Unambiguous identification of the split-vacancy configuration of the SnV^{-} defect in diamond

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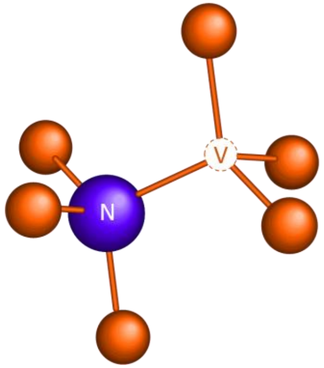
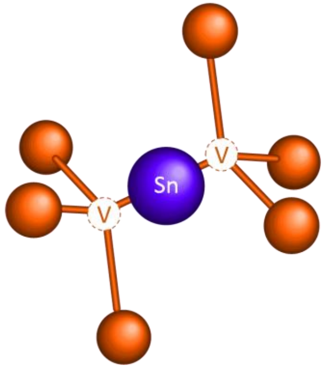
Motivation: Sn*V* in diamond as single photon center

- N*V*, Si*V*, Ge*V*, Sn*V* [1,2] and Pb*V* centers in diamond are intensively investigated for their applications in processing and communication of quantum information and metrology.
- Two possible configurations for impurity-vacancy centers:

“full-vacancy”, assumed for **N*V***





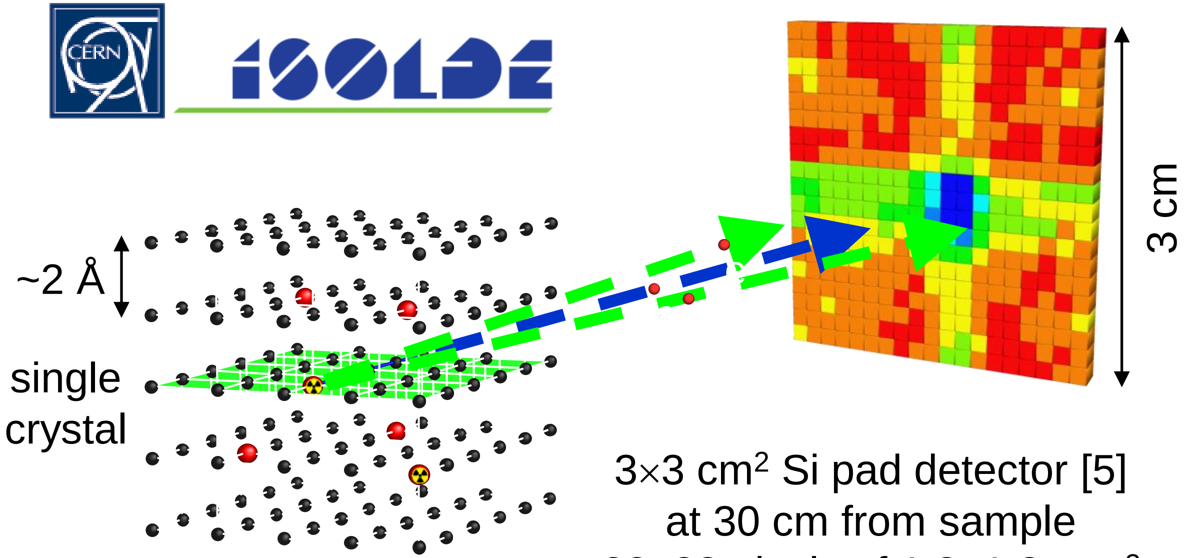
“split-vacancy” [3,4], assumed for **Si*V*, Ge*V*, Sn*V*, Pb*V***

- Superior optical properties of the group-IV-vacancy centers are to a large extent a consequence of the D_{3d} inversion (mirror) symmetry.
- So far, no direct ***structural*** evidence available that these configurations are actually formed.
- Emission channeling lattice location experiments are uniquely suited to study this problem.

