First principles simulation of resistivity recovery in irradiated beryllium

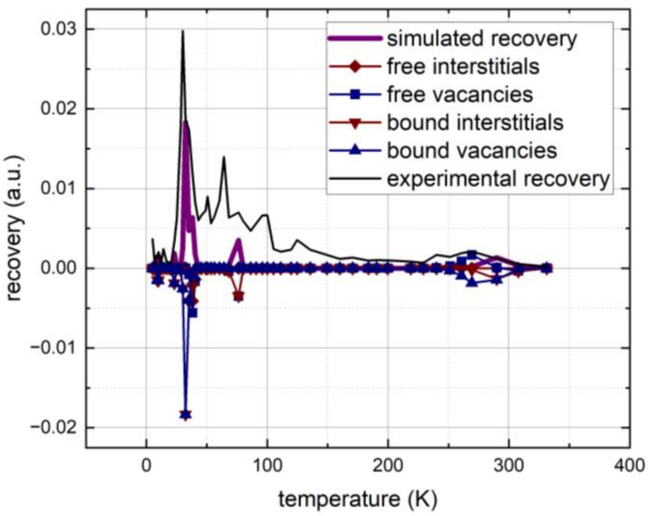
Christopher Stihl[1], Pavel Vladimirov[1]

[1]*Karlsruhe Institute of Technology, Institute for Applied Materials – Applied Materials Physics*

Future fusion devices like ITER or DEMO require closed fuel cycles. These vitally depend on neutron multiplying materials as part of a breeding blanket module like beryllium pebbles in the European Helium-Cooled Pebble-Bed. During operation the beryllium pebbles will accumulate point defects, tritium, and helium due to inevitable exposure to highly energetic neutron irradiation as emitted by the fusion plasma. A detailed knowledge of the characteristics of point defects is decisive for reliable simulations of microstructure evolution in irradiated beryllium. Such models are a prerequisite for predicting tritium inventory during operation as well as after the blanket´s end of life since tritium retention and release is the paramount safety concern.

A well-established experimental approach to assess the dynamics of relevant atomic defects consists in measuring electrical resistivity recovery (ERR) after irradiation during annealing. Within this approach, temperatures corresponding to electrical recovery steps are correlated with activation energies which are associated with different type of reactions between defects.

In this work, results of our ongoing efforts to model and understand the ERR of beryllium are presented. To that end, we introduce a rate equation-based approach to model ERR spectra (see picture below) utilizing density functional theory results as input. Within this approach, electrical resistivity recovery models comprising the volume of spontaneous recombination of monovacancies and self-interstitial atoms in beryllium as well as various additional defects are considered. As a result, an intricate interplay between different defect dynamics is uncovered, suggesting a clear route for further research to obtain systematically improved electrical resistivity recovery models.



Corresponding Author:
Dr. Christopher Stihl
Christopher.stihl@kit.edu
Hermann-von-Helmholtz-Platz 1,
76344 Eggenstein-Leopoldshafen

Karlsruhe Institute of Technology

First principles simulation of resistivity recovery in irradiated beryllium

Christopher Stihl

KIT – The Research University in the Helmholtz Association

www.kit.edu

Outline

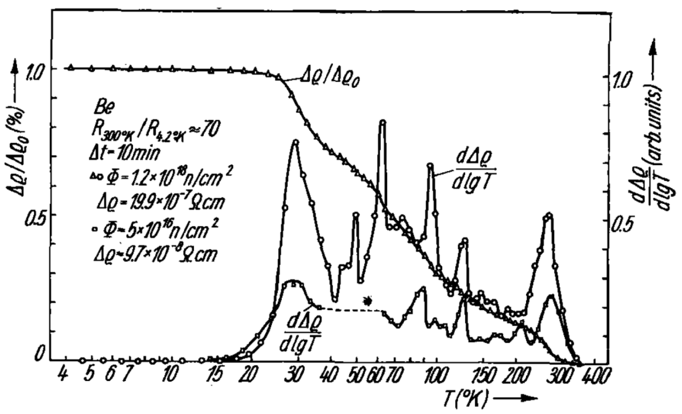
- Resistivity Recovery Experiments
- Resistivity Recovery Modelling
  - Relevant Be properties
  - Model derivation
  - Various models
- Conclusions & outlook

2 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

Resistivity recovery experiment(s by example)

