IS668 EC-SLI: First results on lattice location of implanted ^{27}Mg, ^{45}Ca and ^{89}Sr in diamond

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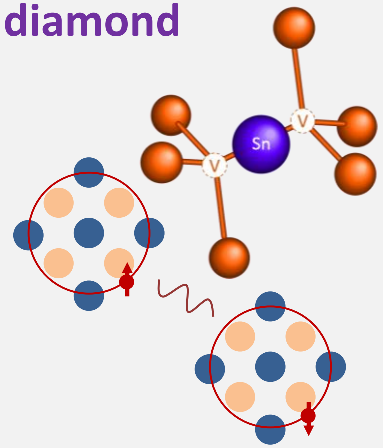
- **Introduction: quantum colour centers in diamond**
- **Emission channeling method**
- **EC results: ^{27}Mg, ^{45}Ca, ^{89}Sr**
- **Conclusions**

Quantum centers suitable for studies with radioactive ion beams

- Our research interest: impurities in solids which exhibit quantum properties useful for future applications: “quantum centers”
- General characteristics:
  Dilute impurity atoms embedded in a solid
- Useful quantum properties are related to spin interactions, (stimulated) photon emission, coherence, entanglement, polarization of photons…
- Quantum properties emerge from the electronic/nuclear interaction of the impurity with the crystal host
- Microscopic structure of centers determines their quantum properties

Examples

IS668: Colour centers in diamond



q-bits and single-photon emitters

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quantum coherence and entanglement

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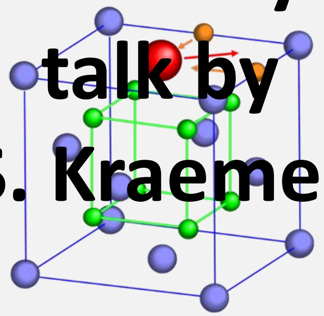
quantum communication, computation, metrology

IS658: ^{229m}Th nuclear isomer in CaF_2

Yesterday’s talk by S. Kraemer



nucleus with lowest isomeric energy: ~8.27 eV

⇩

stimulated photon emission

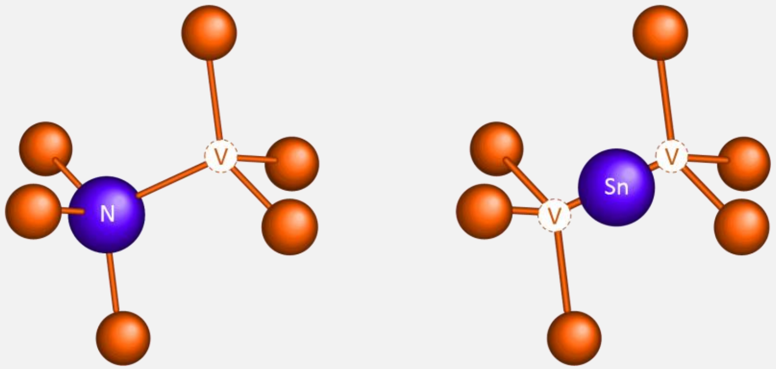
⇩

nuclear clock

Split-*vacancy* centers in diamond

- Colour centers in diamond are intensively investigated for their applications in processing and communication of quantum information and metrology.
- Diamond has a very tight lattice, so it is common that impurity atoms pair with a *vacancy V*.
- Two possible configurations for impurity-*vacancy* centers in diamond:

