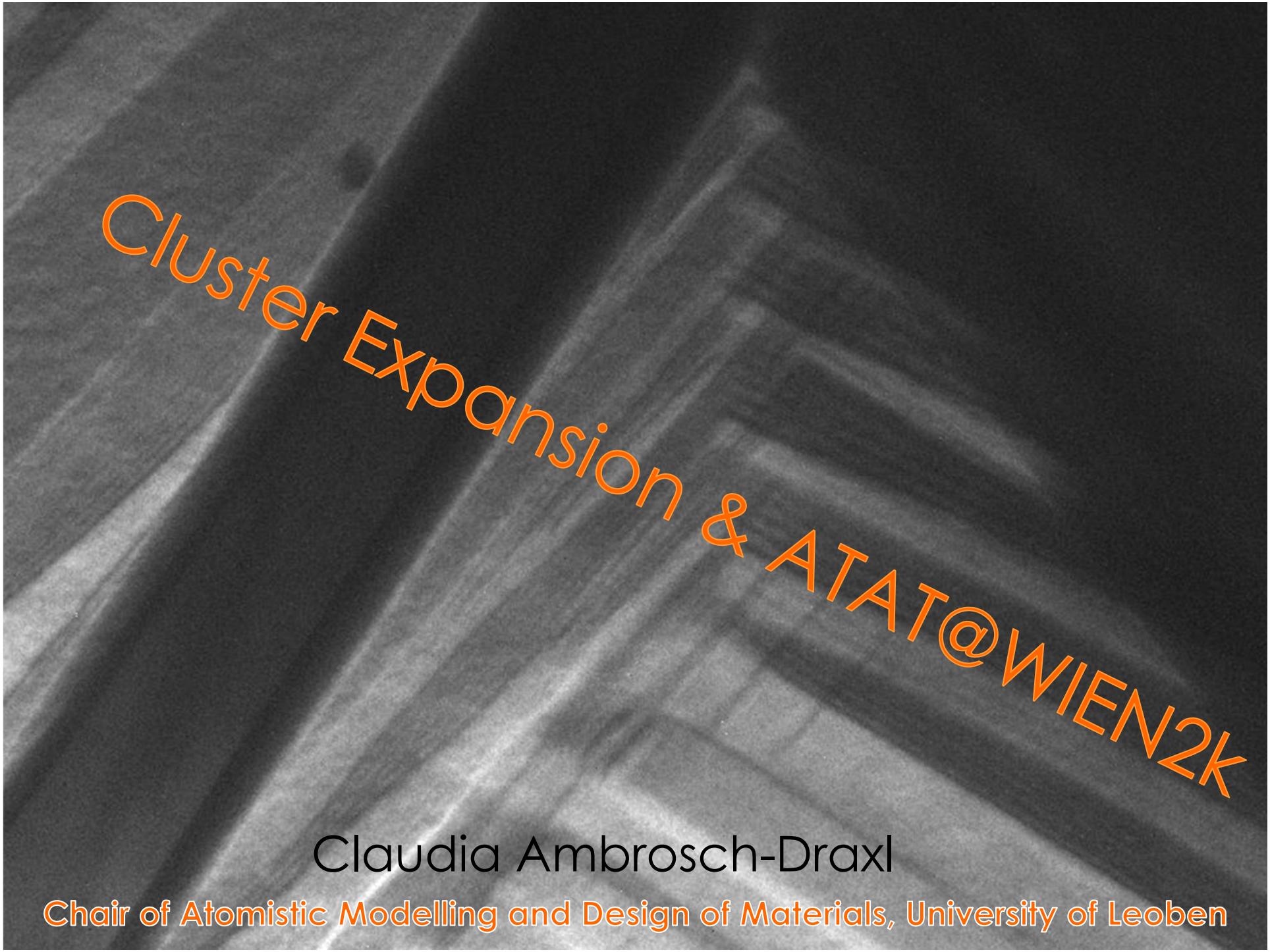
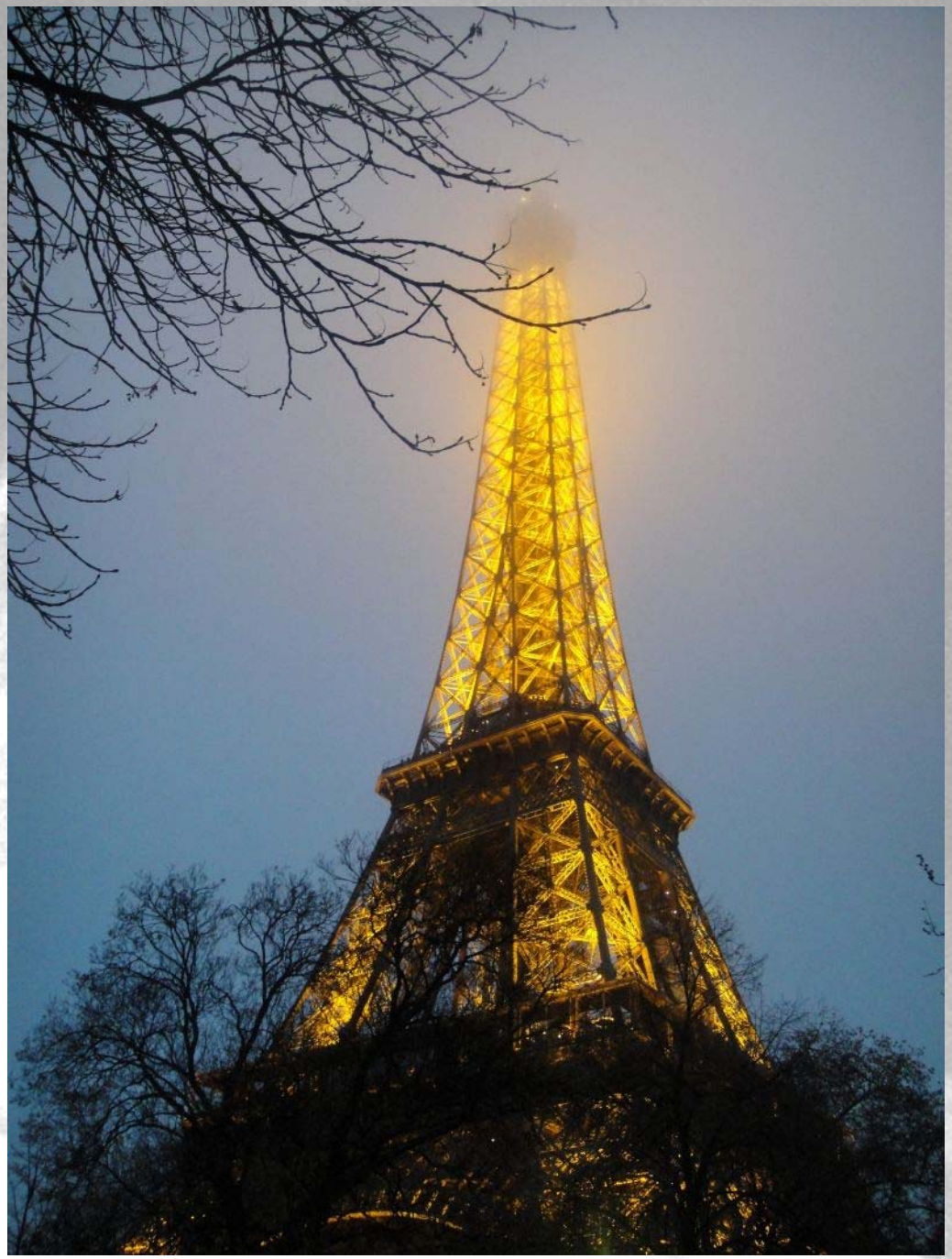


