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Modeling positron response from computer generated samples of materials

J. Kuriplach
Charles University
Prague
Czech Republic

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Jan Kuriplach

Department of Low Temperature Physics
Faculty of Mathematics and Physics
Charles University, Prague, Czech Republic
E-mail: jan.kuriplach@mff.cuni.cz

Joint ICTP-IAEA Advanced Workshop on Multi-Scale Modelling
for Characterization and Basic Understanding of Radiation Damage Mechanisms in Materials
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Outline

- Motivation
- Non-periodic boxes in positron calculations
- Positron affinity treatment in ATSUP method
- Conclusions

Motivation

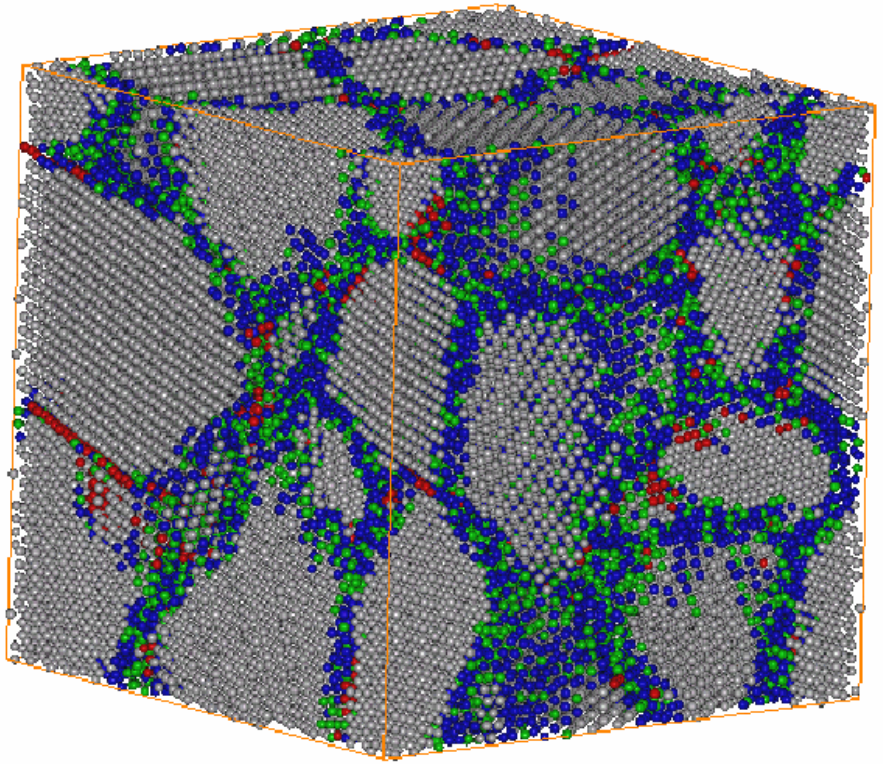
- Realistic atomistic models of various materials under different conditions have become available.
- These models include defects that could be studied with positrons.
- Modelled (computer) samples contain thousands of atoms (and more) and are not suitable for ab initio positron calculations and use of non-selfconsistent methods is preferable.
- It is computationally more convenient to select ‘regions of interest’ where positrons can be trapped and to process such regions in a series of separate positron calculations.

Motivation

- There are two fundamental problems connected with this approach:
  - Selected regions do not need to be periodical and a modification of the ATSUP method is required to handle non-periodic boxes.
  - When studying precipitates, the difference of positron affinities between the host and precipitate needs to be treated properly as the ATSUP method does not include charge transfer responsible for the alignment of Fermi levels of the host and precipitate.

