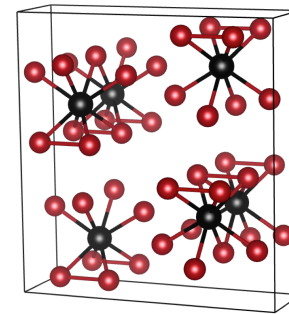
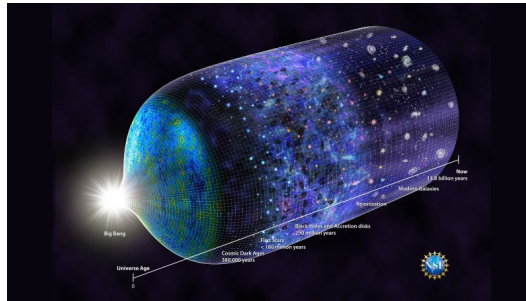


Optical Phonon R&D

Or

What Condensed Matter Theory/Computation Can do For You

Light years

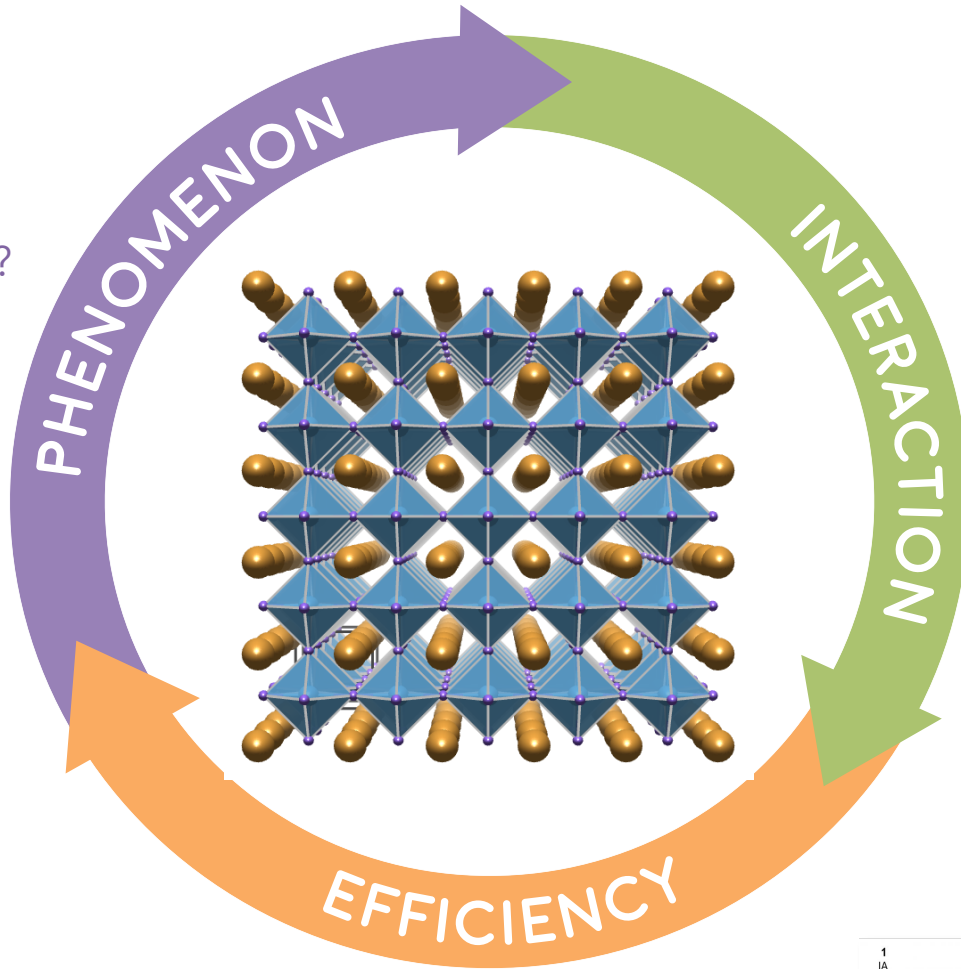
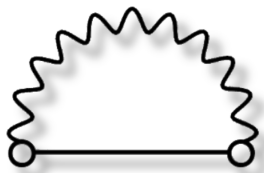


Nanometers

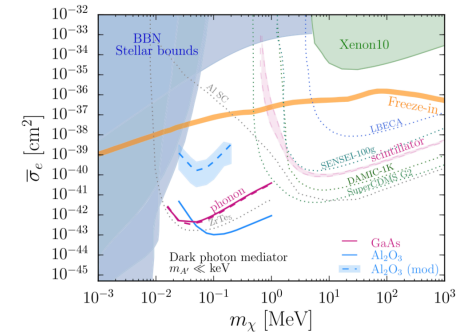


Target Theory and Design

What phenomena?
What materials?