Claudia Ambrosch-Draxl

Chair of Atomistic Modelling and Design of Materials, University of Leoben

Eiffeltower

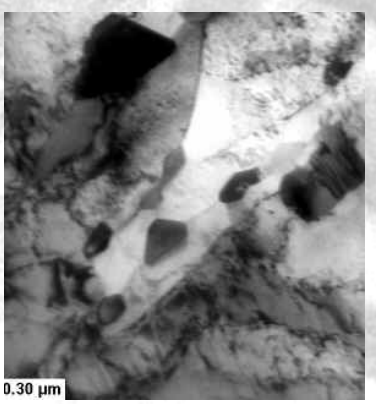
- Built 1887-89
- 10 000 t steel
- 300 m high



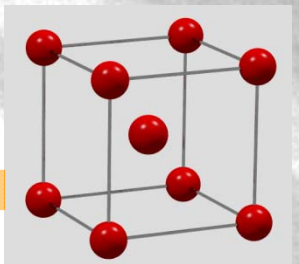
Point of view …

3×10^{2} m

3×10^{-2} m



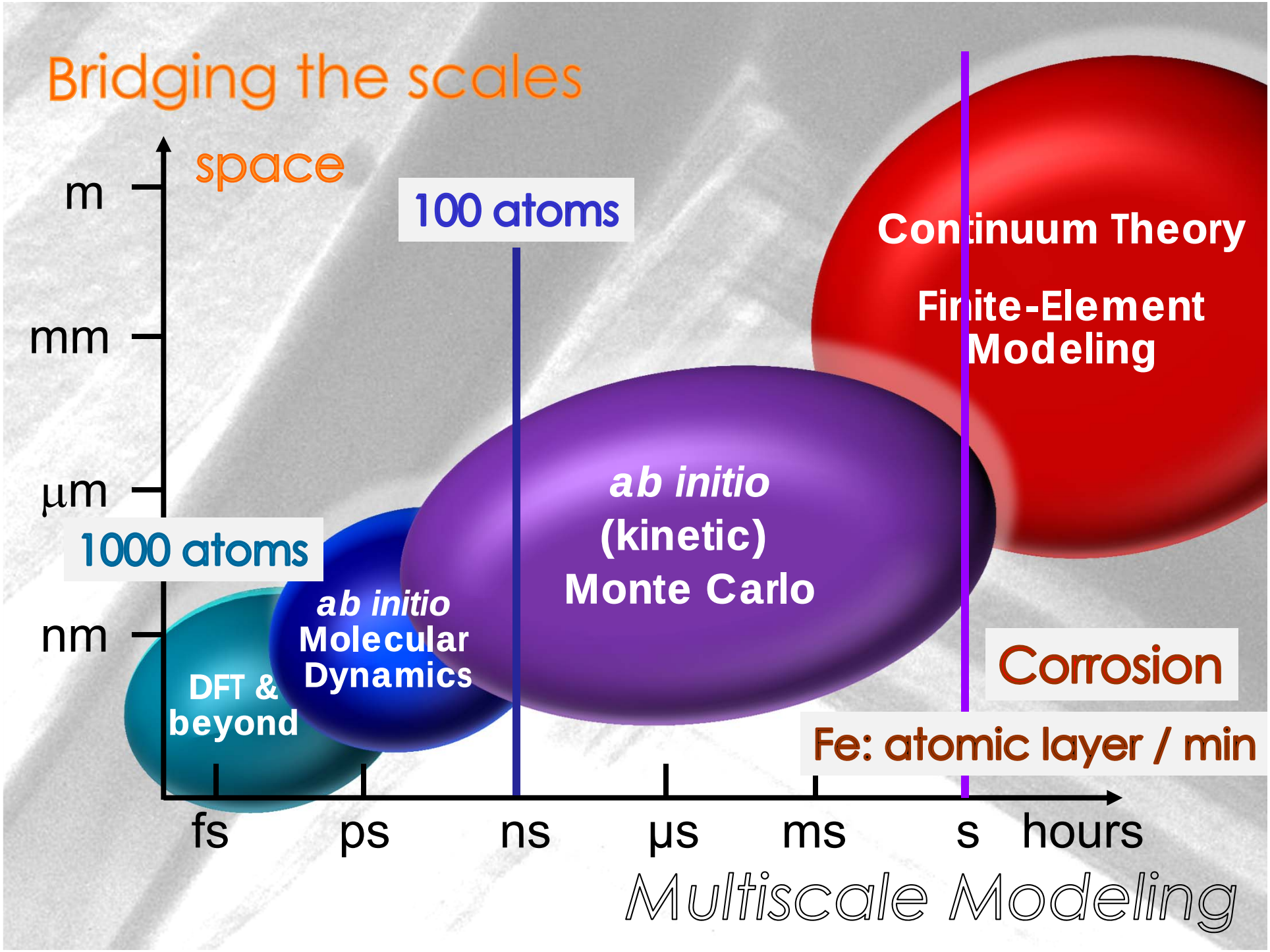
3×10^{-7} m



3×10^{-10} m

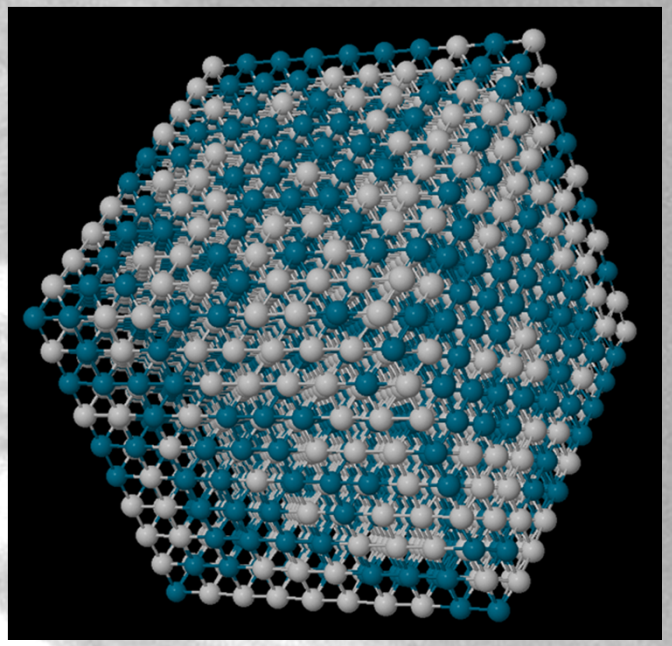
Macroscopic scale (pre)determined by the atomic/electronic scale

Length Scales


Bridging the scales
space
m
mm
μm
nm
100 atoms
1000 atoms
Continuum Theory
Finite-Element Modeling
ab initio (kinetic) Monte Carlo
ab initio Molecular Dynamics
DFT & beyond
Corrosion
Fe: atomic layer / min
fs
ps
ns
μs
ms
s
hours
Multiscale Modeling

Ab initio phase diagrams

- How to treat random alloys?
  - No translational symmetry
  - Large unit cells
  - Temperature as variable
- Stability of alloy phases
  - Intermetallic phases
  - Order / disorder
  - Mixed phases
  - Phase separation



Wanted ...

Ab *initio* thermodynamics

- Free energy from statistical physics

$$H \longrightarrow E_r(V,N) \longrightarrow Z(T,V,N) \longrightarrow F(T,V,N)$$

- Partition function

$$Z(T,V,N) = \sum_r exp\left[-\beta E_r(V,N)\right]$$

- Free energy

$$F(T,V,N) = -k_B T \; ln\left[Z(T,V,N)\right]$$

Wanted ...