C_{3v} "full-*vacancy*", assumed for **N*V***

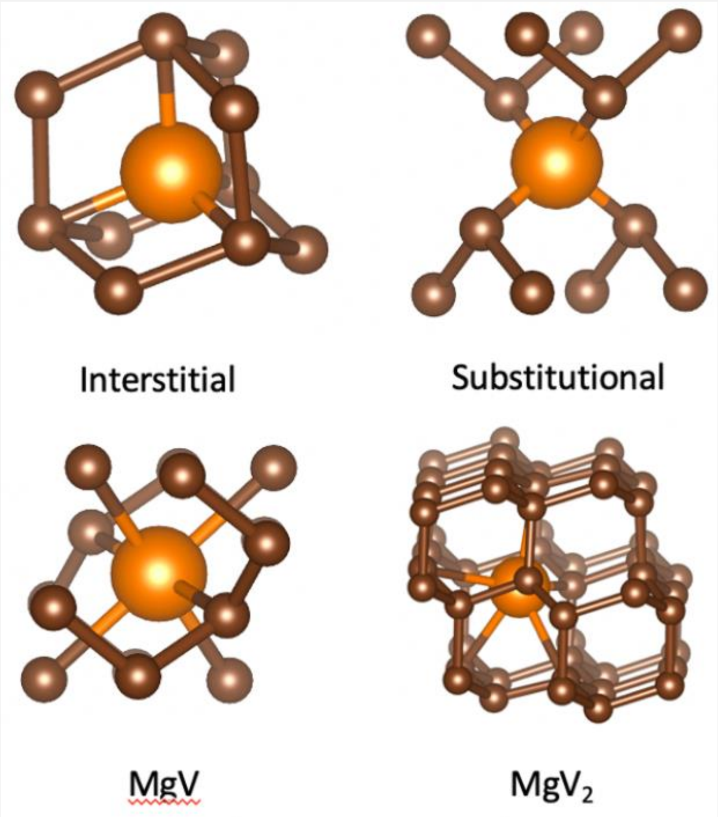


D_{3d} "split-*vacancy*", assumed for group IV-*vacancy*: **Si*V* [1], Ge*V* [2], Sn*V* [3,4], Pb*V* [5],** but also for **Mg*V* [6]**

- Superior optical properties of the centers with split-*vacancy* structure are to a large extent a consequence of their D_{3d} inversion (mirror) symmetry.
- Many colour centers in diamond are commonly produced by ion implantation.
- **How to optimize implantation conditions in order to achieve unperturbed split-*vacancy* configurations?**
- Emission channeling lattice location experiments are uniquely suited to study this problem.

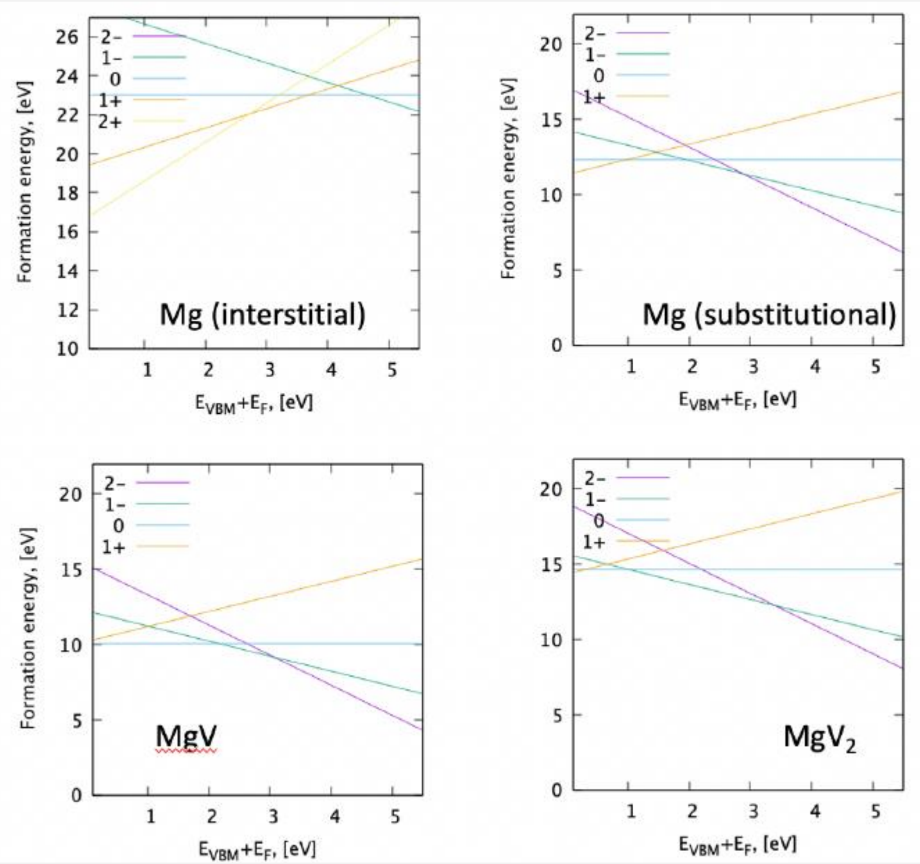
[1] J.P. Goss *et al*., Phys. Rev. Lett. 77 (1996) 3041
[2] T. Iwasaki *et al*., Sci. Rep. 5 (2015) 12882
[3] S.D. Tchernij, … J. Forneris, *et al*., ACS Photonics 4 (2017) 2580
[4] T. Iwasaki, … P. Syushev, *et al*., Phys. Rev. Lett. 119 (2017) 253601
[5] S.D. Tchernij, … J. Forneris, *et al*., ACS Photonics 5 (2018) 4864
[6] A. Pershin et al., npj Quantum Information 7 (2021) 99

