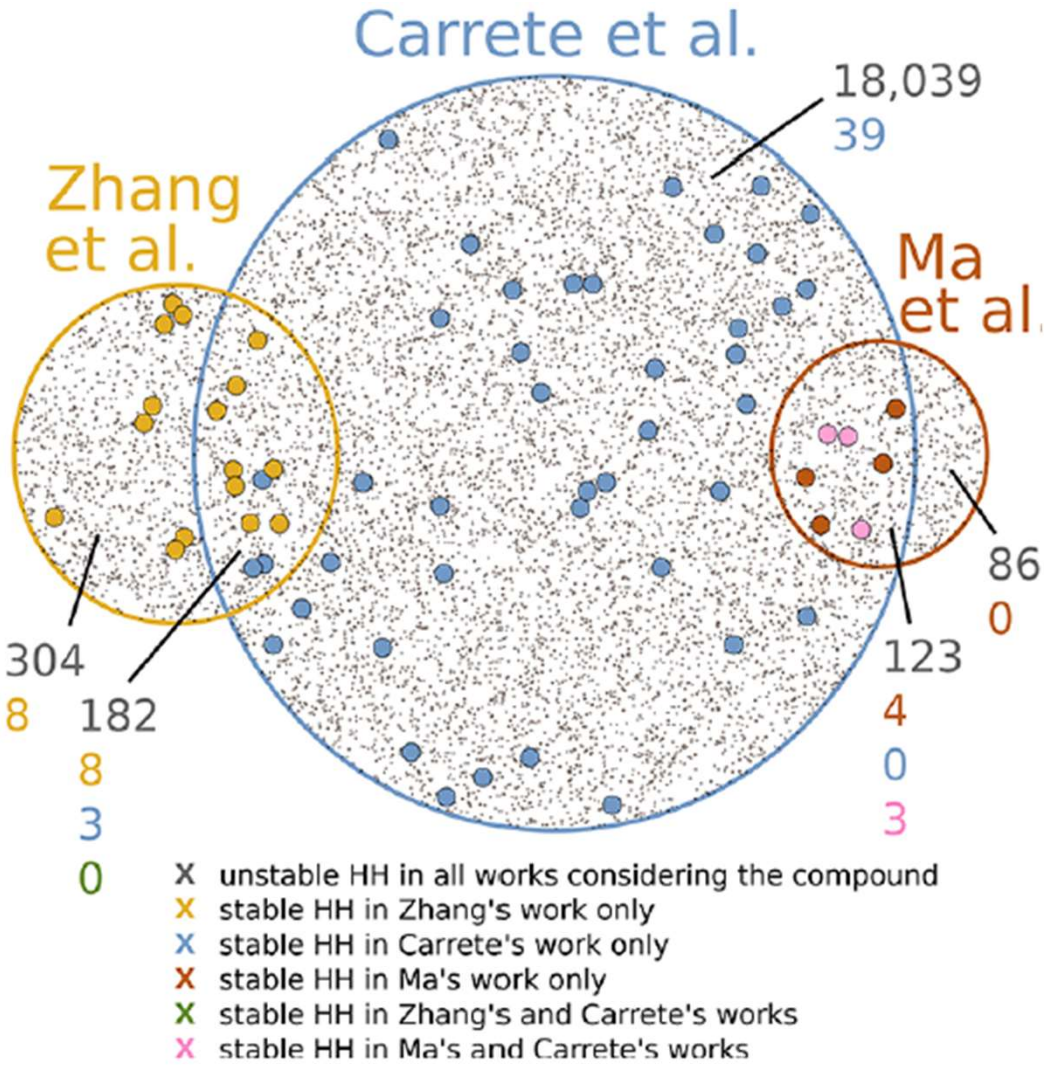
Finding new compounds from experimental data

Chemical intuition with random forests

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

Legrain et al., The Journal of Physical Chemistry B 122, 625 (2018)

Reproducibility and consistency



Legrain et al., The Journal of Physical Chemistry B 122, 625 (2018)

Defects and thermal conductivity

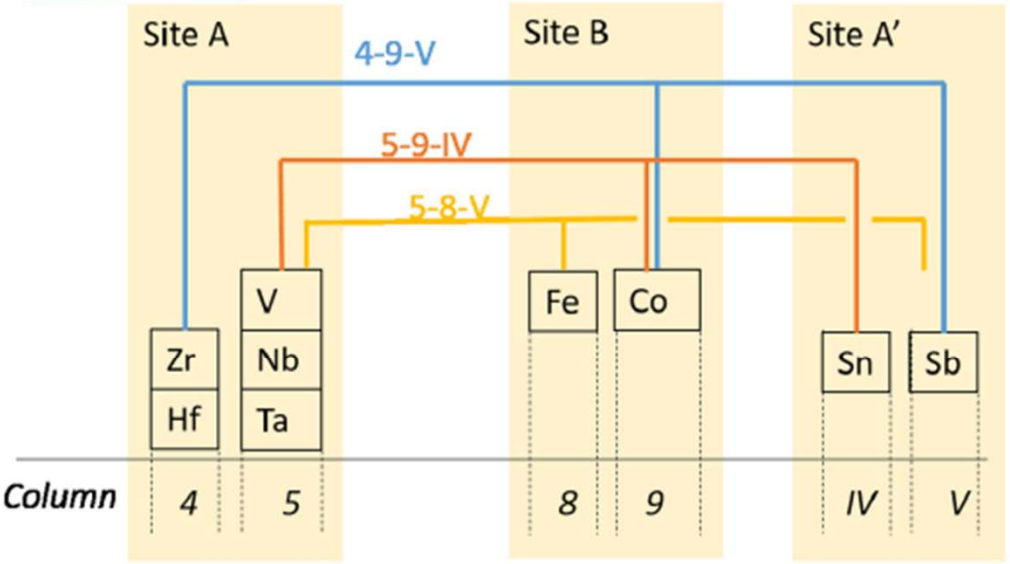
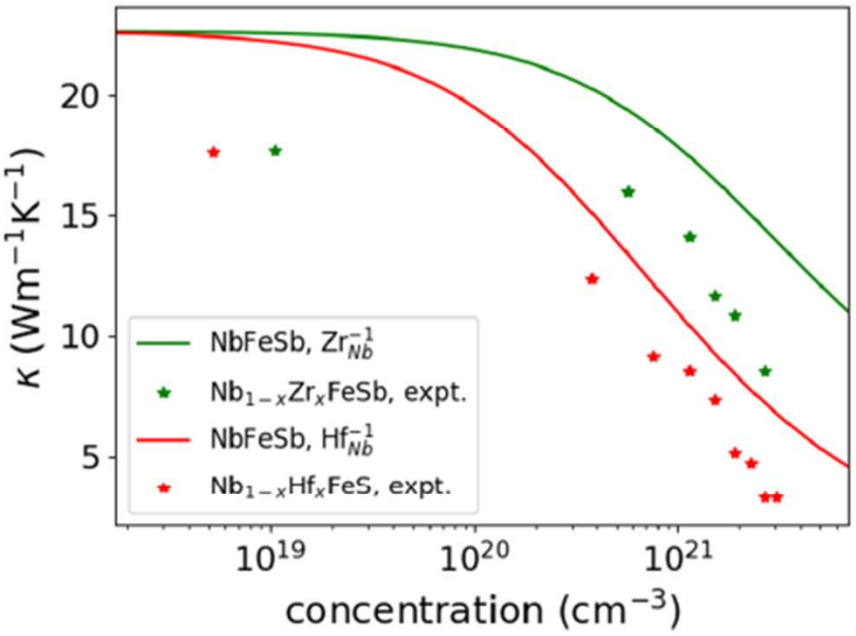