[1] S.D. Tchernij *et al*., ACS Photonics 4 (2017) 2580
[2] T. Iwasaki *et al*, Phys. Rev. Lett. 119 (2017) 253601
[3] J.P. Goss *et al*., Phys. Rev. Lett. 77 (1996) 3041
[4] J.P. Goss *et al*., Phys. Rev. B. 72 (2005) 035214

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

- Radioactive ^{121}Sn ($t_{1/2}$=27 h) probe atoms are produced at CERN's ISOLDE on-line isotope separator facility.
- 60 keV ion implanted ($2{\times}10^{12}$ cm^{-2}) into natural type IIa diamond, measured RT as-implanted and 920ºC annealed
- Position- and energy sensitive detector [5] is used to detect emission channeling [6] effects of β^- decay particles from ^{121}Sn in the vicinity of major crystallographic directions.



3×3 cm^2 Si pad detector [5]
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 ^{121}Sn emitter atoms.

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

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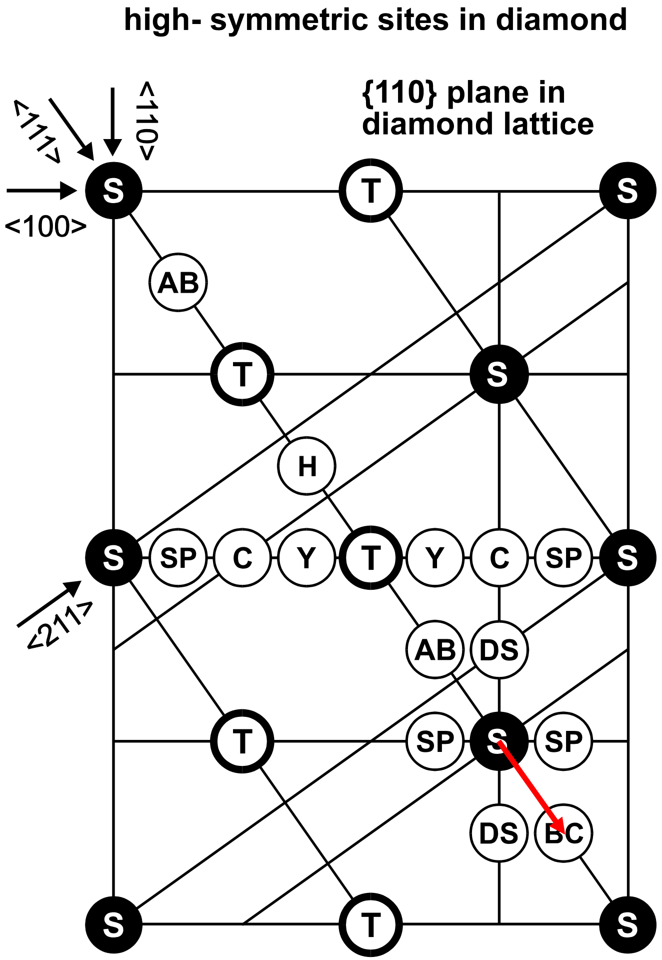
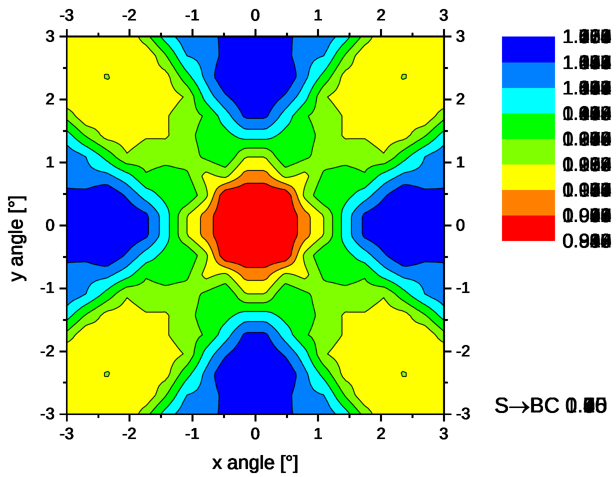
“Many-beam” calculation of β⁻ emission yields