Non-periodic boxes

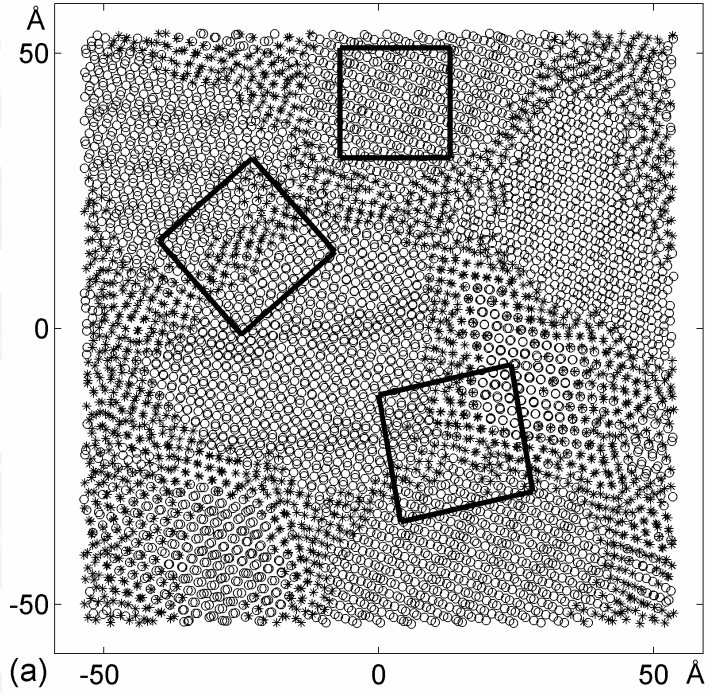
- The solution of this problem will be demonstrated for computer samples of nanocrystalline Ni.
- Samples were produced using molecular dynamics at 300 K and zero pressure starting from a box with 15 grains generated using the Voronoi construction.
- Positron lifetime measurements detect a component corresponding to vacancies which should be located at grain boundaries.



5 nm nc-Ni sample
~100000 atoms

Non-periodic boxes

- The atomic superposition method was modified as follows:
- First a cut from the nc sample containing defects of interest needs to be made.



Non-periodic boxes

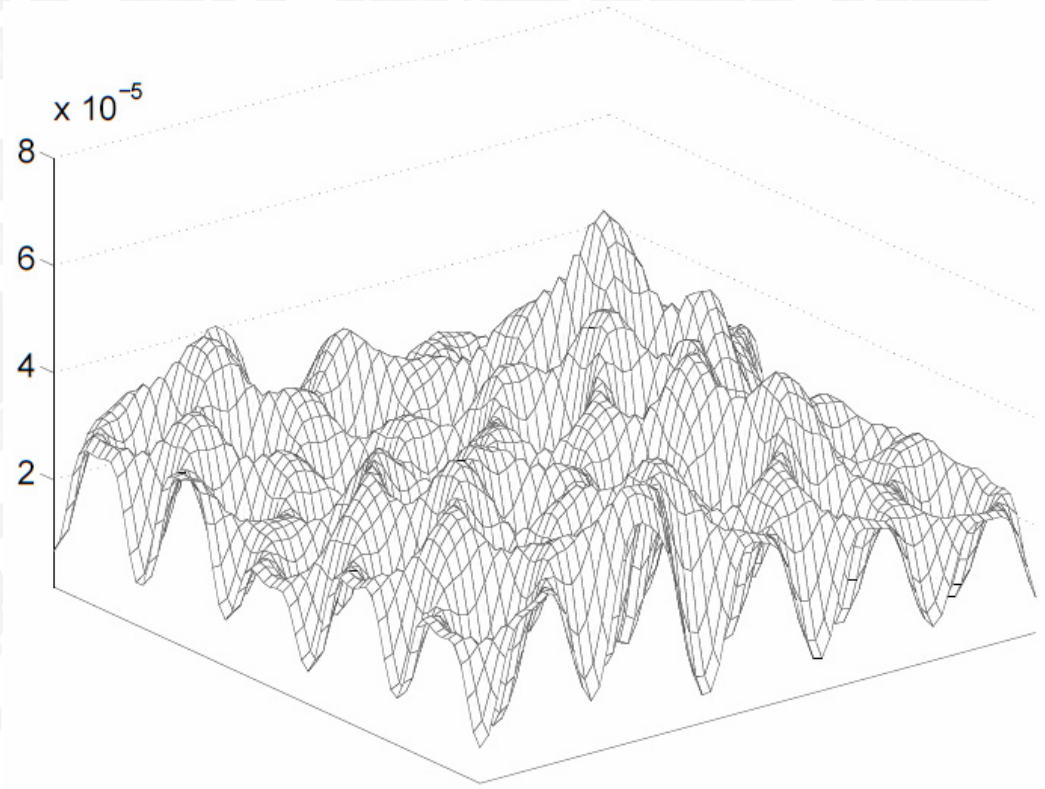
- The atomic electron densities and Coulomb potentials are superimposed as if the box/cut would be periodic.
- Then, the box size is reduced (by ~1 Å) from all sides to avoid an affect of false periodicity imposed in the previous step.

