

NATIONAL
CENTRE
FOR NUCLEAR
RESEARCH
ŚWIERK

NOMATEN

Centre of Excellence in Multifunctional Materials
for Industrial and Medical Applications

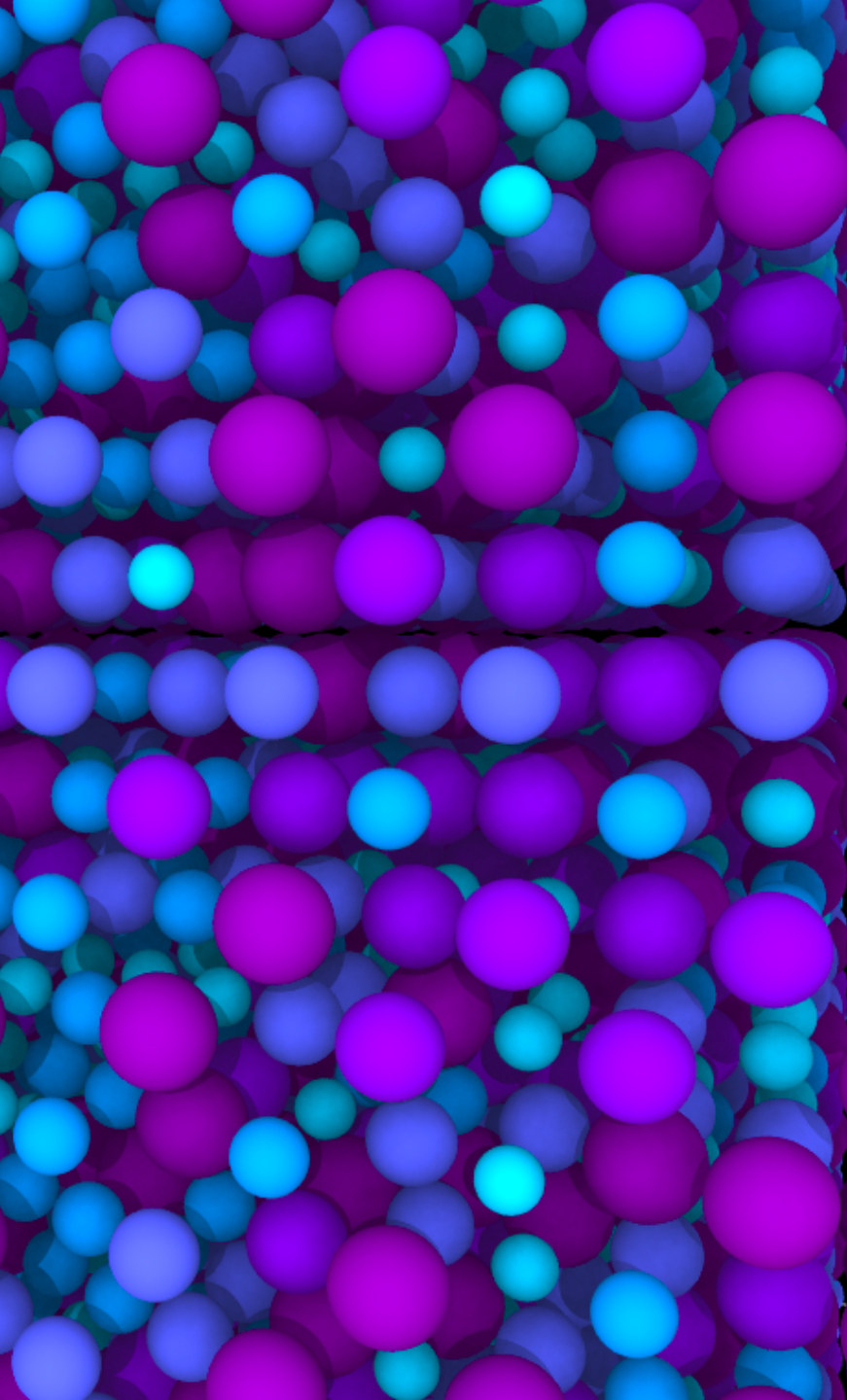
QUASI-LOCALIZED MODES IN CRYSTALLINE HIGH ENTROPY ALLOYS

Silvia Bonfanti

NOMATEN

Interaction, Disorder, Elasticity workshop – École de Physique des Houches – April 2-7, 2023





Outline

- High Entropy Alloys
- Microscopic Structure
- Vibrational Properties
- Disorder
- Future perspectives

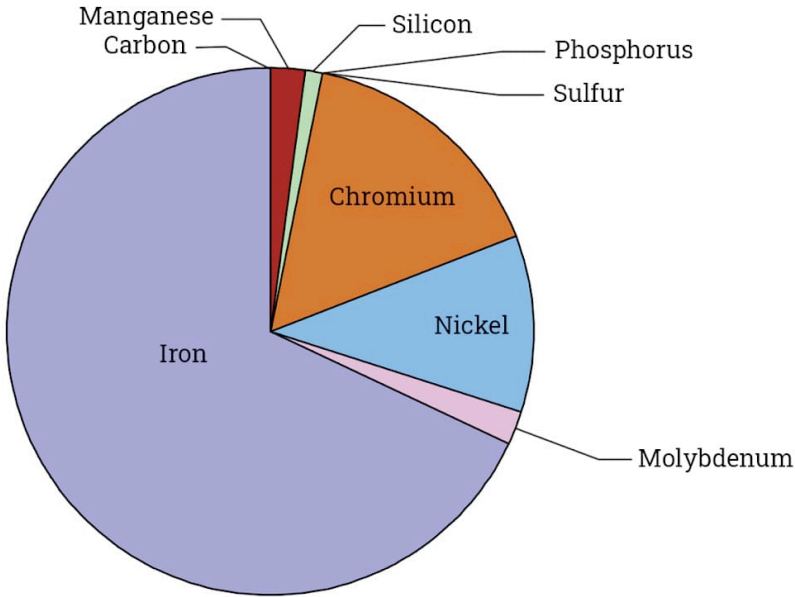
TRADITIONAL ALLOYS

ONE/TWO MAJOR METALLIC ELEMENTS
+ SMALLER AMOUNT OF MINOR ELEMENTS

Different alloy families



E.g.: Stainless Steel Composition



HIGH ENTROPY ALLOYS (2004)

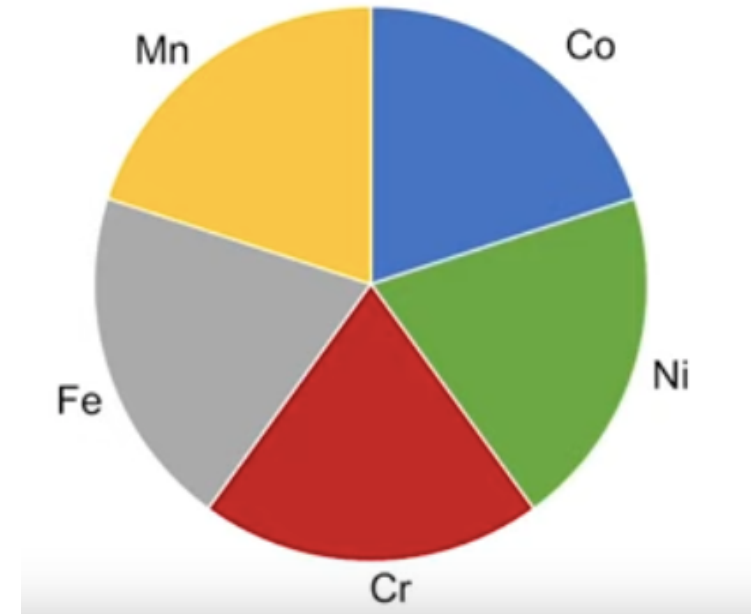
THE IDEA:
MIX SEVERAL (>4) SPECIES
IN SAME PROPORTION!