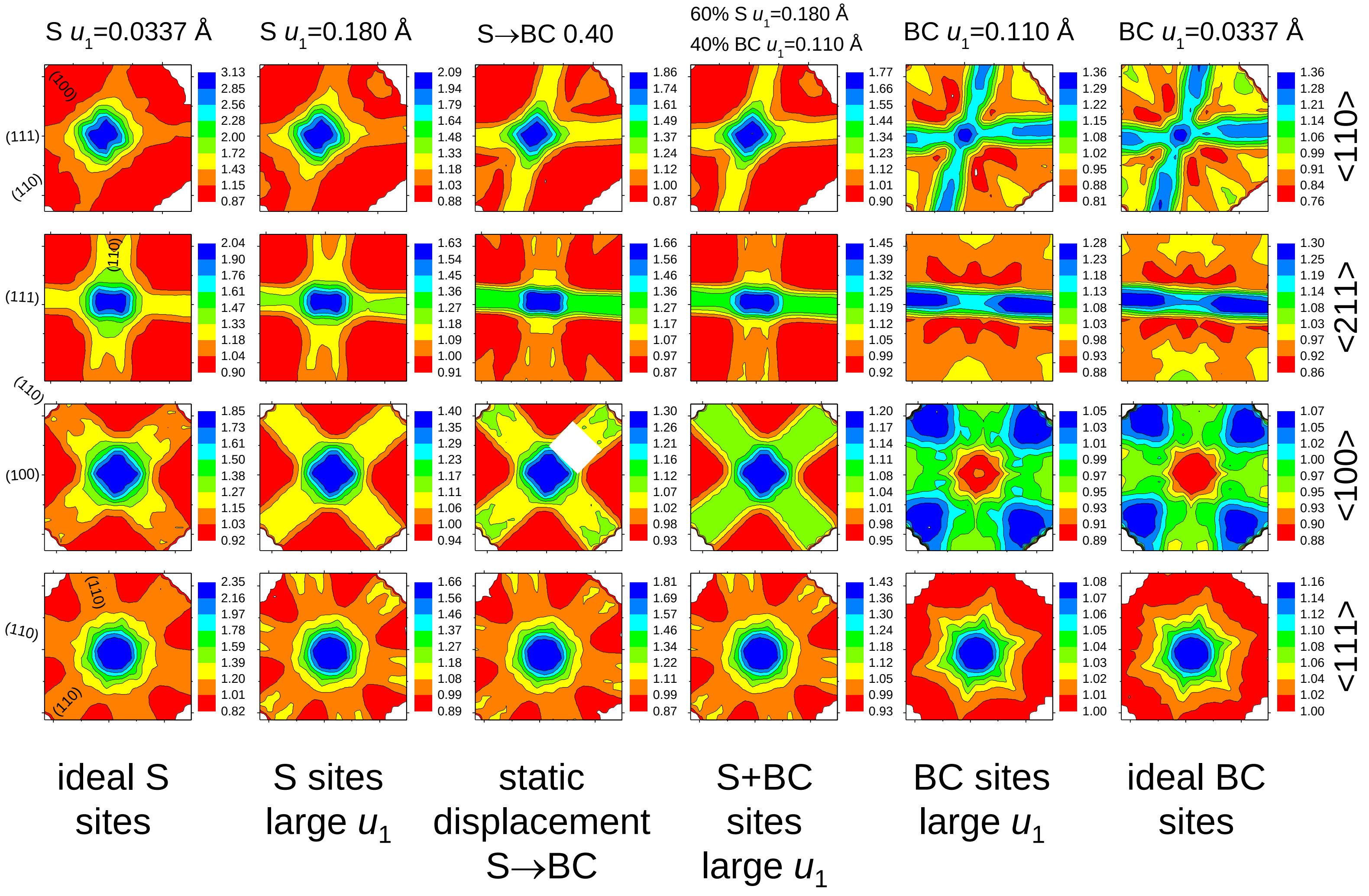
- β⁻ angular emission yield patterns from ^{121}Sn are calculated for ~250 lattice sites in the diamond unit cell using the “many-beam” [6,9] approach.
- Anisotropy and contours of patterns change with position of ^{121}Sn in lattice, e.g. the <100> pattern when moving from S to BC sites:

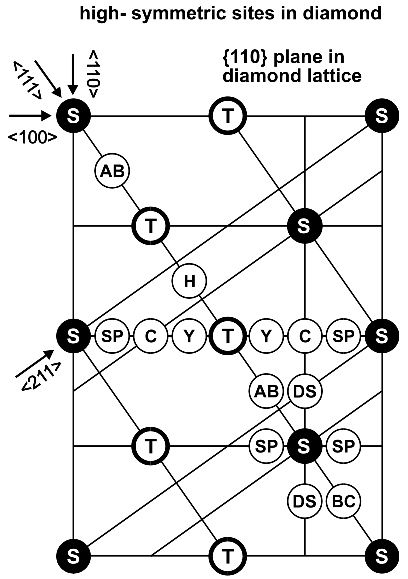


- For S and BC sites patterns also calculated for Gaussian distributions of rms $u_1(^{121}\text{Sn})$= 0.01 … 0.50 Å

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

Theoretical β⁻ emission yields from some relevant sites





- Increasing the rms displacements u_1(^{121}Sn) from the value u_1=0.0337 Å expected for ideal lattice sites or introducing static displacements reduces the anisotropy of patterns and changes their contours.
- Experimental patterns are analyzed by means of fitting linear superpositions [10] of up to 2 theoretical patterns.