Strength of mechanism
Reach estimates



DFT+beyond (Density functional theory)
Symmetry analysis
Effective Hamiltonians

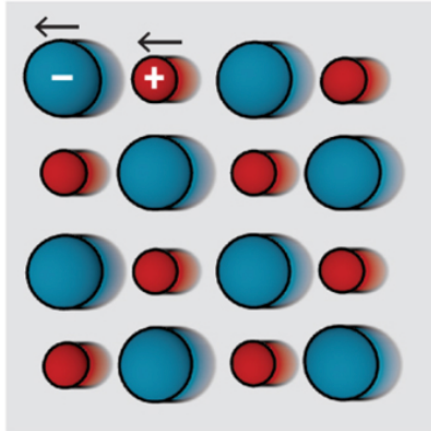
Target efficiency
Optimize interaction/reach

PERIODIC TABLE

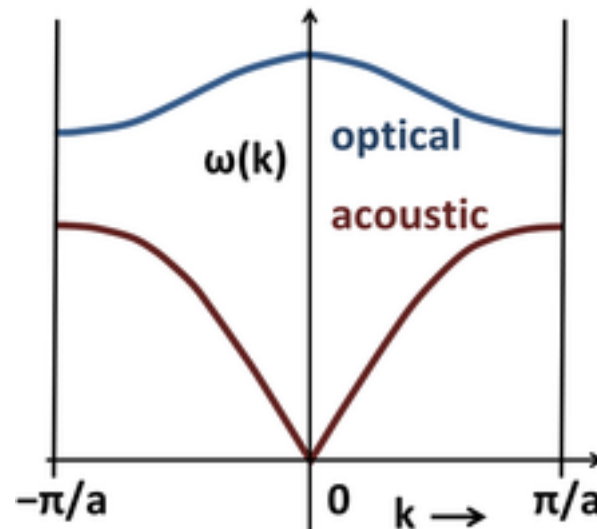
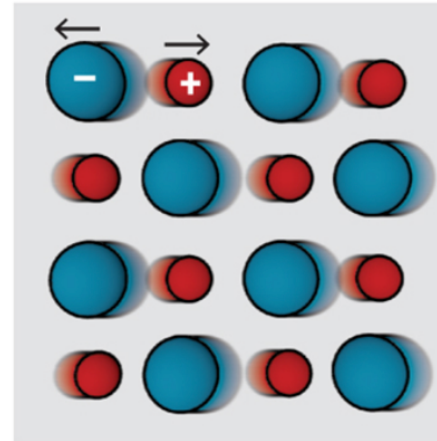
1 IA											14 IVA	Group IUPAC	
H Hydrogen 1.0079											C Carbon 12.011	Group CAS	
Li Lithium 6.941	Be Beryllium 9.012												Selected Oxidation States
Na Sodium 22.990	Mg Magnesium 24.305												Atomic Mass
		3 IIIB	4 IVB	5 VB	6 VIB	7 VIIB	8	9 VIII					
K Potassium 39.098	Ca Calcium 40.078	Sc Scandium 44.956	Ti Titanium 47.867	V Vanadium 50.942	Cr Chromium 51.996	Mn Manganese 54.938	Fe Iron 55.845	Co Cobalt 58.933					
Rh Rhodium 101.07	Sr Strontium 87.62	Y Yttrium 88.906	Zr Zirconium 91.224	Nb Niobium 92.906	Mo Molybdenum 95.94	Tc Technetium 98.906	Ru Ruthenium 101.07	Rh Rhodium 101.07					