PROF. YEH @ UNI TAIWAN



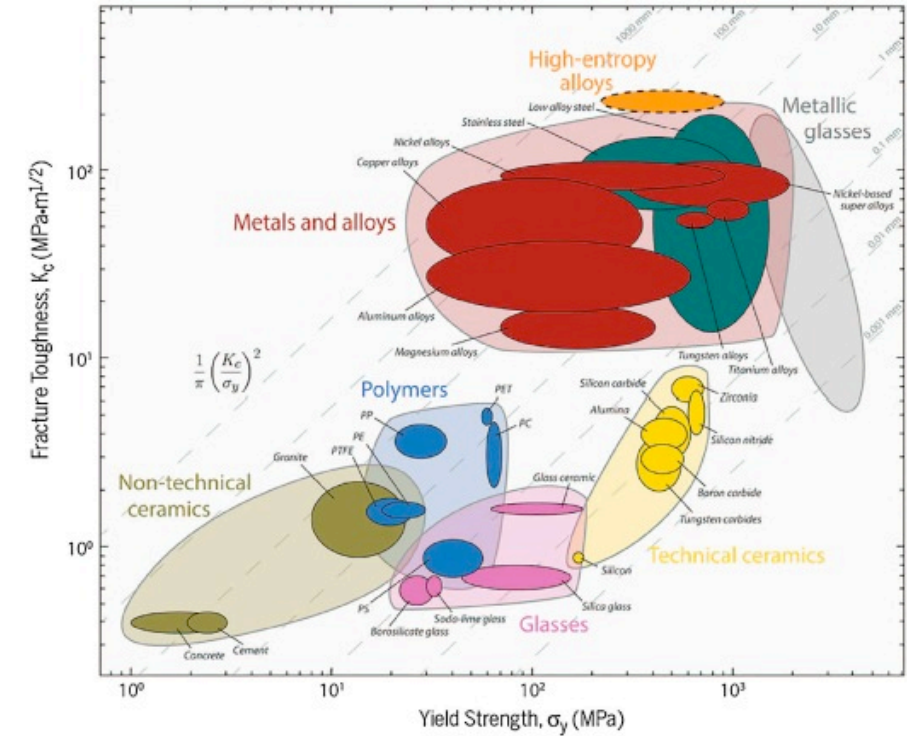
PROF. CANTOR @ CALTECH



HIGH ENTROPY ALLOYS = IMPROVED PROPERTIES

PERIODIC TABLE OF ELEMENTS

1 H Hydrogen	PubChem																2 He Helium
3 Li Lithium	4 Be Beryllium	1 H Hydrogen Nonmetal										5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
11 Na Sodium	12 Mg Magnesium	Name Chemical Group Block										13 Al Aluminum	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
19 K Potassium	20 Ca Calcium	21 Sc Scandium	22 Ti Titanium	23 V Vanadium	24 Cr Chromium	25 Mn Manganese	26 Fe Iron	27 Co Cobalt	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
37 Rb Rubidium	38 Sr Strontium	39 Y Yttrium	40 Zr Zirconium	41 Nb Niobium	42 Mo Molybdenum	43 Tc Technetium	44 Ru Ruthenium	45 Rh Rhodium	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
55 Cs Cesium	56 Ba Barium		72 Hf Hafnium	73 Ta Tantalum	74 W Tungsten	75 Re Rhenium	76 Os Osmium	77 Ir Iridium	78 Pt Platinum	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
87 Fr Francium	88 Ra Radium		104 Rf Rutherfordium	105 Db Dubnium	106 Sg Seaborgium	107 Bh Bohrium	108 Hs Hassium	109 Mt Meitnerium	110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson
		57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium	
		89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium	



50 METALS:
INFINITE POSSIBILITIES
OF COMBINATION

HIGH ENTROPY OF MIXING
ALTER THE INTERACTION
OF ELEMENTS:
COCKTAIL EFFECT

MECHANICAL
THERMODYNAMIC
CORROSION RESISTANCE...

HIGH ENTROPY ALLOYS = IMPROVED PROPERTIES

PERIODIC TABLE OF ELEMENTS

PubChem																		
1 Atomic Number																		
H Symbol																		
Name																		
Chemical Group Block																		
1 H Hydrogen Nonmetal																	2 He Helium Noble gas	
3 Li Lithium Alkali metal	4 Be Beryllium Alkaline earth metal																	10 Ne Neon Noble gas
11 Na Sodium Alkali metal	12 Mg Magnesium Alkaline earth metal																	18 Ar Argon Noble gas
19 K Potassium Alkali metal	20 Ca Calcium Alkaline earth metal	21 Sc Scandium Transition metal	22 Ti Titanium Transition metal	23 V Vanadium Transition metal	24 Cr Chromium Transition metal	25 Mn Manganese Transition metal	26 Fe Iron Transition metal	27 Co Cobalt Transition metal	28 Ni Nickel Transition metal	29 Cu Copper Transition metal	30 Zn Zinc Transition metal	31 Ga Gallium Post-transition metal	32 Ge Germanium Metalloid	33 As Arsenic Metalloid	34 Se Selenium Nonmetal	35 Br Bromine Nonmetal	36 Kr Krypton Noble gas	
37 Rb Rubidium Alkali metal	38 Sr Strontium Alkaline earth metal	39 Y Yttrium Transition metal	40 Zr Zirconium Transition metal	41 Nb Niobium Transition metal	42 Mo Molybdenum Transition metal	43 Tc Technetium Transition metal	44 Ru Ruthenium Transition metal	45 Rh Rhodium Transition metal	46 Pd Palladium Transition metal	47 Ag Silver Transition metal	48 Cd Cadmium Transition metal	49 In Indium Post-transition metal	50 Sn Tin Post-transition metal	51 Sb Bismuth Post-transition metal	52 Te Tellurium Nonmetal	53 I Iodine Nonmetal	54 Xe Xenon Noble gas	
55 Cs Cesium Alkali metal	56 Ba Barium Alkaline earth metal		72 Hf Hafnium Transition metal	73 Ta Tantalum Transition metal	74 W Tungsten Transition metal	75 Re Rhenium Transition metal	76 Os Osmium Transition metal	77 Ir Iridium Transition metal	78 Pt Platinum Transition metal	79 Au Gold Transition metal	80 Hg Mercury Transition metal	81 Tl Thallium Post-transition metal	82 Pb Lead Post-transition metal	83 Bi Bismuth Post-transition metal	84 Po Polonium Nonmetal	85 At Astatine Nonmetal	86 Rn Radon Noble gas	
87 Fr Francium Alkali metal	88 Ra Radium Alkaline earth metal		104 Rf Rutherfordium Transition metal	105 Db Dubnium Transition metal	106 Sg Seaborgium Transition metal	107 Bh Bohrium Transition metal	108 Hs Hassium Transition metal	109 Mt Meitnerium Transition metal	110 Ds Darmstadtium Transition metal	111 Rg Roentgenium Transition metal	112 Cn Copernicium Transition metal	113 Nh Nihonium Post-transition metal	114 Fl Flerovium Post-transition metal	115 Mc Moscovium Post-transition metal	116 Lv Livermorium Nonmetal	117 Ts Tennessine Nonmetal	118 Og Oganesson Noble gas	
	57 La Lanthanum Lanthanide	58 Ce Cerium Lanthanide	59 Pr Praseodymium Lanthanide	60 Nd Neodymium Lanthanide	61 Pm Promethium Lanthanide	62 Sm Samarium Lanthanide	63 Eu Europium Lanthanide	64 Gd Gadolinium Lanthanide	65 Tb Terbium Lanthanide	66 Dy Dysprosium Lanthanide	67 Ho Holmium Lanthanide	68 Er Erbium Lanthanide	69 Tm Thulium Lanthanide	70 Yb Ytterbium Lanthanide	71 Lu Lutetium Lanthanide			
	89 Ac Actinium Actinide	90 Th Thorium Actinide	91 Pa Protactinium Actinide	92 U Uranium Actinide	93 Np Neptunium Actinide	94 Pu Plutonium Actinide	95 Am Americium Actinide	96 Cm Curium Actinide	97 Bk Berkelium Actinide	98 Cf Californium Actinide	99 Es Einsteinium Actinide	100 Fm Fermium Actinide	101 Md Mendelevium Actinide	102 No Nobelium Actinide	103 Lr Lawrencium Actinide			



50 METALS:
INFINITE POSSIBILITIES
OF COMBINATION

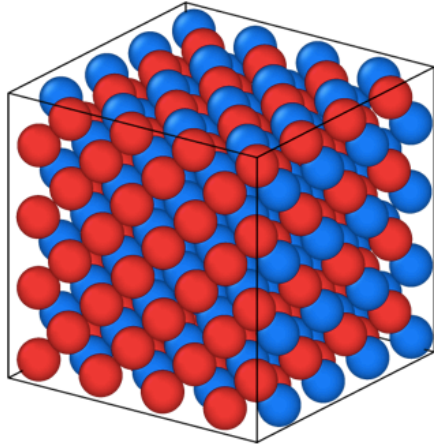
HIGH ENTROPY OF MIXING
ALTER THE INTERACTION
OF ELEMENTS:
COCKTAIL EFFECT

US: STRUCTURE-
PROPERTIES
HOW? MODELING (MD)

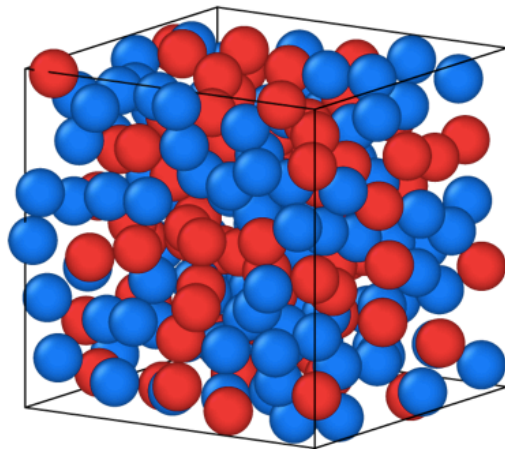
MICROSCOPIC STRUCTURE

CONVENTIONAL ALLOYS

CRYSTAL



GLASS

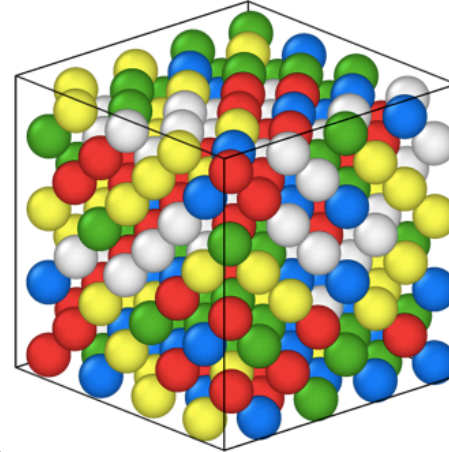
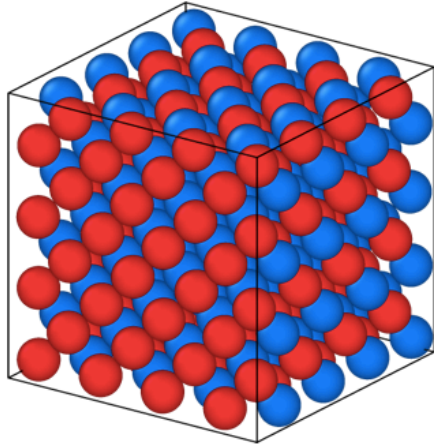


