The Henryk Niewodniczański
Institute of Nuclear Physics
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Ab initio modeling of materials with defects

Przemysław Piekarz, Krzysztof Parlinski, Jan Łażewski, Paweł T. Jochym, Małgorzata Sternik, Andrzej Ptok

Computational Materials Science
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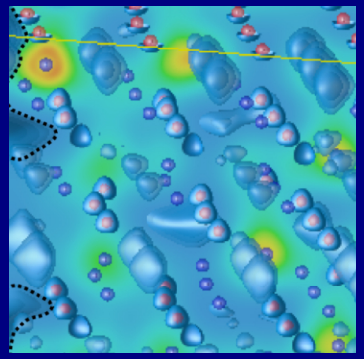

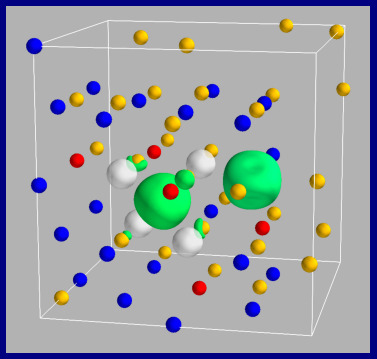

Outline

- Method - density functional theory
- Defects in crystals
- Influence on materials properties
- Irradiation defects

Density functional theory (DFT)

$$E_{tot}[n]=E_K[n]+E_{ext}[n]+E_H[n]+E_{xc}[n]=\min$$

$$\left(-\frac{\hbar^2\nabla^2}{2m}+V_{KS}\right)\psi_i=\epsilon_i\psi_i$$

$$n(\vec{r})=\sum_i|\psi_i(\vec{r})|^2$$

$$V_{KS}=V_{ext}+V_H+V_{xc}$$

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$$n(\vec{r}) = \sum_i |\psi_i(\vec{r})|^2$$

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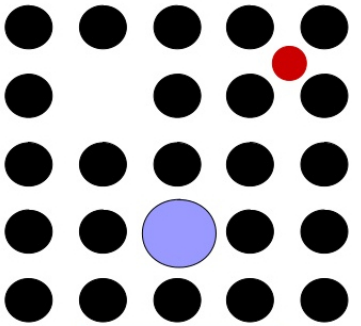
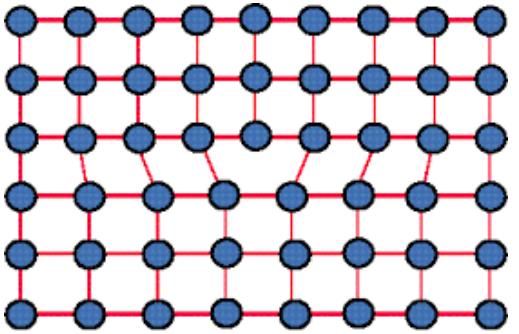
- Electronic structure and magnetic properties
- Lattice parameters and atomic positions
- Interatomic forces and stress tensor
- Elastic and mechanical properties
- Lattice dynamical properties
- Molecular dynamics

Crystal defects

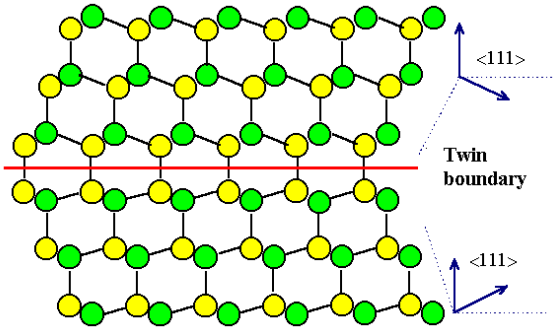
- Point defects
  - vacancy (Schottky defect)
  - interstitial atom
  - Frenkel defect (vacancy and interstitial)
  - impurity
  - antisite



- Linear defects
  - dislocation (edge, screw)



- Planar defects
  - grain boundary
  - antiphase boundary
  - stacking fault
  - twin boundary



- Bulk defects
  - pores, cracks, inclusions
  - clusters of vacancies (voids)
  - clusters of impurities (precipitates)

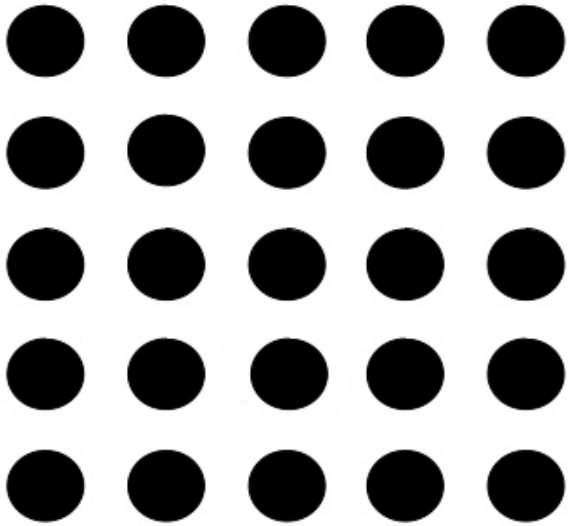
Point defects

Vacancy formation energy

$$E_f = E_{N-1} - \frac{N-1}{N} E_N$$

E_N – total energy of N atoms

E_{N-1} – total energy of N – 1 atoms



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	Al	Cu	Au	Ni	Pd	Pt	Pu
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E_m	0.57[m]	0.72[d]	–	1.285,[e] 1.08[r]	–	1.51[j]	–
	V	**Nb**	**Ta**	**Cr**	**Mo**	**W**	**Fe**
E_f	2.51[l]	2.99[l]	3.14[l]	2.64[l]	2.96,[j] 2.96[l]	3.56[l]	2.02,[b] 2.07,[k] 2.15[l]
E_m	0.62[l]	0.91[l]	1.48[l]	0.91[l]	1.28[l]	1.78[l]	0.65,[b] 0.67,[k] 0.64[l]
	C	**Si**	**Ge**	**Be**	**Ti**	**Zr**	**Hf**
E_f	8.2[f]	3.17,[c] 3.29[g]	2.3[h]	0.81,[n] 1.09[o]	1.97,[p] 2.13[q]	2.17,[q] 1.86[s]	2.22[q]
E_m	1.7[f]	0.4[g]	–	0.72B, 0.89NB[o]	0.47B, 0.61NB[p]	0.51B, 0.67NB[q]	0.79B, 0.91NB[q]

S. L. Dudarev, Annu. Rev. Mater. Res. 43, 35 (2013)

Point defects

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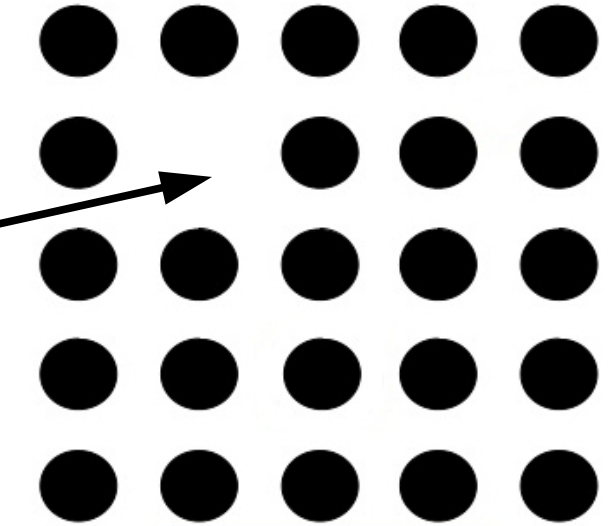