Non-periodic boxes

- Finally, the positron potential is adjusted at the sides of the reduced box as follows:
  - If the original potential value is smaller than V_0, then the potential is set to V_0.
  - If the original potential value is larger or equal than V_0, then the this value is left unchanged.
- The value of V_0 needs to be found and usually is close to the positron energy in the defect free material.
- The purpose of this adjustment is to remove artificial open volume defects at box sides.
- These ‘defects’ originate from cuts taken at arbitrary position.
- In this way positrons are forced to stay in the cut and physically reasonable behavior of the positron wave function is ensured.

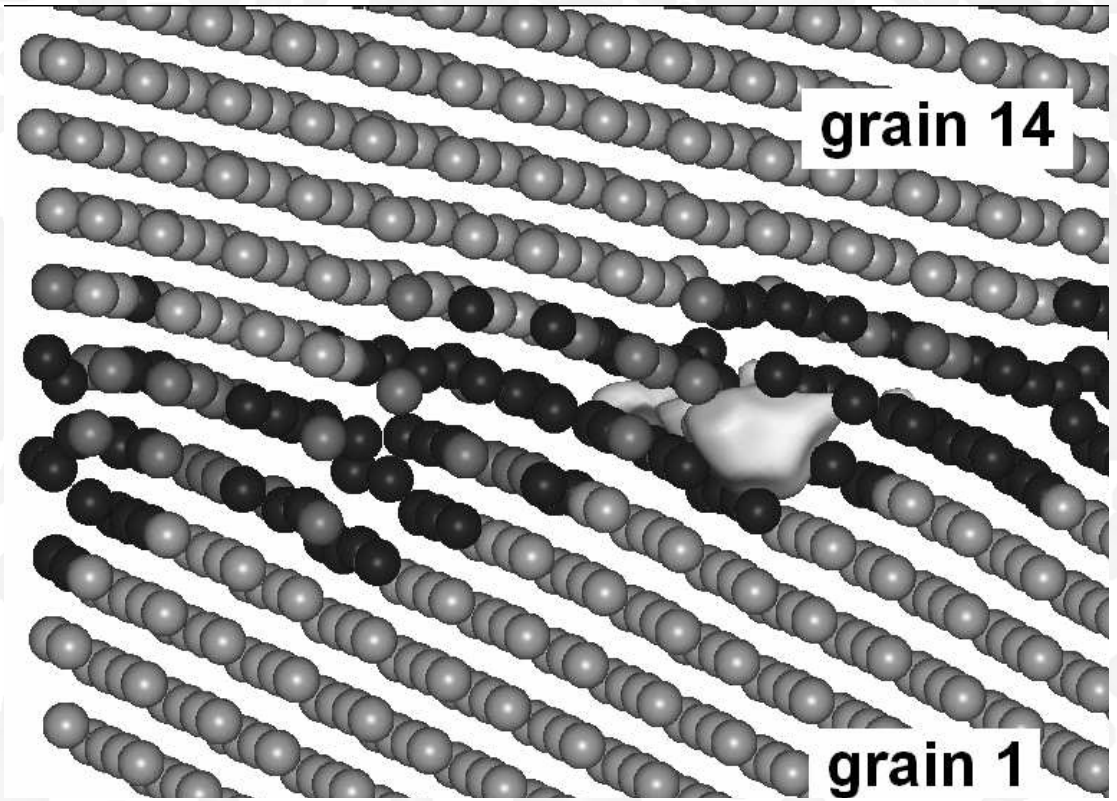
Non-periodic boxes

- Positron wave function in a cut from the center of a grain has rather regular behavior:



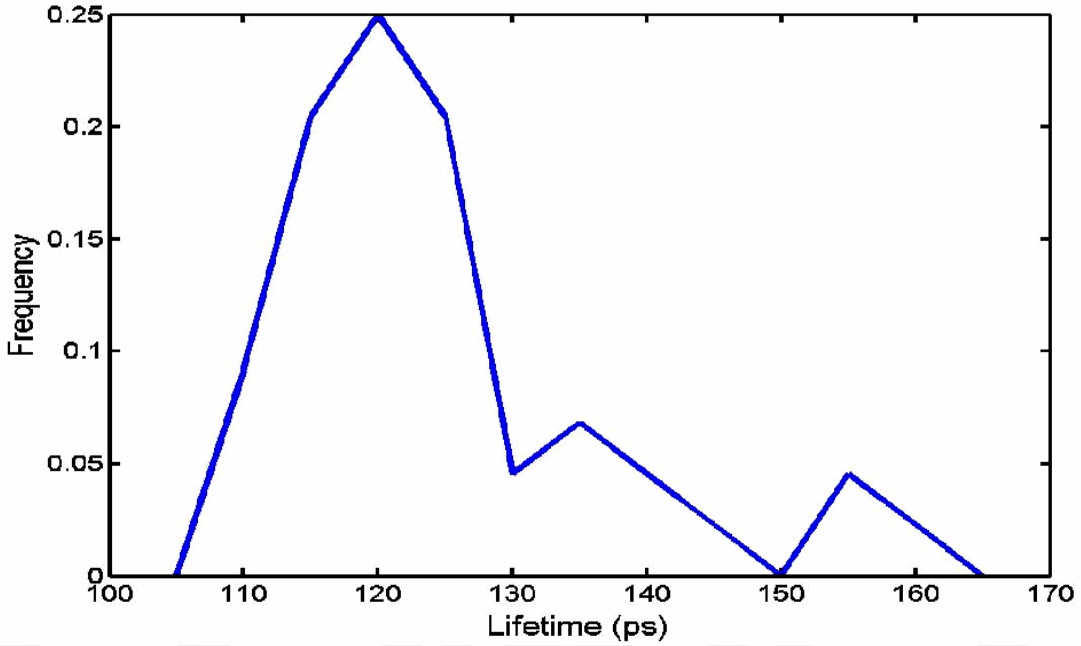
Non-periodic boxes

- Isosurface of the positron density at a shallow defect in the grain boundary of nc-Ni:



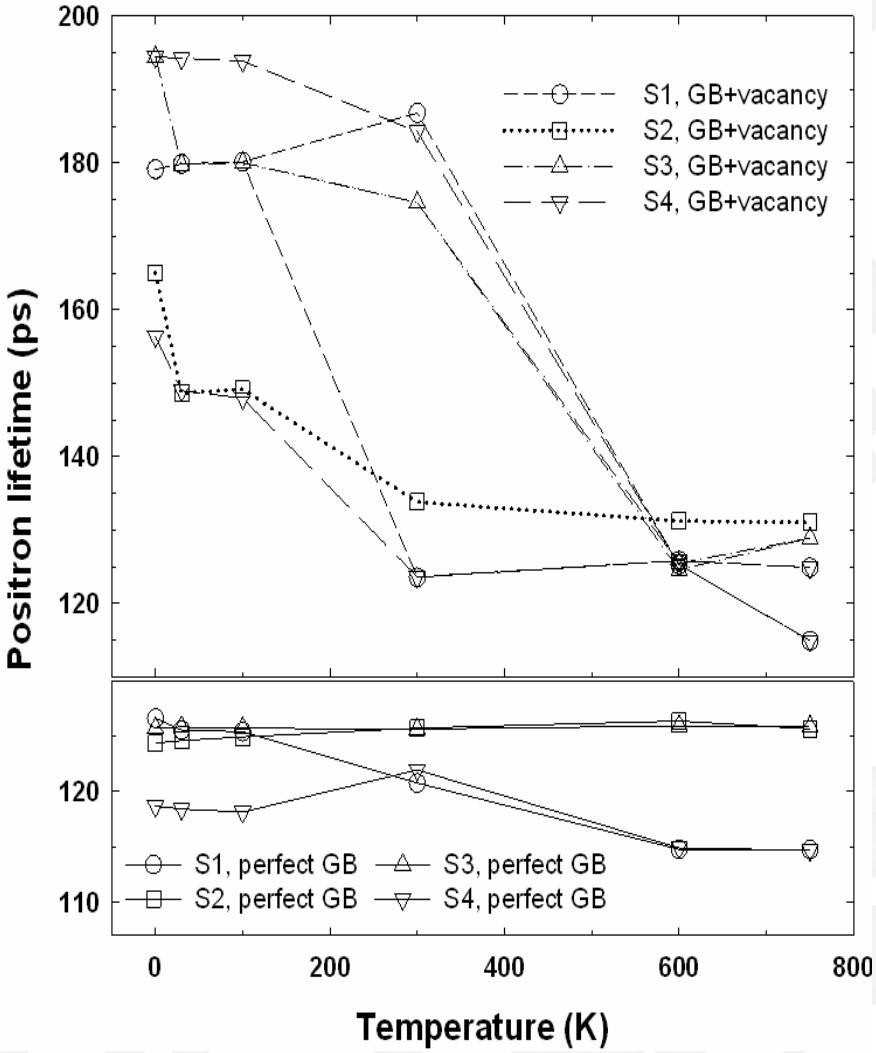
Non-periodic boxes

- The analysis of free volumes gives about 40 defects (3 vacancies) in nc-Ni (5 nm sample).
- The distribution of lifetimes is as follows:



Non-periodic boxes

- Comment on specific grain boundaries (GBs) in Ni:
- Four configurations of the tilt Σ=19 (331) GB studied with and without vacancies.
- Free volume associated with vacancies disappears latest at 600 K.
- Vacancy 'delocalization' occurs.
- Similar behavior observed for other GBs in Ni.
- This explains that GBs serve as a sink for vacancies.
- But are vacancies in nc-Ni different?



Non-periodic boxes

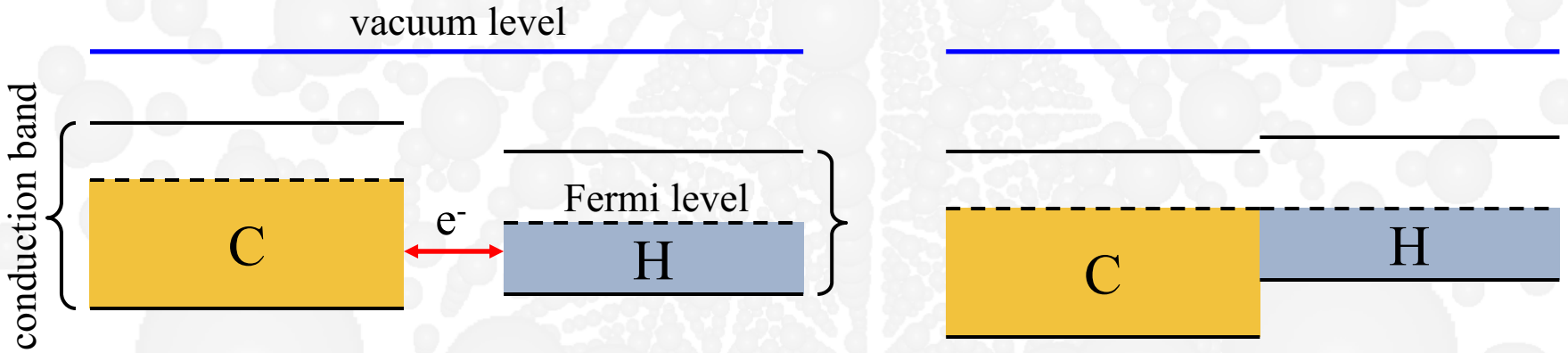
- Simulation of positron lifetimes from cascades in Fe and Fe-Cr will be presented in the next lecture.
- Behavior of vacancies at GBs in Fe will be also briefly discussed.

Positron affinity treatment

- The ATSUP method does not handle charge transfer in materials.
- Still it is possible to use it to study theoretically interaction of positrons with precipitates.
- The correction to have proper positron affinity difference between host and precipitate needs to be done `manually'.

Positron affinity treatment

- Two metals in contact (H=host, C=cluster):



- Positron affinity: $A_+ = \mu_- + \mu_+ = -(\Phi_- + \Phi_+)$.
- $\Delta A_+ = A_+^C - A_+^H$ determines the difference of positron levels of the host and cluster.
- If $\Delta A_+ < 0$, the cluster is attractive for positrons.
- If $\Delta A_+ = 0$, positron levels are equal.

