Emission channeling and ab initio calculations for the study of Ca color centers in diamond

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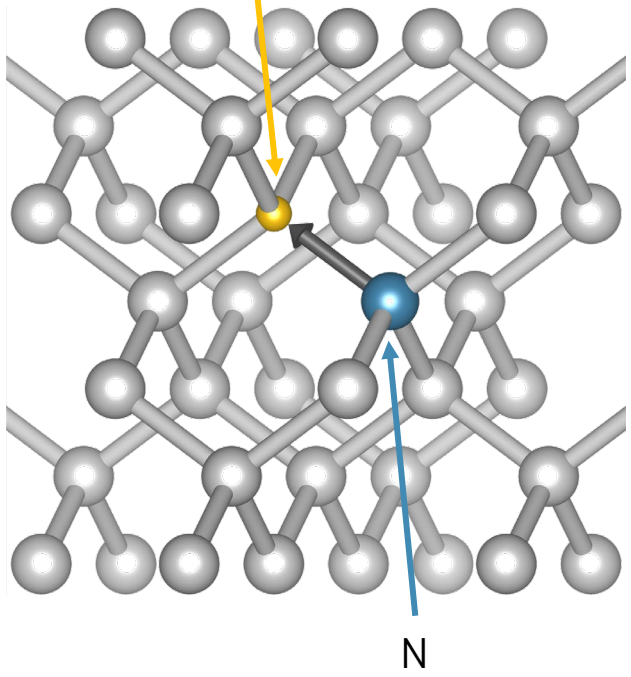
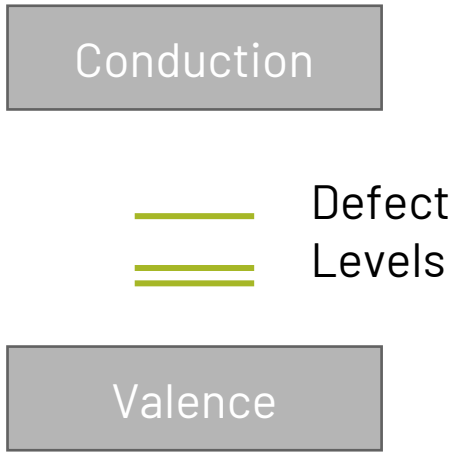
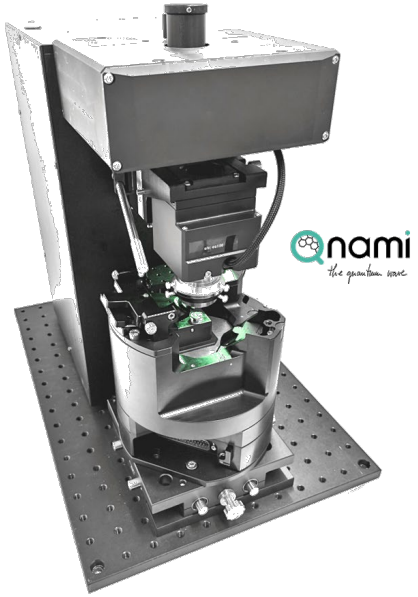
Color Centers in Diamond





Useful properties



Horiba ProteusQ[1]
Scanning NV magnetometry

[1] https://www.horiba.com/en_en/products/detail/action/show/Product/proteusq-2011/

Group II defects in diamond

ARTICLE OPEN

Highly tunable magneto-optical response from magnesium-vacancy color centers in diamond

Anton Pershin[1], Gergely Barcza[1], Örs Legeza[1,2] and Adam Gali[1,3]✉

Defect quantum bits (qubits) constitute an important emerging technology. However, it is necessary to explore new types of defects to enable large-scale applications. In this article, we examine the potential of magnesium-vacancy (MgV) in diamond to operate as a qubit by computing the key electronic- and spin properties with robust theoretical methods. We find that the electronic structure of MgV permits the coexistence of two loosely separated spin-states, where both can emerge as a ground state and be interconverted depending on the temperature and external strain. These results demonstrate a route to control the magneto-optical response of a qubit by modulating the operational conditions.

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Side-by-side approach

Density functional theory (DFT)

Convergence testing

Structural optimization

Formation energy

Binding energy

Spin polarization and charge density

Emission Channeling

Impurity localization

Fraction of each contribution

Behavior with annealing

Density functional theory

Many-body Schrödinger Equation → Infeasible to solve for most practical systems →

Hohenberg-Kohn theorem
Use the charge density (n) instead of wavefunction
+
Kohn-Sham system
auxiliary non-interacting system with wavefunctions ψ_i (Kohn-Sham orbitals)