Coarse graining

$$H|\Psi\rangle = E_r|\Psi\rangle$$

- Hamiltonian contains all degrees of freedom

Electronic	10^{-15} sec
Magnetic	10^{-13} sec
Vibrational	10^{-12} sec
Configurational	$>10^{-10}$ sec

- Make use of different time scales of excitations

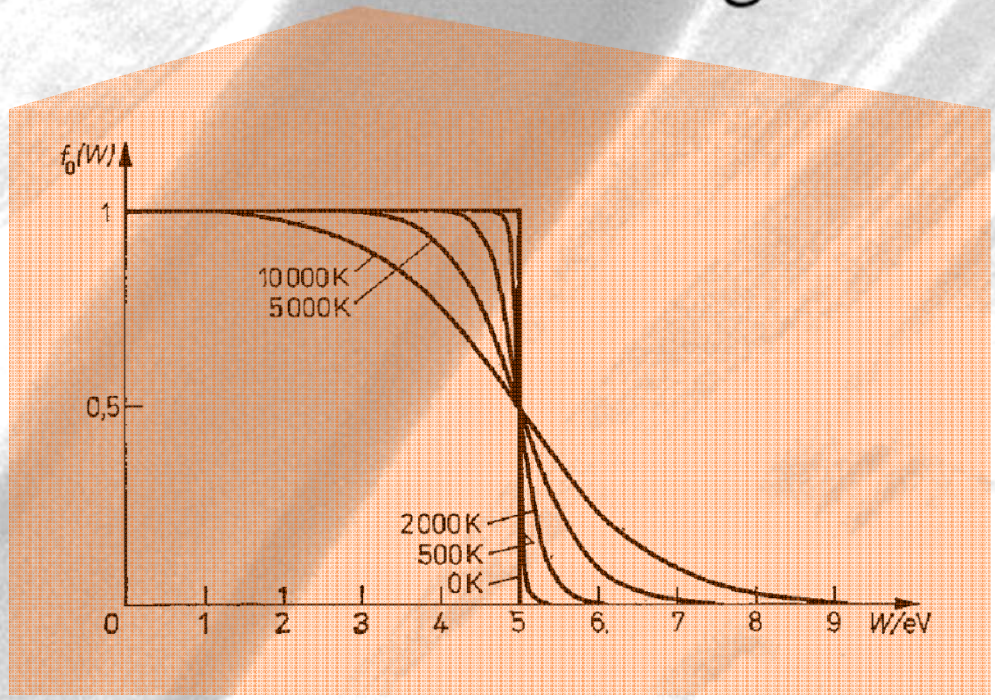
Treat them separately !

Thermodynamics

Entropy

$$F(T,V,N) = E_0(V,N) - TS$$

$$S_{el} + S_{mag} + S_{vib} + S_{conf}$$

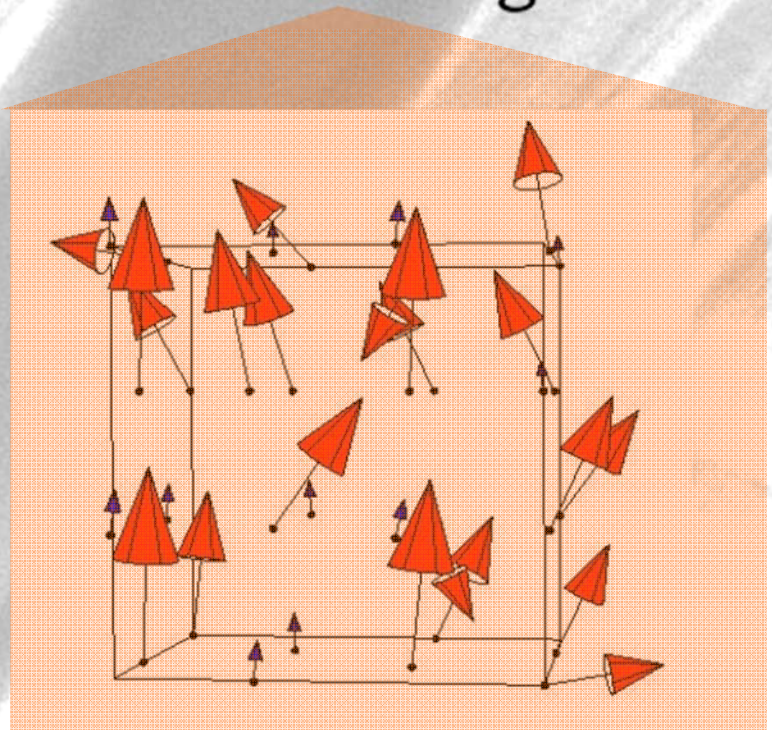


Thermodynamics

Entropy

$$F(T,V,N) = E_0(V,N) - TS$$

$$S_{el} + S_{mag} + S_{vib} + S_{conf}$$



Thermodynamics

Entropy

$$F(T,V,N) = E_0(V,N) - TS$$

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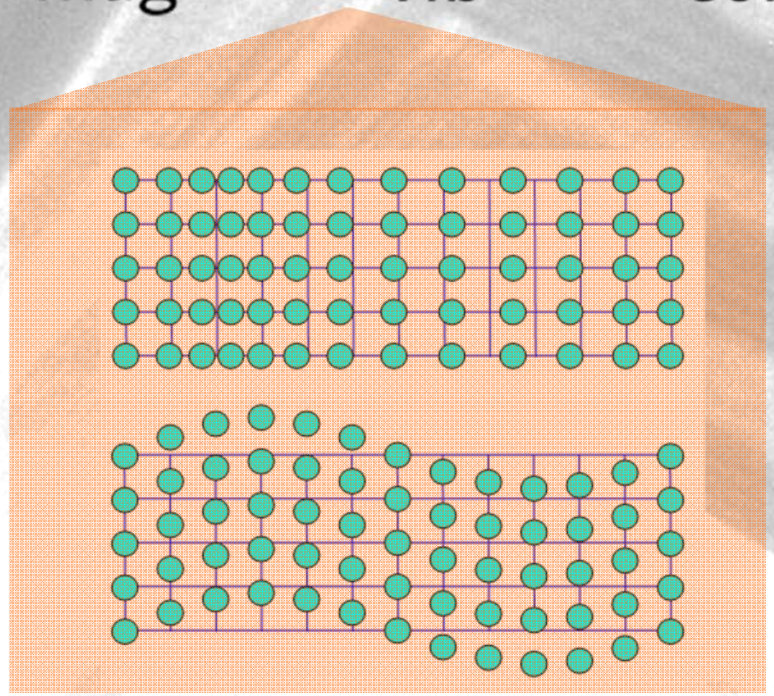
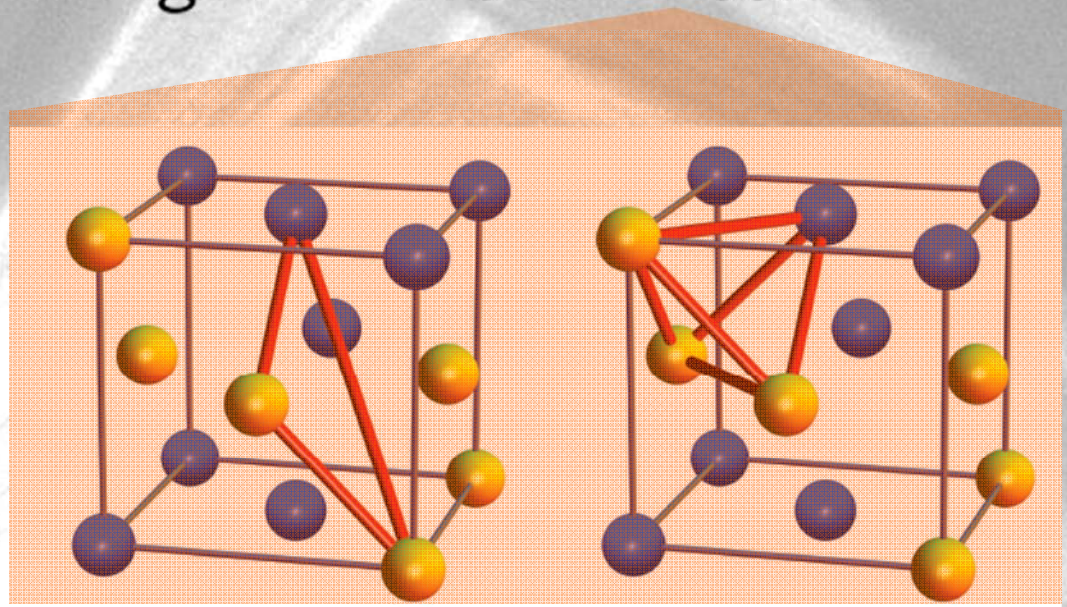


Thermodynamics

Entropy

$$F(T,V,N) = E_0(V,N) - TS$$

$$S_{el} + S_{mag} + S_{vib} + S_{conf}$$



Thermodynamics

Configurational entropy only …

Challanges

- Partition function

$$Z(T,V,N) = \sum_r exp\left[-\beta E_r(V,N)\right]$$

Need energy of huge number of states (~10^{30})

- How to sample so many configurations?