Phonons are Lattice Vibrations

Acoustic Phonons



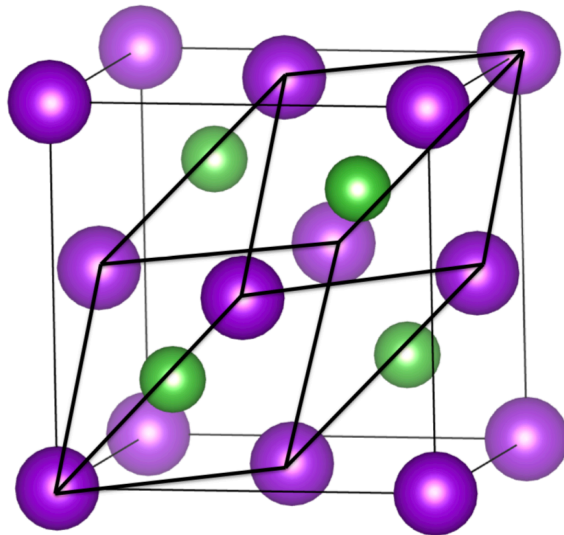
Optical Phonons



What are Polar Materials?

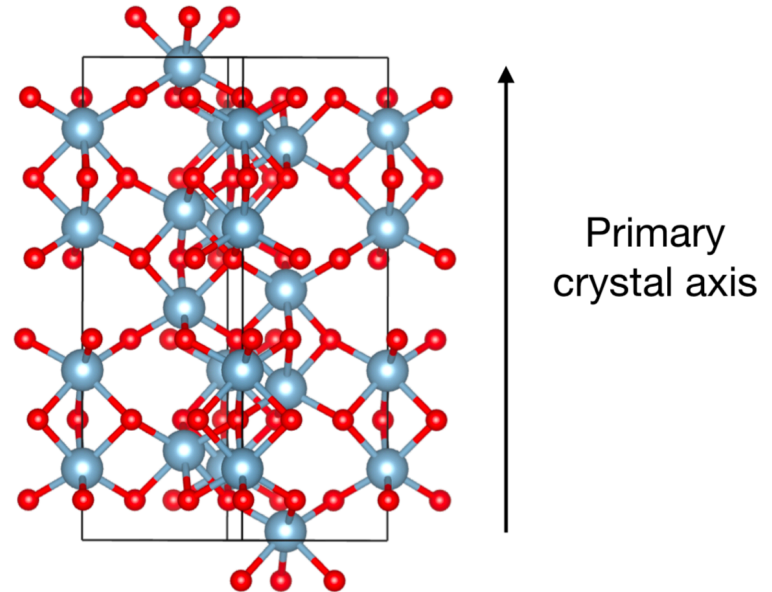
- At least two different ionic species
- Local dipole moment

GaAs



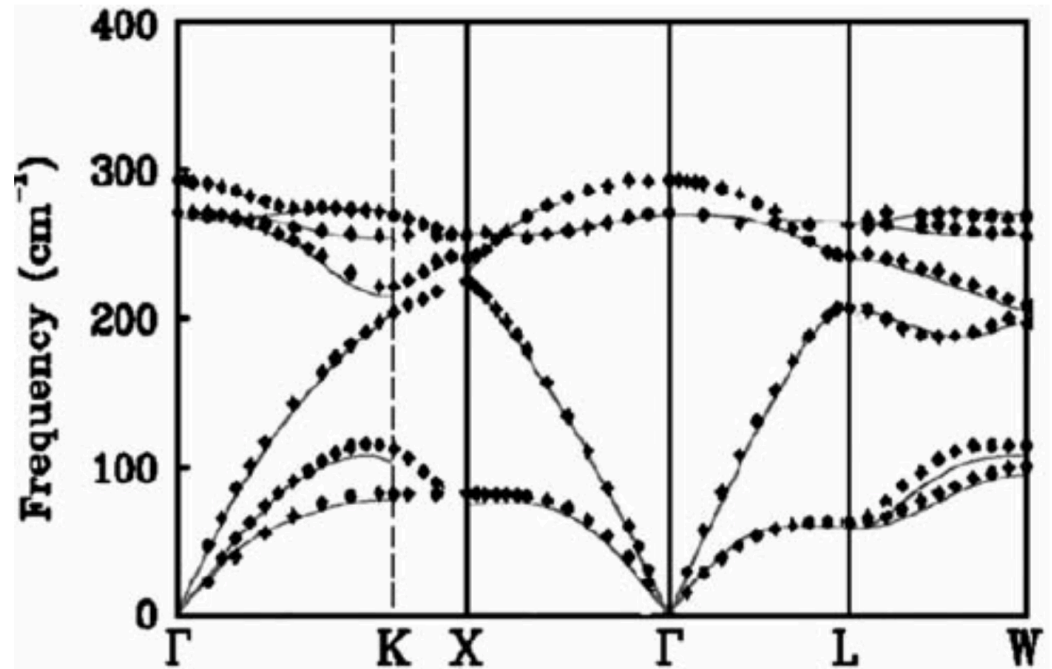
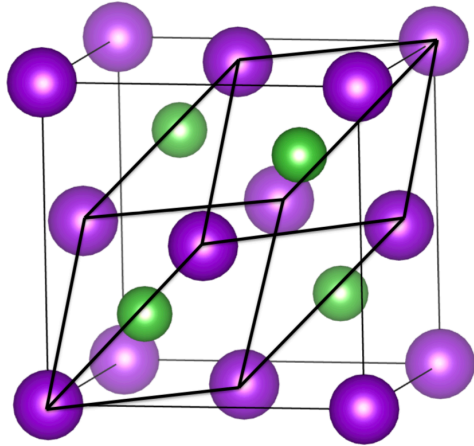
2 atoms in primitive cell