Kohn-Sham equations

$$\left(-\frac{\hbar^2}{2m}\nabla^2 + \underbrace{V_{\text{external}}(\boldsymbol{r}) + V_{\text{Hartree}}(\boldsymbol{r}) + V_{\text{xc}}(\boldsymbol{r})}_{\text{Functionals of } n(\boldsymbol{r}) = \sum_i |\psi_i(\boldsymbol{r})|^2}\right)\psi_i(\boldsymbol{r}) = H^{KS}\psi_i(\boldsymbol{r}) = \epsilon_i\psi_i(\boldsymbol{r})$$

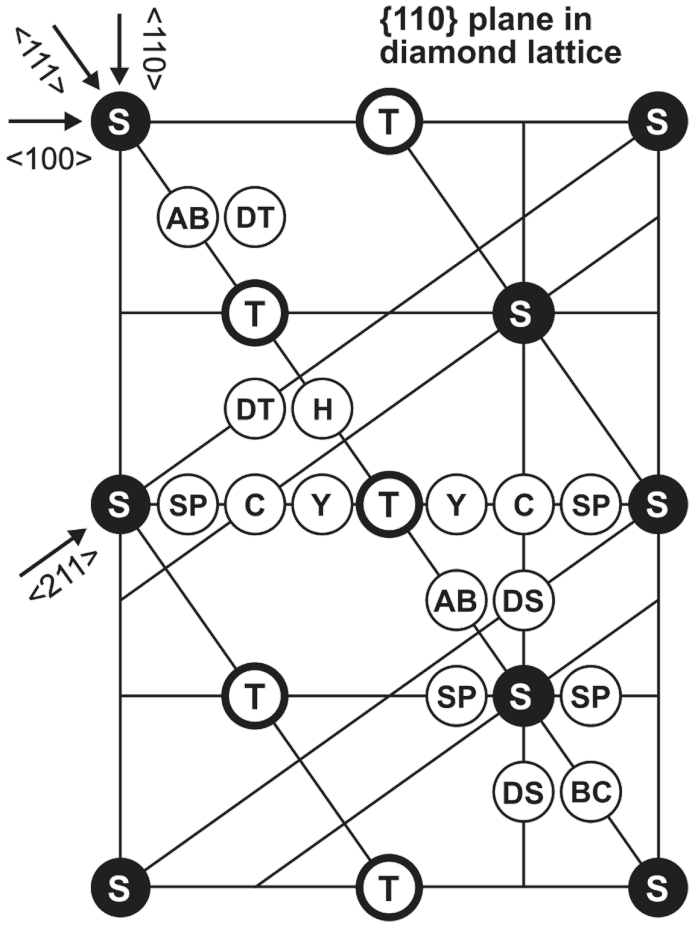
Unknown functional form → multiple approximations
- Perdew-Burke-Ernzerhof (PBE)
- Heyd-Scuseria-Ernzerhof 2006 (HSE06)