Predicted structures of Mg defects in diamond



- Theoretically predicted structures of Mg-related complexes in diamond [6]:

- Interstitial Mg_i: T_d symmetry
- Substitutional Mg_S: T_d symmetry
- Mg*V*: split-*vacancy* configuration with Mg on BC sites, D_{3d} symmetry <111>
- MgV_2: C_1 symmetry <100>



Formation energy *vs* Fermi-level [6]

Formation energies favour Mg*V*, Mg_S, possibly MgV_2, rule out Mg_i

[6] A. Pershin et al., "Highly tunable magneto-optical response from Mg*V* color centers in diamond", npj Quantum Information 7 (2021) 99

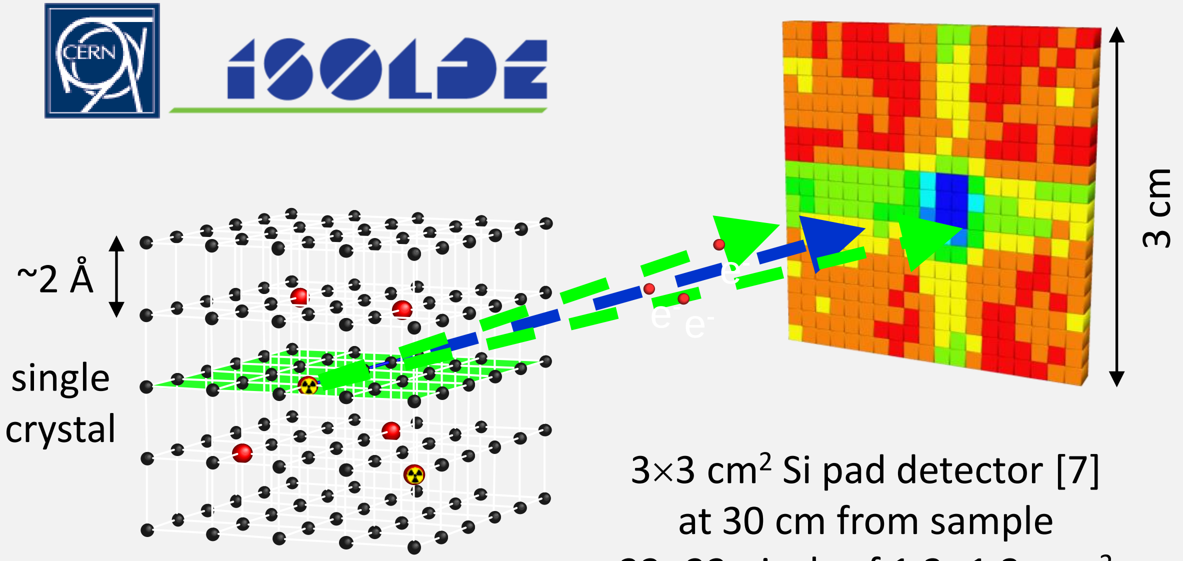
Emission Channeling with Short-Lived Isotopes (EC-SLI)

- Radioactive ^{27}Mg ($t_{1/2}$=9.5 min), ^{45}Ca ($t_{1/2}$=164 d), ^{89}Sr ($t_{1/2}$=50.5 d) probe atoms are produced at ISOLDE.