MICROSCOPIC STRUCTURE

CONVENTIONAL ALLOYS

HIGH ENTROPY ALLOYS

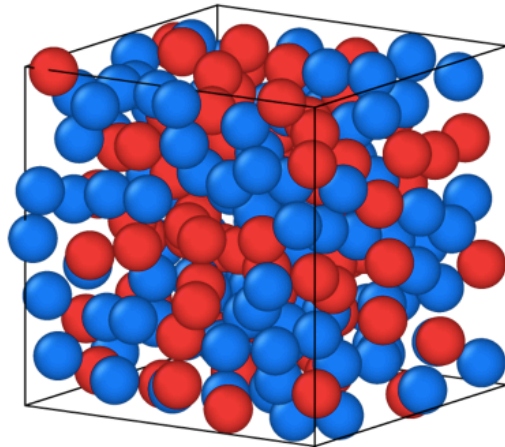
CRYSTAL



- POSITIONAL ORDER
- **COMPOSITIONAL DISORDER**

PARADIGM SHIFT

GLASS

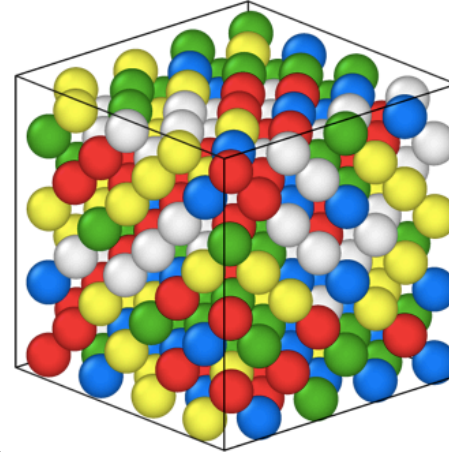
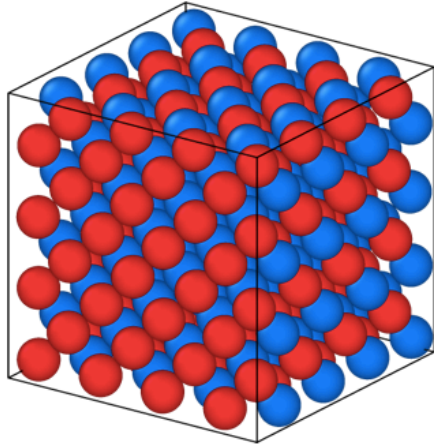


MICROSCOPIC STRUCTURE

CONVENTIONAL ALLOYS

HIGH ENTROPY ALLOYS

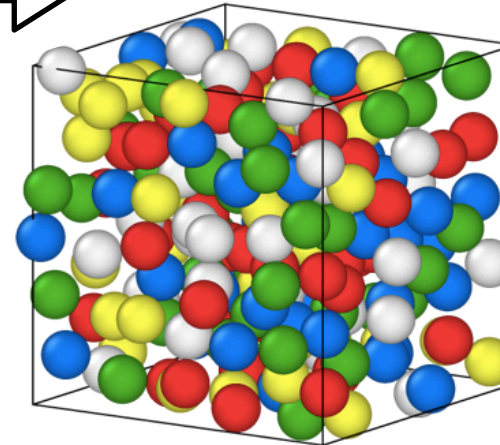
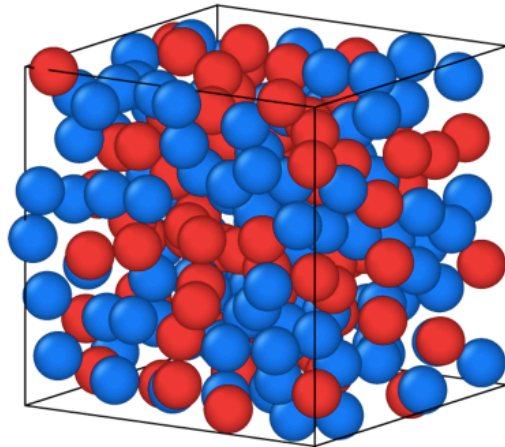
CRYSTAL



- POSITIONAL ORDER
- **COMPOSITIONAL DISORDER**

PARADIGM SHIFT

GLASS



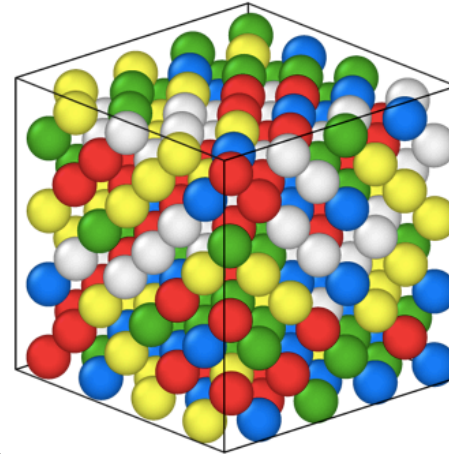
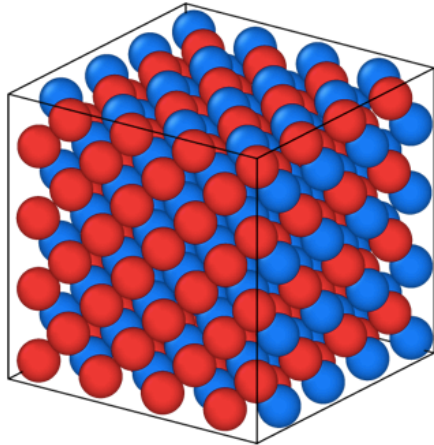
- POSITIONAL DISORDER

MICROSCOPIC STRUCTURE

CONVENTIONAL ALLOYS

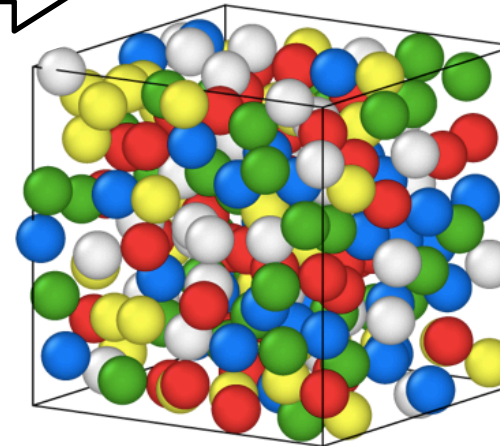
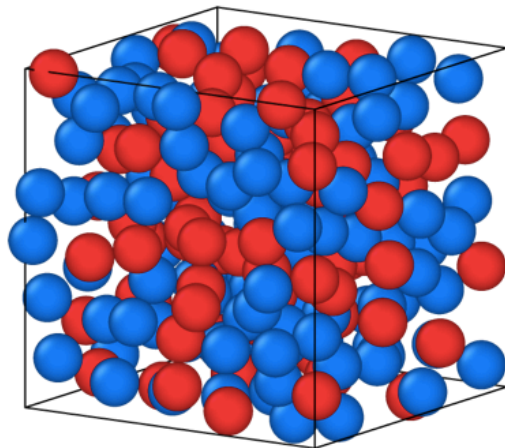
HIGH ENTROPY ALLOYS

CRYSTAL



PARADIGM SHIFT

GLASS



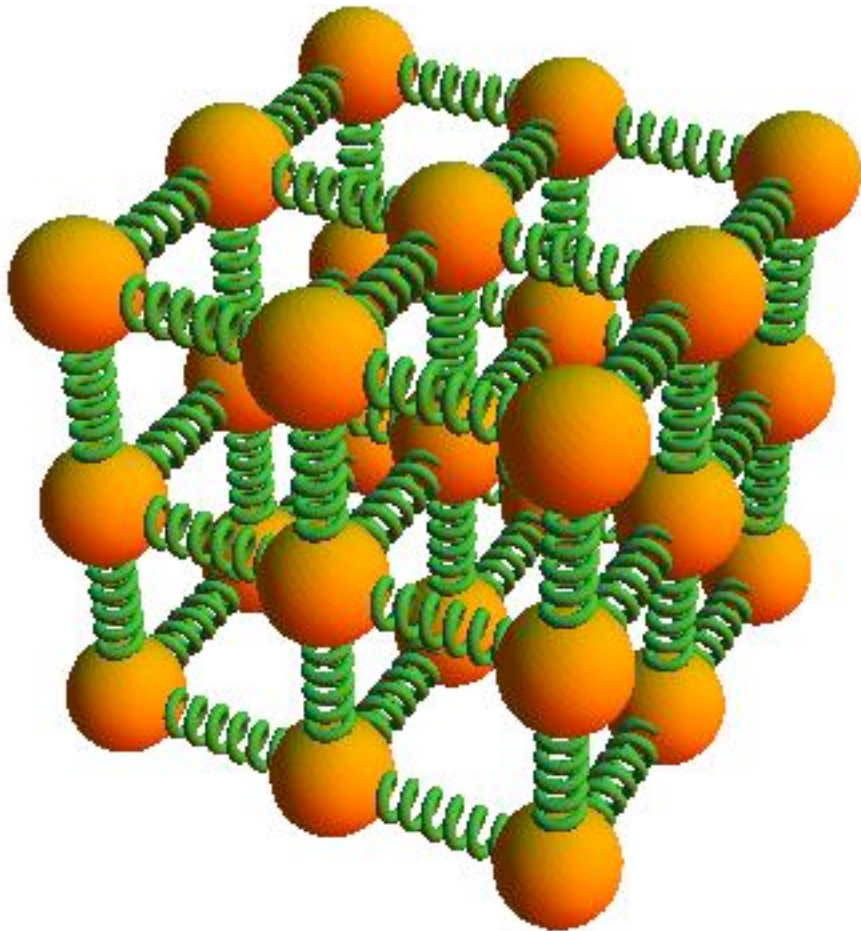
GLASSCOTT NAT. COMM. 2019

....