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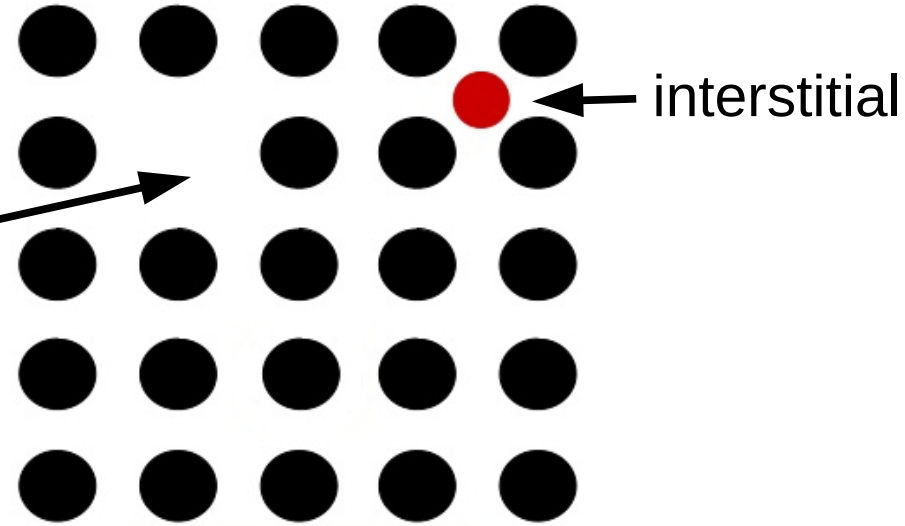

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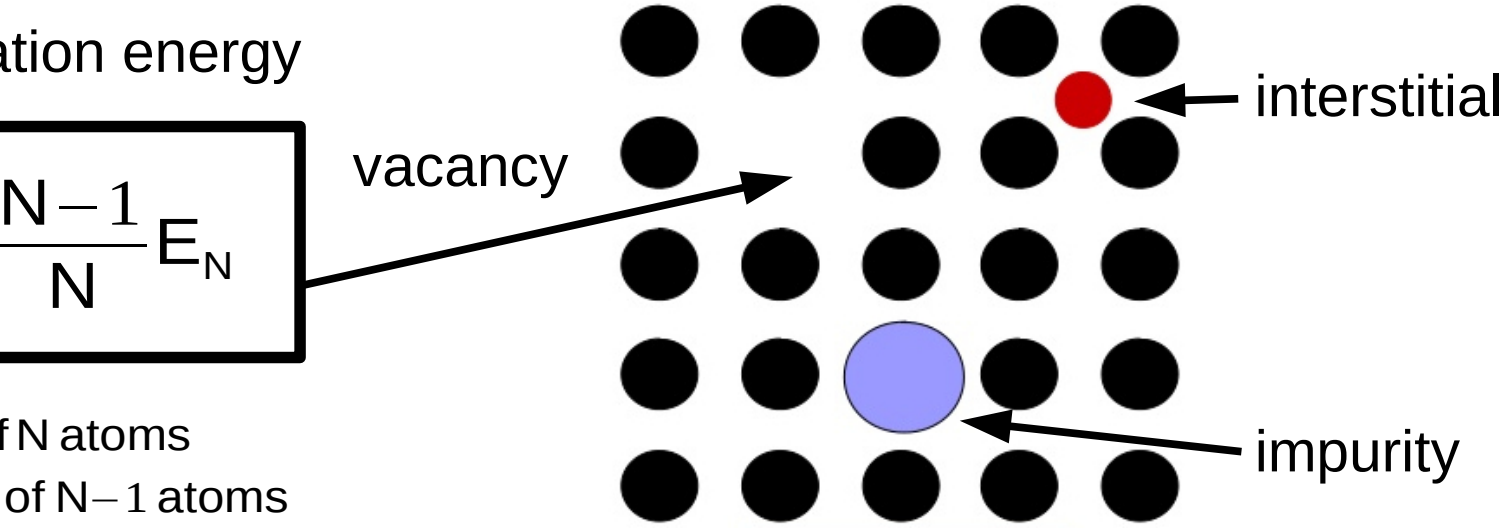

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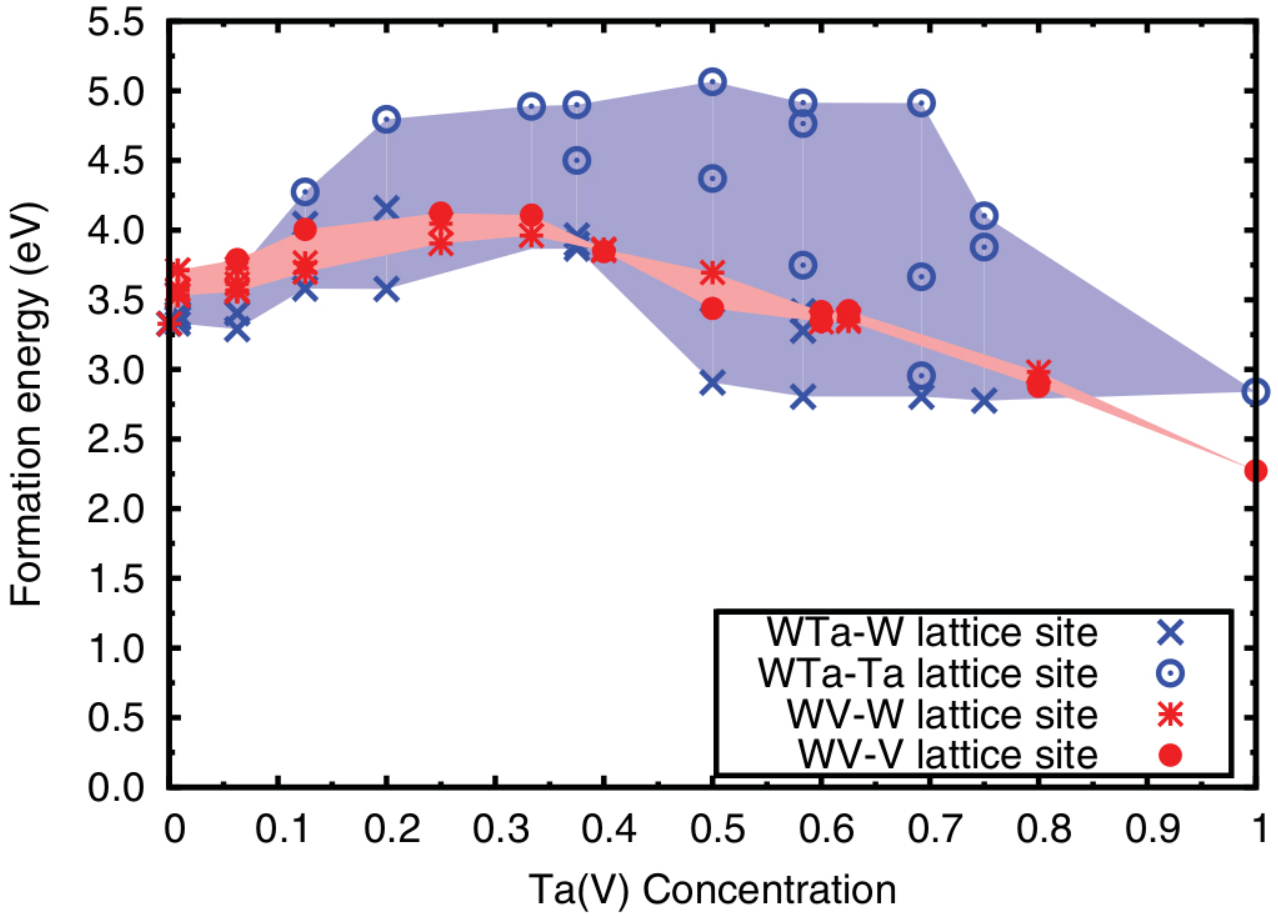
S. L. Dudarev, Annu. Rev. Mater. Res. 43, 35 (2013)