Structure Optimization

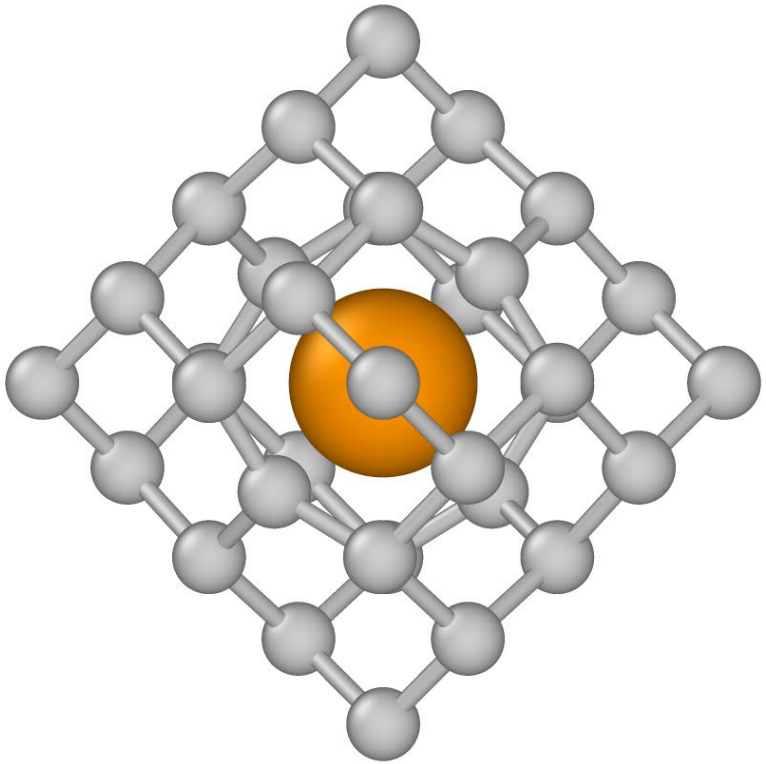
Calculations with 3x3x3 supercell

270 starting configurations of calcium defects

High symmetry sites
+
Varying number of vacancies
+
-2 to +2 charge



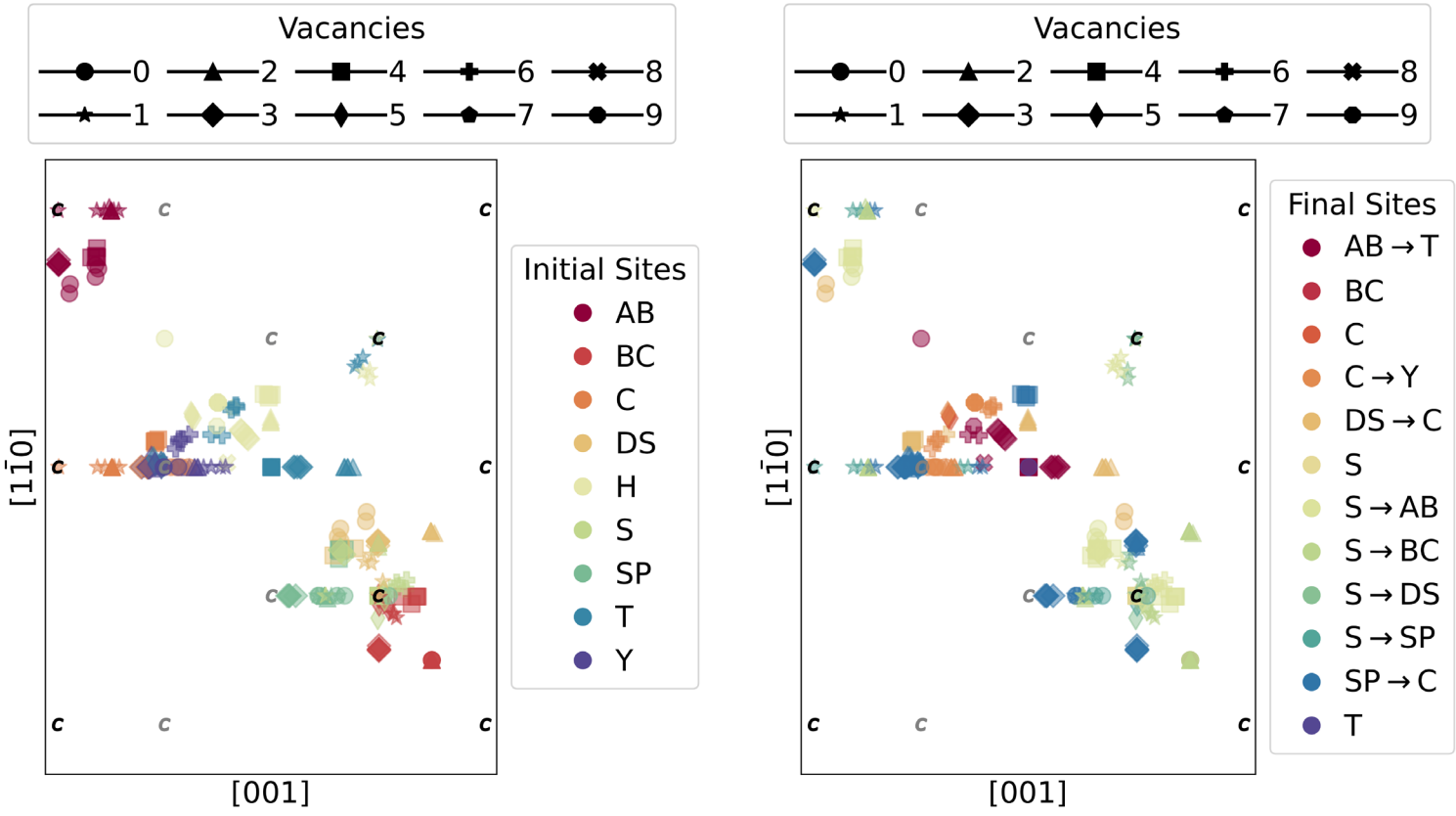
Created by Dr Ulrich Wahl



Relaxed structures examples

Calcium Carbon

Structure Optimization



Relaxed calcium position project into the (110) plane for all configurations

Equivalent relaxed structures

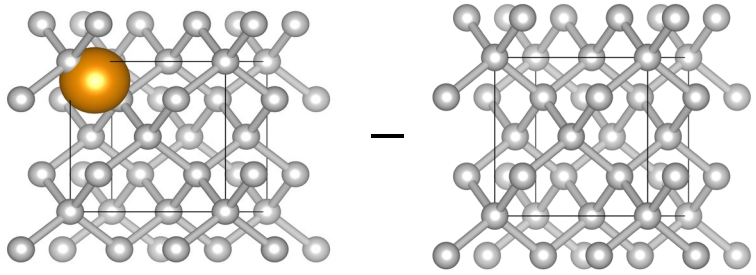
- S + 1 vacancy ⟺BC + 2 vacancies
- S + 2 vacancies ⟺ SP + 3 vacancies
- Most Interstitial + 1 vacancy ⟺ S