VIBRATIONAL PROPERTIES

VIBRATION OF CONFIG. AROUND MINIMA

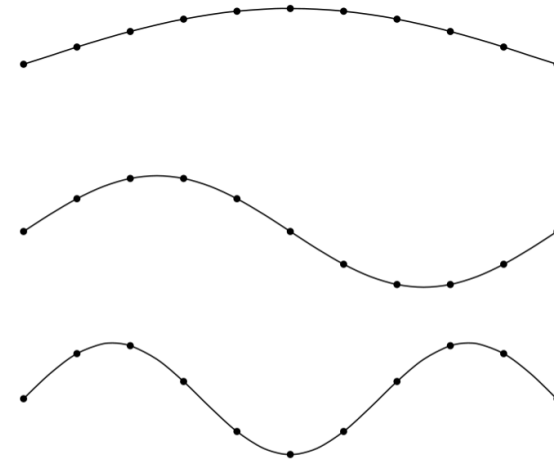
CRYSTALS, PERIODIC LATTICE:



DEBYE'S THEORY PREDICTS

$$D(\omega) \propto \omega^{d-1}$$

“EXTENDED” PHONON MODES





**VIBRATIONAL PROPERTIES
OF GLASSES: NON PERIODIC, AMORPHOUS?**

RECENT NUMERICAL STUDIES

$$D(\omega) = A\omega^{d-1} + B\omega^4$$

↑
PHONON MODES PREDICTED BY
THE DEBYE THEORY

↑
**NON-PHONON MODES ORIGINATE
FROM GLASSY CONFIGURATION**

LERNER, DURING, AND BOUCHBINDER, PRL 2016

MIZUNO, SHIBA, AND IKEDA, PNAS 2017

LERNER, AND BOUCHBINDER, J. CHEM. PHYS. 2021

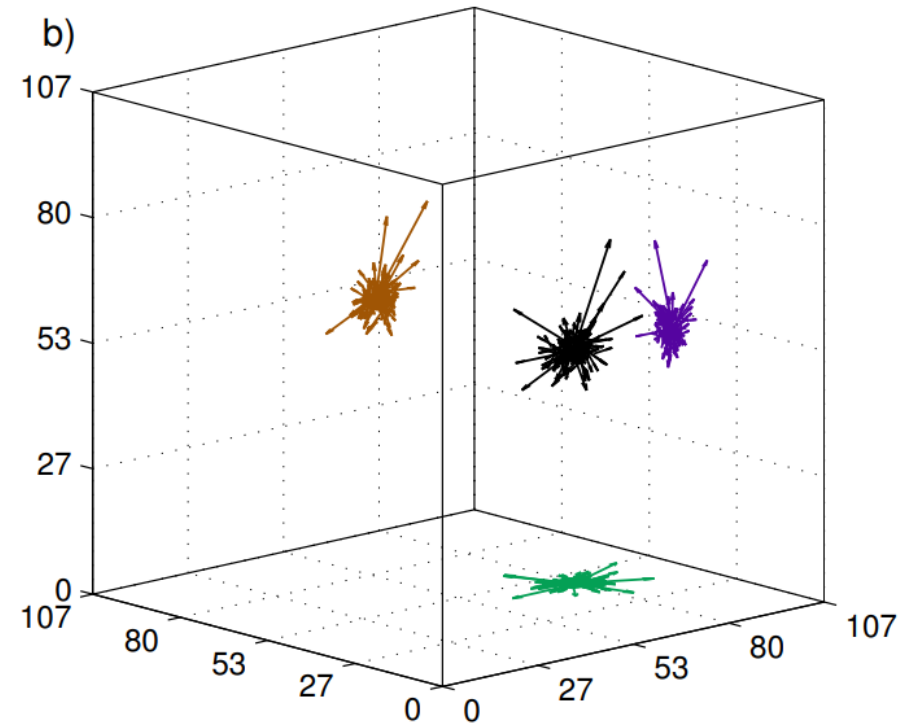
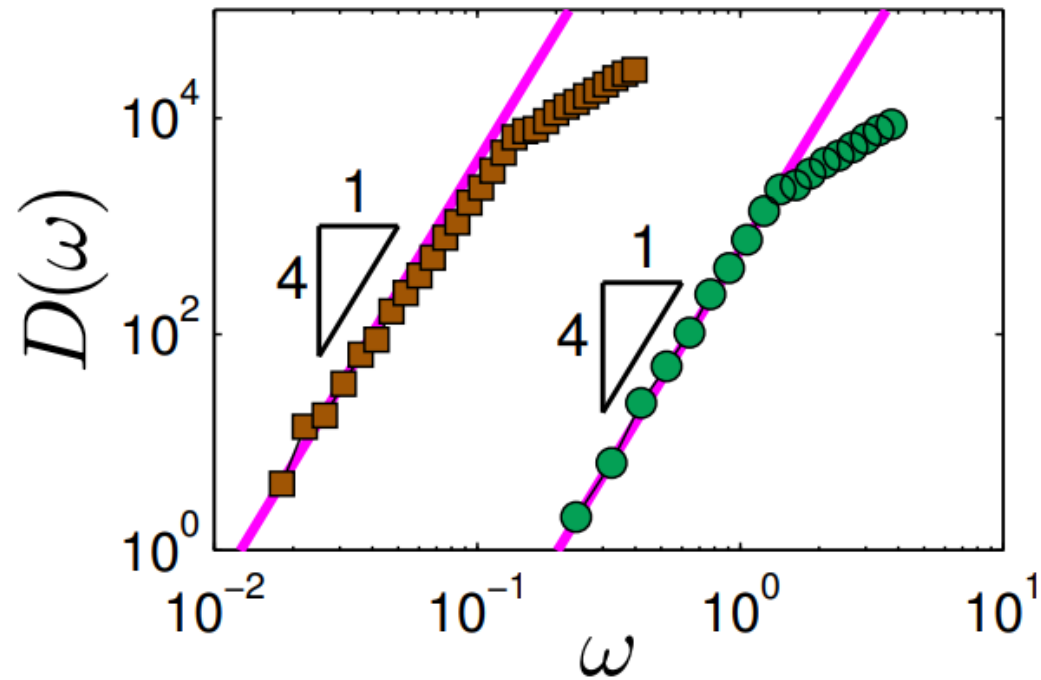
HYBRIDIZATION OF MODES!

HOW TO DISENTANGLE PHONON MODES AND NON-PHONON MODES?