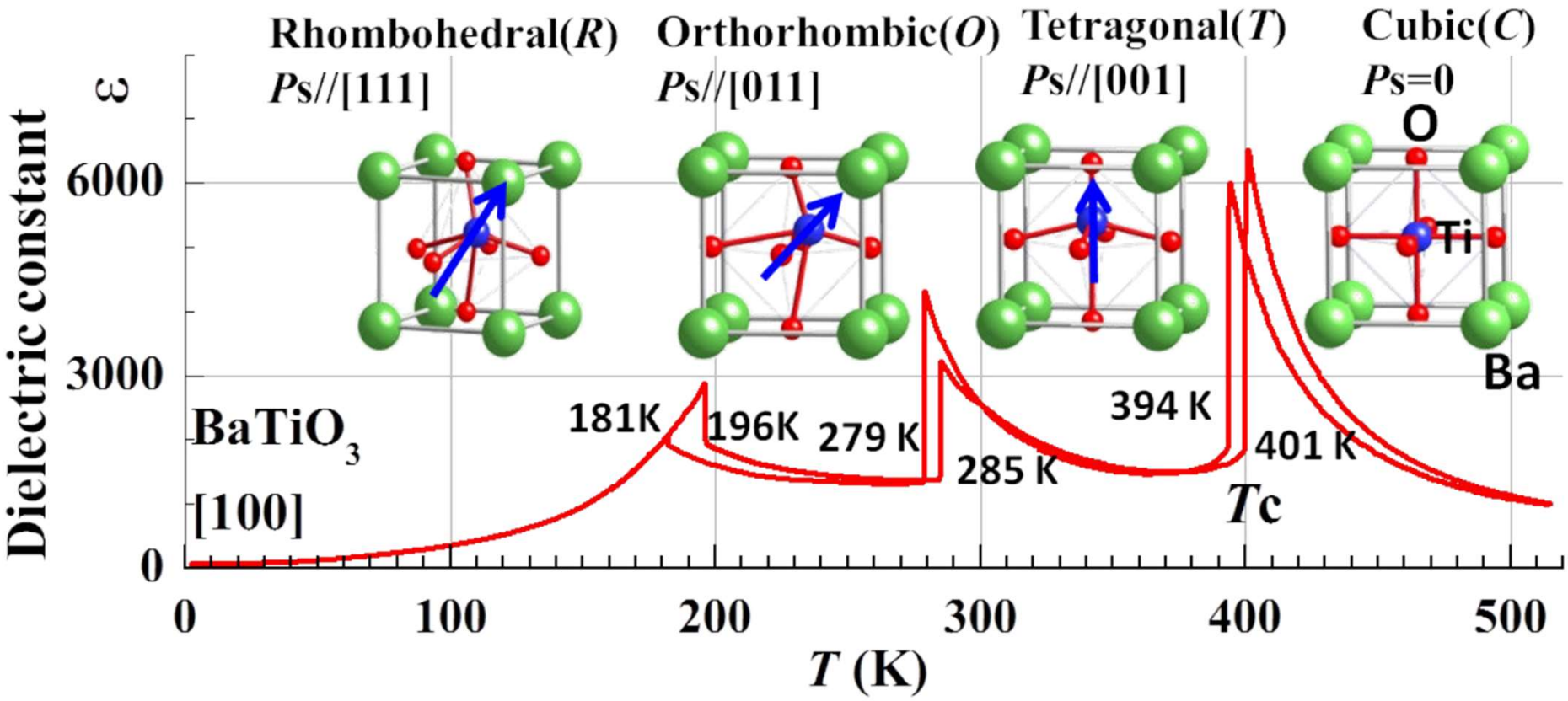
TABLE I. Stable substitution schemes considered.

Substitution scheme	Cases
3_4-9-V	$Sc_{(Zr,Hf)}$-Co-Sb
4_5-{9-IV, 8-V}	$(Ti, Zr, Hf)_{(V,Nb,Ta)}$-{Fe-Sb, Co-Sn}
5-7_8-V	(V, Nb, Ta)-Mn_{Fe}-Sb
{4-9, 5-8}-IV_V	{(Zr, Hf)-Co, (V, Nb, Ta)-Fe}-Sn_{Sb}



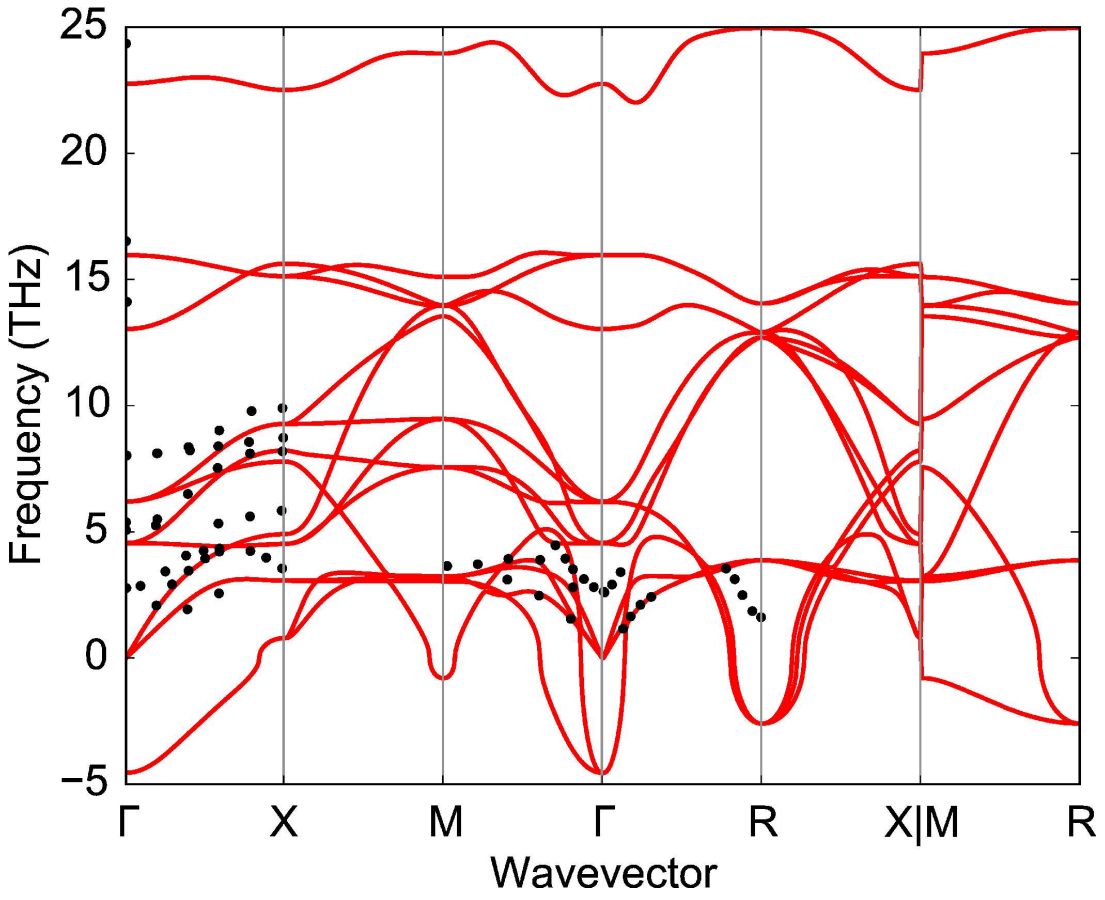
Fava et al., Phys. Rev. B 103, 174112 (2021)

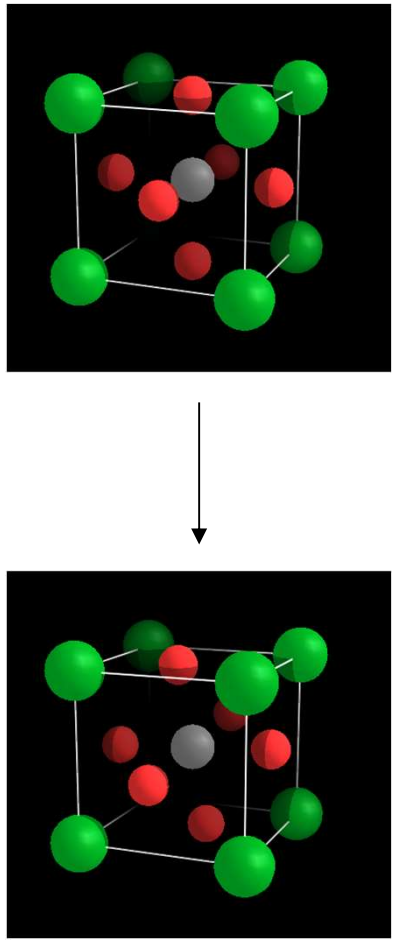
Distortions in perovskites: the example of BaTiO3



D. Fu and M Itoh, Ferroelectrics Materials – Synthesis and Characterization (2015)

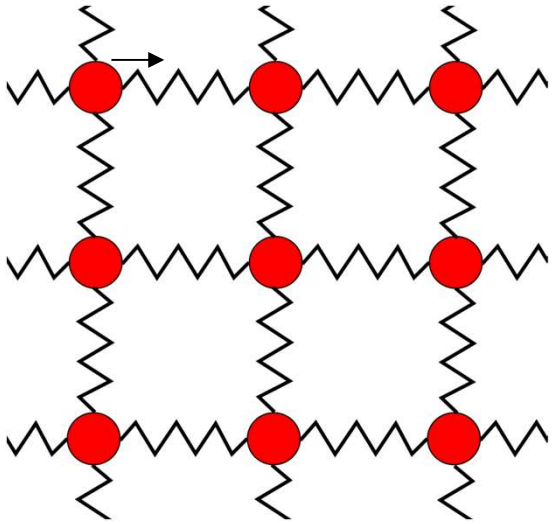
Distortions in perovskites: the example of SrTiO3