Point groups

- C_1 : 27
- C_2 : 6
- C_s : 43
- C_{2v} : 82
- C_{3v} : 36
- D_{3d} : 42
- T_d : 39

Formation Energy

Defect X in charge state q

$$E_f[X^q] = \text{  } - \sum_i n_i\mu_i + q(E_{\text{vbm}} + E_F) + E_{\text{corr}}$$

From interaction between defect replicas

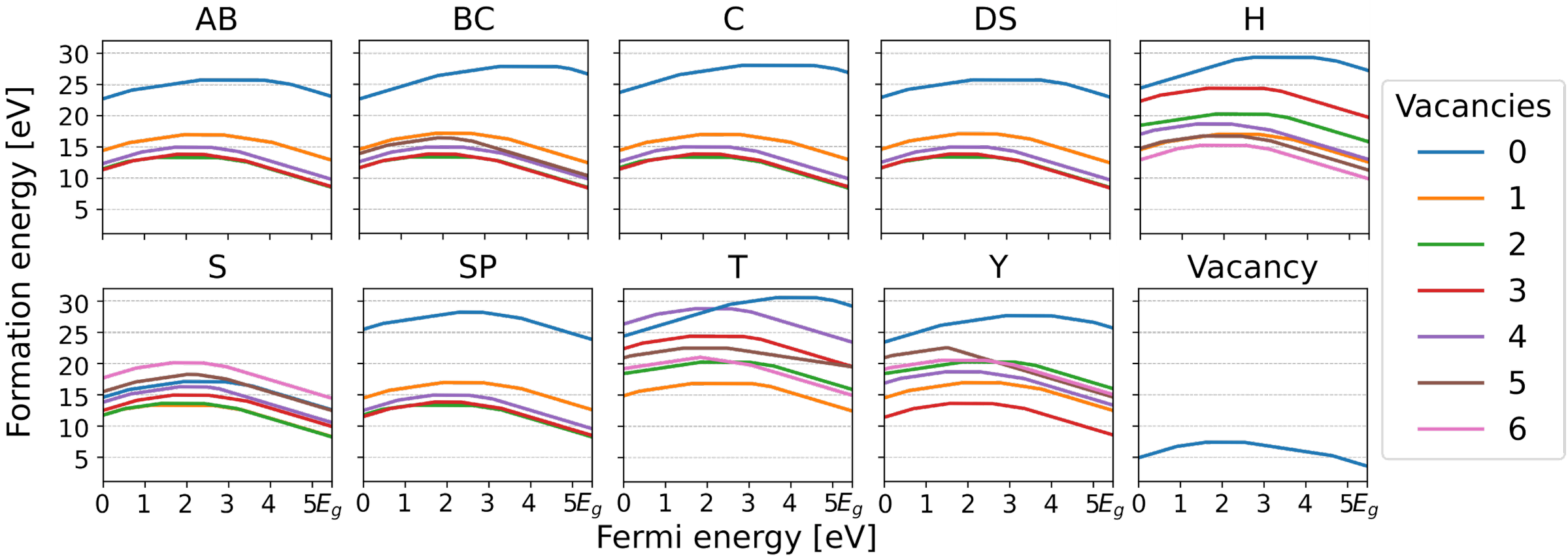
n_i Added atoms of specie i
μ_i Chemical potential of specie i