- 30 keV ion implanted (1-5×10^{12} cm^{-2}) into diamond, measured as function of implantation or annealing temperature.

- Position- and energy sensitive detector [7] is used to detect emission channeling [8] effects of β^- decay particles in the vicinity of major crystallographic directions.

CERN ISOLDE



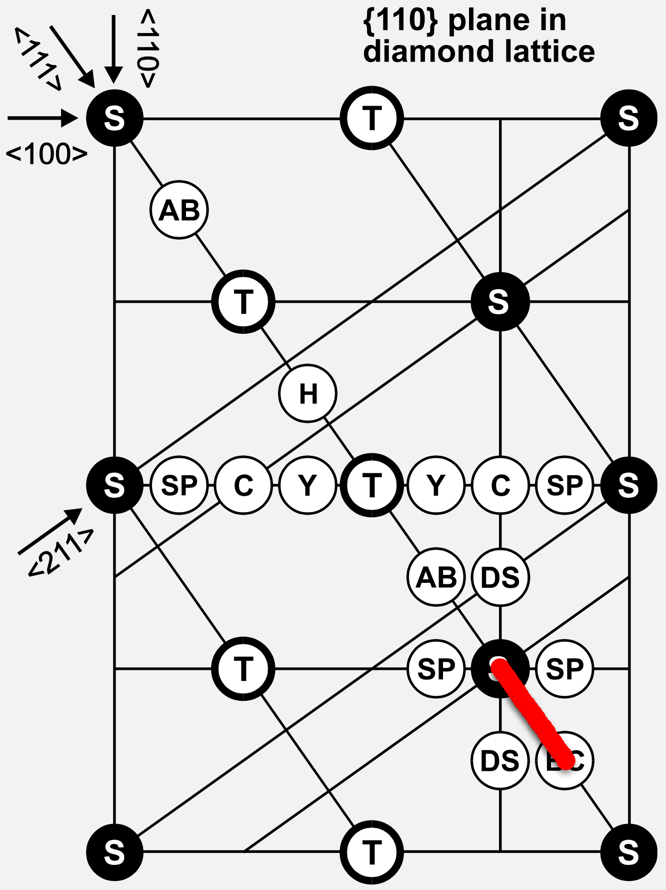
3×3 cm^2 Si pad detector [7]
at 30 cm from sample
22×22 pixels of 1.3×1.3 mm^2

Angular dependent β^- emission patterns characterize the lattice site distribution of the radioactive probe atoms.

[7] U. Wahl *et al.*, Nucl. Instr. Meth. A 524 (2004) 245
[8] H. Hofsäss, G. Lindner, Phys. Rep. 201 (1991) 121

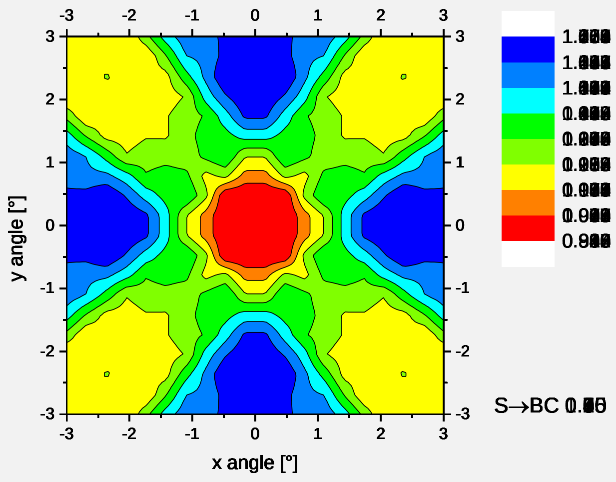
“Many-beam” calculation of β^- emission yields