Point defects in alloys

Formation energies of vacancies in alloys depend on chemical composition, local atomic environment and lattice site

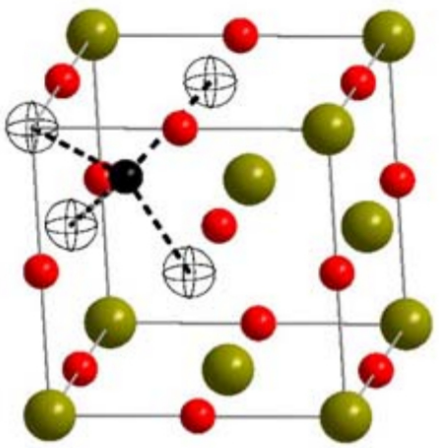


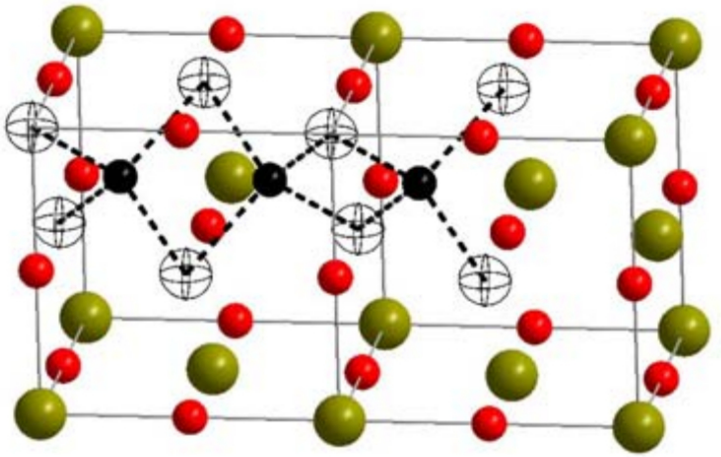
M. Muzyk, D. Nguyen-Manh, K. J. Kurzydłowski, N. L. Baluc, S. L. Dudarev, Phys. Rev. B 84, 104115 (2011)

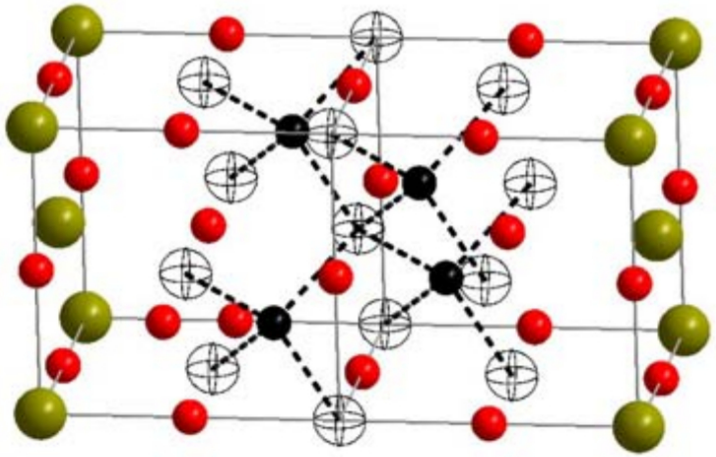
Iron oxide – defect clusters

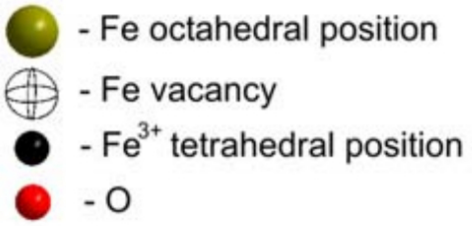
$Fe_{1-x}O$ x~0.05-0.15

Energy formation of Fe vacancies and clusters $E_f < 0$









Iron oxide – defect clusters

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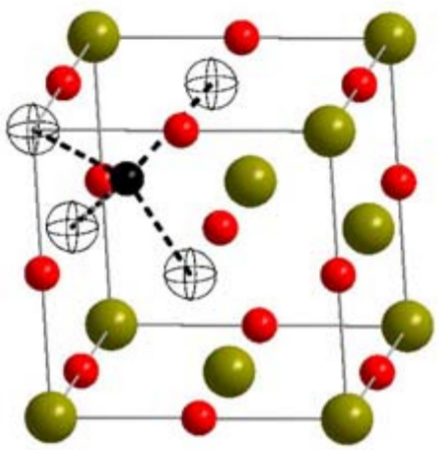
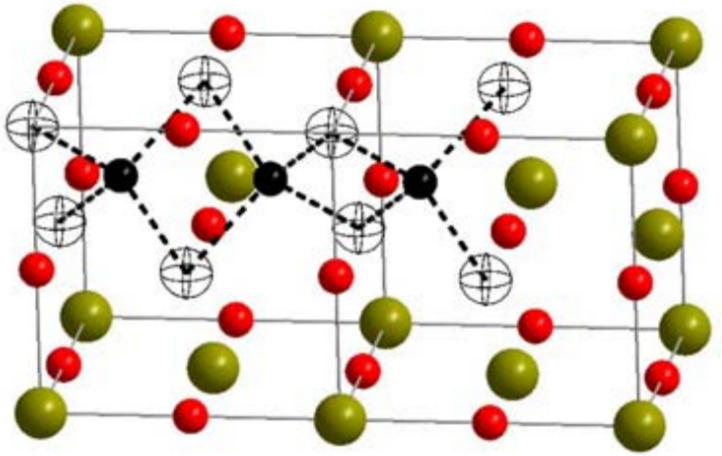
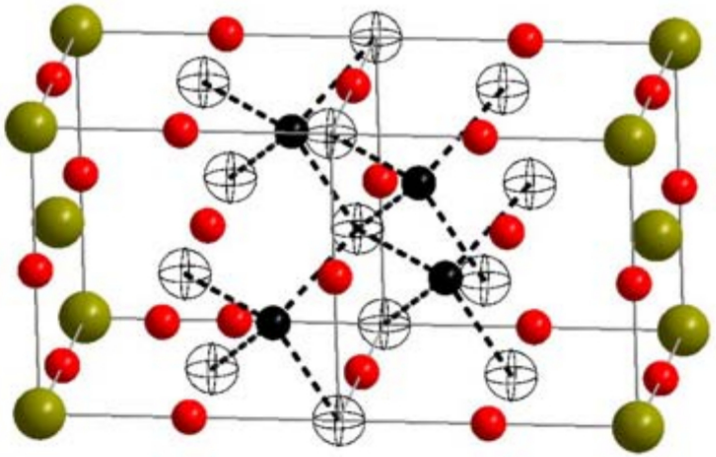
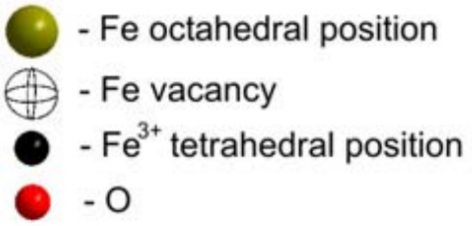
Cluster 4:1
4 Fe vacancies
1 Fe interstitial