E_{vbm} Energy of the valence band maximum
E_F Fermi energy

E_{corr} *A posteriori* correction energy

Formation Energy

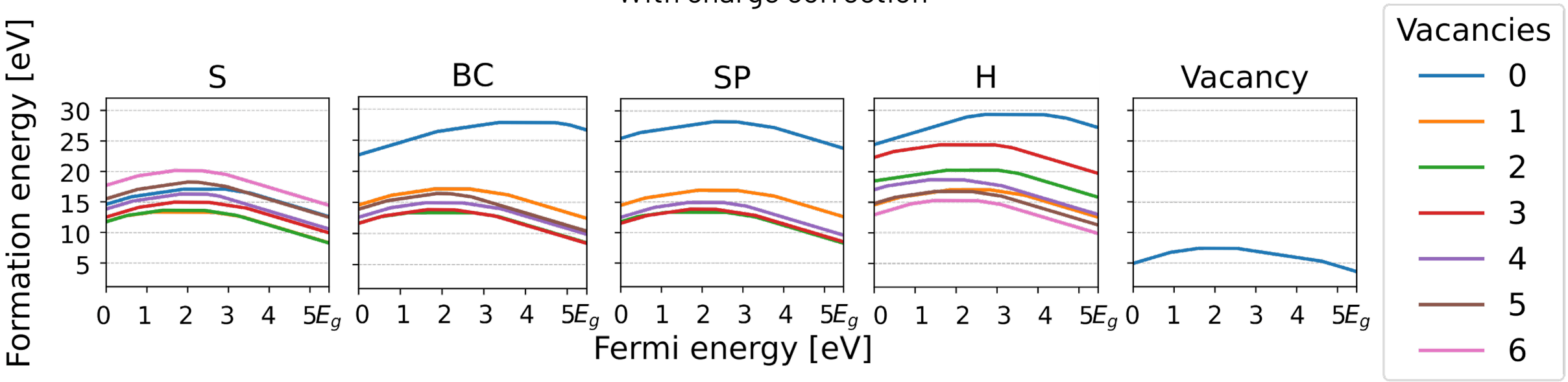
With charge correction



Charge:ΔHybrid	-2: 0.992 eV	-1: 1.180 eV	0: 1.099 eV	1: 1.145 eV	2: 1.299 eV

Formation Energy

With charge correction



⟩ Vacancy formation energy ~0.5 eV of literature

⟩ Different configurations with similar formation energy

⟩ Lower formation energy with some vacancies

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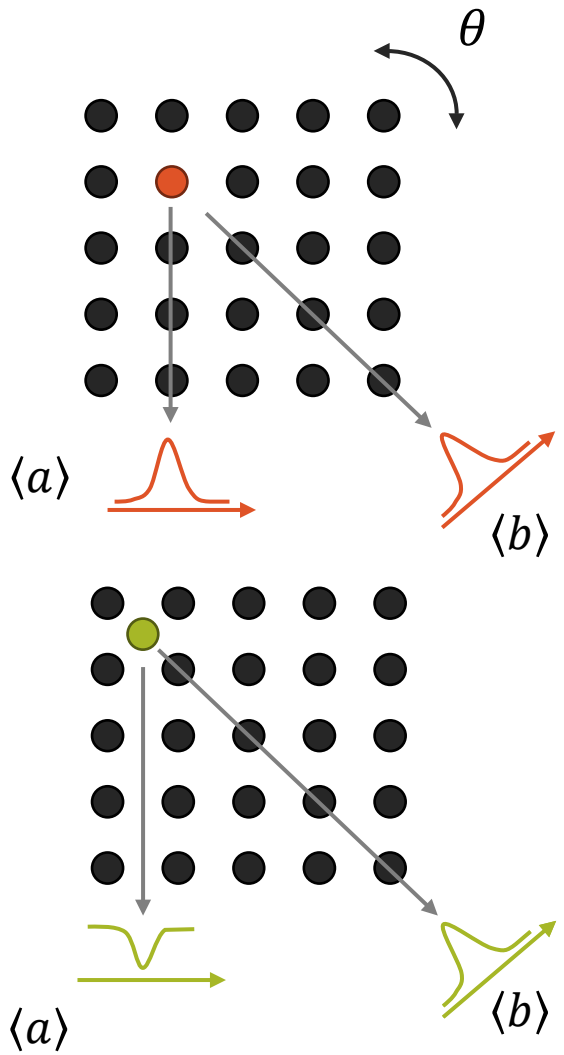
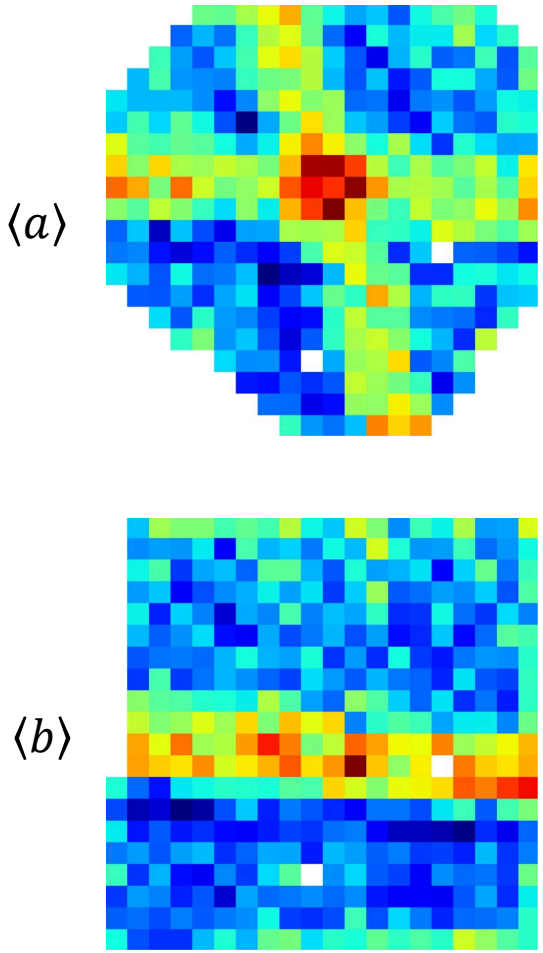
Emission Channeling