RECENT NUMERICAL STUDIES

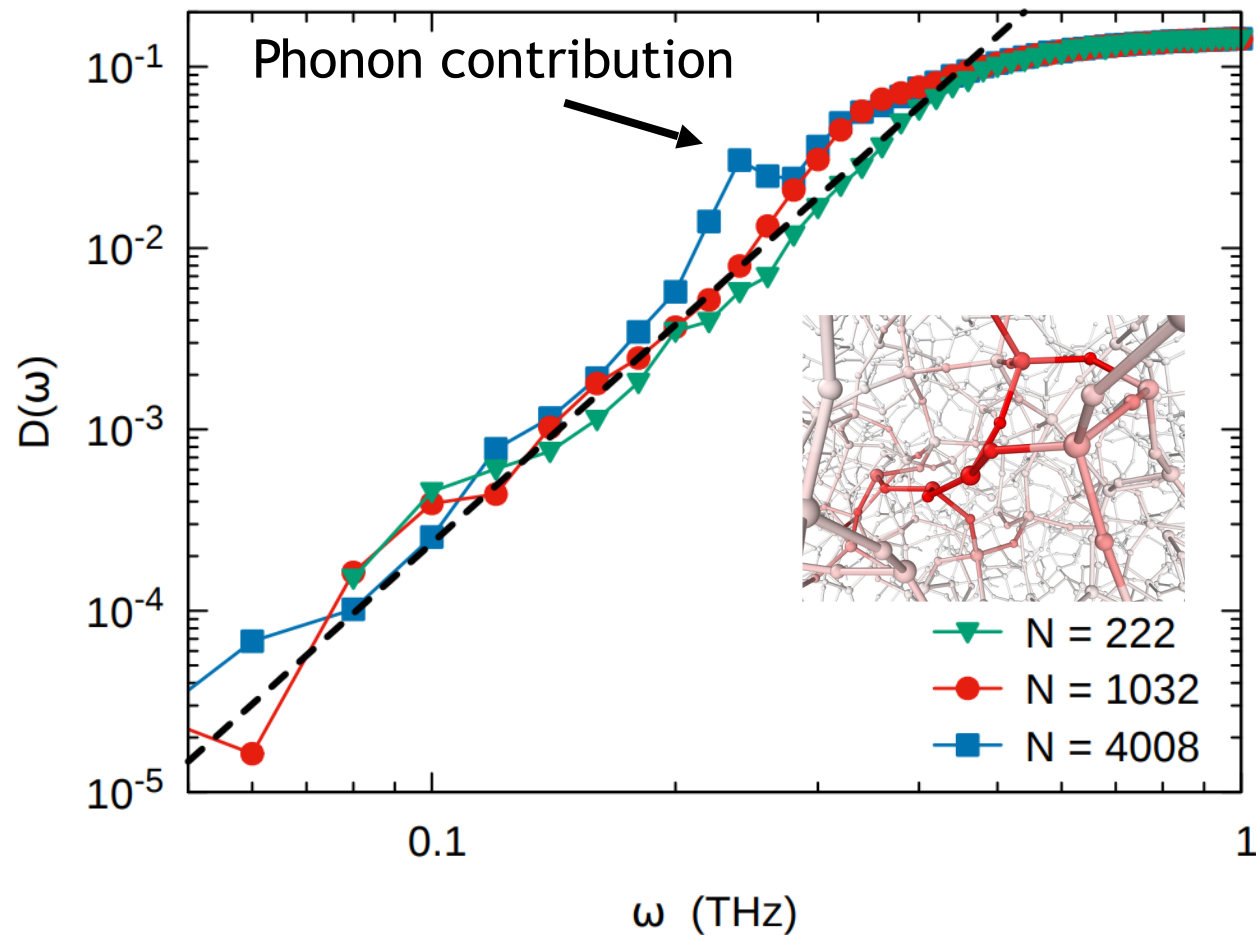
LERNER, DURING, AND BOUCHBINDER, PRL 2016

THE LOWEST PHONON MODE SCALES WITH L^{-1}



THE NON-PHONON MODES ARE MAINLY OBSERVED IN SIMPLE POTENTIAL SYSTEMS.
REALISTIC MODELS?

THE CASE OF SILICA GLASS



WE NUMERICALLY CONFIRMED $D(\omega) \propto \omega^4$

BONFANTI, GUERRA, MONDAL, PROCACCIA, & ZAPPERI, PRL 2020

THE CASE OF HEAS

WHAT IS THE ROLE OF [COMPOSITIONAL] DISORDER?

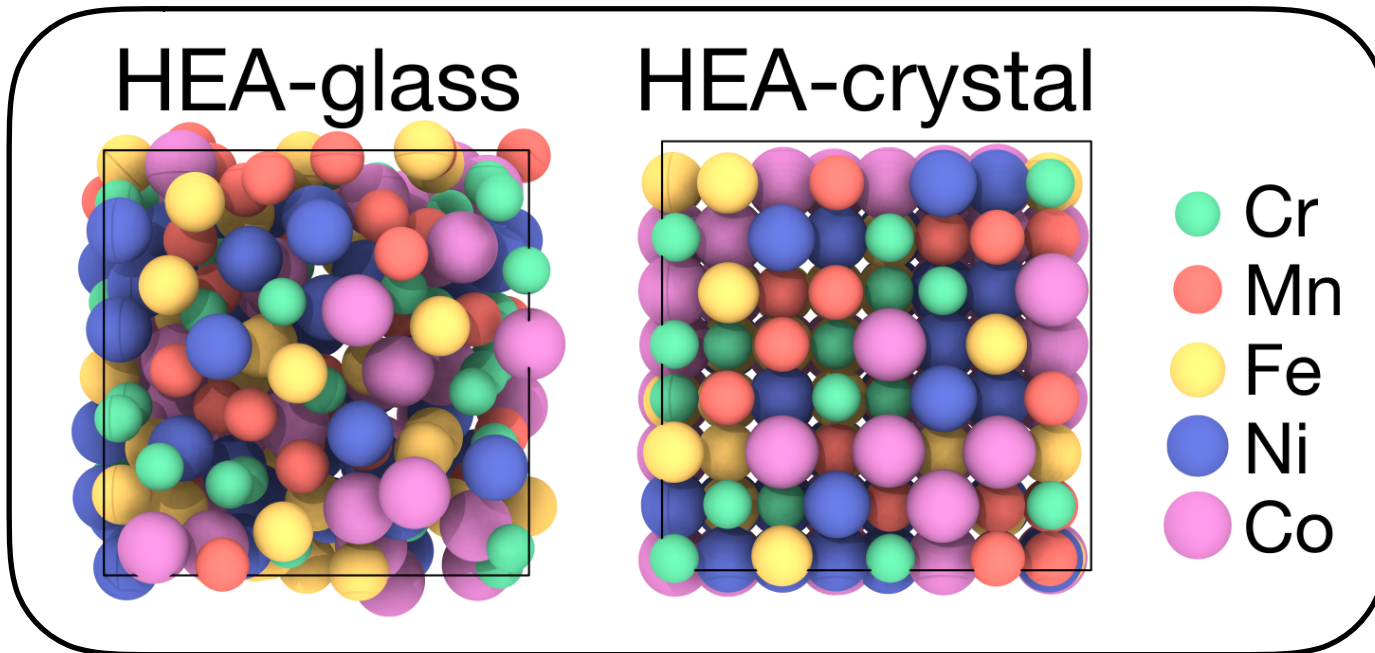
THE CASE OF HEAS

WHAT IS THE ROLE OF [COMPOSITIONAL] DISORDER?

MODEL OF DISORDERED BINARY ALLOYS:
LERNER, AND BOUCHBINDER, PRL 2022

THE CASE OF HEAS

WHAT IS THE ROLE OF [COMPOSITIONAL] DISORDER?

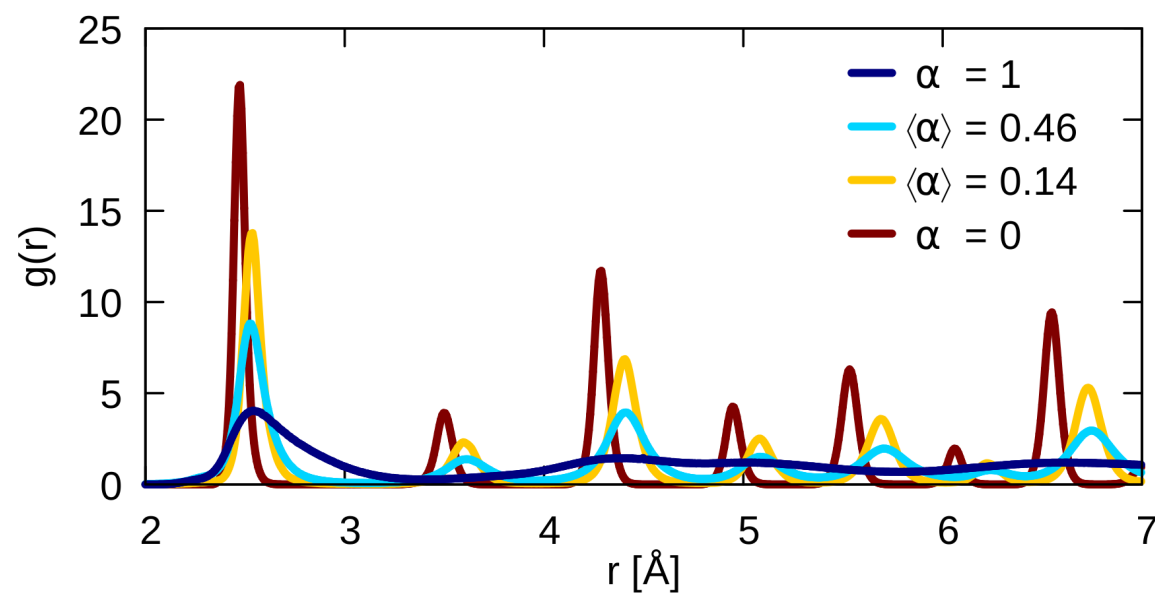
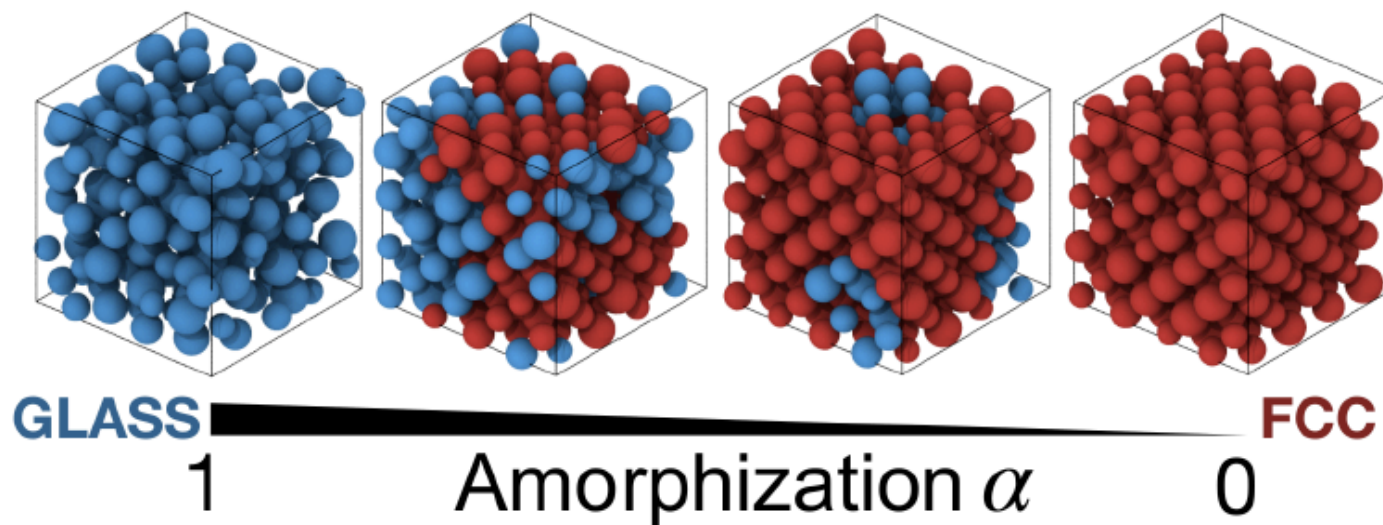