high- symmetric sites in diamond



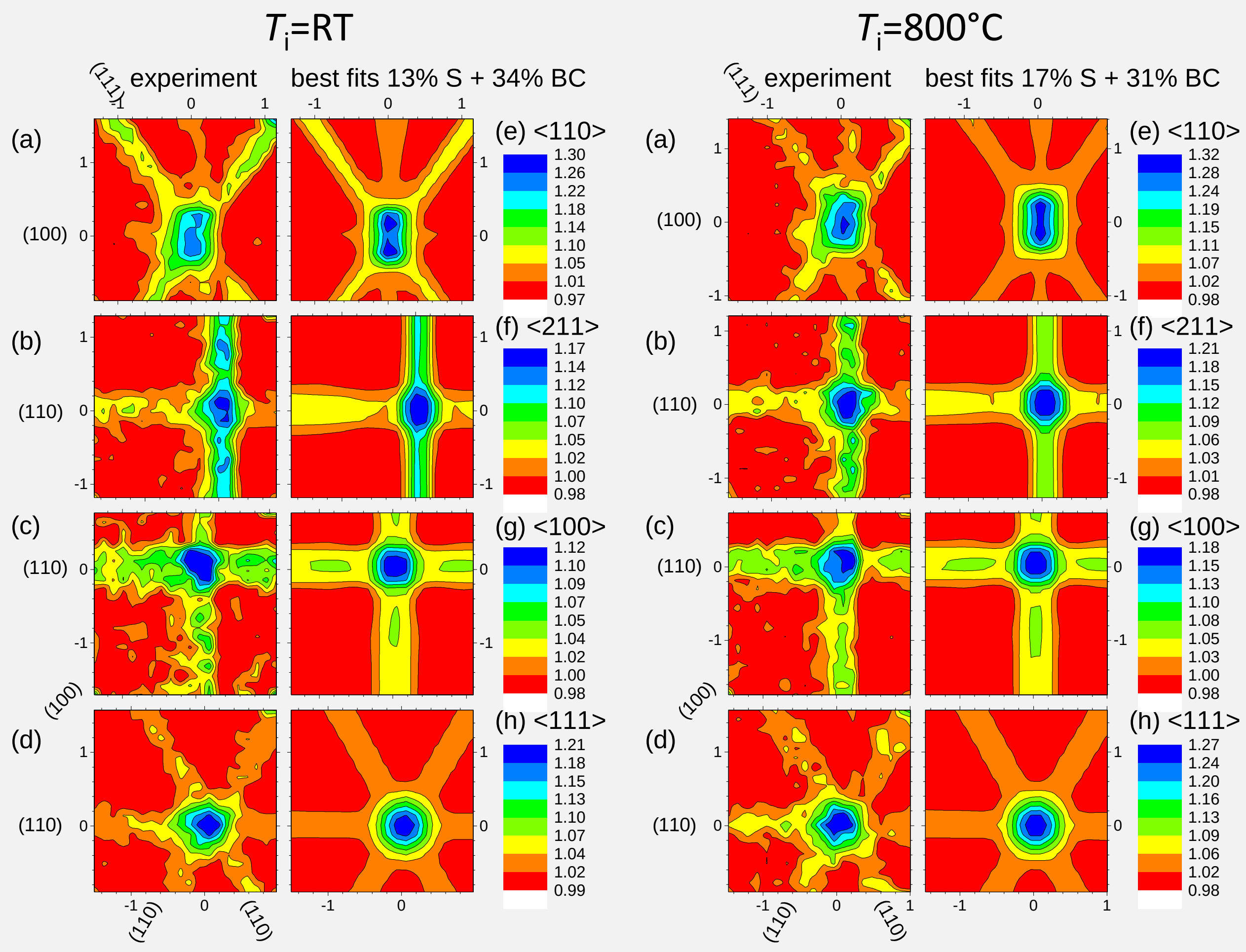
- β^- angular emission yield patterns are calculated for ~250 lattice sites in the diamond unit cell using the “many-beam” [8,9] approach. Example: ^{121}Sn.