Monte Carlo simulations

- How to compute their total energy?

Cluster expansion

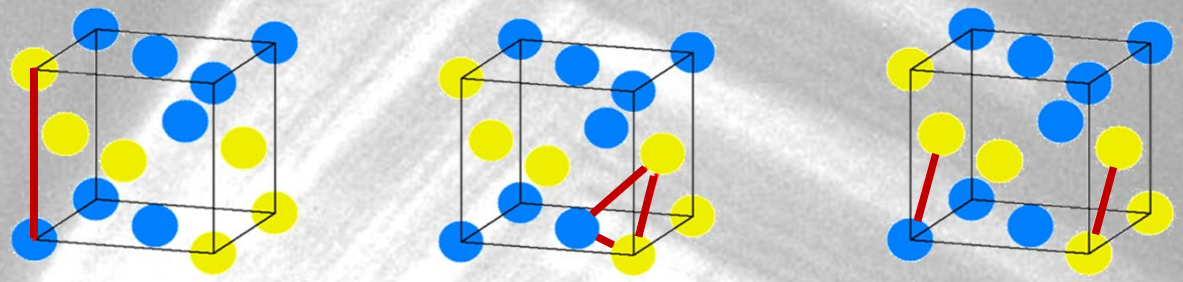
Thermodynamics

Concept

- Express total energy as a function of configurations

  Atomic and lattice relaxation treated implicitly

- Expand energy in terms of interactions inside clusters:



$$E = \sum_{\alpha} V_{\alpha} \xi_{\alpha}$$

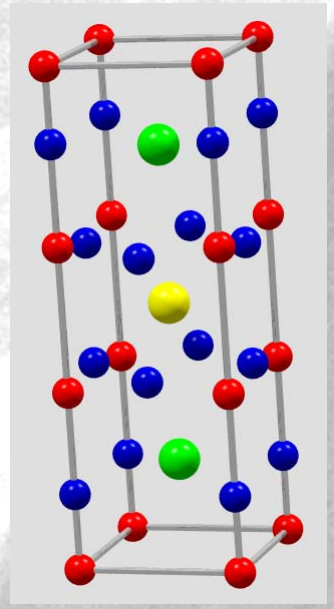
Cluster function

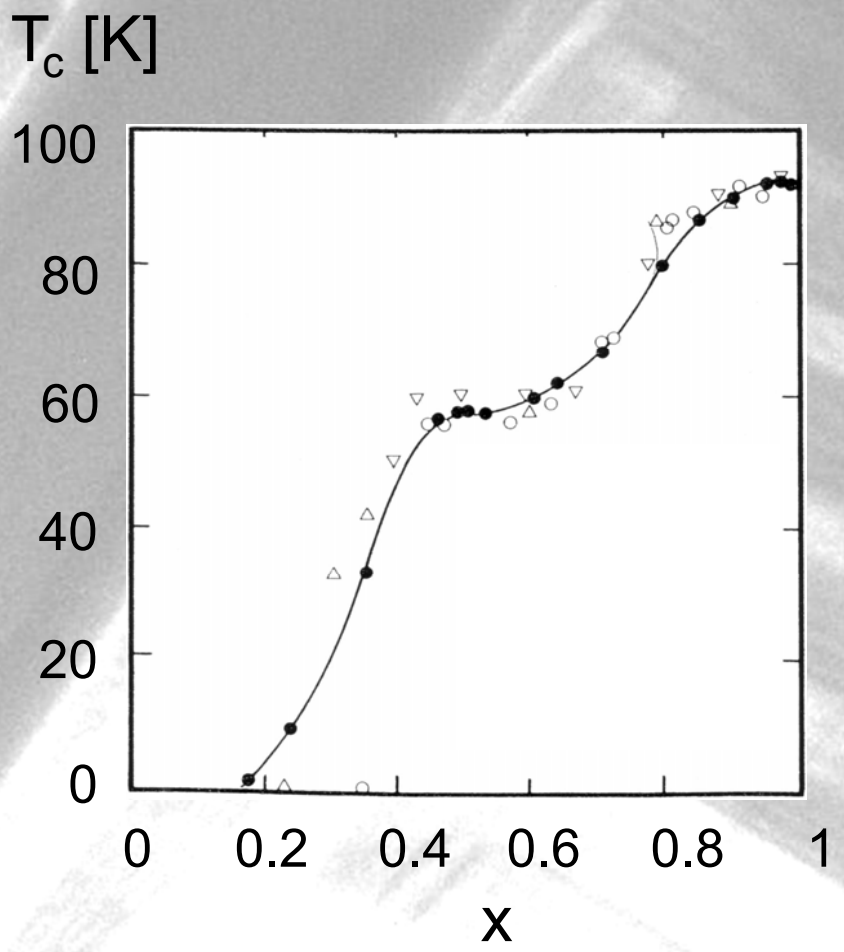
Effective cluster interaction (ECI)