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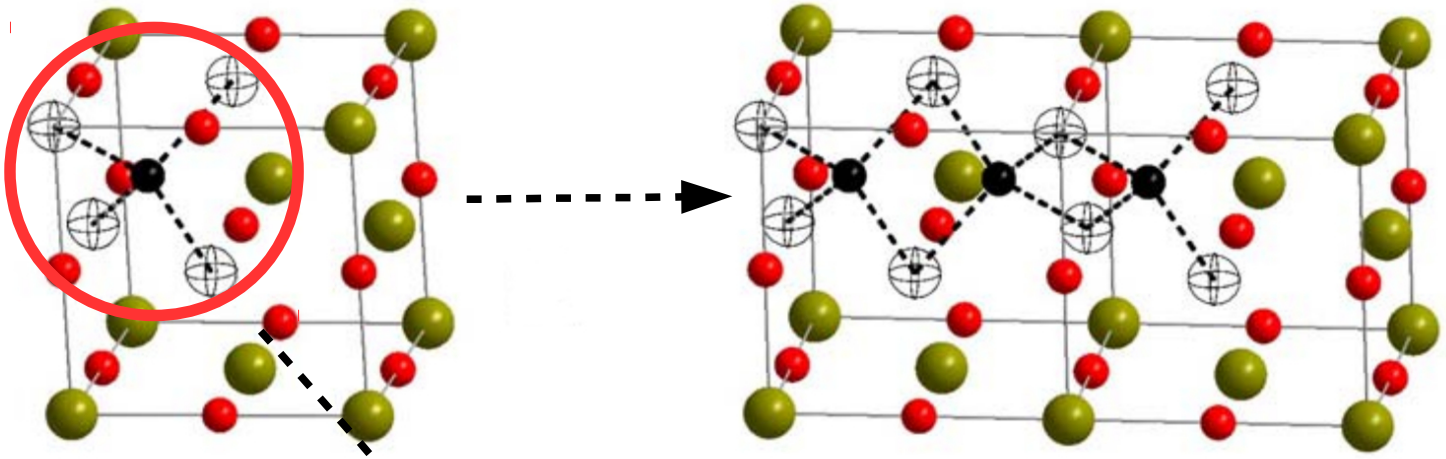
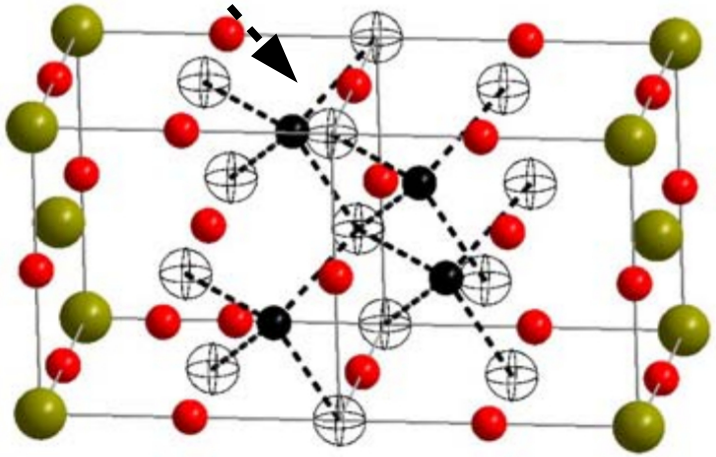
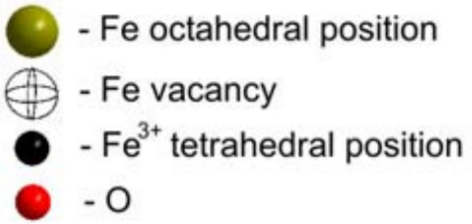


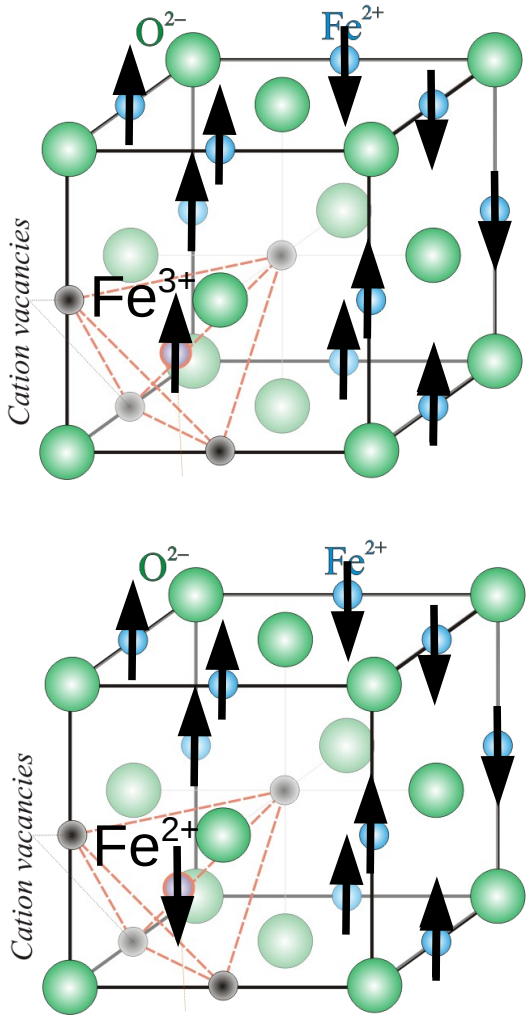
Clusters accumulate and creat larger defect structures