- Anisotropy and contours of patterns change with position of impurity in the lattice,
e.g. the <100> pattern when moving from S to BC sites:



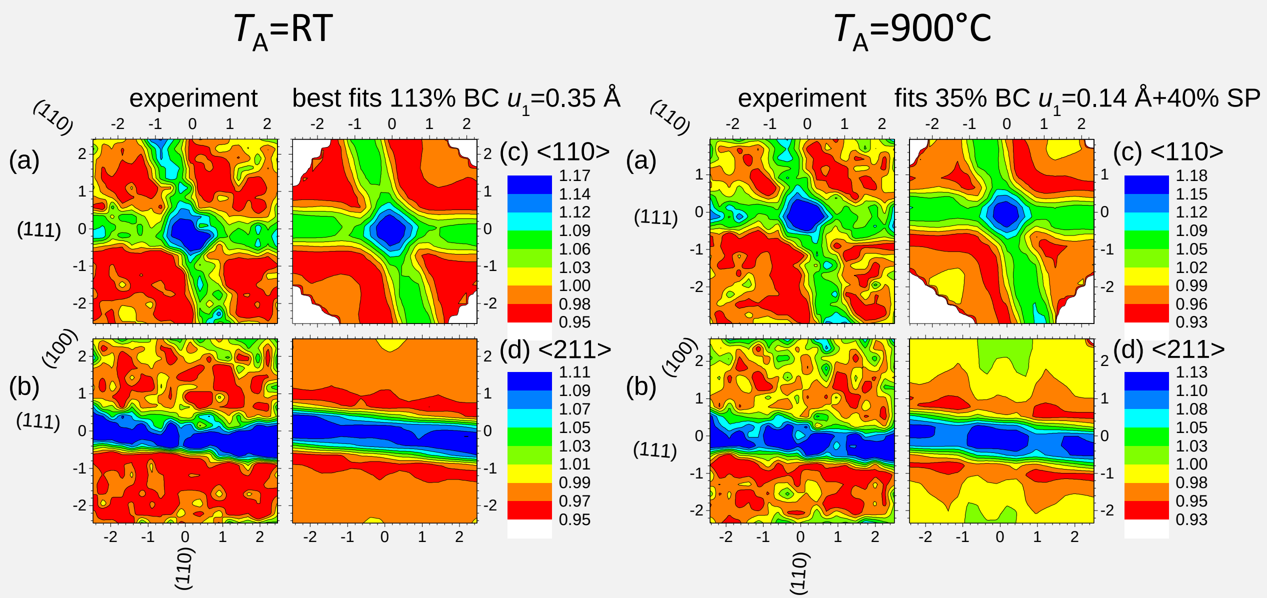
[8] H. Hofsäss, G. Lindner, Phys. Rep. 201 (1991) 121
[9] U. Wahl *et al.*, Hyperf. Interactions (2000) 129 349

Emission channeling ^{27}Mg in diamond



- **Preliminary results July 2021 beam time**
- **Ongoing analysis**
- Only small differences between RT and 800°C
- Patterns were fitted assuming ideal S and BC sites (u_1=0.034 Å)
- T_i=RT:
  13% S
  34% BC
- T_i=800°C:
  17% S
  31% BC
- Mg_S and $Mg_{BC}V$ complex
- High fraction (>50%) of "random" sites
- Other lattice sites must be present
- Large fraction in MgV_2 or MgV_3 complexes?