Al_2O_3 (Sapphire)



10 atoms in primitive cell

Why Polar Materials?



From Strauch & Dorner, JPCM 2 1457 (1990)

1. Kinematic matching with optical phonons
2. Small screening (insulating or semiconducting)
3. Can have anisotropic crystal structure (directional detection)
4. High-quality crystals available now

Discussing DM absorption via dark phonon with optical phonons

Fröhlich Hamiltonian – DM/phonon interaction

$$H = i \frac{\kappa e^2}{V} \sum_{j,v,q} \sum_{\mathbf{G} \neq \mathbf{q}} \frac{1}{\sqrt{2Nm_j \omega_{v,q}}} \frac{(\mathbf{q} + \mathbf{G}) \cdot \mathbf{Z}_j \cdot \mathbf{e}_{j,u}(\mathbf{q})}{(\mathbf{q} + \mathbf{G}) \cdot \boldsymbol{\varepsilon}_{\text{inf}} \cdot (\mathbf{q} + \mathbf{G})} e^{i(\mathbf{q} + \mathbf{G}) \cdot (\mathbf{r} + \boldsymbol{\tau}_j)}$$

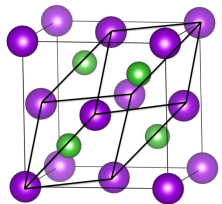
Sum over BZ (points to the summation indices)

Born effective charge (points to \mathbf{Z}_j)

Phonon energy (points to $\omega_{v,q}$)

Phonon eigenvectors (points to $\mathbf{e}_{j,u}(\mathbf{q})$)

High-frequency dielectric constant (points to $\boldsymbol{\varepsilon}_{\text{inf}}$)