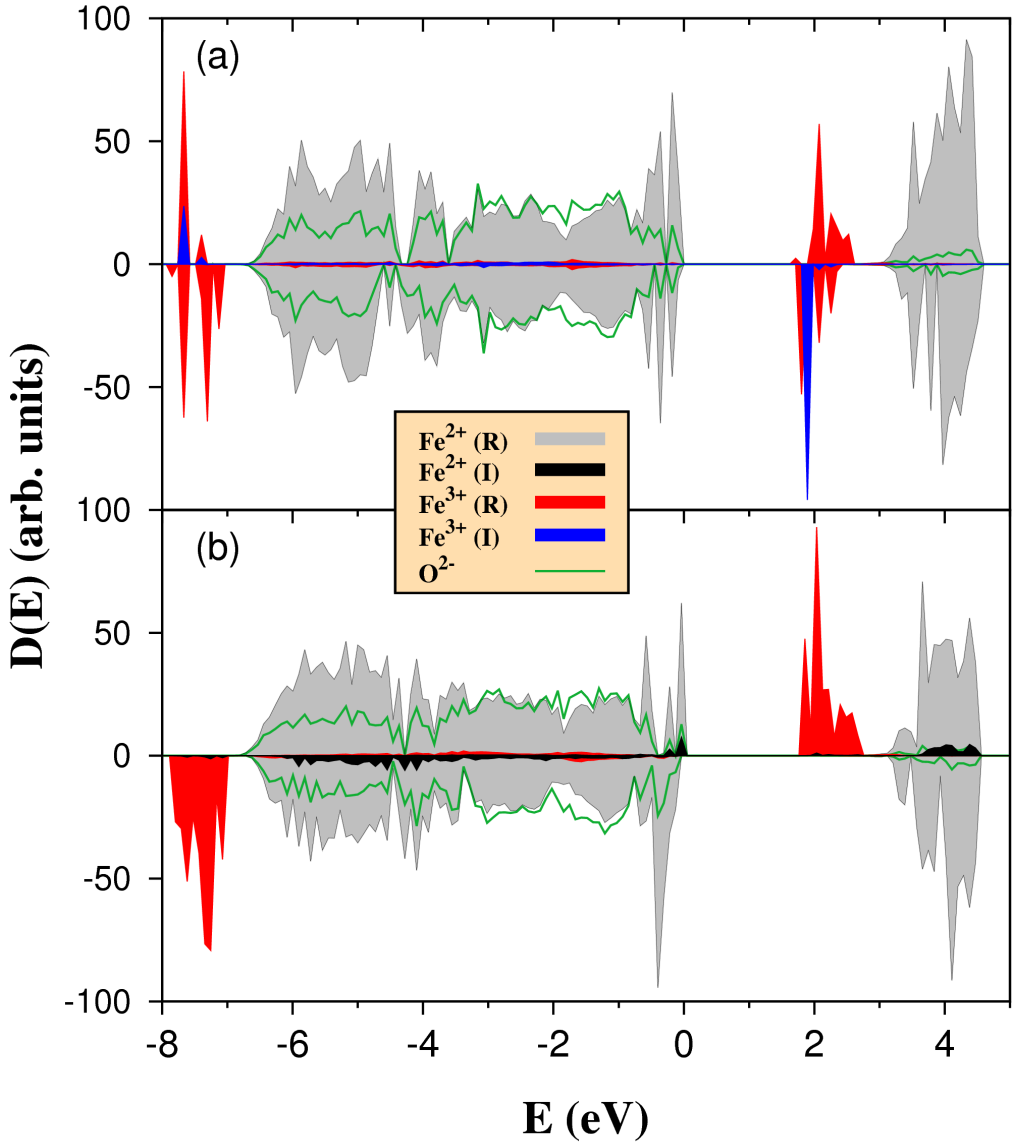




Iron oxide – electronic structure

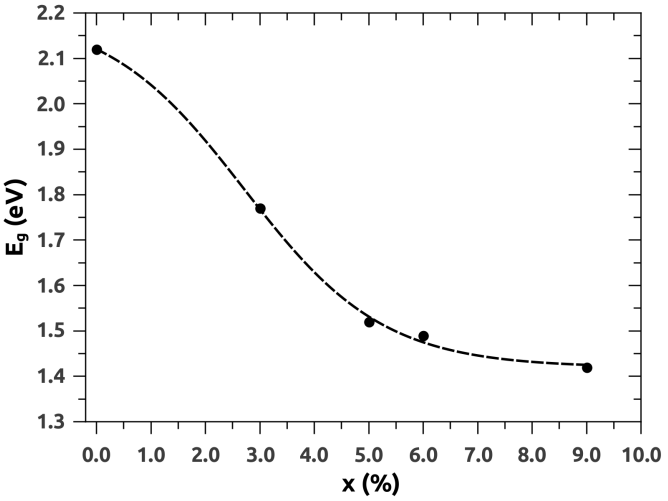
Vacancies and interstitial Fe atoms introduce additional electronic states and strongly modify the band structure