Emission channeling ^{45}Ca in diamond

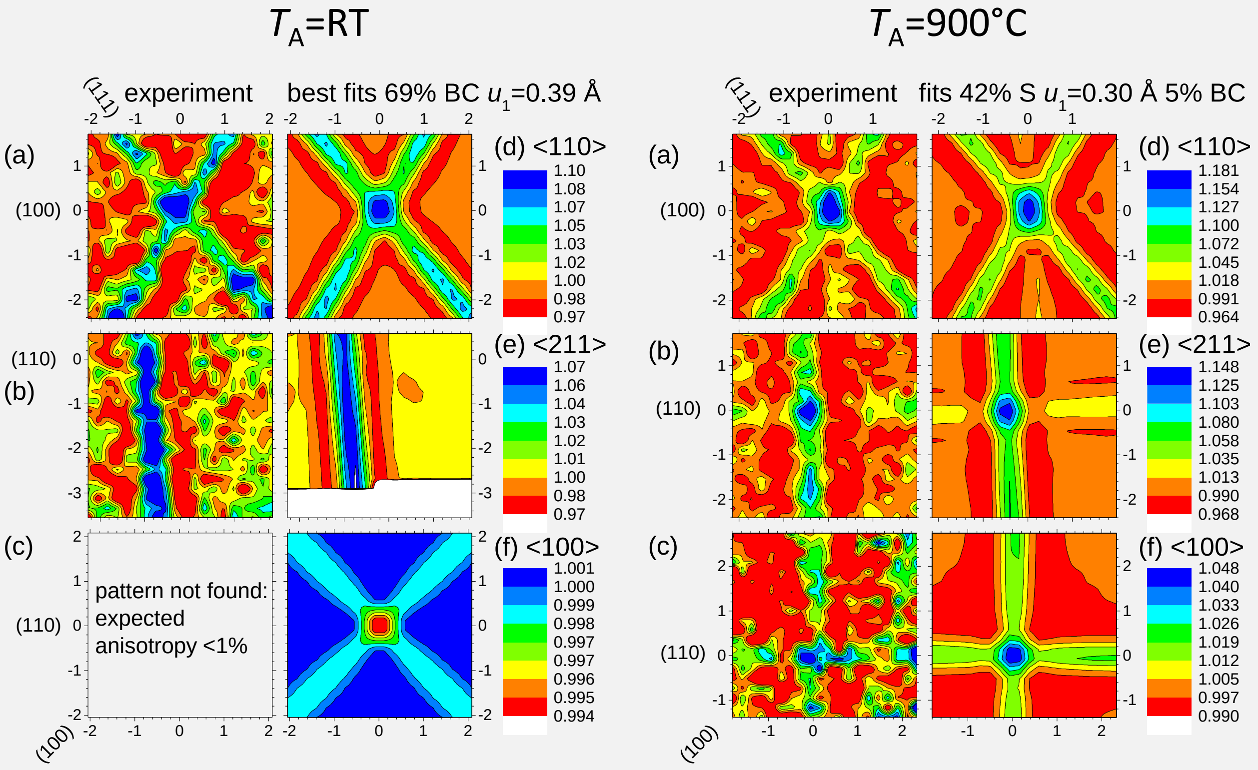


<100> and <111> patterns not found because their anisotropy is too weak ⇒ absence of S-sites

- ^{45}Ca ($t_{1/2}$= 164 d) long-lived isotope
- **Preliminary results + ongoing analysis**
- Changes in fine structure of patterns
- Assuming ideal S and BC sites (u_1=0.034 Å), patterns could NOT be well fitted
- Possible scenarios:
- RT as-implanted: 113% BC with u_1=0.35 Å
- T_A=900°C: 35% BC with u_1=0.14 Å 40% SP
- No substitutional Ca_S !
- Only $Ca_{BC}V$ and $Ca_{SP}V_2$ complexes?

Addressed in more detail in following talk of Afonso Lamelas

Emission channeling ^{89}Sr in diamond



- ^{89}Sr ($t_{1/2}$= 50.5 d) long-lived isotope
- **Preliminary results + ongoing analysis**
- Anisotropy doubled after 900°C annealing
- Assuming ideal S and BC sites (u_1=0.034 Å), patterns could NOT be well fitted
- Possible scenarios:
- RT as-implanted:
  69% BC with u_1=0.39 Å
- T_A=900°C:
  42% S with u_1=0.30 Å
  5% BC with u_1=0.05 Å
- As-implanted: mostly $Sr_{BC}V$ but NO Sr_S
- For T_A=900°C converted largely to Sr_S with large u_1
- What is the nature of Sr_S with large u_1?

Conclusions

- ^{27}Mg: ~30% $Mg_{BC}V$ "split-*vacancy*" configuration + ~15% Mg_S + indications that $Mg_{SP}V_2$ complexes are also likely to be found
- ^{45}Ca: negligible occupation of S sites, probably only $Ca_{BC}V$ and $Ca_{SP}V_2$
- ^{89}Sr: at RT $Sr_{BC}V$ but for T_A=900°C Sr_S, both with large u_1
- "Split-*vacancy*" configurations found but no obvious correlation with atomic size of impurities
- Complicated defect situation (more complex than e.g. for ^{121}Sn)
- DFT calculations may be helpful to identify the possible configurations