- CANTOR ALLOY
- REALISTIC POTENTIAL APPROPRIATE FOR METALLIC SYSTEMS: EAM POTENTIAL
- SMALL SYSTEM SIZE ~ 250 ATOMS
- VIBRATIONAL MODES CALCULATION:

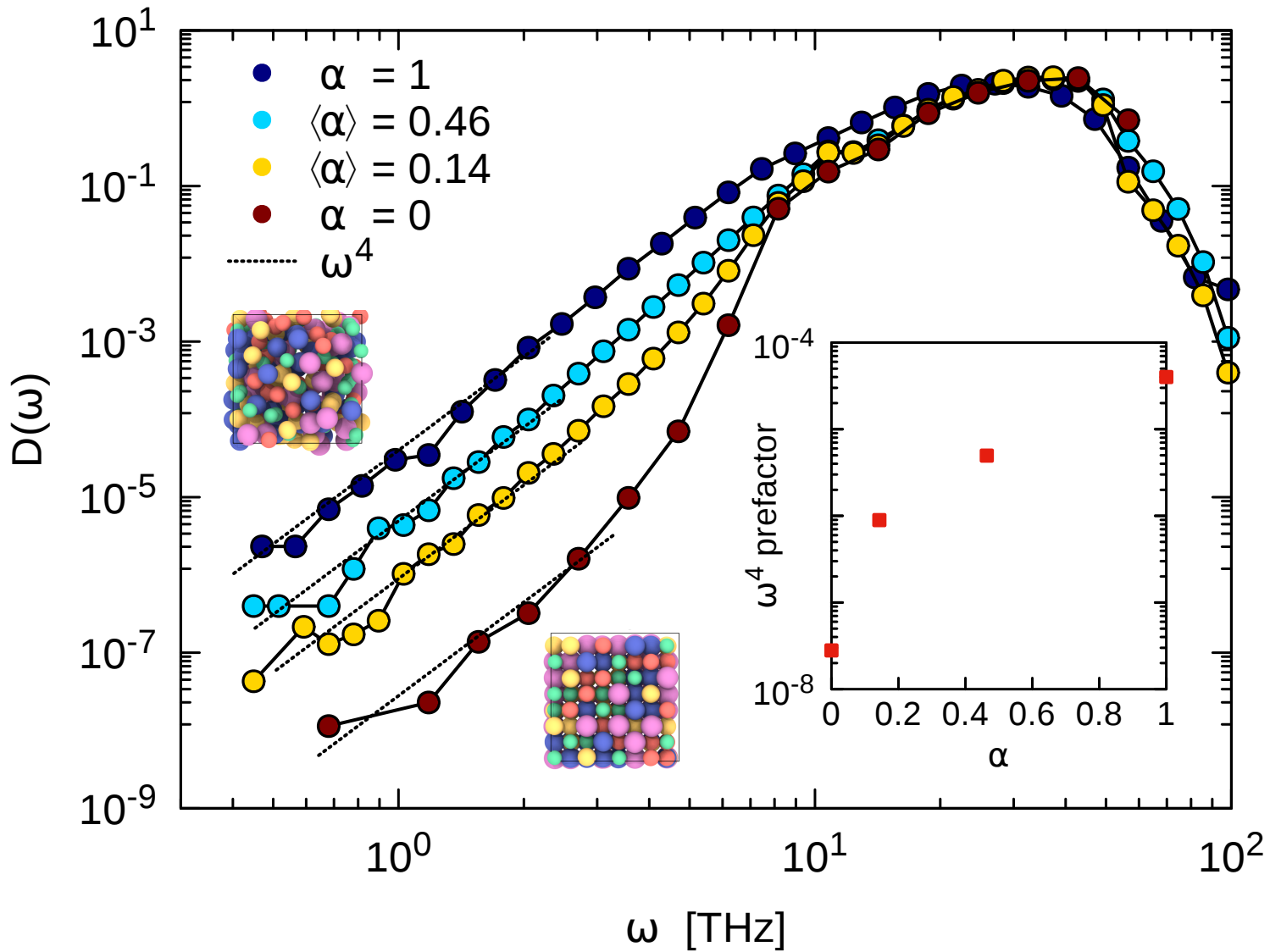
$$H_{ij}^{\alpha\beta} \equiv \frac{1}{\sqrt{m_i m_j}} \frac{\partial^2 U(\mathbf{r}_1, \dots, \mathbf{r}_N)}{\partial r_i^\alpha \partial r_j^\beta}$$