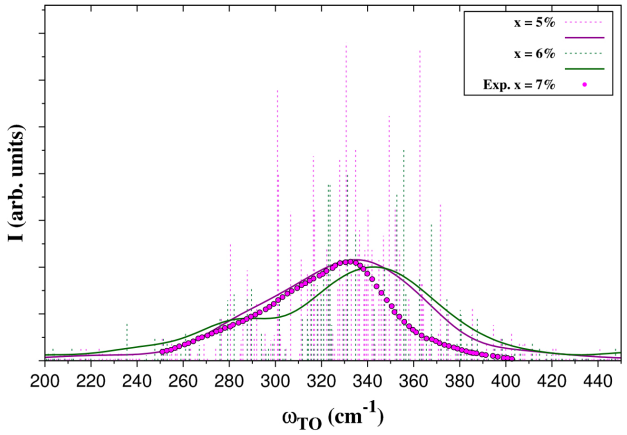


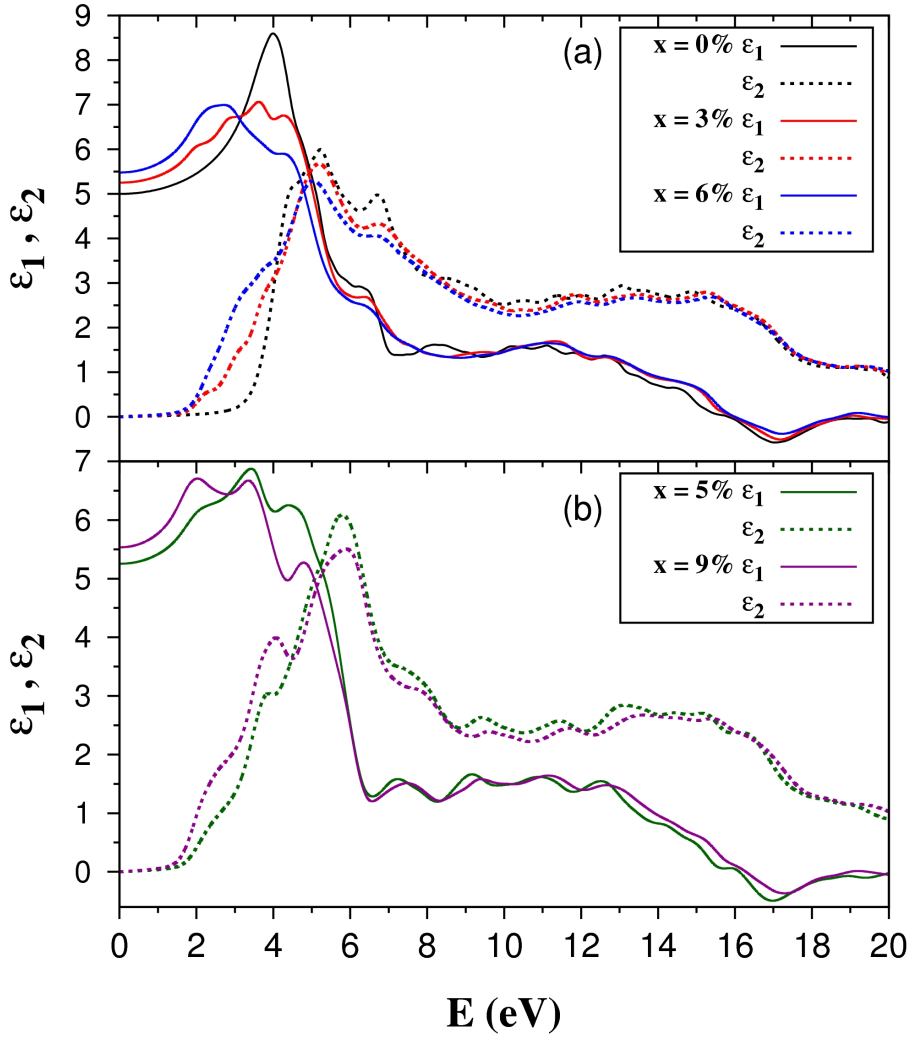


Iron oxide - dielectric properties

The optical gap, dielectric functions and phonon infrared absorption depend on the concentration of defects *x*





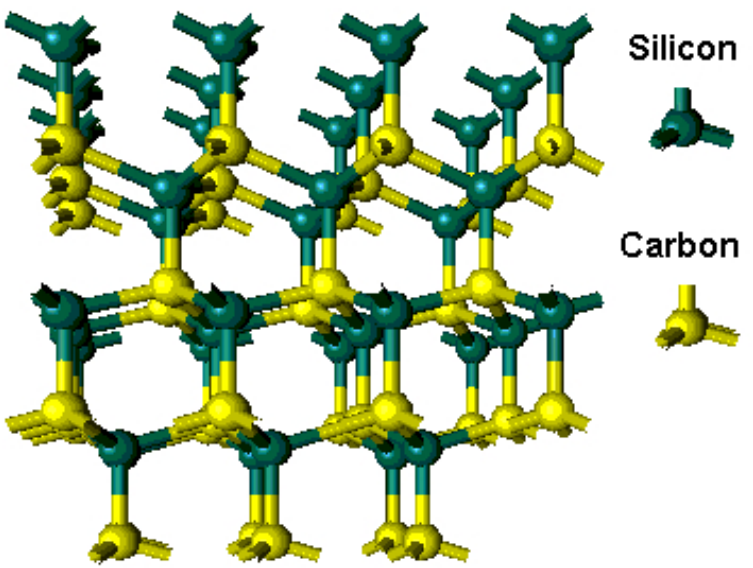


U. D. Wdowik, P. Piekarz, P. T. Jochym, K. Parlinski, A. M. Oleś, Phys. Rev. B 91, 195111 (2015)

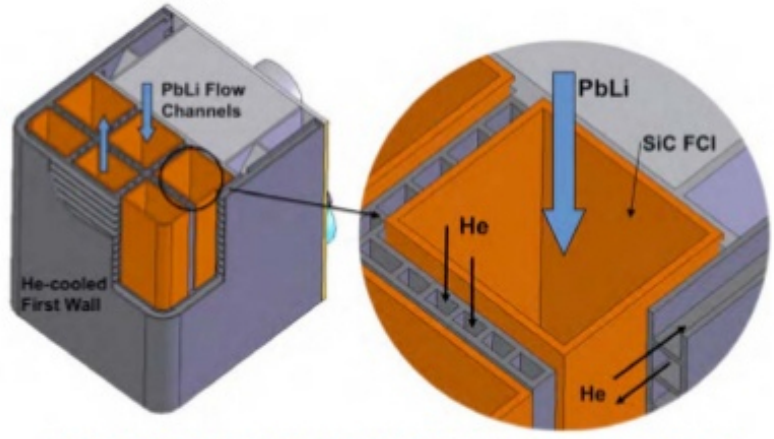
Silicon carbide SiC

SiC - a wide band gap semiconductor is a promising material for high-voltage and high-frequency nanoelectronic devices:

- high values of breakdown voltage
- high charge carrier mobility
- high temperature stability
- high thermal conductivity
- very good mechanical properties
- resistance to radiation damage



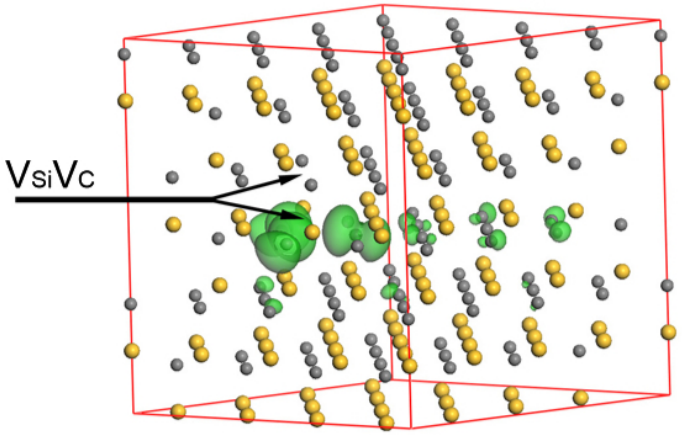
The electronic properties of epitaxial layers strongly depend on the material quality. The presence of intrinsic defects and impurities which arise during crystal growth process substantially limit applications of SiC