Simulated pattern library

N = 1

N site(s) fit
for all
combinations

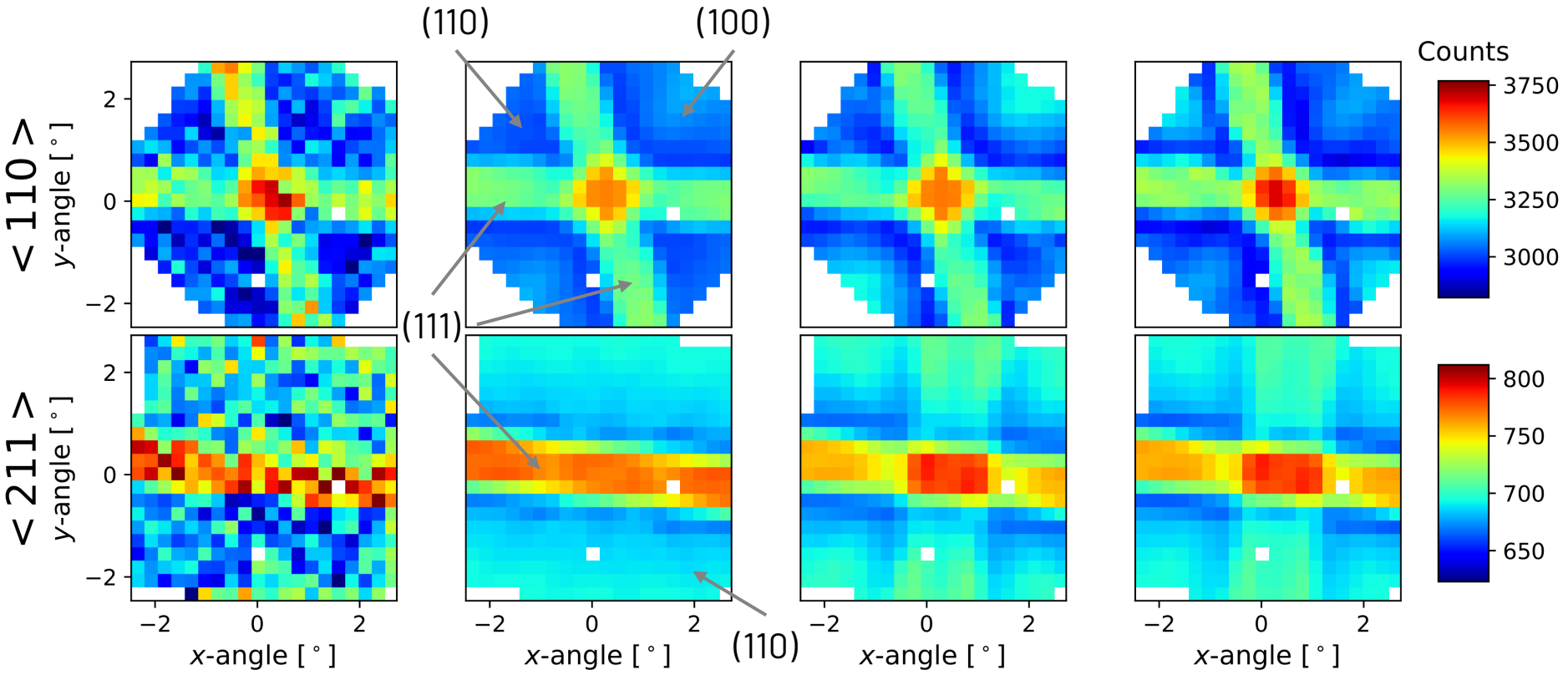
N = N + 1

Best M groups as
an additional site

Filter
consistent fraction for all directions
$0 <$ fractions < 1

Score groups
scoring function $f([m_1, \ldots, m_g])$
where m_i is the quality metric of
element i of the group