Karlsruhe Institute of Technology

- sample preparation
  - cool to 4.2 K
  - measure resistivity
  - irradiate
  - measure resistivity
- measurements
  - do annealing cycle
    - heat
    - hold
    - cool
  - measure resistivity
  - repeat at higher T
- process spectrum

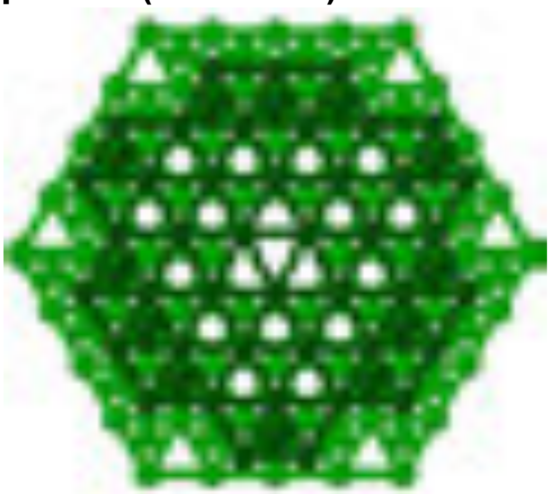


3 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

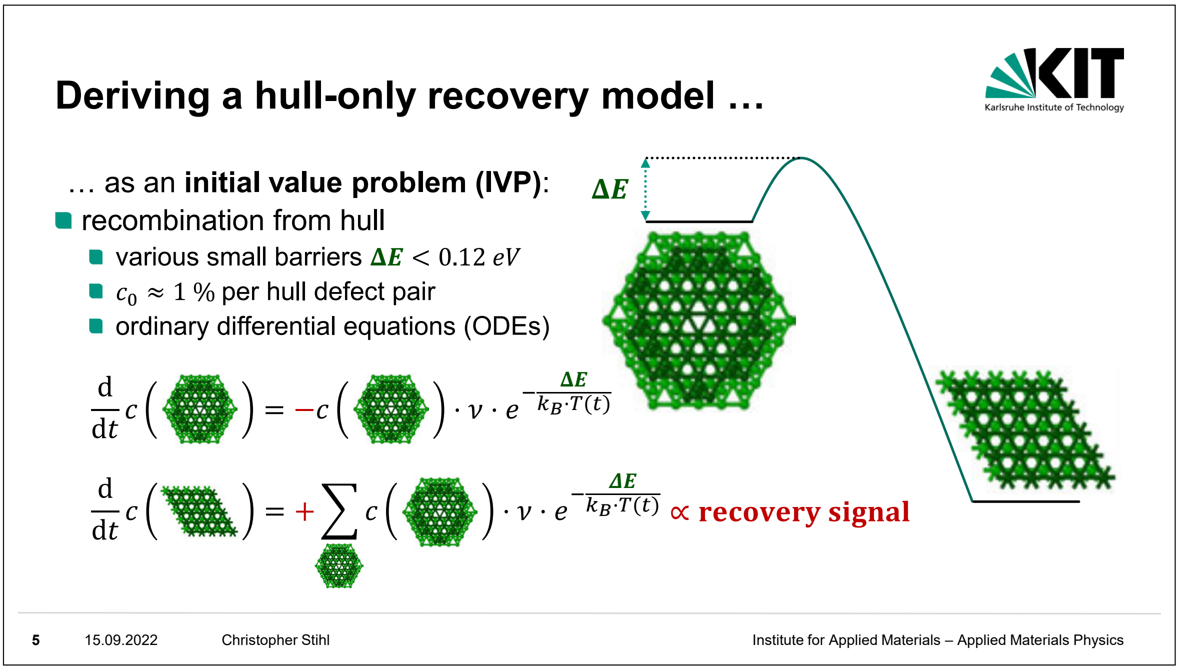
Model-relevant Be properties (from DFT)