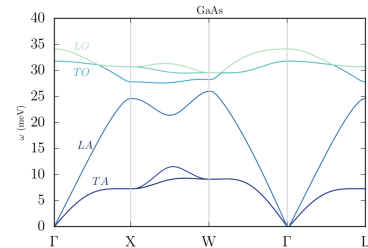
VASP

DFT
E, wavefunction



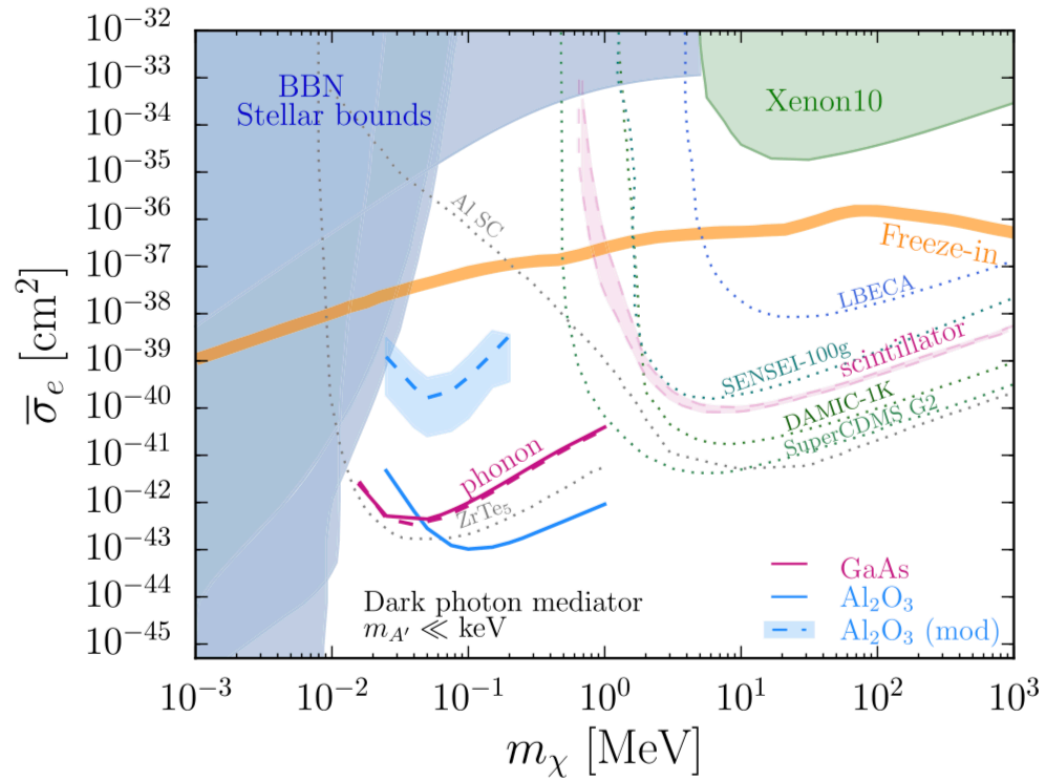
phonopy

Force constant
matrix



Theoretical reach of optical phonons

1kg year exposure

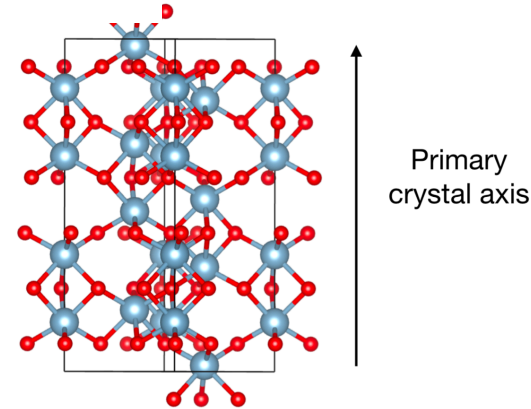


Tongyan Lin, UCSD

Simon Knapen, IAS

Kathryn Zurek, LBL

Hidden bonus:
directionality!



Can we do better?

$$Q = \frac{Z^{*2}}{A_1 A_2 \epsilon_\infty^2 \omega_{LO}}$$

Z*= Born effective charges

A₁, A₂= atomic mass numbers

ϵ_{inf} = high-frequency dielectric constant

w_{LO} = optical phonon frequency

	Z*	A ₁	A ₂	Eps_inf	w _{LO} (meV)	Q (E-7)
GaAs	2.27	69.7	74.9	10.89 (14.8)	25, 35	2.4
Al ₂ O ₃	2.98, 1.34	27.0	16.0	3.3	30, 106	80



Kevin Zhang, LBL

Tanner Trickle, LBL

Kathryn Zurek, LBL

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ω_{LO} = optical phonon frequency

Smaller ions?

A periodic table of elements with red circles highlighting Lithium (Li) and Fluorine (F). The table is color-coded by groups: Group 1 (yellow), Group 2 (orange), Groups 3-10 (brown), Group 11 (pink), Group 12 (grey), Groups 13-16 (green), Group 17 (cyan), and Group 18 (blue). The highlighted elements are in the first and second columns of the second row.

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	Xe		
Cs	Ba	*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn		
Fr	Ra	**	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og		

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Al ₂ O ₃	2.98, 1.34	27.0	16.0	3.3	30, 106	80
LiF	1.05	6.9	19.0	2.02	77	268

Can we do better?