As implanted single-site fits



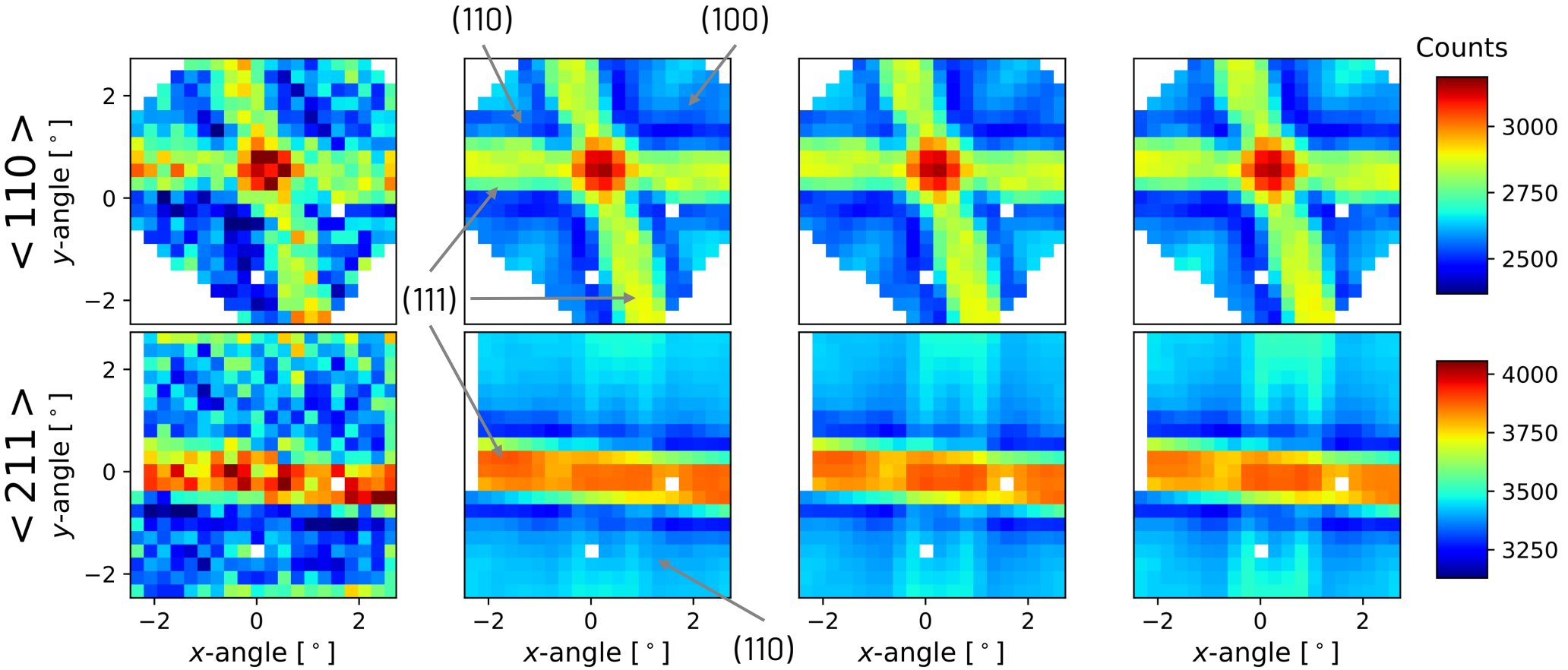
Experimental | BC (u_1) | 0.1 · SP → C | 0.6 · S → BC