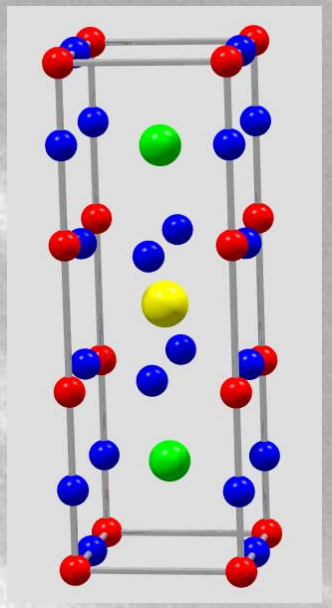
Cluster Expansion

A historic example …

Example: $YBa_2Cu_3O_{6+x}$



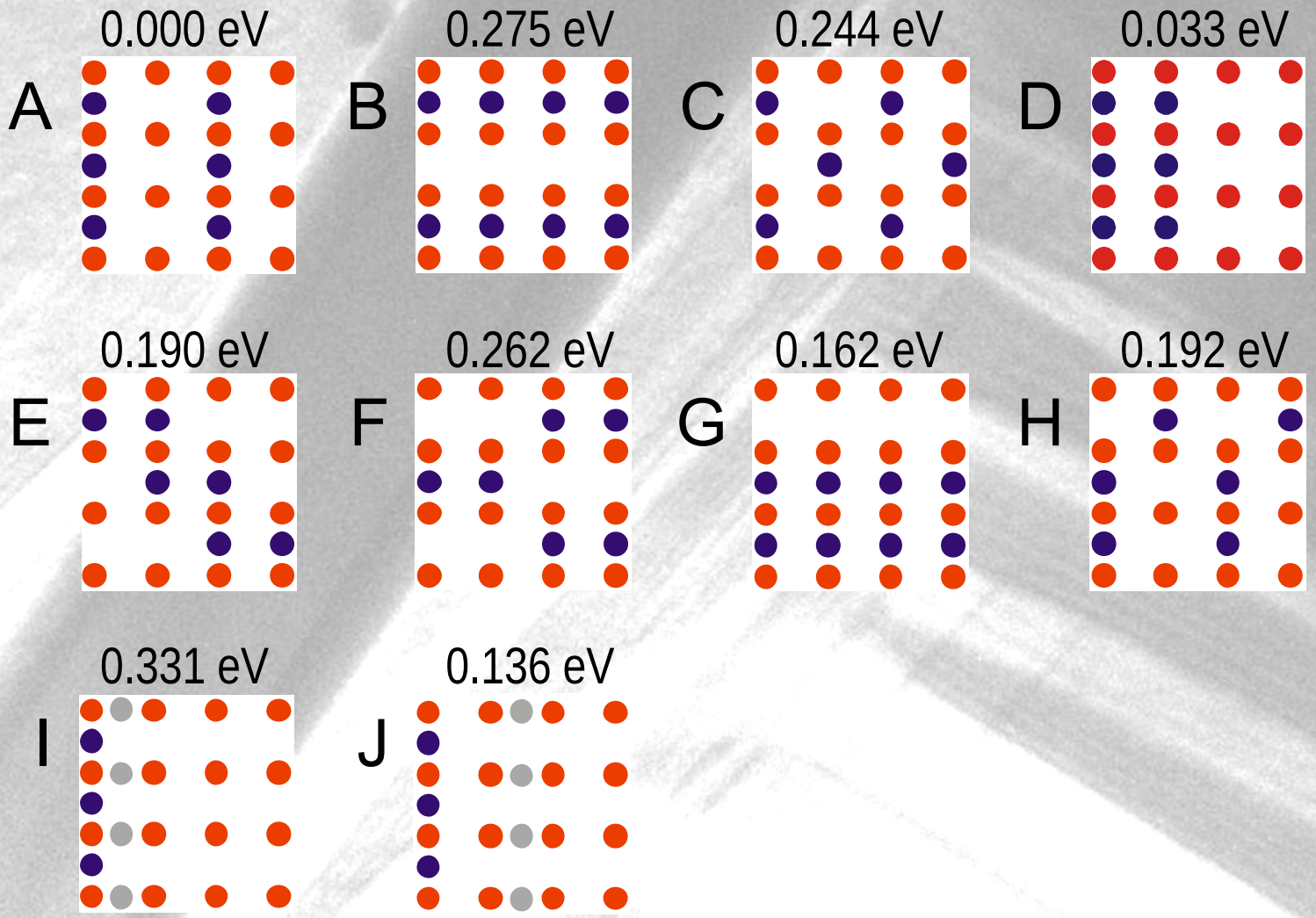




H. F. Poulsen et al., Nature 349, 594 (1991).
R. J. Cava et al., Physica C165, 419 (1990).

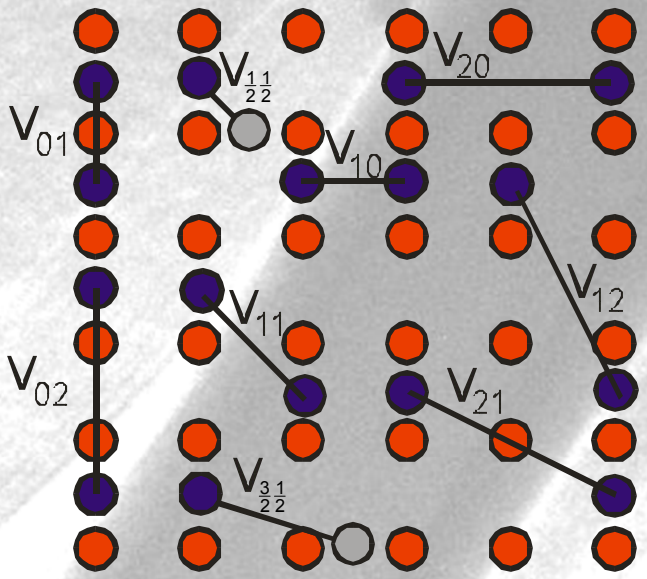
Cluster Expansion

Example: $YBa_2Cu_3O_{6.5}$



Cluster Expansion

Example: $YBa_2Cu_3O_{6.5}$



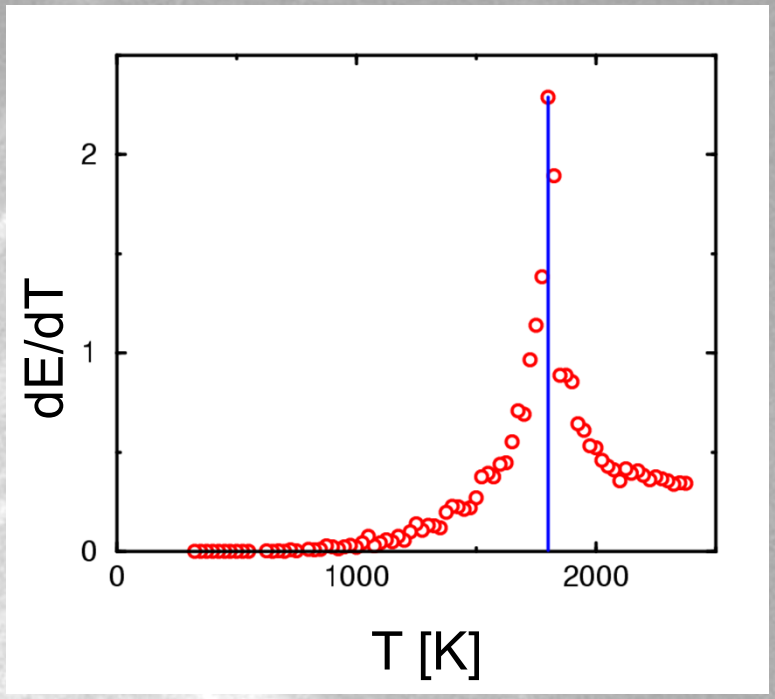
	V_α [eV]
$V_{1/2\ 1/2}$	0.098
V_{10}	-0.114
V_{01}	0.119
V_{11}	-0.001
$V_{3/2\ 1/2}$	-0.017
V_{20}	-0.006
V_{02}	-0.021