Finite-temperature phonon calculations: QSCAILD

Small displacements

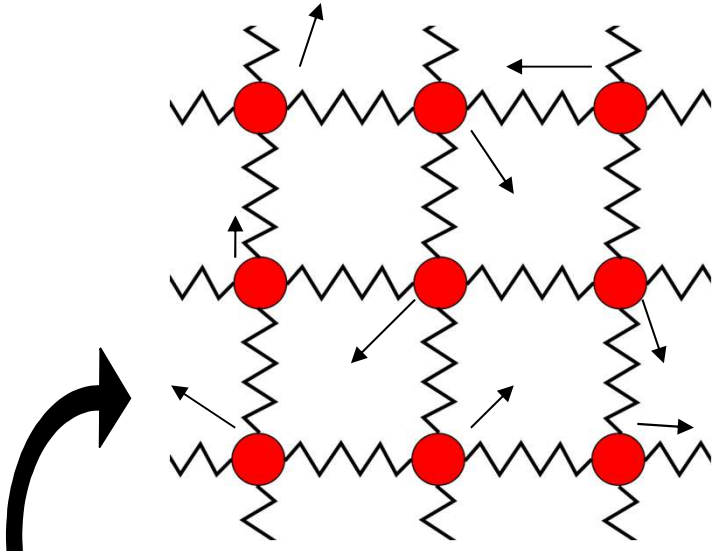


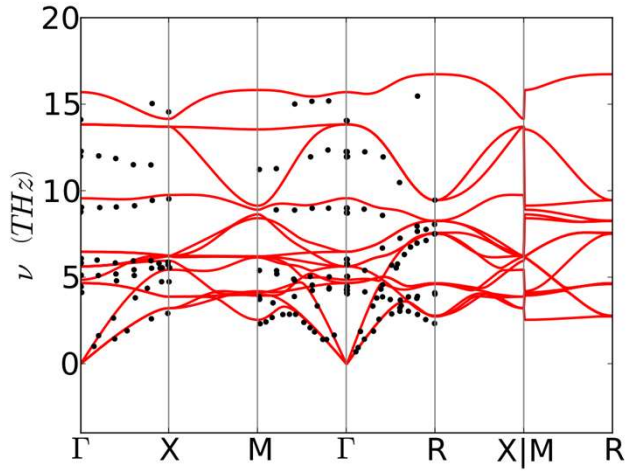
$$F_i^\alpha = \sum_{j\beta} \Phi_{ij}^{\alpha\beta} u_j^\beta + \frac{1}{2} \sum_{jk\beta\gamma} \Psi_{ijk}^{\alpha\beta\gamma} u_j^\beta u_k^\gamma$$

→ 2nd and 3rd order force constants

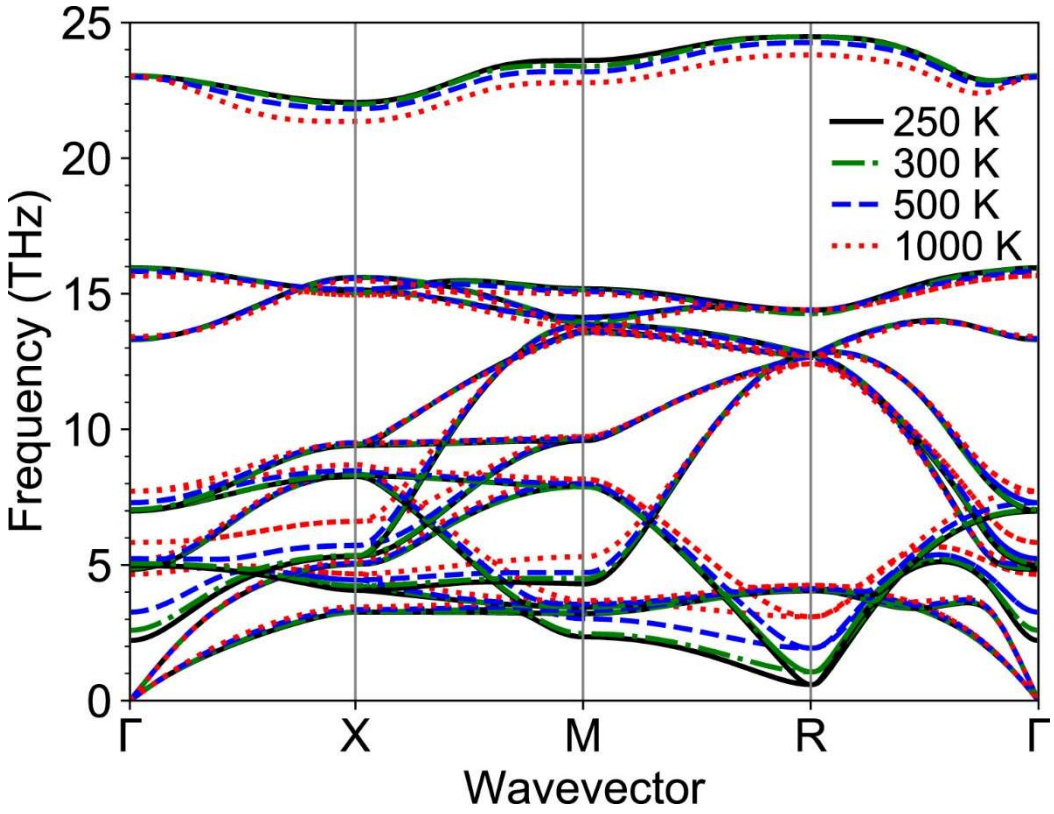
→ Thermal conductivity

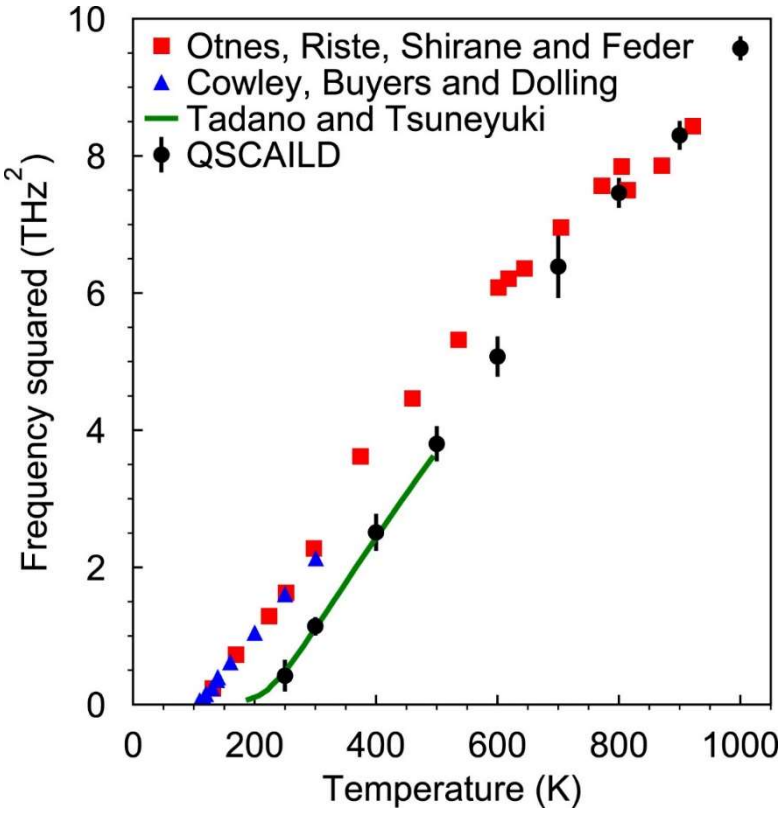
Quantum statistics, finite T





SrTiO3 at finite temperature

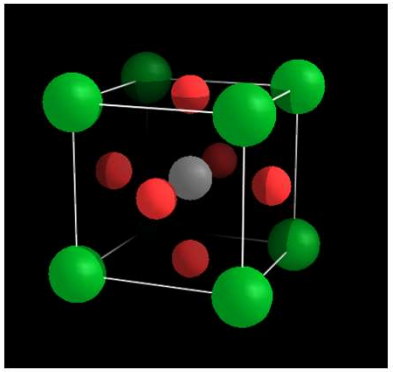




van Roekeghem, Carrete and Mingo, Comp. Phys. Comm. 263, 107945 (2021)

High-throughput screening

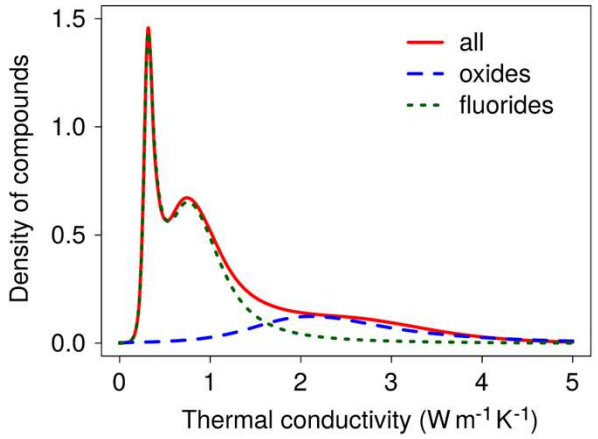
ABX_3 with X=O or F



H																He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
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