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ϵ_{inf} = high-frequency dielectric constant

ω_{LO} = optical phonon frequency

$$Z^* = \frac{V}{e} \frac{\partial P}{\partial u}$$

Materials with large electric polarization?

Can we do better?

$$Q = \frac{Z^{*2}}{A_1 A_2 \epsilon_\infty^2 \omega_{LO}}$$

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A_1, A_2 = atomic mass numbers

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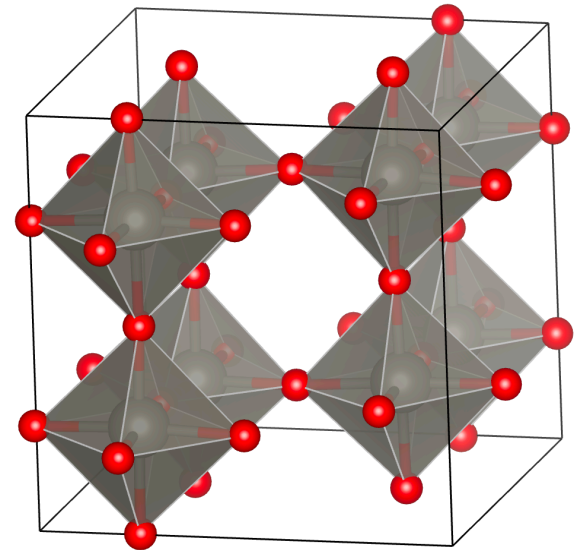
$$Z^* = \frac{V}{e} \frac{\partial P}{\partial u}$$

Materials with large electric polarization?

WO₃ (in P4mm structure)

Calculated polarization of **69 $\mu\text{C}\cdot\text{cm}^{-2}$** (high)

Hamdi et al. PRB 94, 245124 (2016)



Can we do better?

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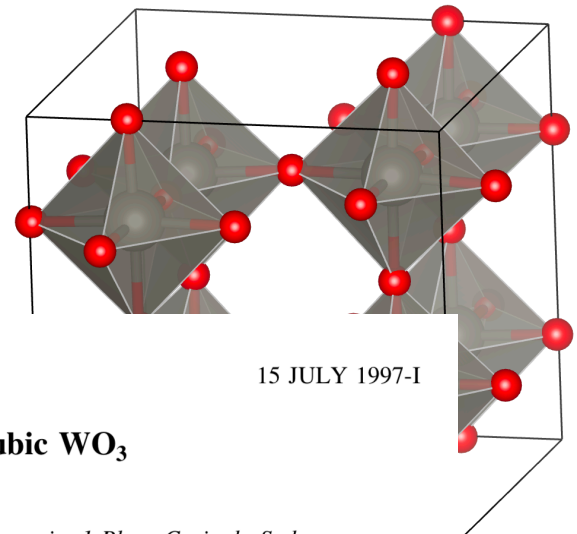
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Materials with large electric polarization?

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Hamdi et al. PRB 94, 245124 (2016)



PHYSICAL REVIEW B

VOLUME 56, NUMBER 3

15 JULY 1997-I

Anomalously large Born effective charges in cubic WO₃

F. Detraux, Ph. Ghosez, and X. Gonze

Unité de Physico-Chimie et de Physique des Matériaux, Université Catholique de Louvain, 1 Place Croix du Sud,

B-1348 Louvain-la-Neuve, Belgium

(Received 15 January 1997)

Can we do better?