[10] E. David-Bosne *et al.*, Nucl. Instr. Meth. B 462 (2020) 102

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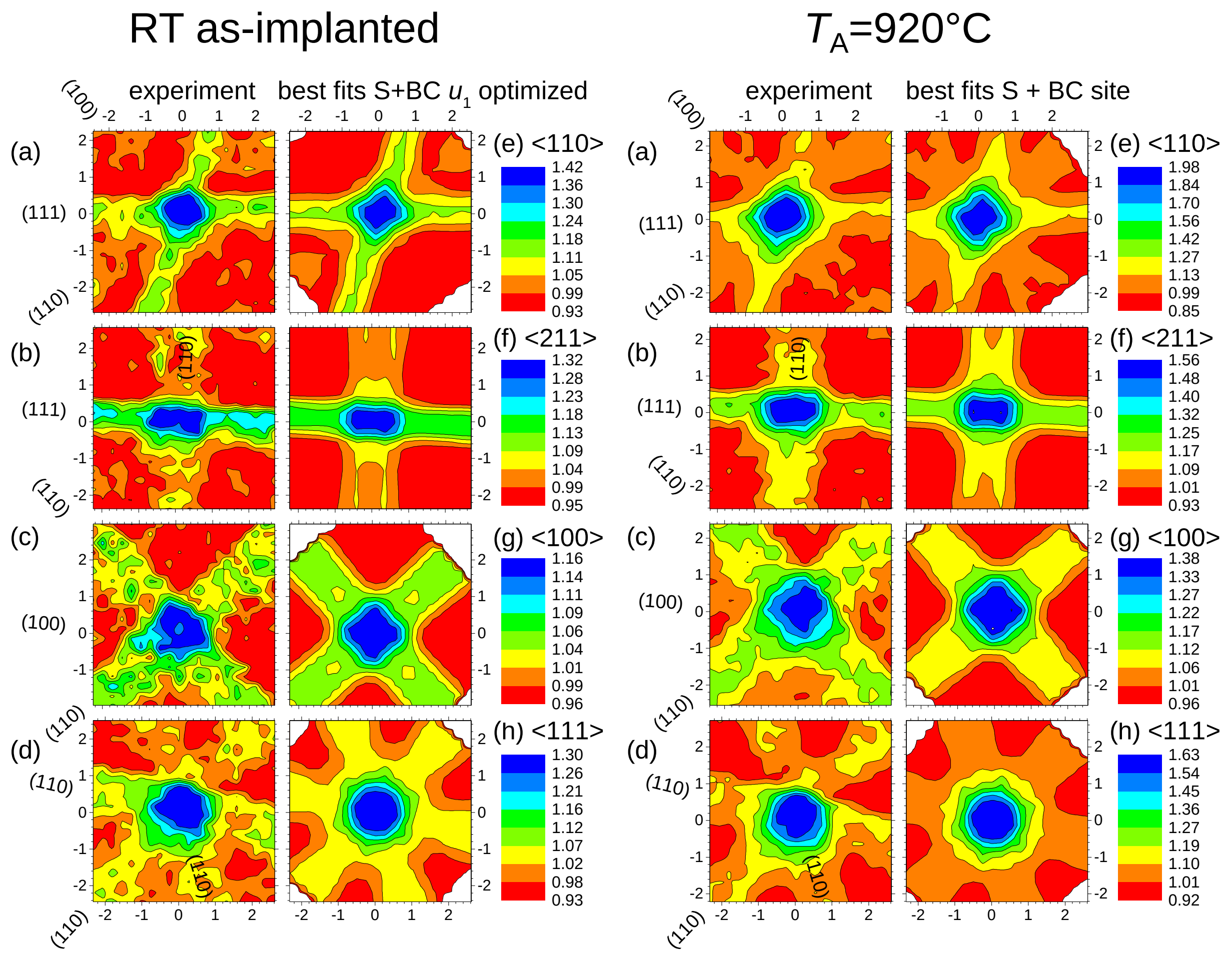
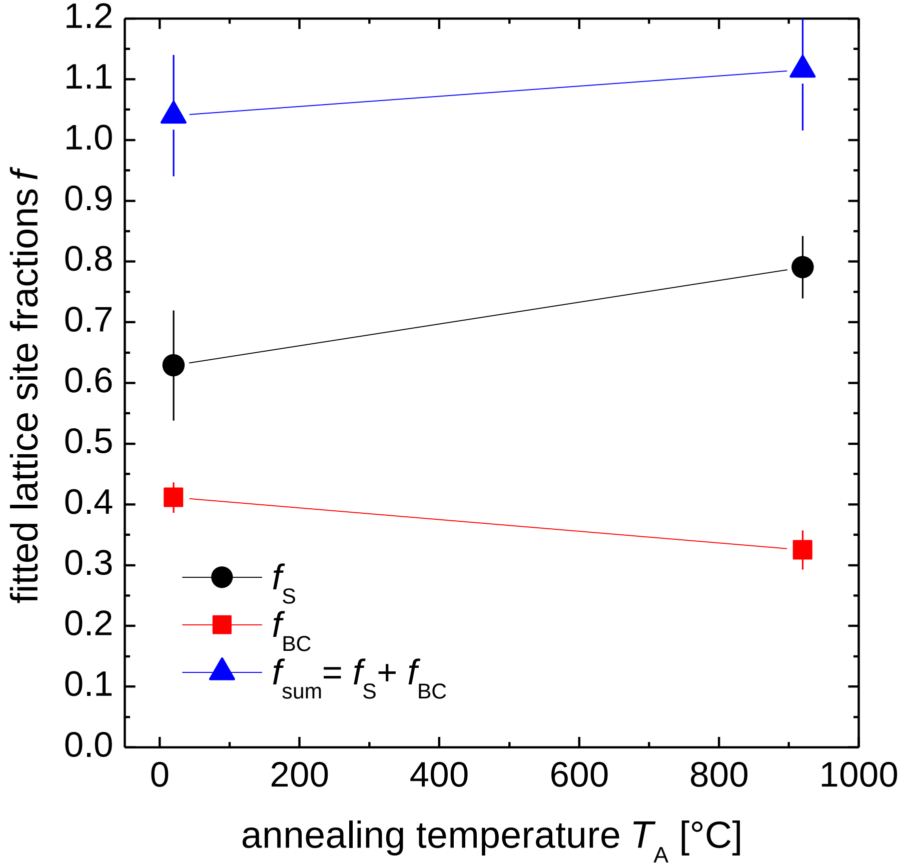
Emission channeling lattice location results