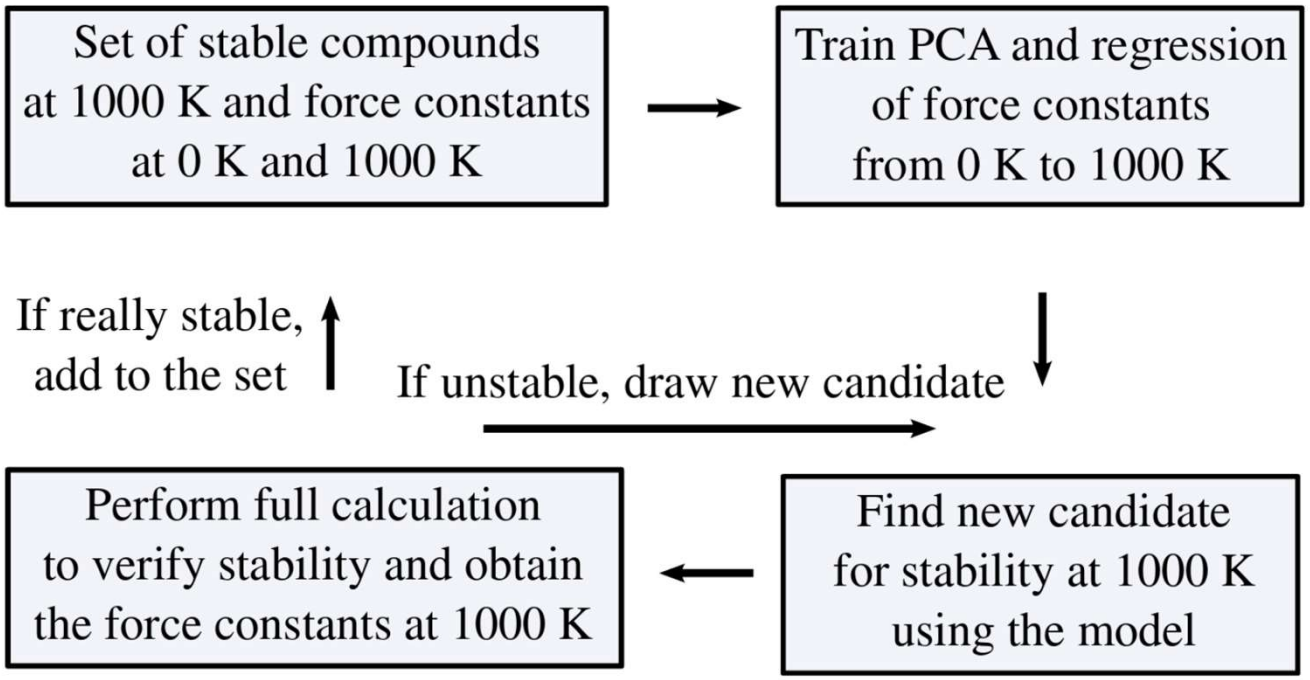
- 8000 possible combinations
- 400 non-magnetic semi-conductors
- 90 found mechanically stable at 1000 K

- 35 already synthesized perovskites
- 17 mentioned only as non-perovskites
- 38 potentially new compounds
- 2 with negative thermal expansion at 300 K



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

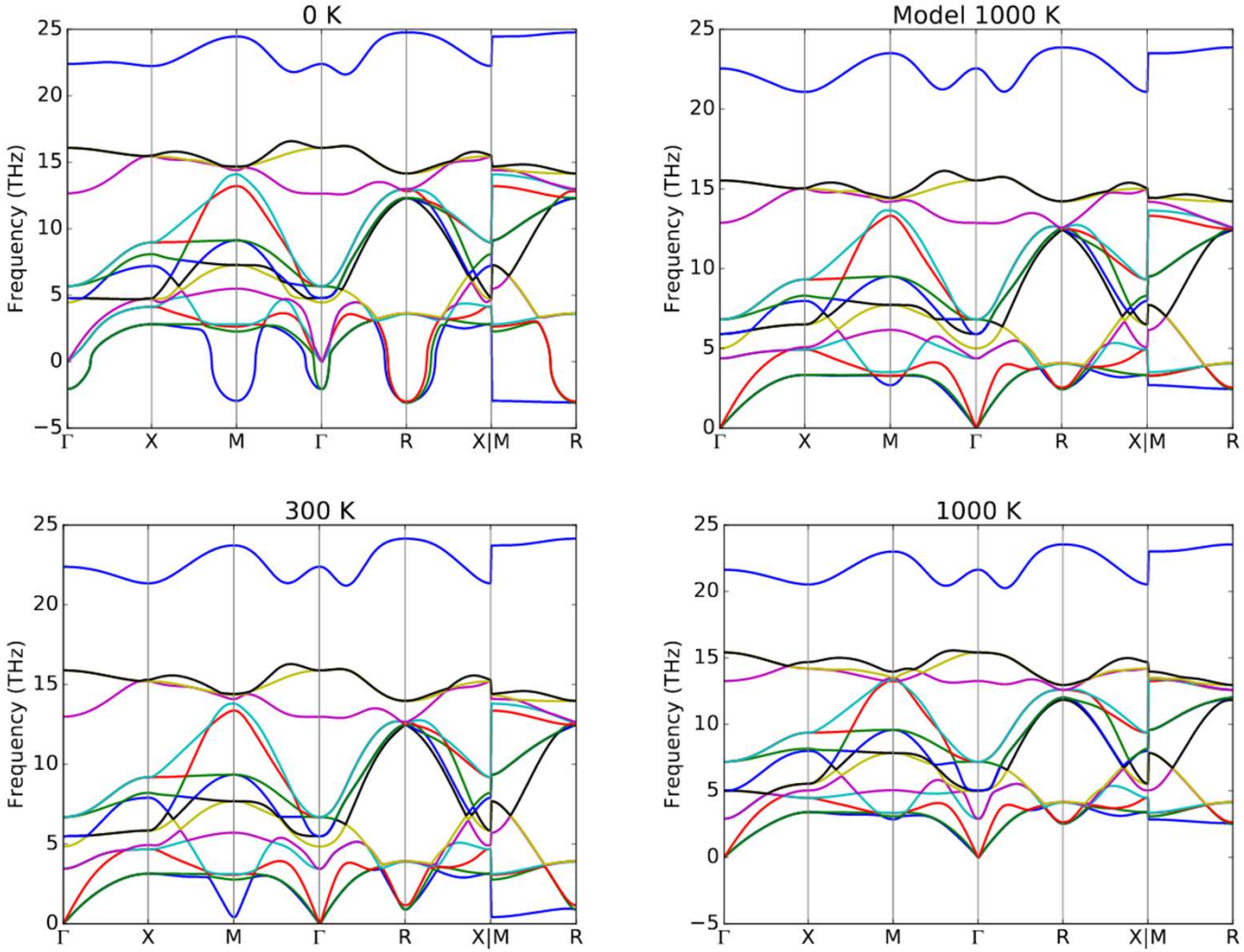
Dimensionality reduction



79 candidates out of about 400 compounds
68 positives vs 92 for the brute force calculation

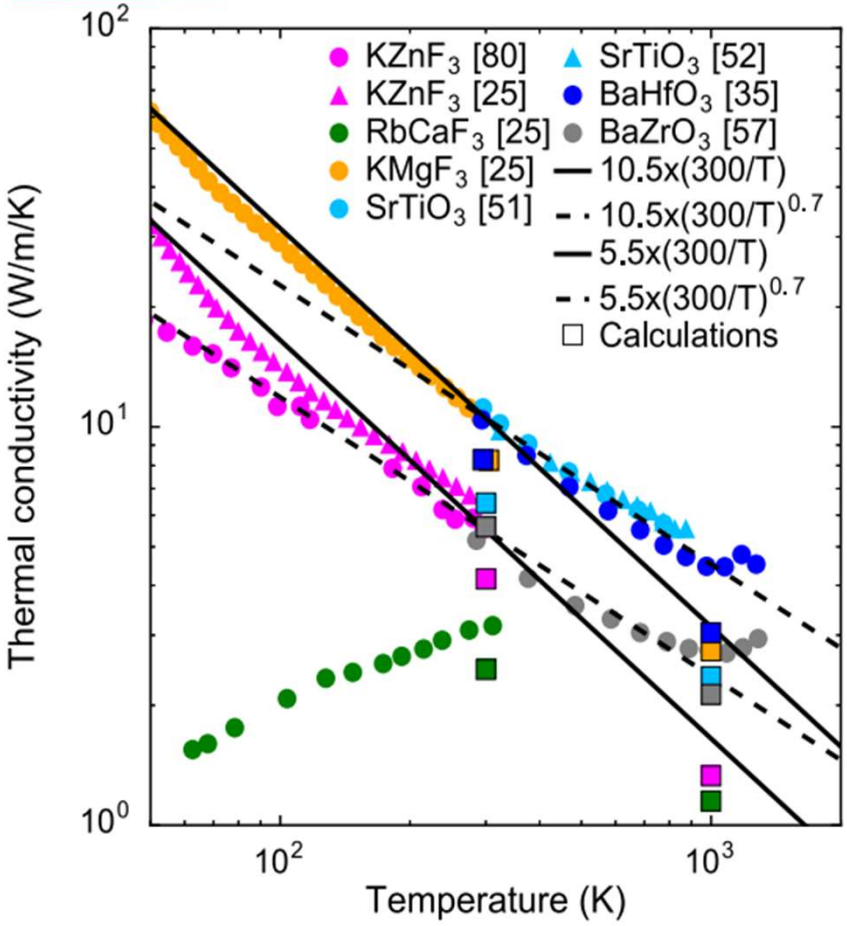
van Roekeghem et al., Physical Review X 6, 041061 (2016)