$$E = \sum_\alpha V_\alpha \xi_\alpha$$

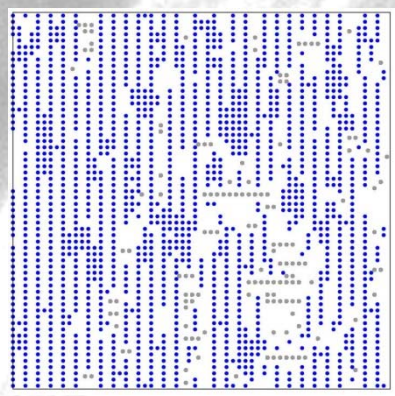
Effective cluster interaction
ECI

Cluster Expansion

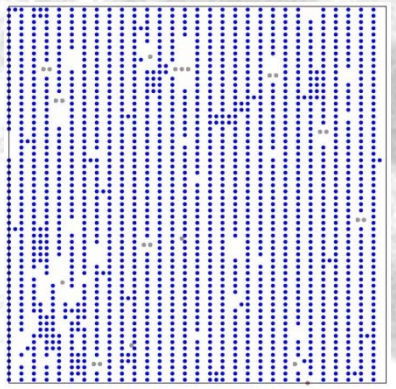
Example: $YBa_2Cu_3O_{6.5}$



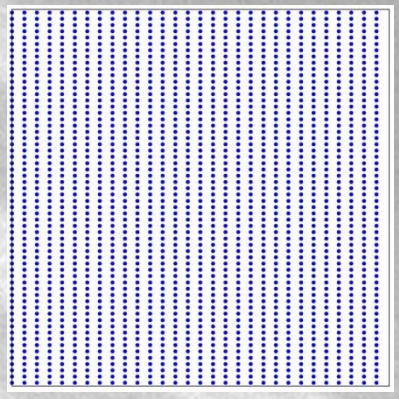
T=1900 K



T=1800 K



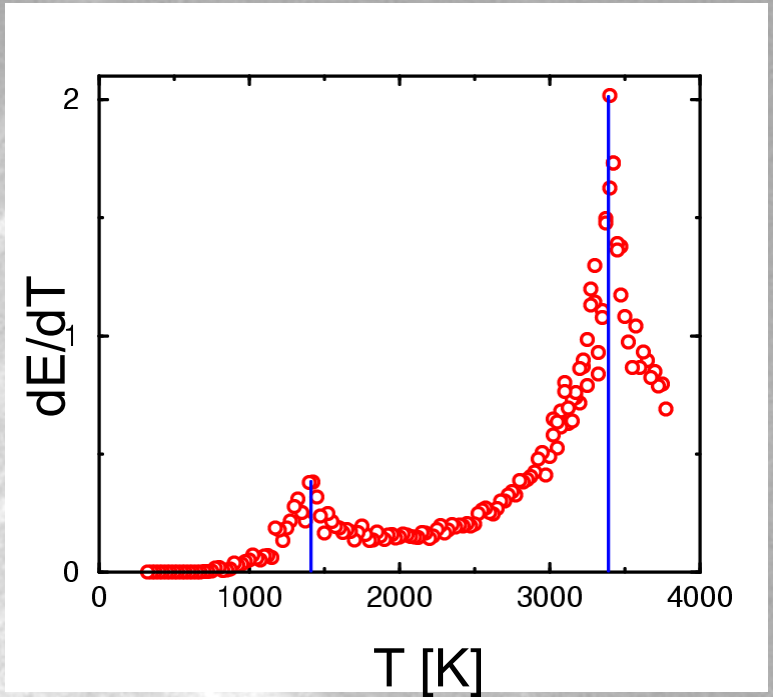
T=1600 K

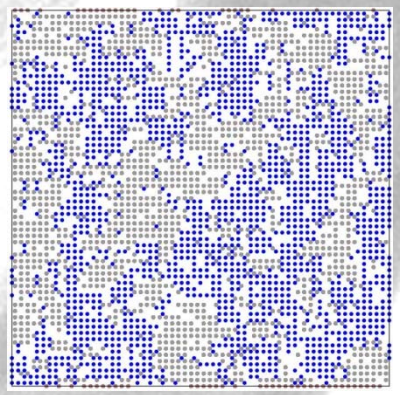


T=900 K

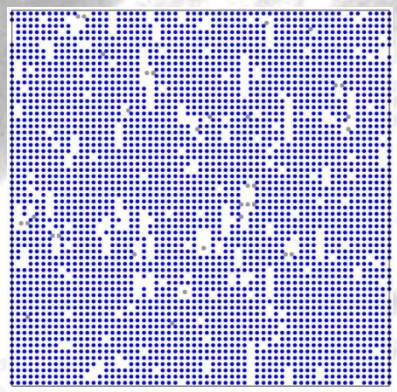
Monte-Carlo Simulation

Example: $YBa_2Cu_3O_{6.9}$

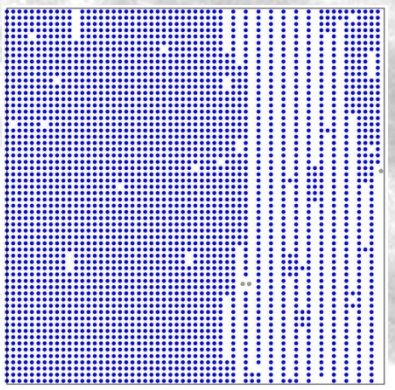




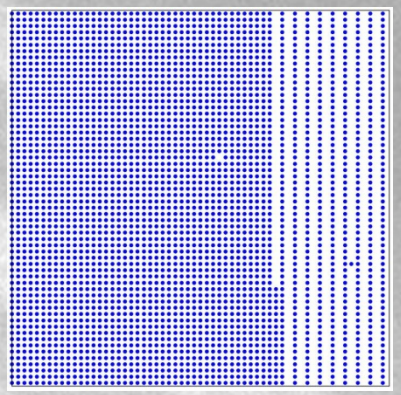
T=3750 K



T=2300 K



T=1300 K



T=900 K

Monte-Carlo-Simulationen


We can stand on other peoples' shoulders …
10 Years Later …

Alloy Theoretic Automated Toolkit

- @ Axel van der Walle
- @ http://www.its.caltech.edu/~avdw/atat/
- @ Tool for cluster expansion & phase diagrams

Alloy Automated Theoretic Toolkit

A. van de Walle, M. Asta, and G. Ceder , Calphad 26, 539 (2002).
A. van de Walle, Calphad 33, 266 (2009).

ATAT@WIEN2k

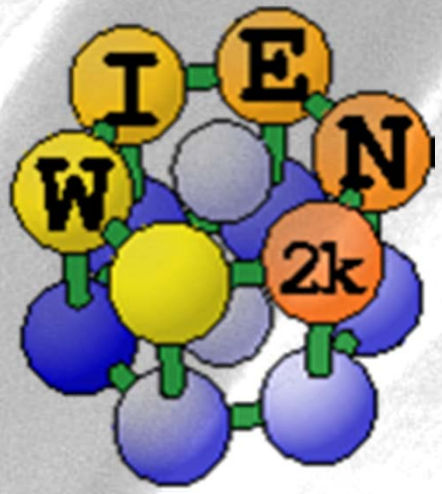
WIEN2k

- http://www.wien2k.at/
- DFT package
- All-electron full-potential code
- Accurate total energies for arbitrary structures