ROLE OF DISORDER

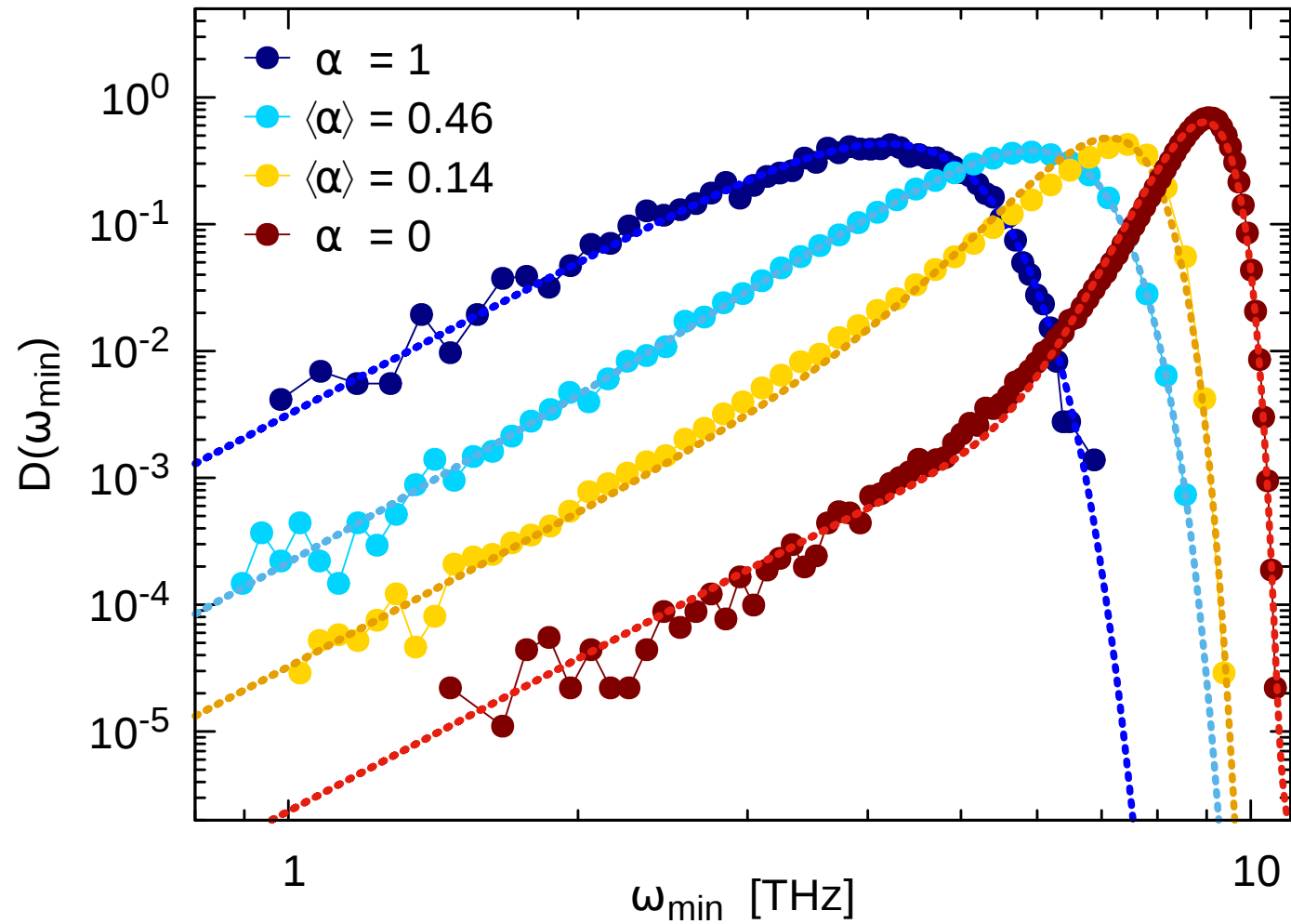
$$\alpha = 1 - \frac{N_{FCC}}{N}$$



DOS of High Entropy Alloys (HEA) follow ω^4 **also** in crystal structures

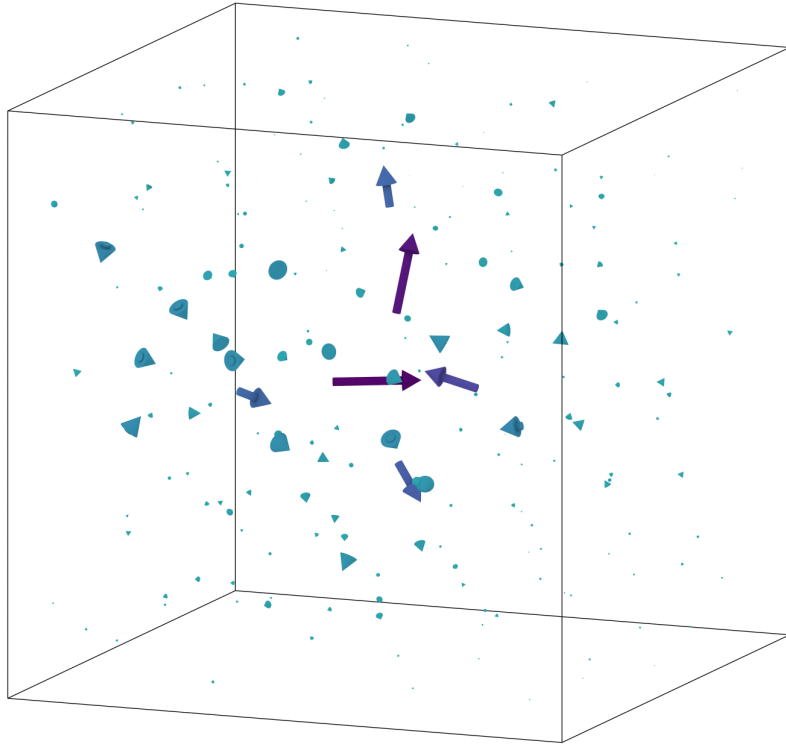


MINIMUM EIGENFREQUENCY

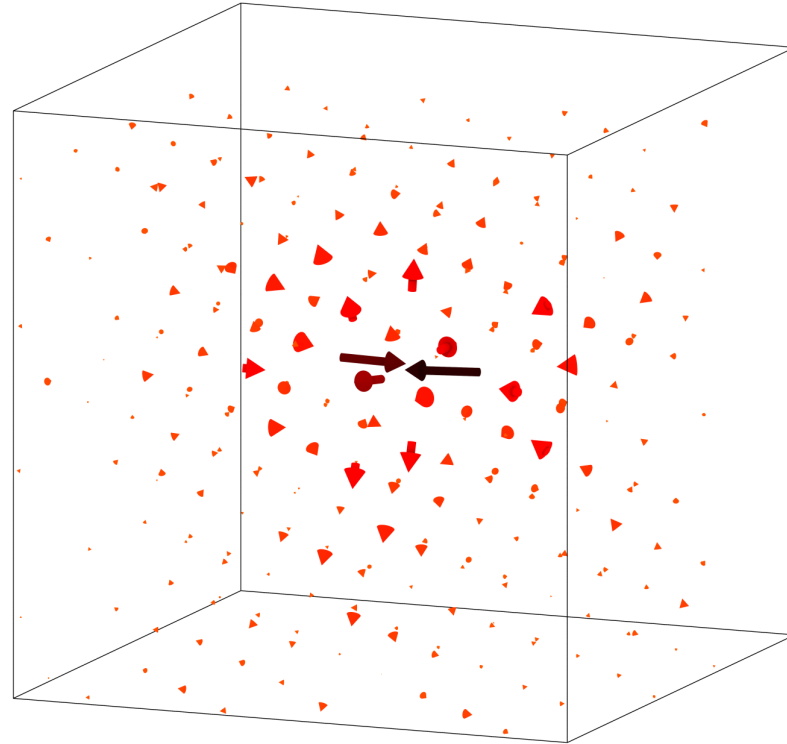


Distribution of ω_{\min}
follows Weibull

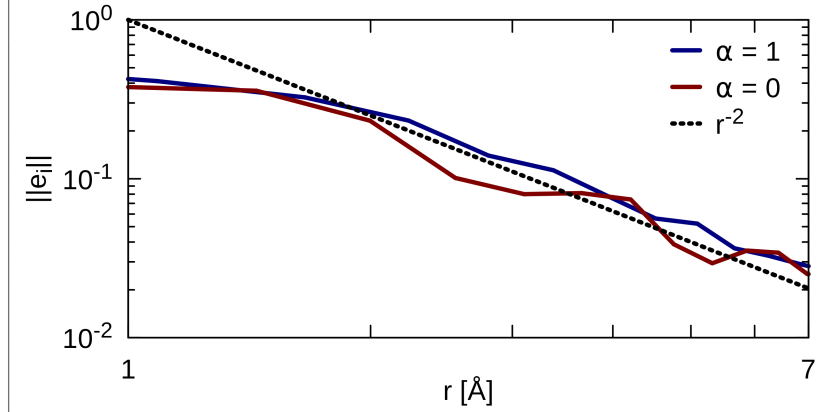
MODES ARE LOCALIZED



HEA-GLASS



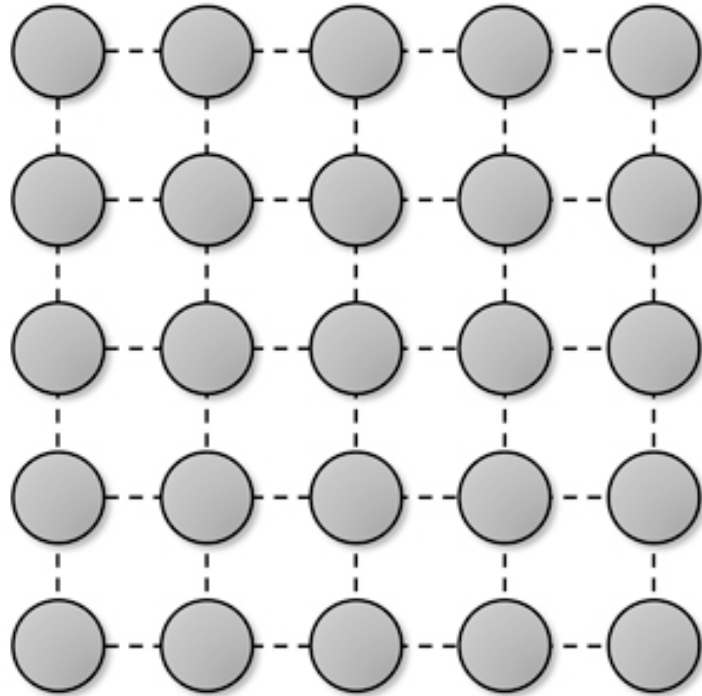
HEA-CRYSTAL



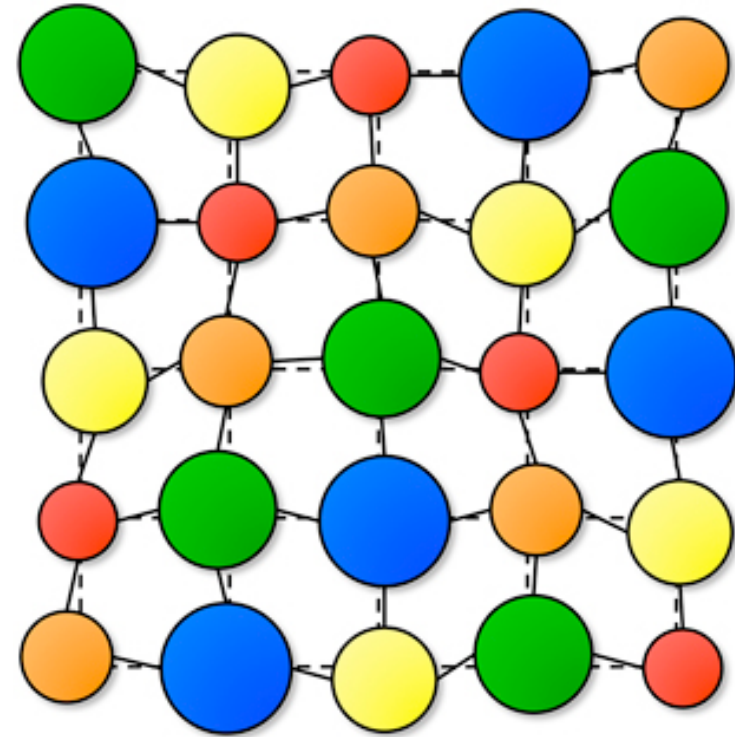
Long range trend

LATTICE DISTORSION

CONVENTIONAL ALLOYS



CRYSTALLINE HEAS



[Submitted on 16 Mar 2023]

Quasi-localized modes in crystalline high entropy alloys

Silvia Bonfanti, Roberto Guerra, Rene Alvarez-Donado, Pawel Sobkowicz, Stefano Zapperi, Mikko Alava

High Entropy Alloys (HEAs) are designed by mixing multiple metallic species in nearly the same amount to obtain crystalline or amorphous materials with exceptional mechanical properties. Here we use molecular dynamics simulations to investigate the role of positional and compositional disorder in determining the low-frequency vibrational properties of CrMnFeCoNi HEAs. Our results show that the expected dependence of the density of states on the frequency as $D(\omega) \sim \omega^4$ is recovered for amorphous HEAs and is also observed for partially crystallized alloys with deviations that depend on the degree of crystallization. We find that the quasi-localized vibrations are still visible in crystalline HEAs, albeit suppressed compared to the corresponding amorphous alloys. Our work offers a unified perspective to describe HEA mechanical properties in terms of their vibrational density of states.