Simple ai on ab initio data



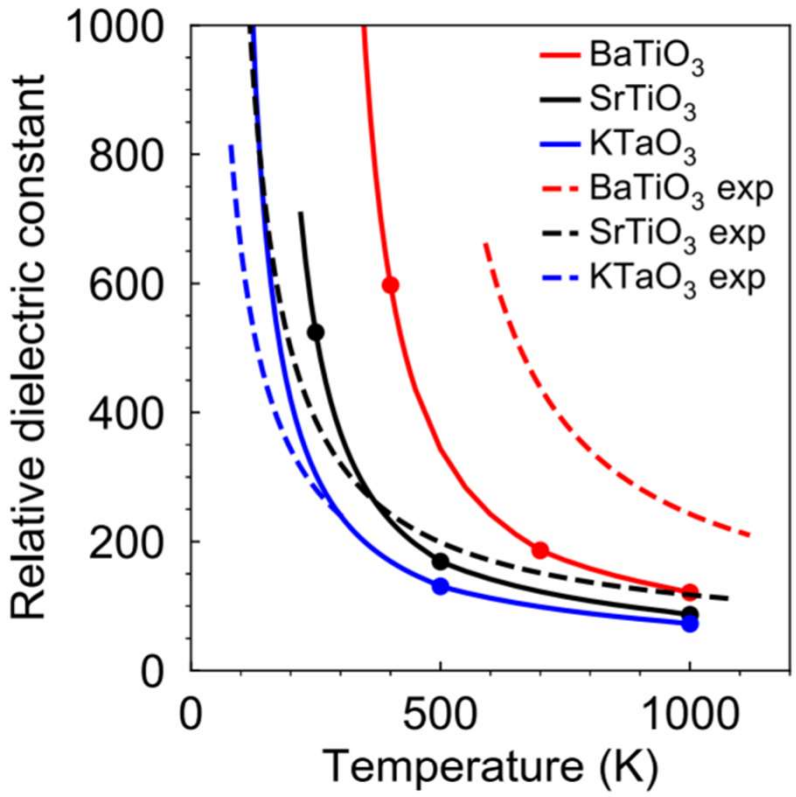
van Roekeghem et al., Physical Review X 6, 041061 (2016)

Thermal conductivities and dielectric constants



$$\kappa \propto T^{-\alpha} \qquad \text{Mean } \alpha = 0.8$$

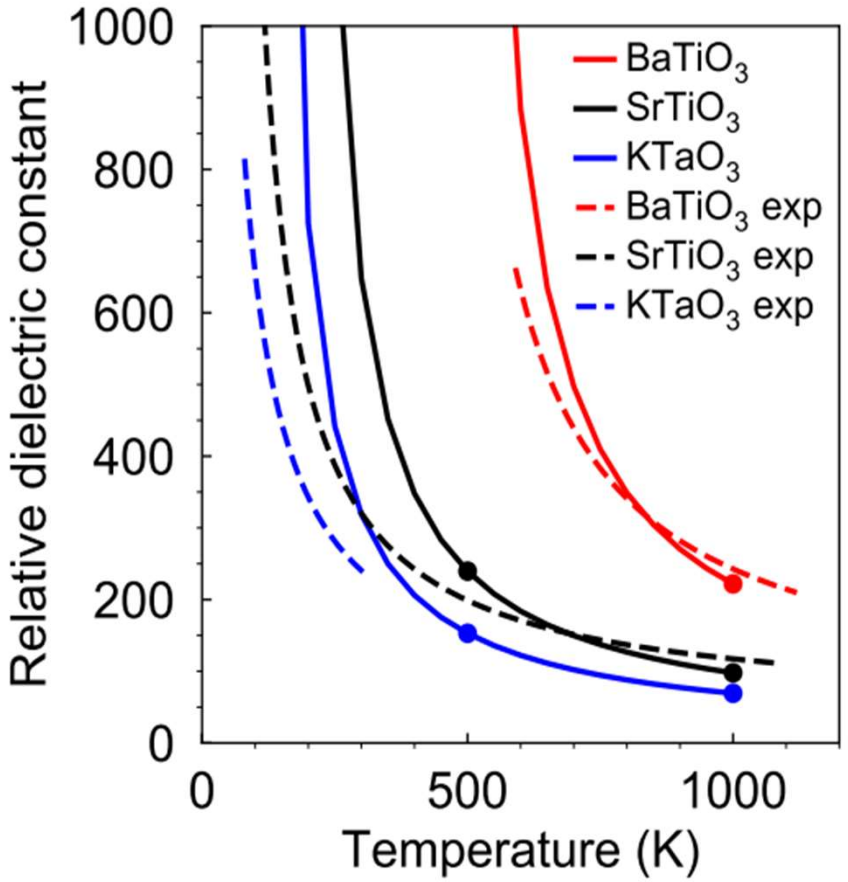
van Roekeghem, Carrete, Oses, Curtarolo and Mingo
Phys. Rev. X 6, 041061 (2016)

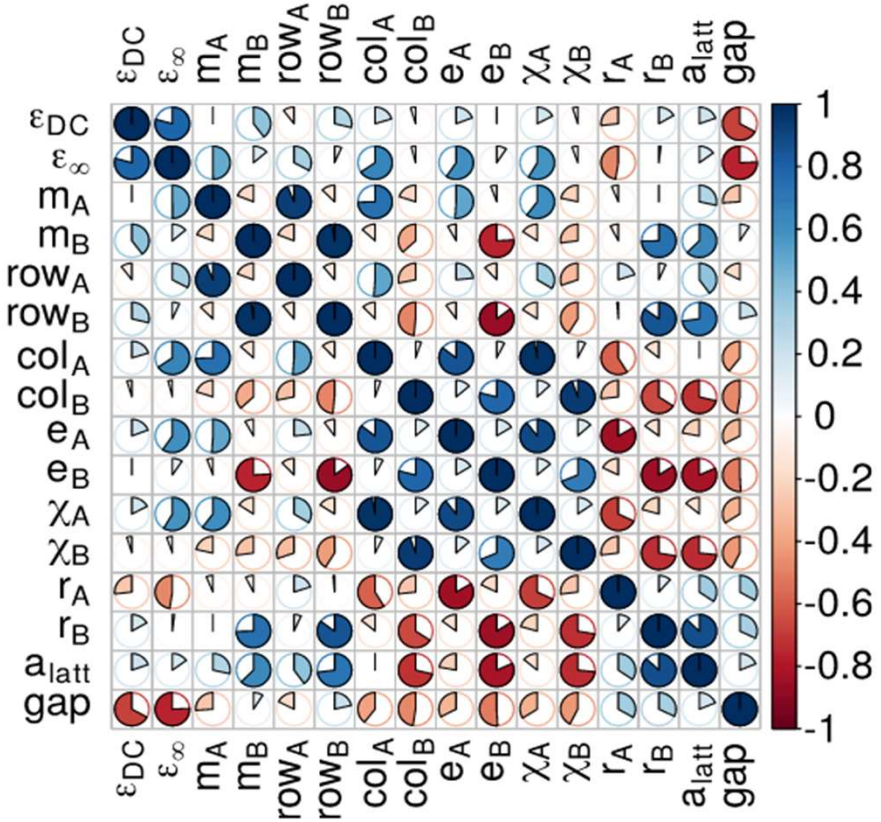


$$\frac{\epsilon_{DC}}{\epsilon_{\infty}} = \prod_j \left(\frac{|\omega_{Lj}|}{\omega_{Tj}} \right)^2$$

van Roekeghem, Carrete, Curtarolo and Mingo
Phys. Rev. Materials 4, 113804 (2020)

High-throughput screening





van Roekeghem, Carrete, Curtarolo and Mingol, arXiv:1805.09199, submitted to PRM (2018)