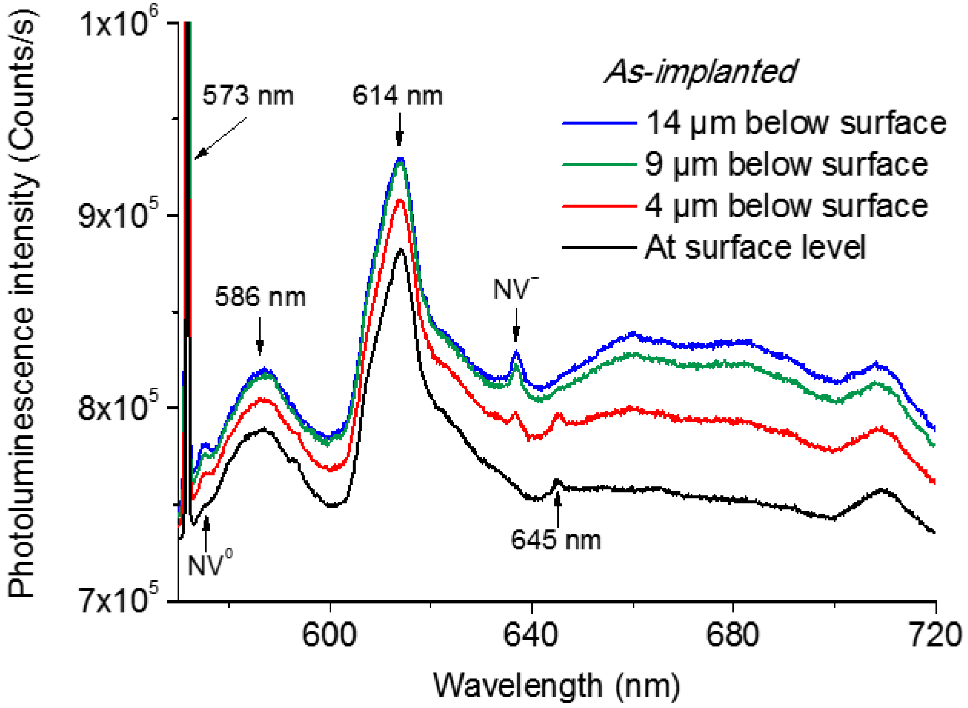
- Strong channeling effects along all axial and planar directions indicate that ^{121}Sn substitutional sites must be involved.
- 920ºC annealing ~doubles the maximum yield (β⁻ anisotropy) of all patterns. A minority fraction is found on BC sites.
- <u>RT as-implanted:</u> best fits obtained for
  63% S with u_1=0.18 Å
  41% BC with u_1=0.11 Å
- <u>T_A=920°C:</u> best fits obtained for
  79% ideal S with u_1=0.034 Å
  32% ideal BC with u_1=0.034 Å

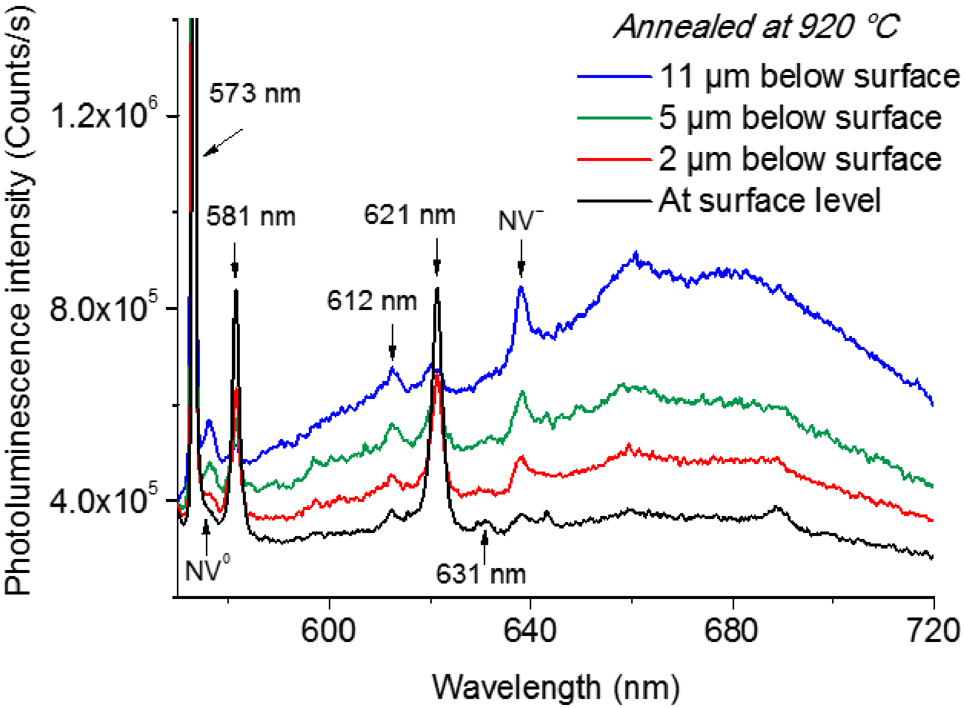
^{121}Sn site fractions as function of annealing temperature