ATAT@WIEN2k

A happy marriage …

- Interface between the two programs
- Translates ATAT output into WIEN2k input

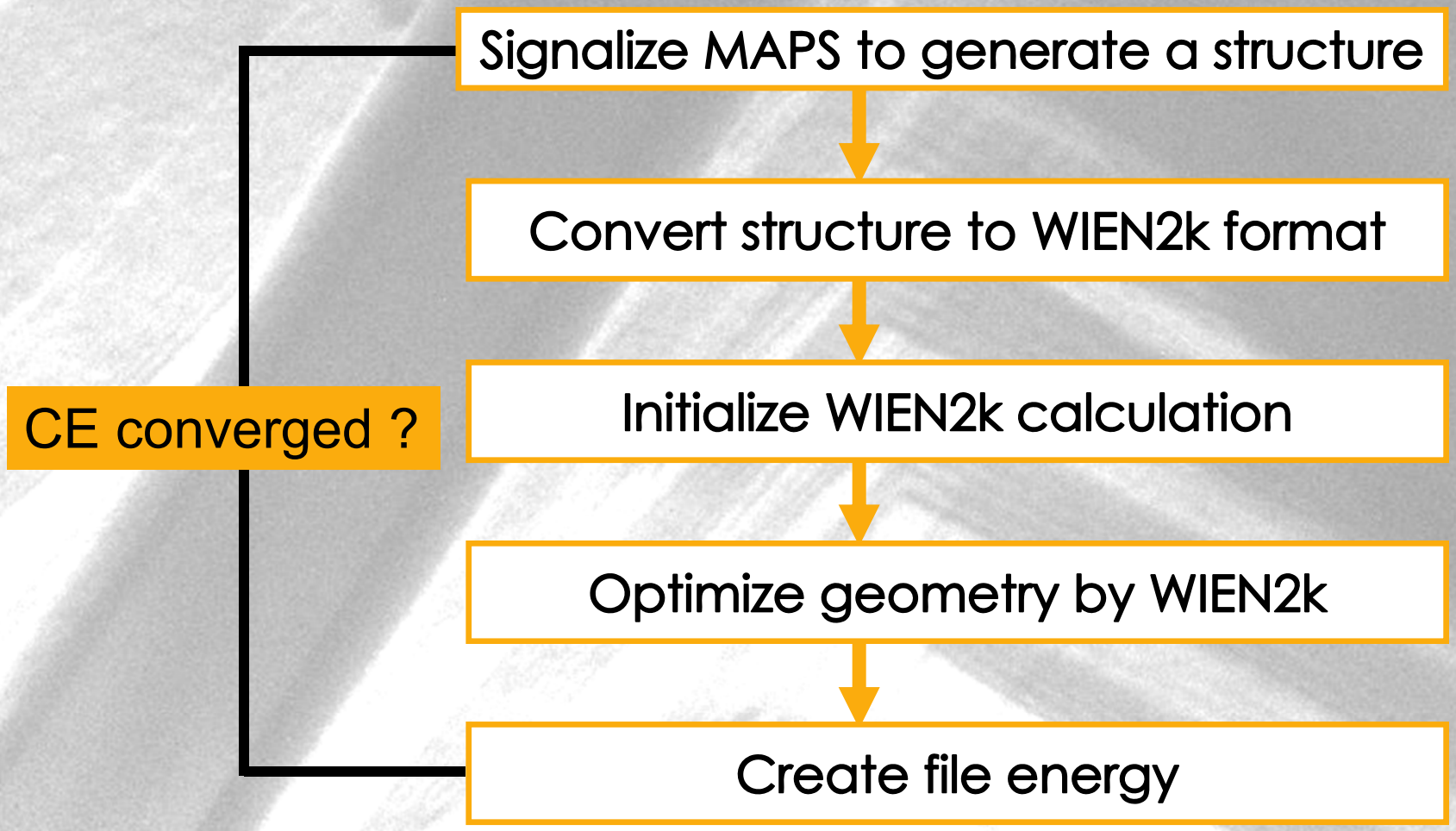


Alloy Automated Theoretic Toolkit

M. Chakraborty, J. Spitaler, P. Puschnig, and CAD
Comp. Phys. Commun. 181, 913 (2010).

ATAT@WIEN2k

Program flow



ATAT@WIEN2k

Scripts and programs

Script/Programme	Purpose
cluster_expand.csh	master script
read.csh	creates inputs files from wien.wrap
latt_gen.f90	creates inputs files for Vegard
vegard2.csh	implements Vegard averaging
str.out2xsf.sh [25]	convert str.out to WIEN2k struct file
correct.f90	
init_ATAT.csh	
in1_create.f90	create the case.in1 files for individual element
in1_create.csh	
in1.f90	create the file case.in1 (or case.in1c)
in1.csh	
read_rmt.csh	
vol_opt.csh	shape optimization
vol_opt.f90	
run_ATAT.csh	
poly_fit.f	

ATAT@WIEN2k

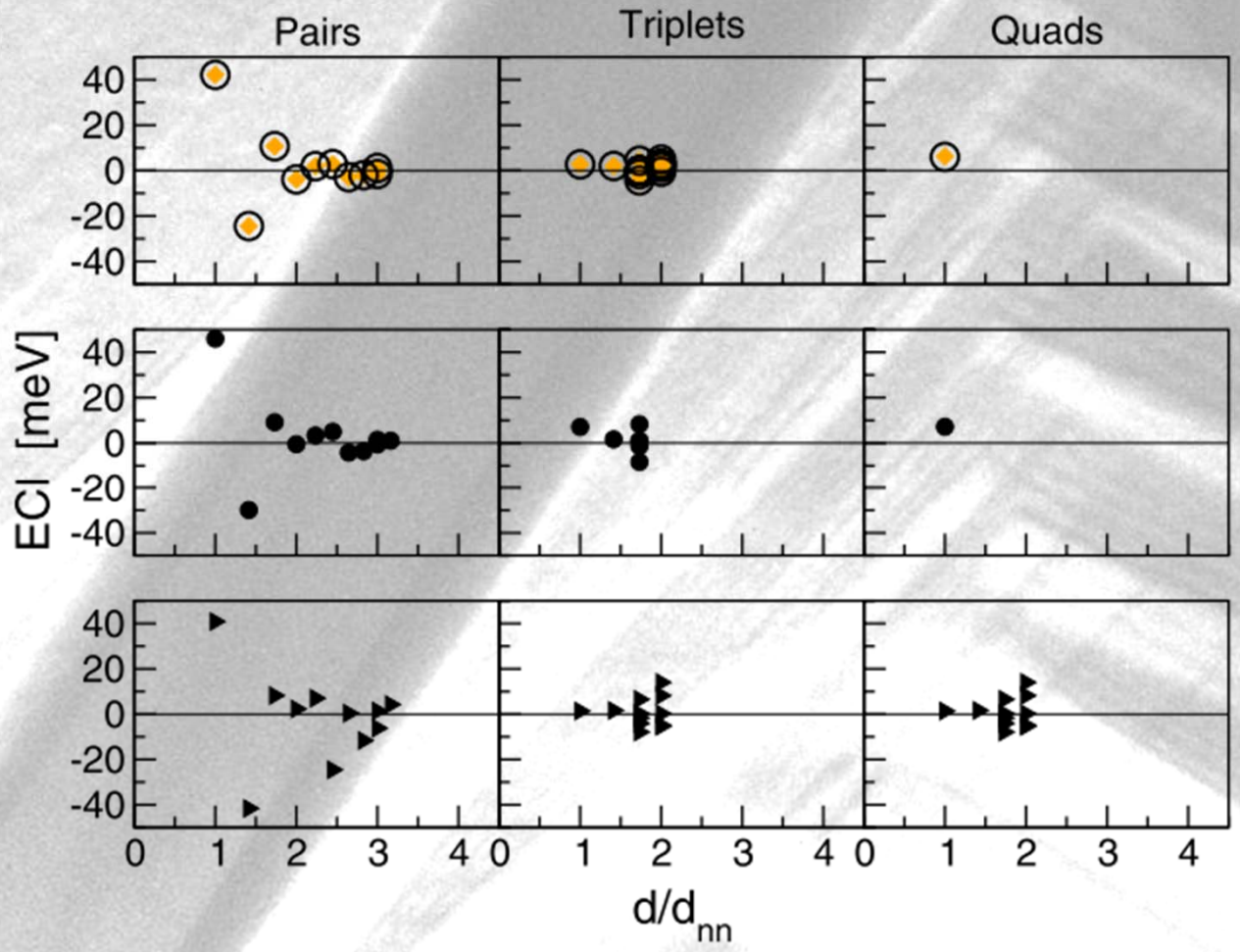
Input: `wien.wrap`

Variables	Status
RKmax = 9.0	required
Ncomponents = 2	required
Nspecies = 2	required
Element = Al	required
Z = 13 RMT = 2.2	required
Element = Ti	required
Z = 22 RMT = 2.2	required
Nk = 13 824	required
Error_flag = 0	required
Nshape = 3	required
Njobs = 12	required
Vegard = 1	optional
In1_flag = 1	optional
VASP_flag = 0	optional
Pre-relax = 0	optional