Quantum paraelectricity in KTaO3

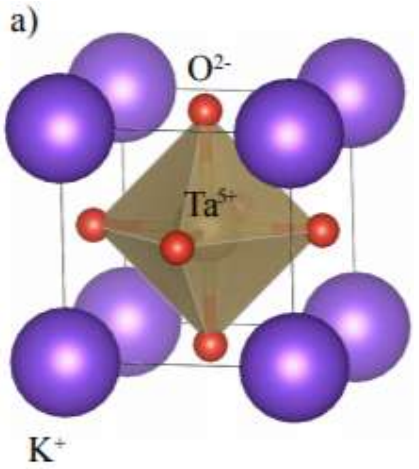
$KTaO_3$ crystallizes in the perovskite structure

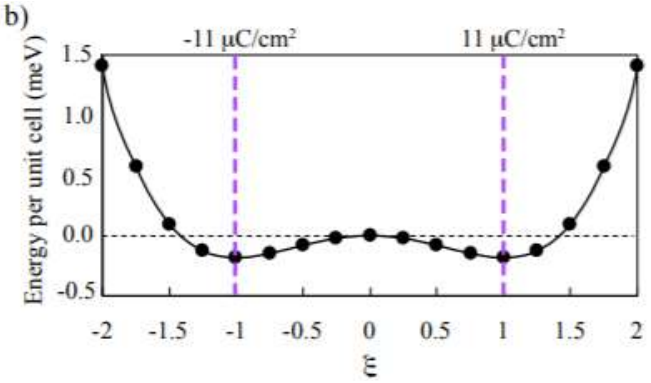
Quantum paraelectric:

existence of a ferroelectric instability

No ferroelectric phase, because quantum fluctuations stabilize unstable phonon branch even at 0 K

Very soft polar branch -> large bandgap but huge dielectric constant



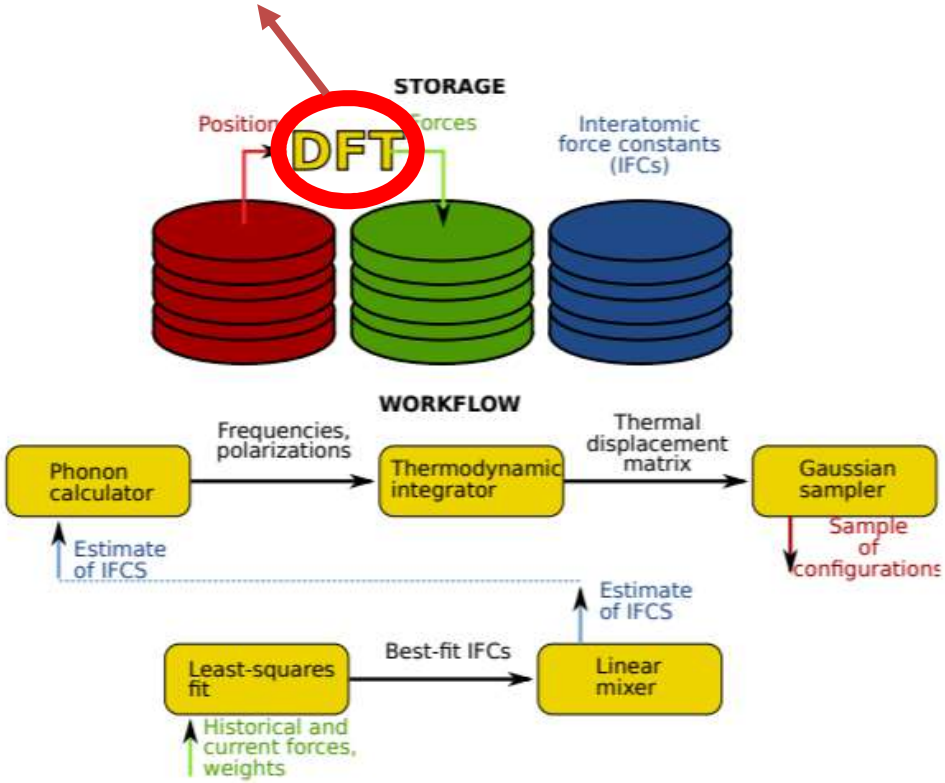


Gattinoni et al, arXiv:2108.10207

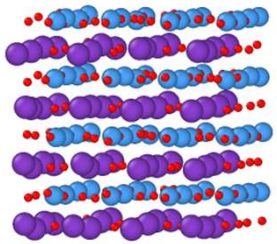
Speeding up QSCAILD using ML potentials

- Acquiring forces requires many DFT calculations -> slow and expensive
- We have to sample many similar configurations -> ideal to combine with machine learning
- We interface QSCAILD with active learning of Moment Tensor Potentials

Bottleneck! -> DFT + active learning



Machine learning force fields/potentials



atomic positions

(ML) FF → Energy, forces, stress

Reference data: energy, forces, stress from Density Functional Theory

Atomic positions are used to build descriptors of the local environments

Machine learning methods can be for instance:

- Neural Networks
- Gaussian processes
- Linear methods

PRL 104, 136403 (2010) PHYSICAL REVIEW LETTERS week ending 2 APRIL 2010

Gaussian Approximation Potentials: The Accuracy of Quantum Mechanics, without the Electrons

Albert P. Bartók and Mike C. Payne
Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge, CB3 0HE, United Kingdom

Risi Kondor
Center for the Mathematics of Information, California Institute of Technology, MC 305-16, Pasadena, California 91125, USA

Gábor Csányi
Engineering Laboratory, University of Cambridge, Trumpington Street, Cambridge, CB2 1PZ, United Kingdom
(Received 1 October 2009; published 1 April 2010)

PRL 98, 146401 (2007) PHYSICAL REVIEW LETTERS week ending 6 APRIL 2007

Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces

Jörg Behler and Michele Parrinello
Department of Chemistry and Applied Biosciences, ETH Zurich, USI-Campus, Via Giuseppe Buffi 13, CH-6900 Lugano, Switzerland
(Received 27 September 2006; published 2 April 2007)

MULTISCALE MODEL. SIMUL.
Vol. 14, No. 3, pp. 1153–1173

MOMENT TENSOR POTENTIALS: A CLASS OF SYSTEMATICALLY IMPROVABLE INTERATOMIC POTENTIALS*

ALEXANDER V. SHAPEEV†

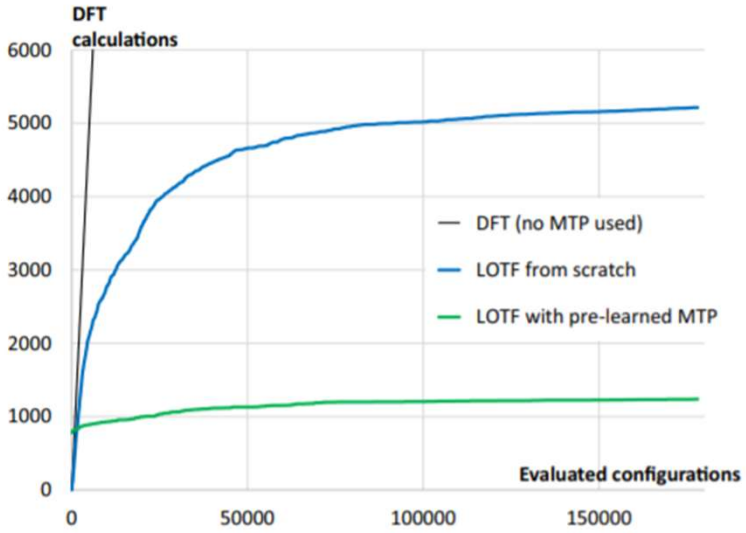
Moment Tensor Potentials (A. Shapeev)

Cutoff: typically of the order of 5-10 Angstroms
Loss function: energies, forces, stress…

$$V(\mathrm{n}_i) = \sum_{\alpha} \xi_{\alpha} B_{\alpha}(\mathrm{n}_i),$$

$$M_{\mu,\nu}(\mathrm{n}_i) = \sum_{j} f_{\mu}(|r_{ij}|, z_i, z_j)\, \underbrace{r_{ij} \otimes \ldots \otimes r_{ij}}_{\nu \text{ times}},$$

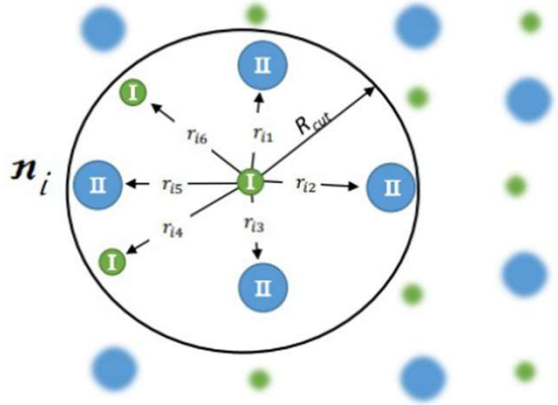
Active learning



Podryabinkin et al., Phys. Rev. B 99, 064114 (2019)