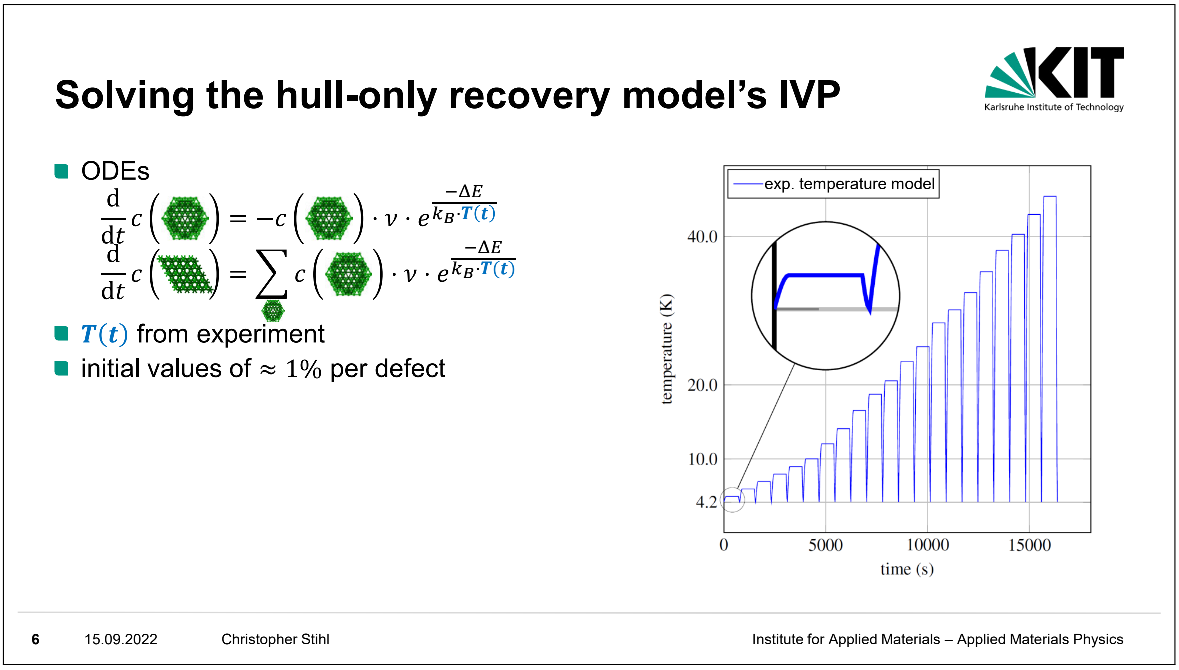
Karlsruhe Institute of Technology

- HCP structure
- **AB**-stacked layers
- mono vacancies
  - immobile (0.8 eV)
- basal interstitials
  - mobile (0.12 eV)
- spontaneous recombination hull
  - oblate spheroid
  - with (very) fast recombination



4 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics


Deriving a hull-only recovery model …
Karlsruhe Institute of Technology
… as an initial value problem (IVP):
recombination from hull
various small barriers $\Delta E < 0.12\ eV$
$c_0 \approx 1\ \%$ per hull defect pair
ordinary differential equations (ODEs)
$\frac{\mathrm{d}}{\mathrm{d}t} c(\ ) = -c(\ ) \cdot \nu \cdot e^{-\frac{\Delta E}{k_B \cdot T(t)}}$
$\frac{\mathrm{d}}{\mathrm{d}t} c(\ ) = +\sum c(\ ) \cdot \nu \cdot e^{-\frac{\Delta E}{k_B \cdot T(t)}}$ ∝ recovery signal
ΔE
5 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics


Solving the hull-only recovery model's IVP
Karlsruhe Institute of Technology
ODEs
$\frac{\mathrm{d}}{\mathrm{d}t} c(\ ) = -c(\ ) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$
$\frac{\mathrm{d}}{\mathrm{d}t} c(\ ) = \sum c(\ ) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$
$T(t)$ from experiment
initial values of ≈ 1% per defect
exp. temperature model
temperature (K)
40.0
20.0
10.0
4.2
0
5000
10000
15000
time (s)
6 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

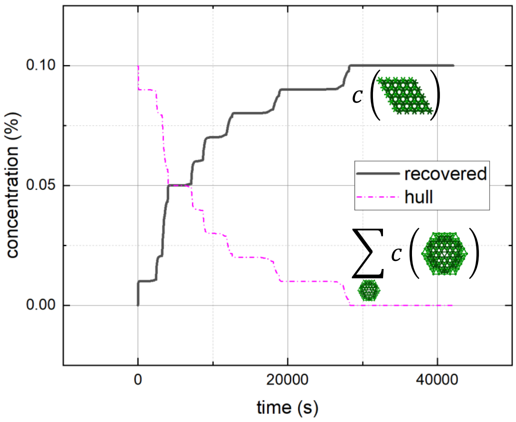
Solving the hull-only recovery model's IVP