ATAT@WIEN2k

Typical results

@ Effective interaction parameters for TiAl



○ WIEN2k
♦ VASP
no shape relaxation

● WIEN2k
shape relaxation

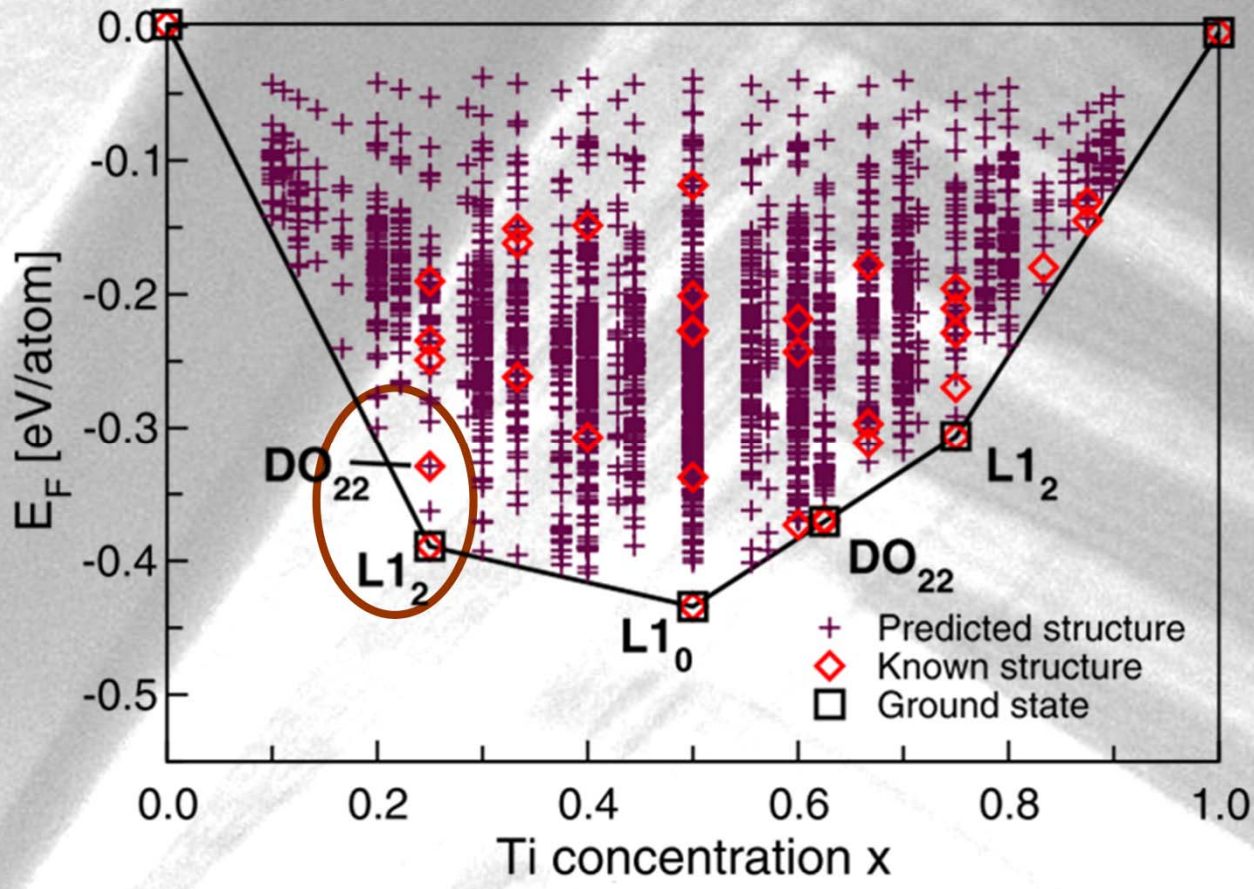
► VASP
Gosh et al.
shape relaxation

ATAT@WIEN2k

Typical results

@ Predicted ground states for TiAl

Without shape relaxation

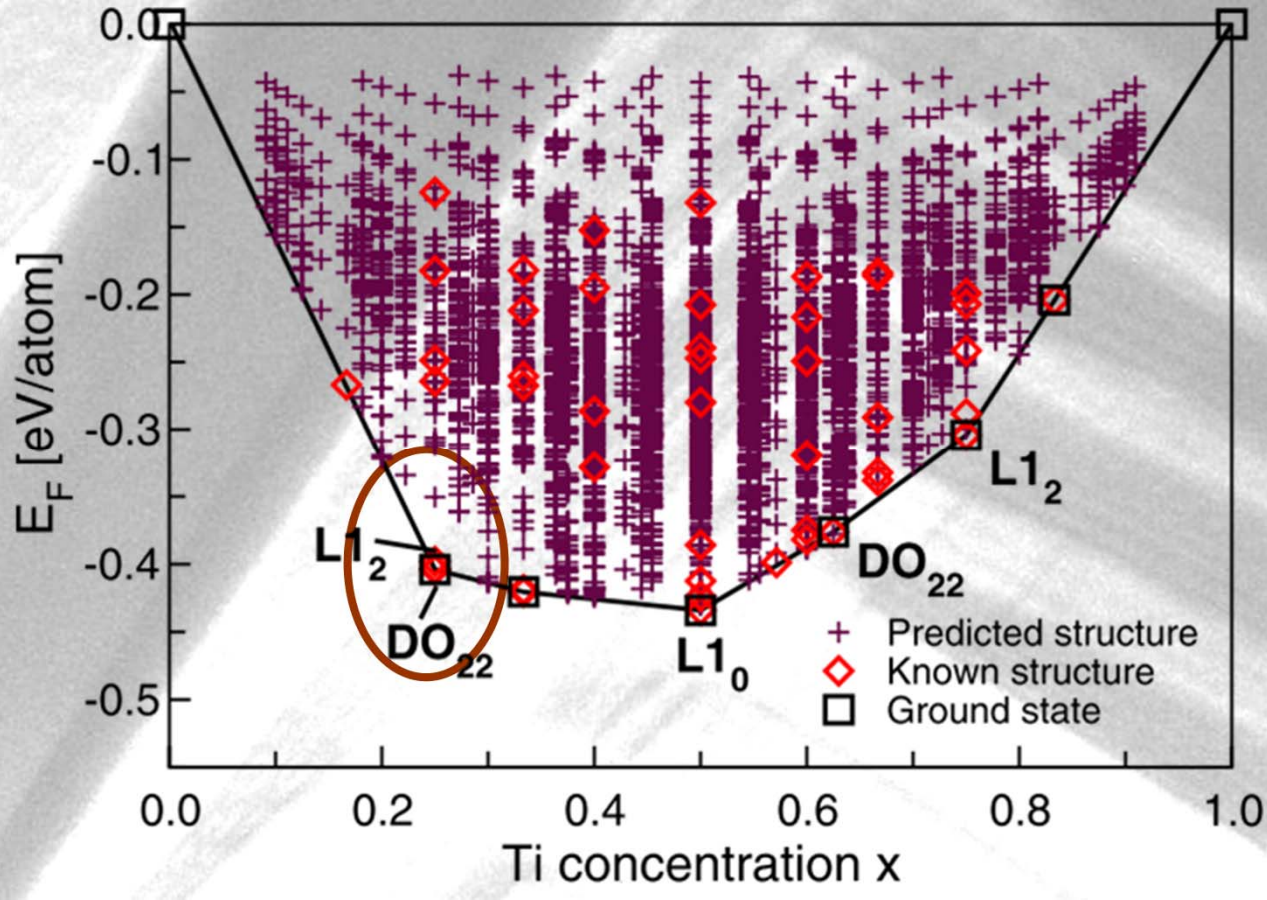


ATAT@WIEN2k

Typical results

@ Predicted ground states for TiAl

With shape relaxation



ATAT@WIEN2k

Summary

- Interface between ATAT and WIEN2k
- Code available (P. Blaha)
- http://amadm.unileoben.ac.at/software.html
- Reference:

  M. Chakraborty, J. Spitaler, P. Puschnig, and CAD
  ATAT@WIEN2k: An interface for cluster expansion based on the linearized augmented planewave method
  Comp. Phys. Commun. 181, 913 (2010).

Have fun with it!

ATAT@WIEN2k

Thanks for your attention !!