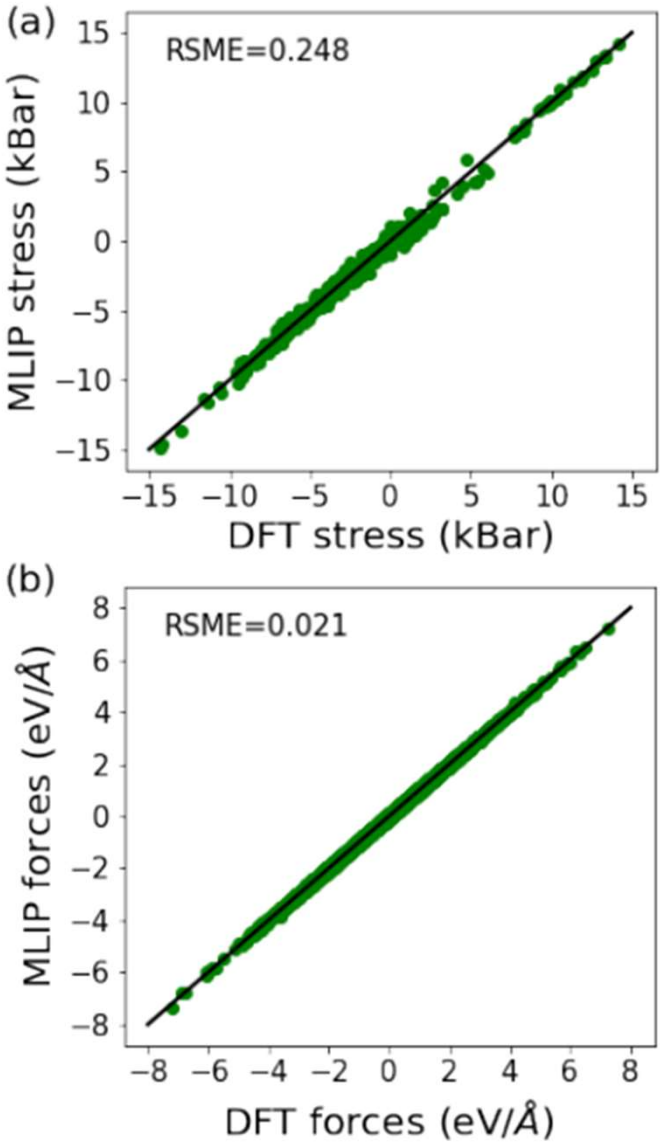
Radial: depends on relative distances and types of atoms

Angular: tensor resembling moments of inertia

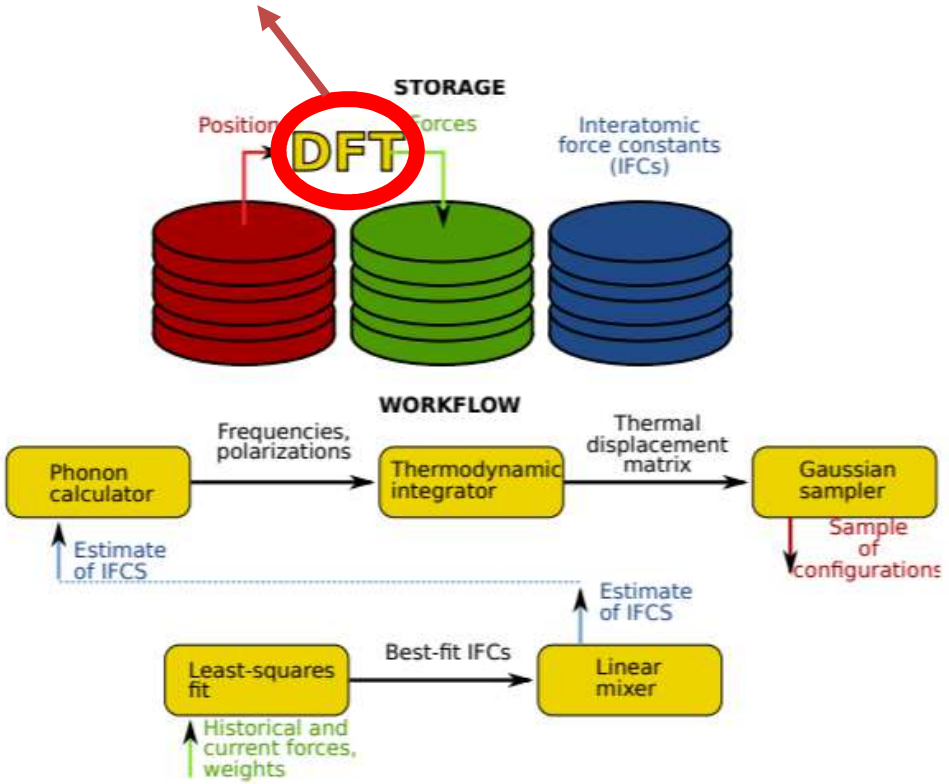


Gubaev et al., Comput. Mater. Sci 156, 148 (2019)

Speeding up QSCAILD using ML potentials

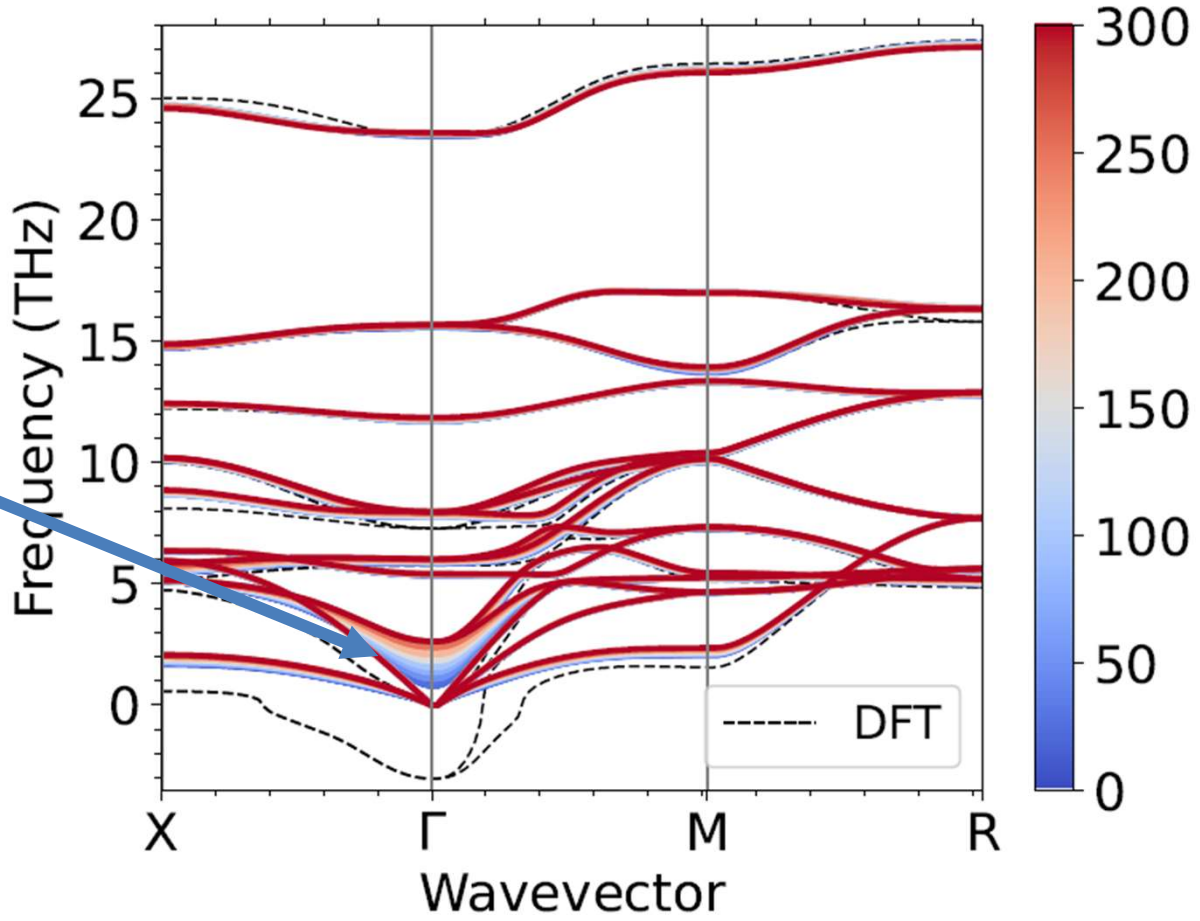


Bottleneck! -> DFT + active learning



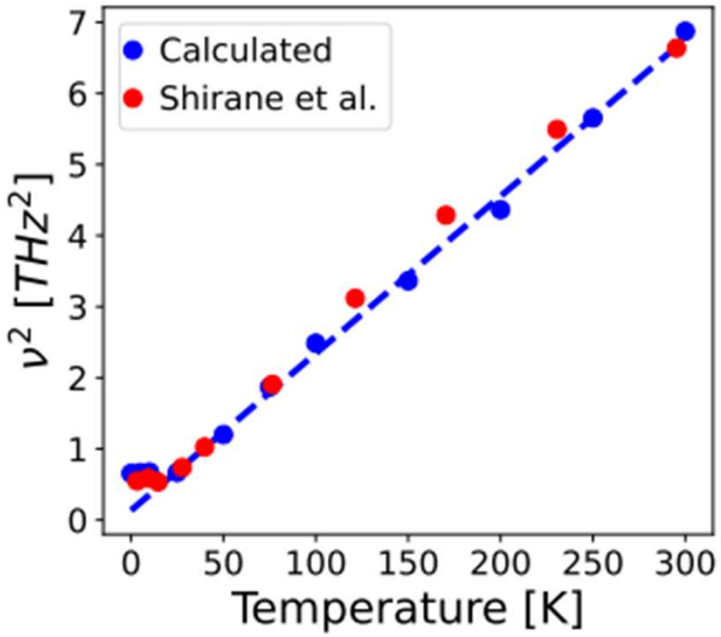
Results: Temperature dependent phonon spectrum of KTaO3