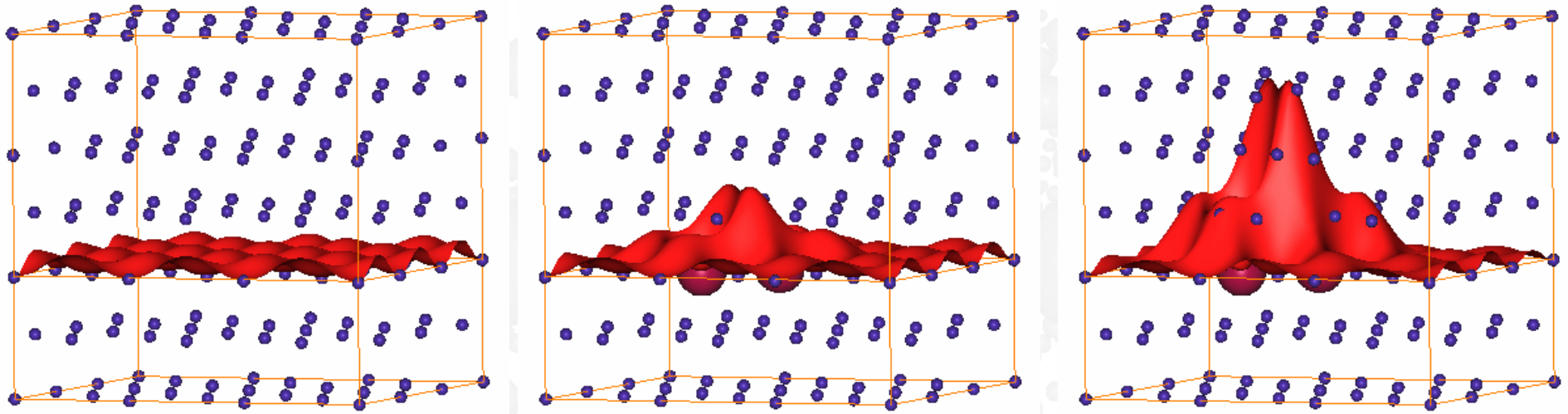
Positron affinity treatment

- Modified ATSUP method – three step procedure:

1. The energy (E_1) of delocalized positrons in the matrix is found.
2. The positron potential in the vicinity of cluster's atoms is shifted in order to get the positron energy equal to E_1.
3. The additional shift equal to ΔA_+ is applied.

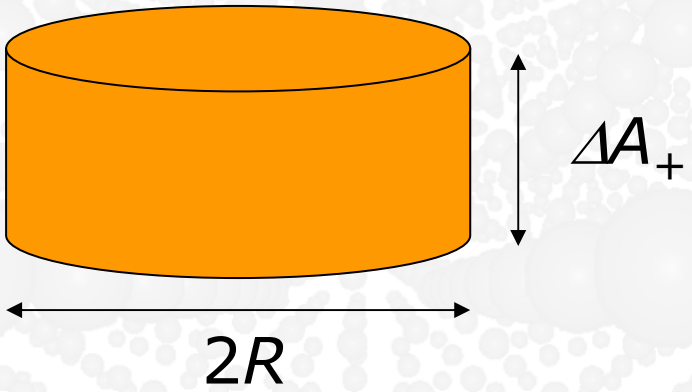
Positron affinity treatment

- Three step procedure:



Positron affinity treatment

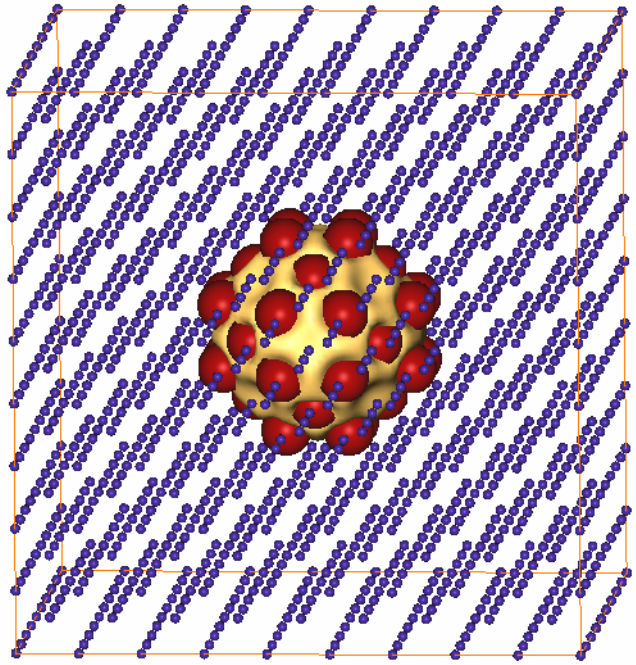
- Choice of model parameters:

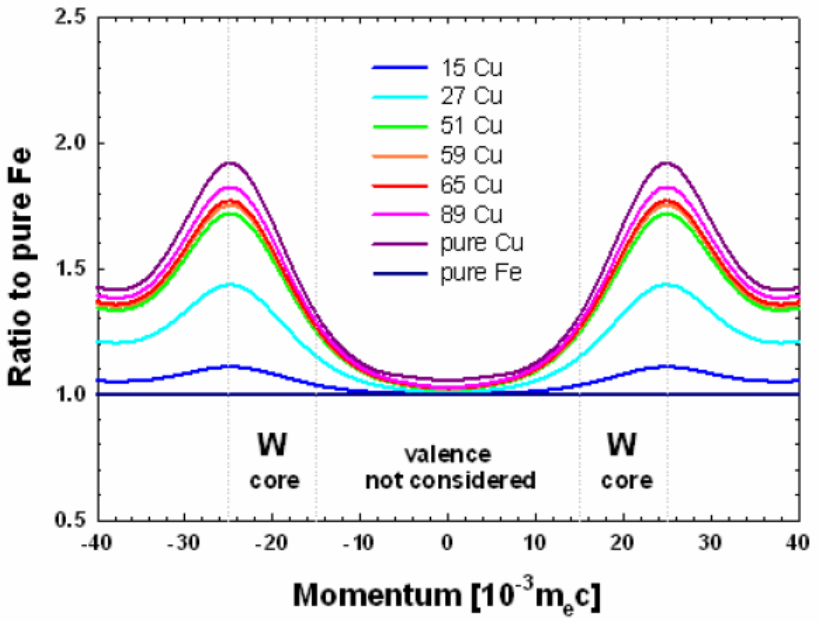


  - Fe-Cu: R= 1.6 Å, ΔA_+= 0.7 eV (LMTO)

Positron affinity treatment

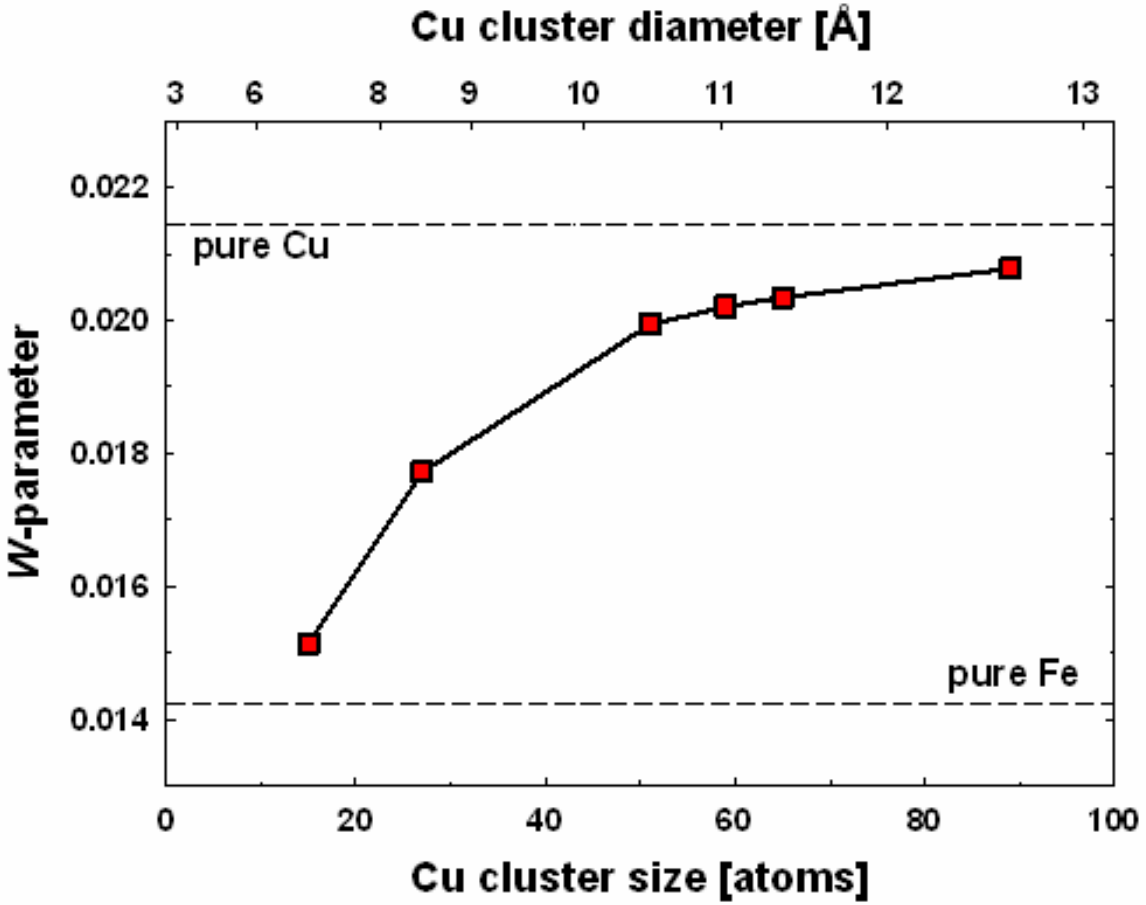
- Test calculation for regular Cu clusters in Fe.
- Cu-Fe alloys serve as a model system to study embrittlement of steels due to Cu clusters