- Surprisingly high fraction of ^{121}Sn found on BC sites (= “split-vacancy” configuration) already in the as-implanted state.
- Besides reducing the rms displacements from S and BC sites, annealing at 920°C also converts some of the ^{121}Sn from BC sites to S sites.

Confocal depth-resolved photoluminescence





- Same sample was implanted with stable ^{120}Sn at KU Leuven (60 keV, 2×10^{12} cm^{-2}).
- PL excitation by 532 nm laser 1 mW

- RT as-implanted state dominated by broad lines around 586 nm and 614 nm (damage-related)
- 920ºC annealed: characteristic sharp PL line (FWHM 2.3 nm) from Sn*V*$^{-}$ at 621 nm [1,2] near the surface
- Sharp line at 581 nm seems to be the so-called L1 center [11] (of yet unidentified origin)

[1] S.D. Tchernij *et al.*, ACS Photonics 4 (2017) 2580
[2] T. Iwasaki *et al.*, Phys. Rev. Lett. 119 (2017) 253601
[11] R. John *et al.*, New J. Phys. 19 (2017) 053008

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Isotopes for possible future EC-SLI experiments on colour centers in diamond

isotope	$t_{1/2}$	yield [ions/μC]	target + ion source	remarks
^{31}Si	157 min	^{31}Al: 2.5×10^{5}	UC_x + Al RILIS	low yield, ^{31}Si recoil implant
^{75}Ge	82.8 min	^{75}Ga: 3×10^{7}	UC_x + Ga RILIS	^{75}Ge recoil implanted
^{209}Pb	3.25 h	?	UC_x + Pb RILIS + LIST?	possible ^{209}Fr contamination
^{27}Mg	9.46 min	1×10^{7}	Ti + Mg RILIS	
^{45}Ca	164 d	^{45}K: 1×10^{7}	UC_x + W	^{45}Ca recoil implanted
^{65}Ni	2.52 h	7×10^{7}	UC_x + Ni RILIS	
^{6}He	807 ms	5×10^{7}	UC_x or BeO	All noble gases
^{23}Ne	37.2 s	1.6×10^{6}	UC_x	from plasma
^{41}Ar	109 min	3.2×10^{7}	TiO_2 or UC_x	ion sources
^{87}Kr	76.3 min	2×10^{8}-2×10^{9}	UC_x or PbBi	
^{133}Xe	5.24 d	6×10^{7}	PbBi	
^{135}Xe	9.14 h	1.5×10^{8}	ThC	

- Yields are mostly “book values” from new ISOLDE yield data base.
- In red: $t_{1/2}$ also suitable for radioactive PL;
- For He and Ne: no PL radiotracers;
- For Mg: ^{28}Mg (20.9 h) for PL.

Conclusions

- First direct structural evidence and quantification for implanted Sn in the Sn*V* "split-vacancy" configuration
- Clarification of the Sn*V* production via ion implantation:
- RT as-implanted state characterized by high fraction of Sn*V* (~40%) but in a distorted configuration;
- 920ºC annealing results in reconfiguration of Sn in ideal S (~70%) and BC sites (Sn*V*, ~30%).
- PL signature of SnV^{-} detected: sharp line at 621 nm.
- Radioactive isotopes for EC-SLI and radiotracer PL can be found for most "colour center elements".
- Colour centers in diamond will be subject of a proposal to INTC in 2020.