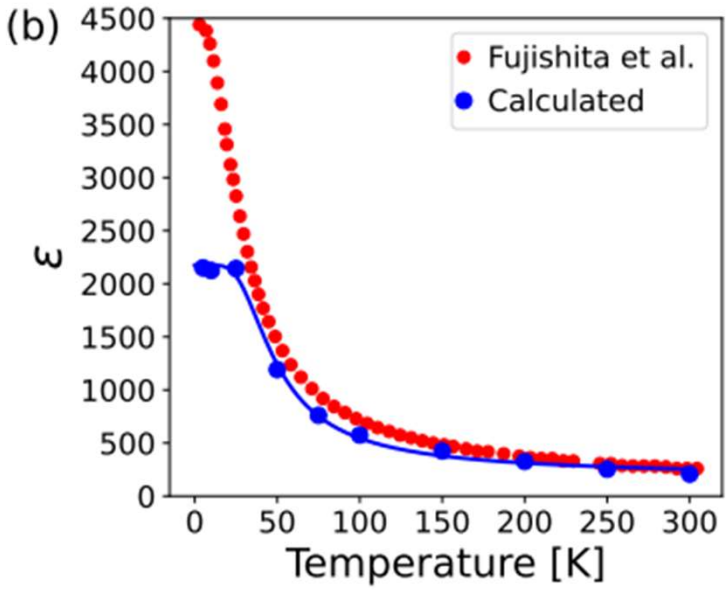
Temperature dependence mostly concentrated on the ferroelectric soft mode



Meier, Mingo and van Roekeghem, arXiv:2206.08296 (2022)

Phonon frequencies of the soft mode and dielectric constant





Excellent agreement of temperature dependent phonon frequencies with experiment

Small difference in temperature of saturation

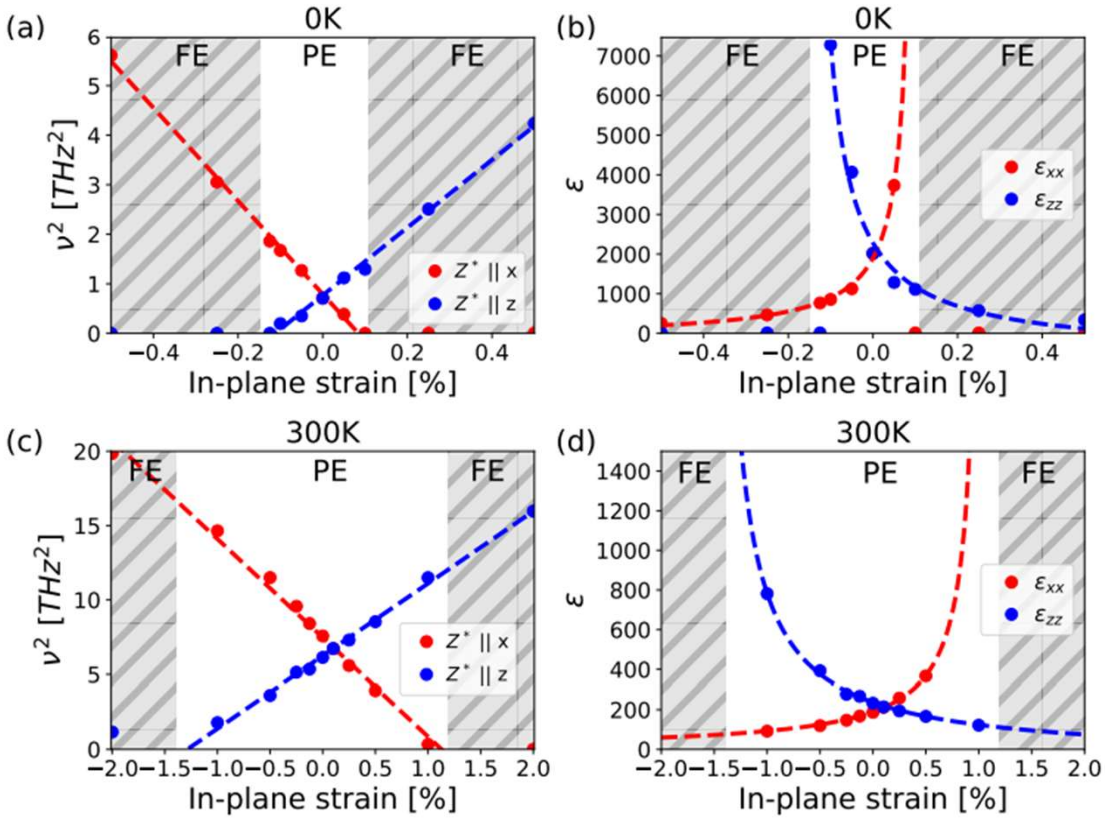
G. Shirane, R. Nathans, and V. J. Minkiewicz
Phys. Rev. **157**, 396 (1967)

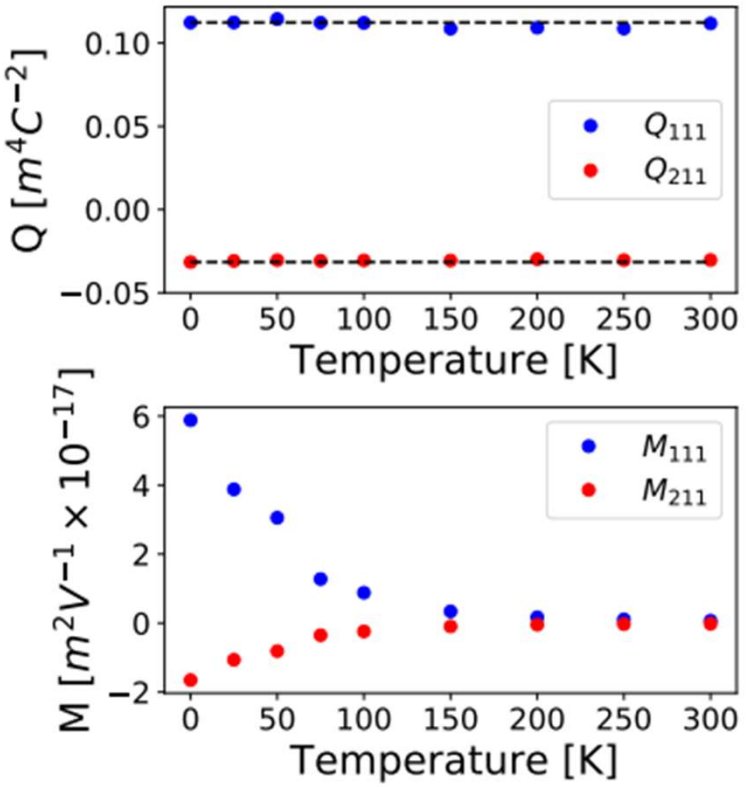
Temperature dependent dielectric constant compared to experiment, excellent agreement up to 25K and observation of Barrett law

J. Barrett, Phys. Rev. 86, 118 (1952)

Meier, Mingo and van Roekeghem, arXiv:2206.08296 (2022)

Temperature-dependent electrostrictive properties





$$x_i = Q_{ikl} P_k P_l$$
$$x_i = M_{ikl} E_i E_k$$
$$M_{ikk} = Q_{ikk} \chi^2_{kk}$$

Meier, Mingo and van Roekeghem, arXiv:2206.08296 (2022)

Conclusions

Predicting vibrational properties and thermal conductivities in a high throughput fashion is nowadays accessible, with satisfying agreement with experiment.

The interplay between theoretical predictions and synthesis of actual compounds is still a major issue.

ML interatomic potentials with DFT accuracy open new possibilities.

Thank you!

And thanks to:

Quintin MEIER
Fleur LEGRAIN
Mauro FAVA
Jesús CARRETE
Stefano CURTAROLO
Alexander SHAPEEV
Georg MADSEN
Natalio MINGO

Commissariat à l'énergie atomique et aux énergies alternatives
17 rue des Martyrs | 38054 Grenoble Cedex
www-liten.cea.fr