KIT Karlsruhe Institute of Technology

- ODEs

$$\frac{\mathrm{d}}{\mathrm{d}t} c\left(\;\right) = -c\left(\;\right) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$$
$$\frac{\mathrm{d}}{\mathrm{d}t} c\left(\;\right) = \sum c\left(\;\right) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$$

- $T(t)$ from experiment
- initial values of $\approx 1\%$ per defect
- symmetric hull and recovery evolution
- sloped step patterns from annealing cycles



6 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

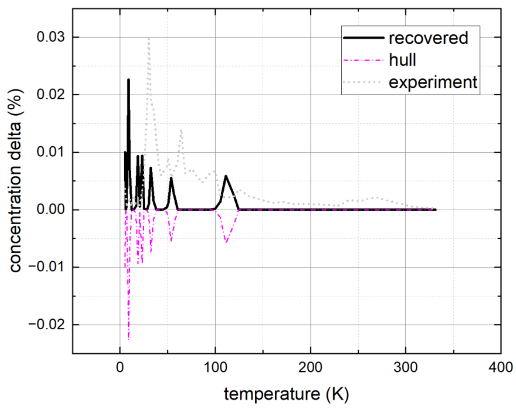
Solving the hull-only recovery model's IVP

KIT Karlsruhe Institute of Technology

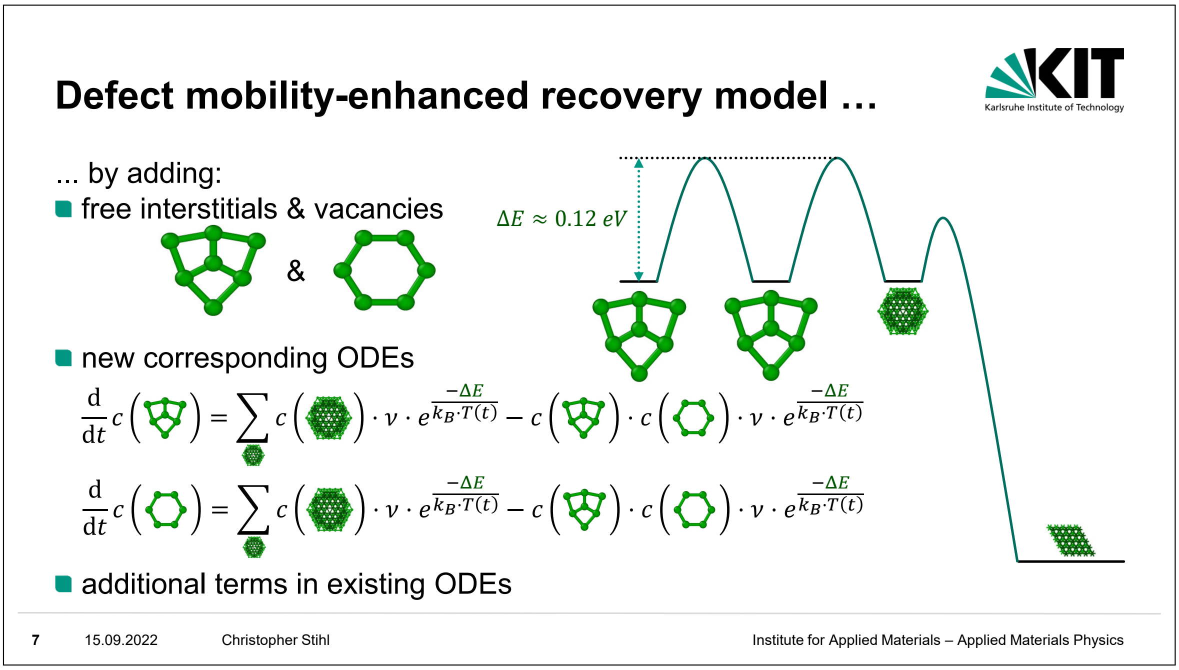
- ODEs

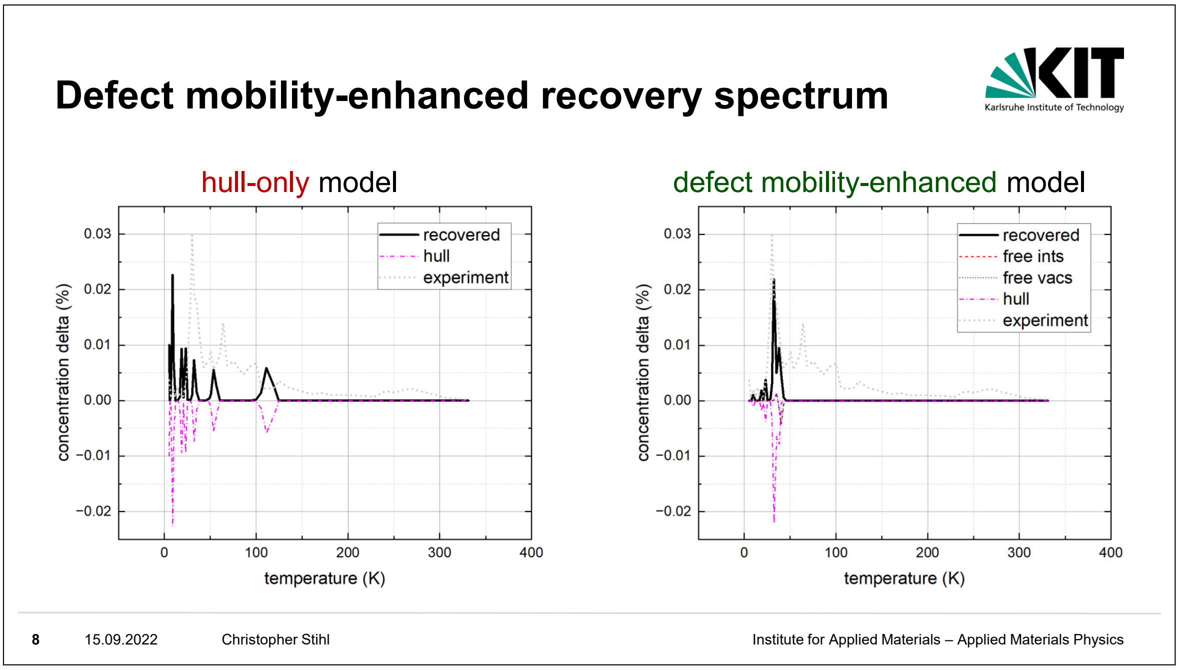
$$\frac{\mathrm{d}}{\mathrm{d}t} c\left(\;\right) = -c\left(\;\right) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$$
$$\frac{\mathrm{d}}{\mathrm{d}t} c\left(\;\right) = \sum c\left(\;\right) \cdot \nu \cdot e^{\frac{-\Delta E}{k_B \cdot T(t)}}$$

- $T(t)$ from experiment