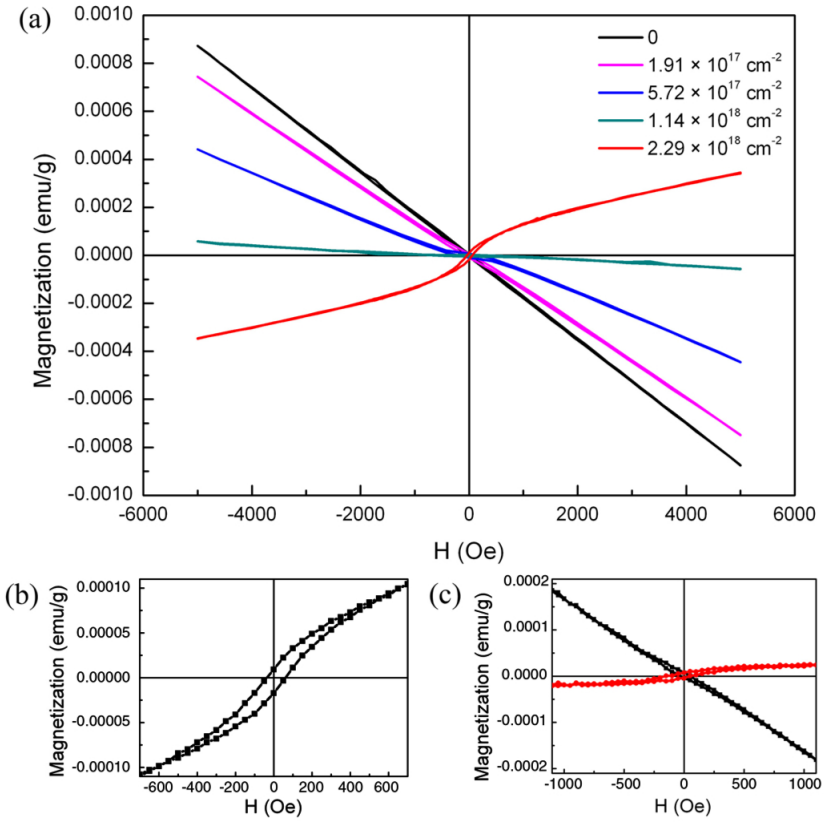


Application in fusion power plant: use of SiC flow channel inserts (FCI) as electrical and thermal insulator in the Dual Coolant Lithium Lead (DCLL) blanket

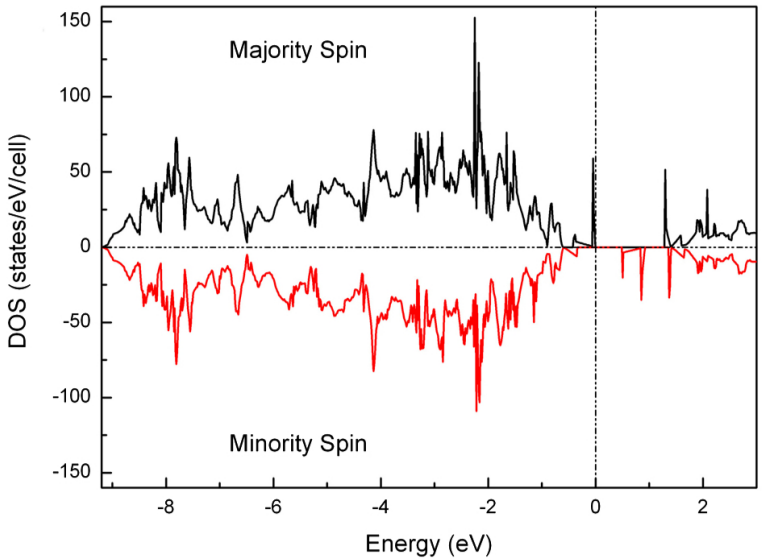
Defect-induced magnetism in neutron irradiated SiC

The intentionally created defects dominated by divacancies ($V_{Si}V_C$) are responsible for the observed magnetism in the SiC single crystal





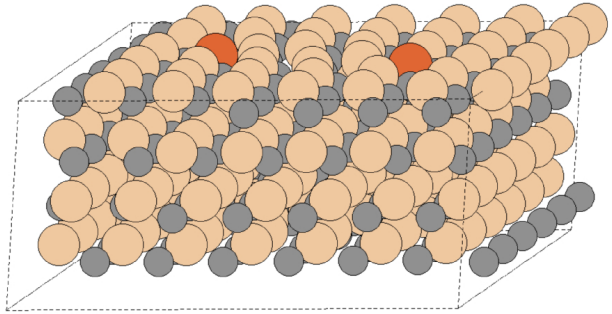
DFT calulations: the vacancy-induced electronic states show spin polarization



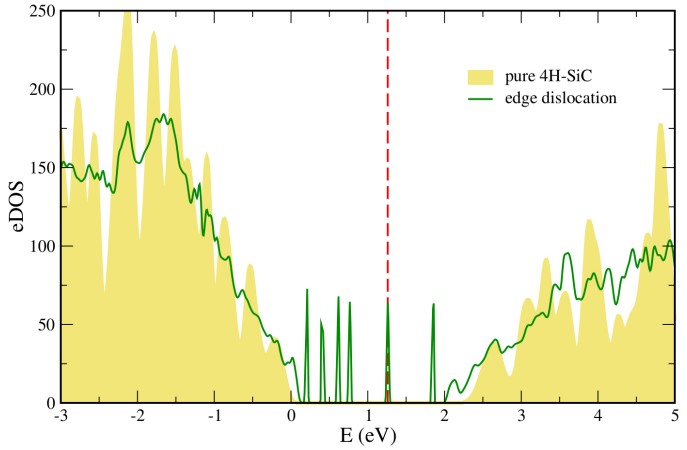
Y. Liu *et al.* Phys. Rev. Lett. 106, 087205 (2011)