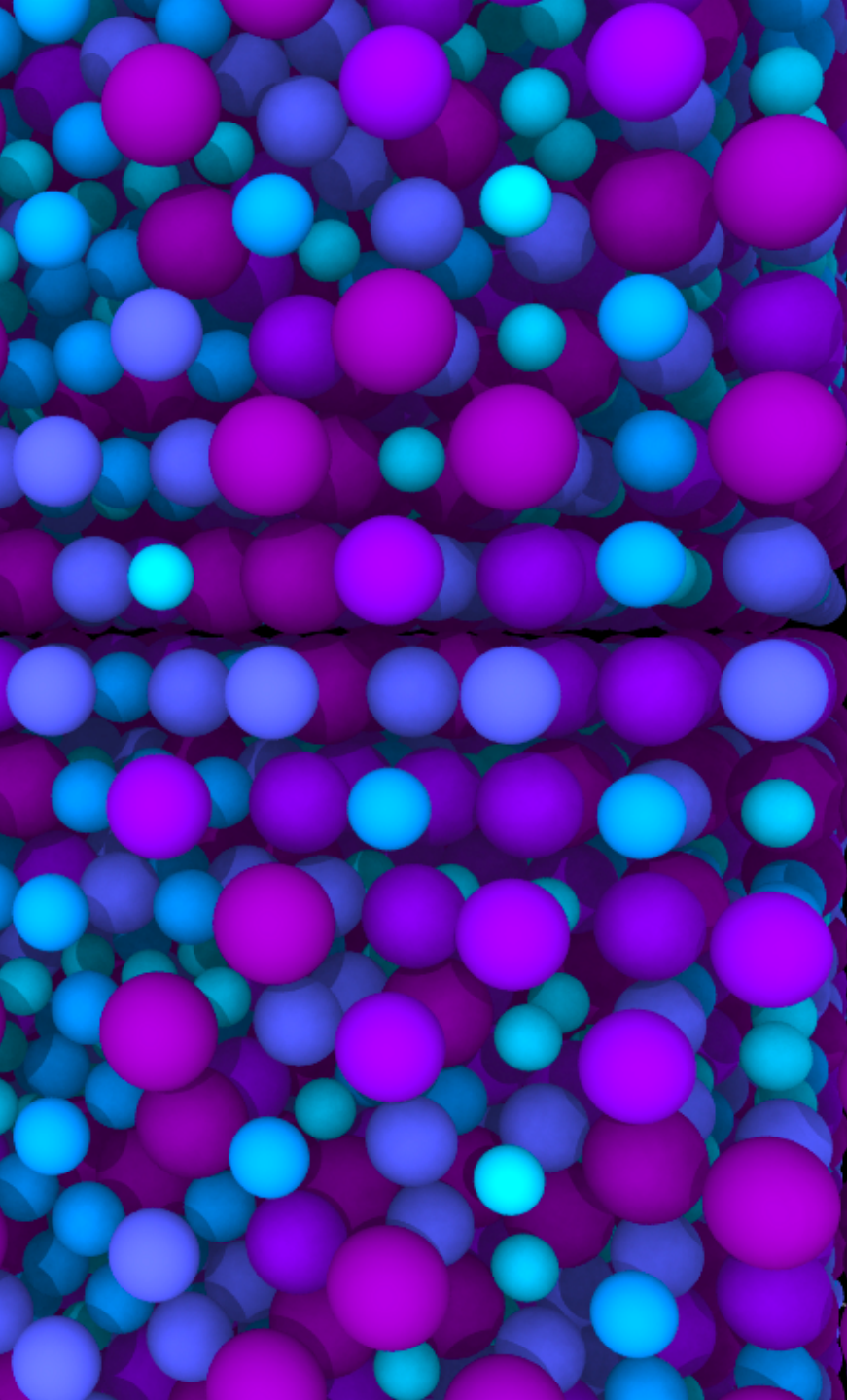
Comments: 8 pages, 7 figures

Subjects: **Disordered Systems and Neural Networks (cond-mat.dis-nn)**; Materials Science (cond-mat.mtrl-sci)

Cite as: [arXiv:2303.09161](https://arxiv.org/abs/2303.09161) [cond-mat.dis-nn]

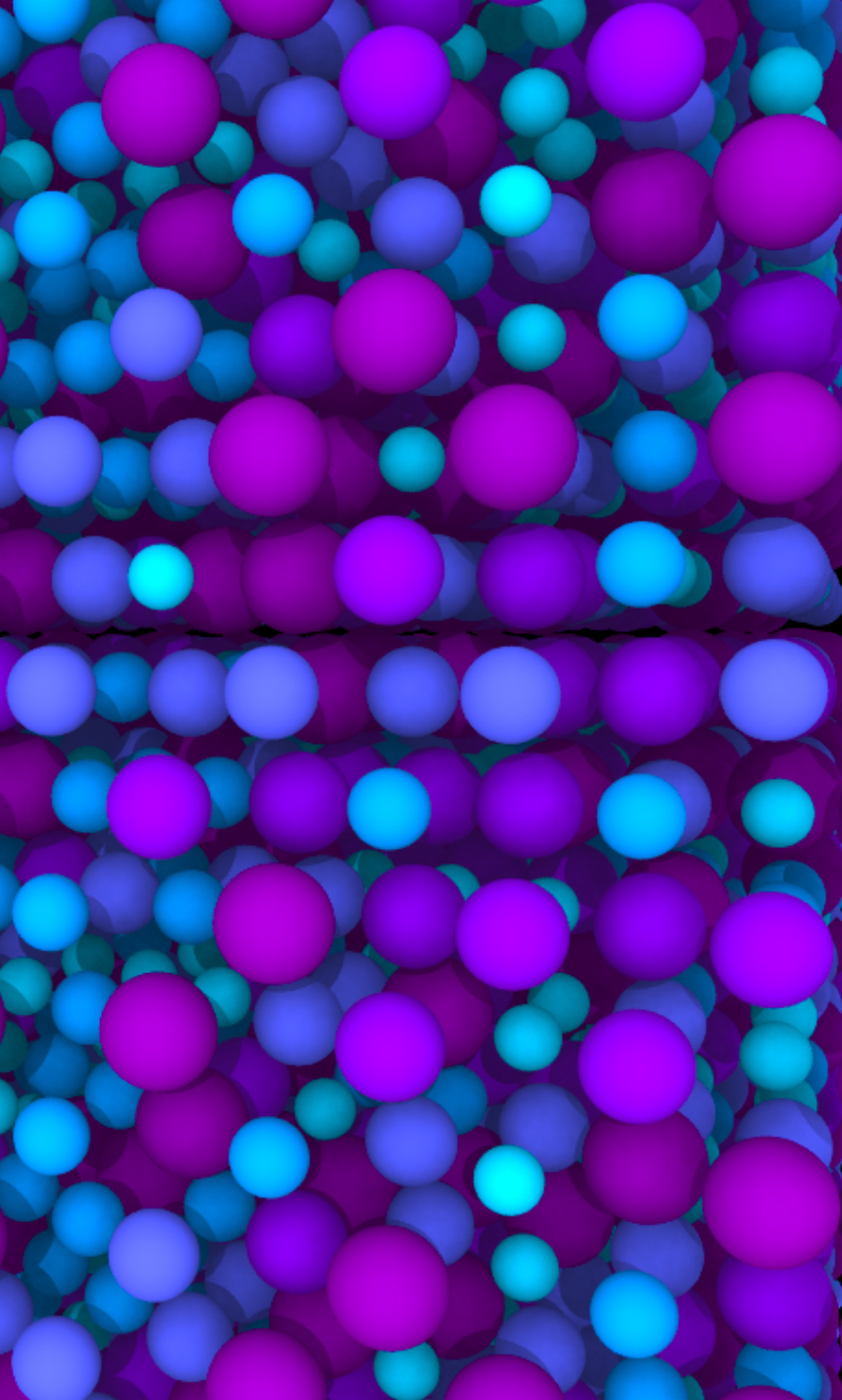
(or [arXiv:2303.09161v1](https://arxiv.org/abs/2303.09161v1) [cond-mat.dis-nn] for this version)

<https://doi.org/10.48550/arXiv.2303.09161> 



FUTURE PERSPECTIVES

- CAN WE INDIRECTLY SPOT QLM IN EXPERIMENTS IN HEAS?
- RELATION WITH THE MECHANICAL PROPERTIES
 - LOW-FREQUENCY ENERGY BARRIERS TAKE PLACE WHERE THE MATERIAL BREAKS. HEAS?



NATIONAL
CENTRE
FOR NUCLEAR
RESEARCH
ŚWIERK

NOMATEN

Centre of Excellence in Multifunctional Materials
for Industrial and Medical Applications

QUASI-LOCALIZED MODES IN CRYSTALLINE HIGH ENTROPY ALLOYS

Silvia Bonfanti

NOMATEN

Interaction, Disorder, Elasticity workshop – École de Physique des Houches – April 2-7, 2023