- initial values of $\approx 1\%$ per defect
- symmetric hull and recovery evolution
- sloped step patterns from annealing cycles
- +/- peaks show in/decreasing concentration
- lowest-energy hull pairs decay early
- initial values are important free parameter



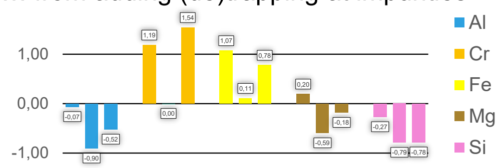
6 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics


Defect mobility-enhanced recovery model …
KIT
Karlsruhe Institute of Technology
... by adding:
free interstitials & vacancies
&
$\Delta E \approx 0.12\ eV$
new corresponding ODEs
additional terms in existing ODEs
7
15.09.2022
Christopher Stihl
Institute for Applied Materials – Applied Materials Physics

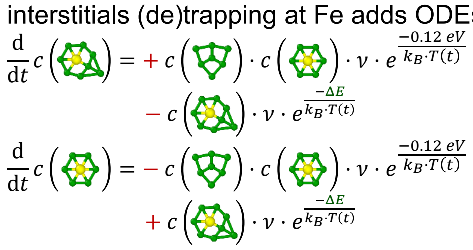

Defect mobility-enhanced recovery spectrum
KIT
Karlsruhe Institute of Technology
hull-only model
defect mobility-enhanced model
recovered
hull
experiment
free ints
free vacs
concentration delta (%)
temperature (K)
8
15.09.2022
Christopher Stihl
Institute for Applied Materials – Applied Materials Physics