Positron affinity treatment

- Dependence of the W parameter on the cluster size:



Positron affinity treatment

- Cu clusters smaller than ~10 atoms do not trap positrons.

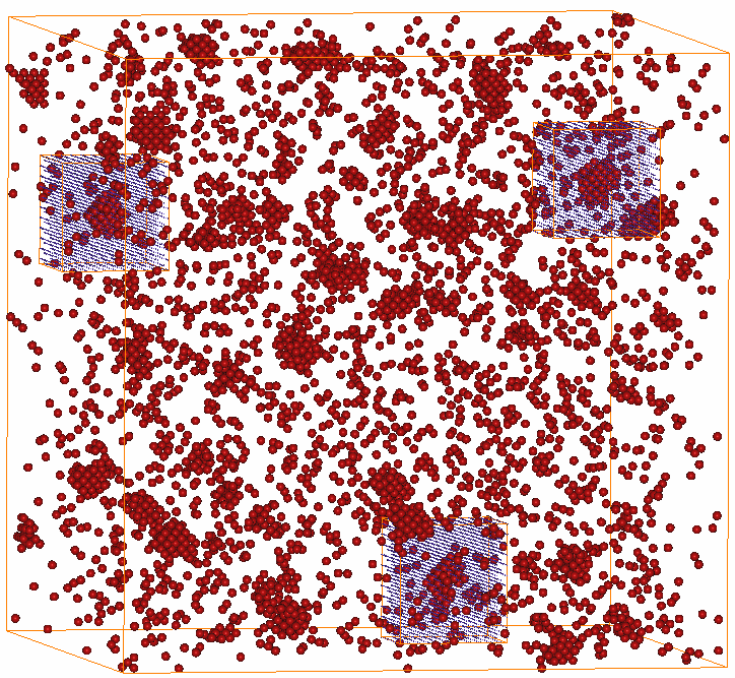
- Even if a cluster has $\Delta A_+ < 0$, there is a condition for minimum cluster radius

$$R > \frac{\pi\hbar}{\sqrt{8m_e|\Delta A_+|}}$$

that comes from quantum mechanics (there must be at least one level in the potential well).

Positron affinity treatment

- In the next lecture results for an AKMC simulated Fe-Cu alloy will be given.



Conclusions

- The atomic superposition method can be modified to handle non-periodic boxes, which enables to do positron calculations for large simulation boxes.
- In addition the difference of positron affinities between the host and precipitates can be also treated within the ATSUP method.
- This opens possibilities for new type of positron calculations/simulations, but not so many things were done in relation to experiment.

Thank you!