$$Q = \frac{Z^{*2}}{A_1 A_2 \epsilon_\infty^2 \omega_{LO}}$$

Z*= Born effective charges

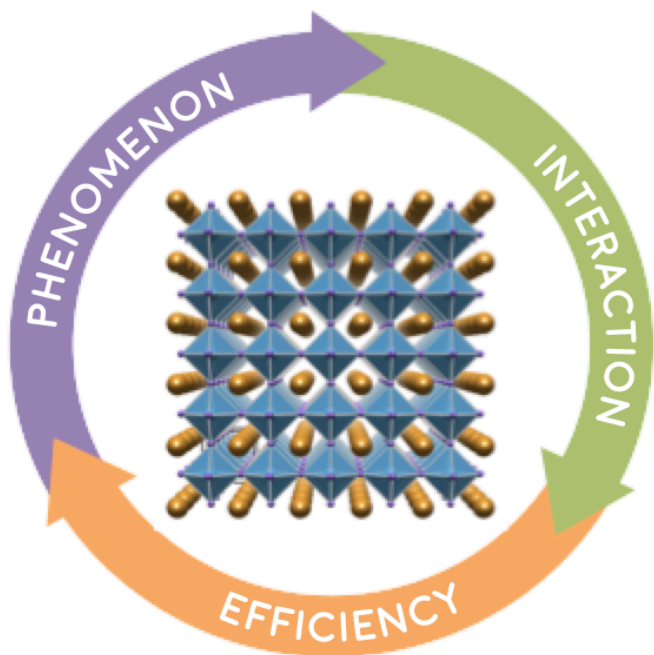
A₁, A₂= atomic mass numbers

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Al ₂ O ₃	2.98, 1.34	27.0	16.0	3.3	30, 106	80
LiF	1.05	6.9	19.0	2.02	77	268
WO ₃	11.73, 8.78, 1.62	183.8	16.0	5	126	556

Optical Phonon Theory and Design – Future



Multi-phonon production

Lin, Knapen

Harrelson, Trickle, Zhang, Zurek

Phonon lifetimes

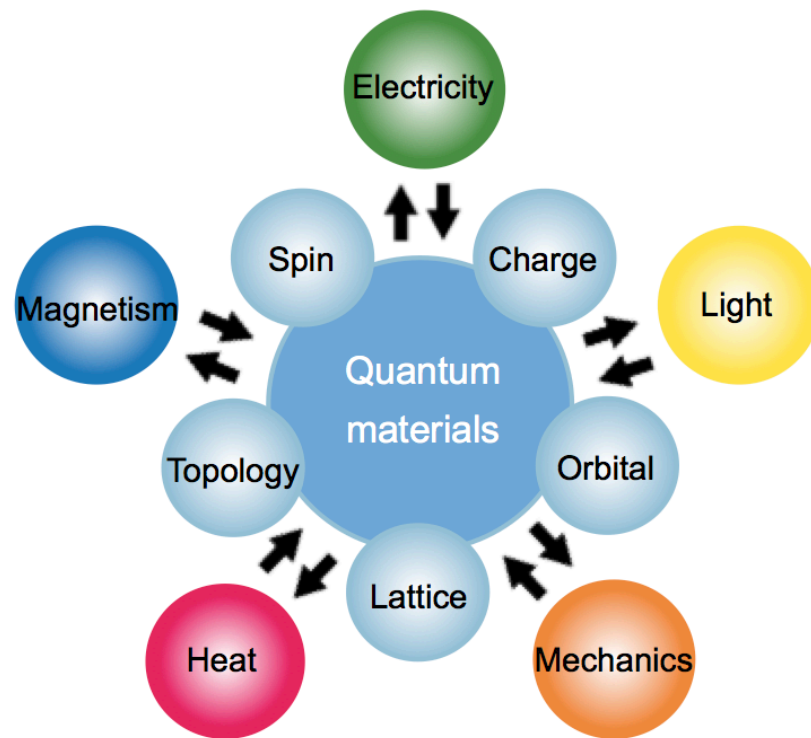
Harrelson

Better polar semiconductors

Theory and experiment

Ferroelectrics

Lin, Knapen





Characterization		Fabrication	Theory	Synthesis		
NCEM National Center for Electron Microscopy	Floor 1 Imaging and Manipulation of Nanostructures	Floor 2 Nano-fabrication	Floor 3 Theory of Nano-structured Materials	Floor 4 Inorganic Nano-structures	Floor 5 Biological Nano-structures	Floor 6 Organic and Macro-molecular Synthesis
Electron microscopy and nano-characterization	Characterization and manipulation of nanostructures	Advanced lithographic and thin-film processing techniques	Guiding understanding of new principles, behavior and experiments	Science of semiconductor, carbon and hybrid nanostructures	Bio-materials; new probes for bio-imaging; synthetic biology techniques	Soft materials: organics, macromolecules, polymers and their assemblies