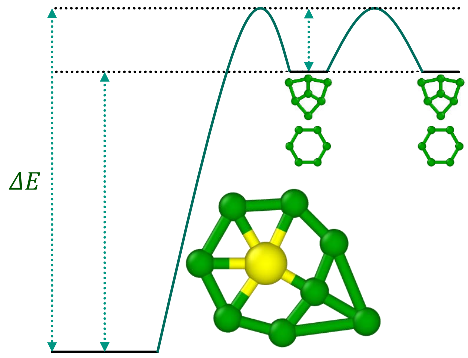
Impurity-enhanced recovery models …

… from adding (de)trapping at impurities



- interstitials (de)trapping at Fe adds ODEs

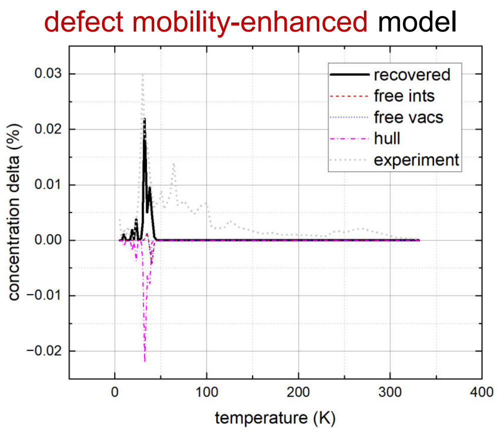


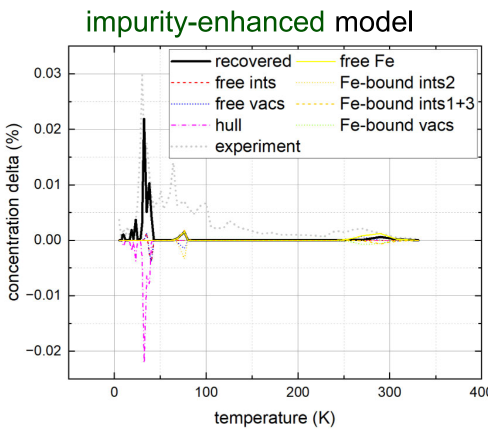


- analogous vacancy (de)trapping ODEs
- additional terms in existing ODEs

9 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

Fe impurity-enhanced recovery spectrum



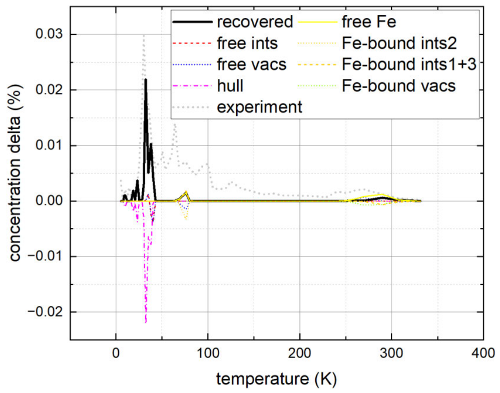


10 15.09.2022 Christopher Stihl Institute for Applied Materials – Applied Materials Physics

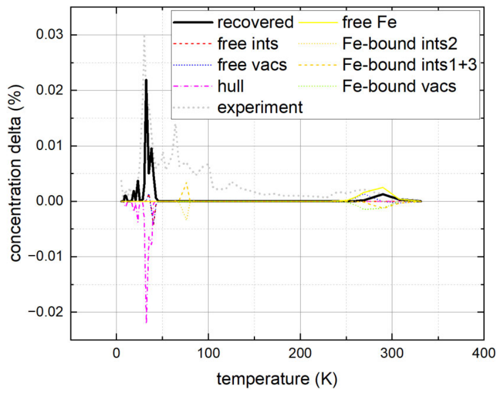
Fe impurity-enhanced recovery spectra

KIT
Karlsruhe Institute of Technology

impurity-enhanced model



impurity-enhanced, vac-starved model



Conclusions & outlook

KIT
Karlsruhe Institute of Technology

- first peaks due to closely correlated pairs
  - reasonable defect concentrations sufficient
  - gaps likely due to non-exhaustive set of pairs
- diffusion onset decisive for later spectrum
- last peaks due to mobile vacancies
  - here from vacs weakly bound to Fe
- interdependent dynamics of different traps
  - e.g. different traps at Fe (and additional ones …)
- intermediate peaks *likely not* due to impurities
  - concentrations *too low*
  - might still contribute some peaks
- additional Be-only defects to be considered
- model can be extended to calorimetry signals