⟨111⟩: Low contrast
⟨211⟩: Little anisotropy
Explain a higher fraction

⟨111⟩: Better contrast
⟨211⟩: High anisotropy

^{45}Ca - 30 keV - 3.5×10^{12} ions/cm^2

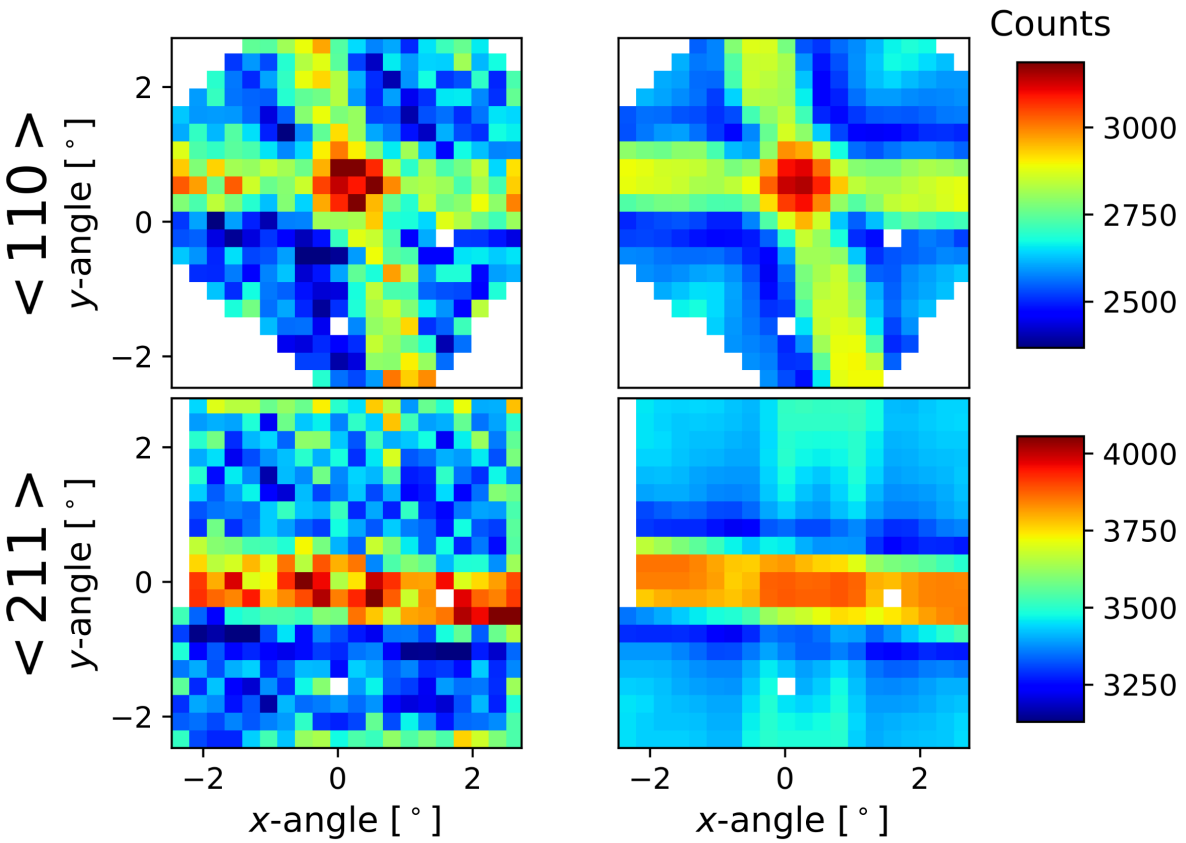
After Annealing (900 °C) two-site fits



Experimental | $0.65 \cdot S \rightarrow BC$ $0.5 \cdot DS \rightarrow C$ | $0.65 \cdot S \rightarrow BC$ DS | SP $BC\,(u_1)$

^{45}Ca - 30 keV - 3.5×10^{12} ions/cm^2

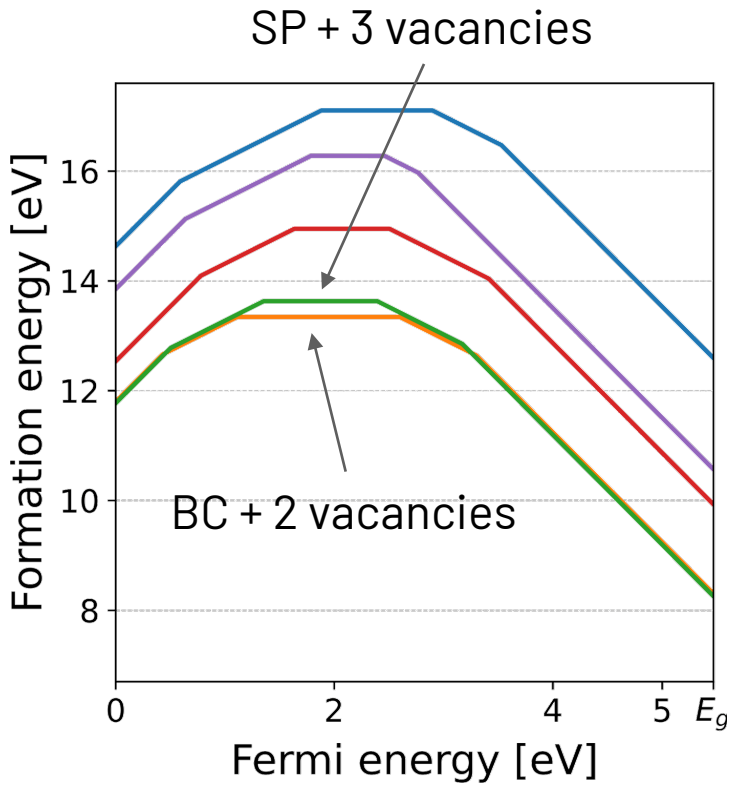
After Annealing (900 °C) two-site fits



Experimental

SP

BC (u_1)



^{45}Ca - 30 keV - 3.5×10^{12} ions/cm^2

Conclusions

DFT
most stable configuration BC + 2 Vacancies ⟷ EC before annealing
Distribution around BC

EC after annealing
Small change in the patterns

EC after annealing
No definite assignment but change from the BC site + Formation energies + Site assignment of relaxed structures → Close to SP site + Tighter distribution around BC

DFT
BC + 2 Vacancies and SP + 3 Vacancies → Similar formation energies → Convertion during annealing

Current and Future Work

- ⟩ DFT calculations with more vacancies → check for higher stability configurations
- ◊ Many-body energy spectrum
  - ◊ Optical excitation → aid in experimental identification of Calcium defects
  - ◊ Determine if similar properties to MgV
- ◊ Minimum energy path between sites → behavior during annealing
- ◊ Systematically test filtering criteria and scoring function in EC filtering
- ◊ Corrected fractions
- ⟩ New EC measurements → clarify the assignment after annealing

⟩ Work in progress
◊ Future work

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