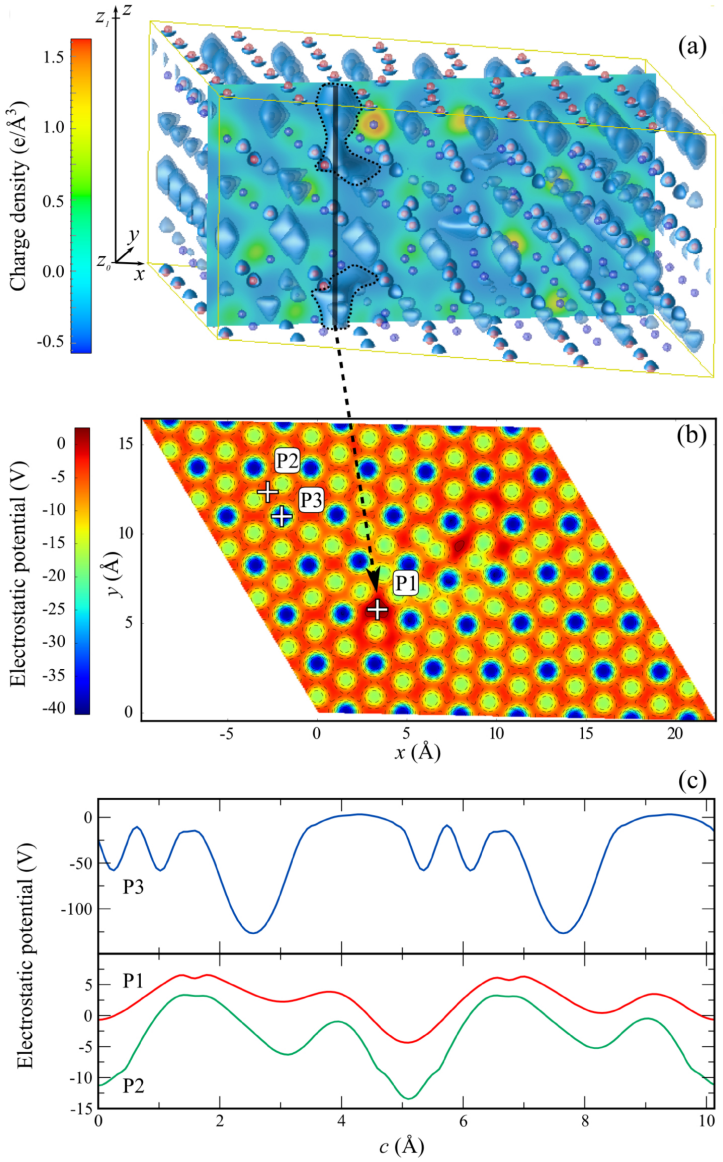
Dislocations in SiC

Pair of edge dislocations in SiC with opposite Burgers vectors



Edge dislocations induce the electronic states in the gap and modify charge density and electrostatic potential

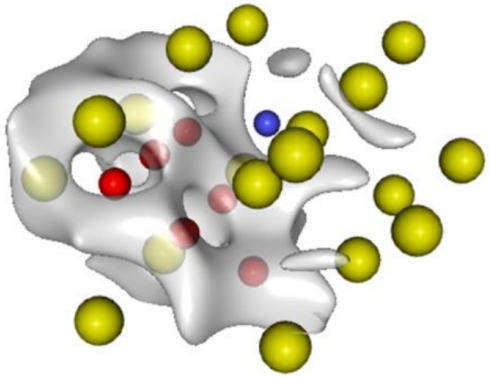




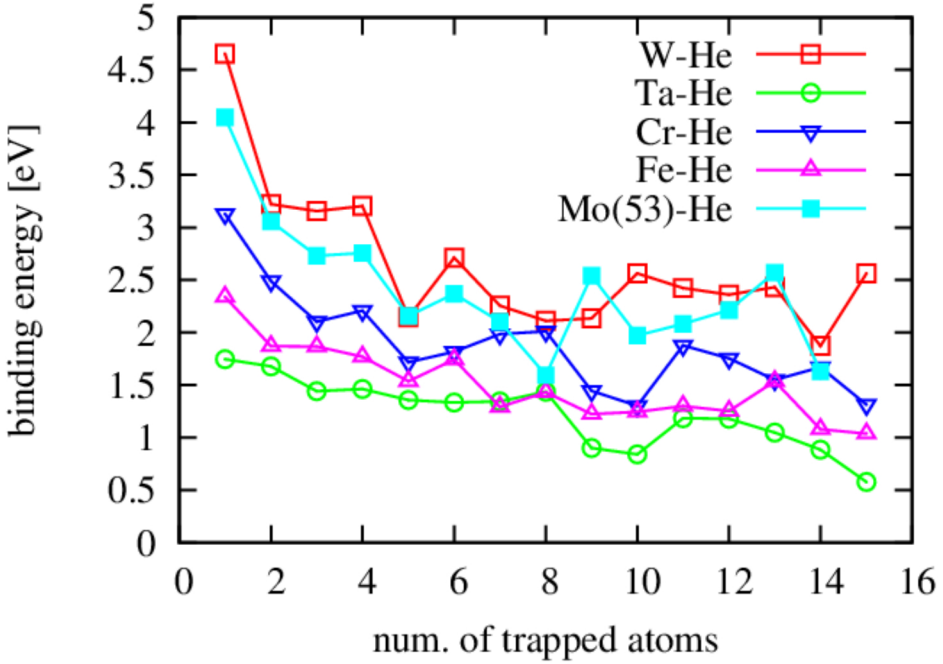
J. Łażewski, P. T. Jochym, P. Piekarz, M. Sternik, K. Parlinski, J. Cholewiński, P. Dłużewski, S. Krukowski, arXiv:1502.00309

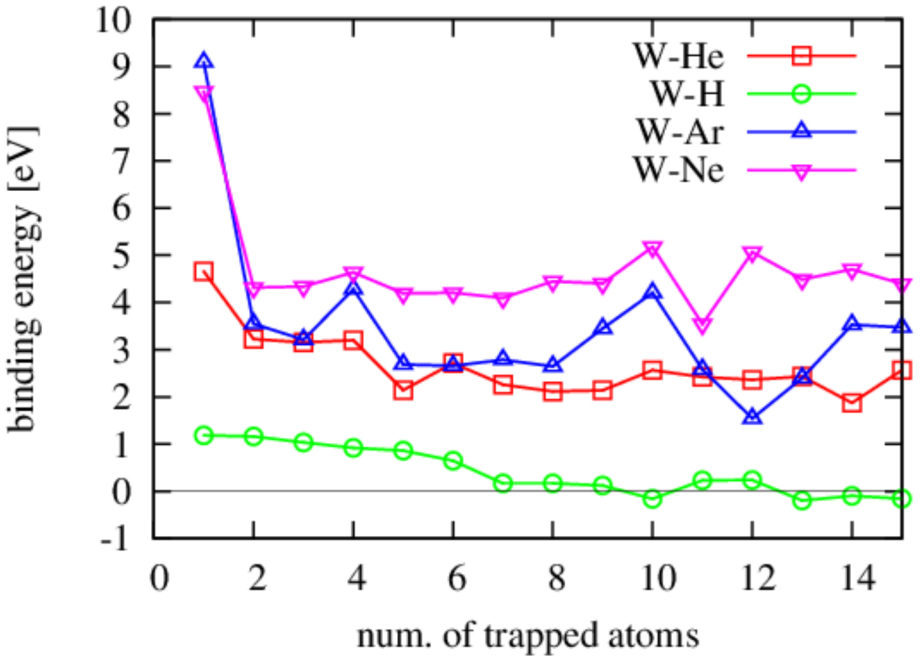
Helium from transmutation reactions

Neutron irradiation with energy 14 MeV produce large amount of helium and hydrogen from transmutation reactions. High He concentrations induce bubble formation, void swelling, and changes in microstructural and mechanical properties such as high temperature embrittlement



Optimized atomic configurations and electron-density isosurfaces for the system containing 6-He and 1-H atoms





T. Tamura, R. Kobayashi, S. Ogata, A. M. Ito, Model. Sim. Mater. Sci. Eng., 22, 015002 (2014)

Conclusions

- Crystal defects modify physical properties of materials: crystal structure, electronic structure, transport properties, lattice dynamics, thermo-elastic properties

- *Ab initio* calculations based on the density functional theory (DFT) provide information about the formation energies, structure of nanoscale defects, short-range interactions, clustering of defects and their migration (often not available from experiments)

- From DFT calculations we can obtain the structure of radiation damage, energy of interaction between radiation defects and impurities, activation energies, dynamics of migration, clustering of radiation defects

- DFT can provide the input parameters to other methods: Monte Carlo, molecular dynamics, cluster expansion, …