Materials science aspects of quantum colour center creation in diamond by means of ion implantation

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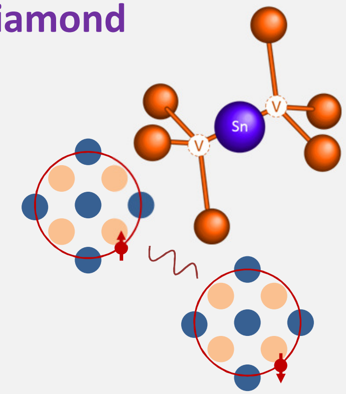
- **Introduction: quantum centers suitable for studies with radioactive ion beams**
- **^{121}Sn*V* colour center in diamond**
- **Conclusions**

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
- Quantum properties emerge from the electronic/nuclear interaction of the impurity with the crystal host
- Useful quantum properties are related to spin interactions, (stimulated) photon emission, coherence, entanglement, polarization of photons…
- Microscopic structure of centers determines their quantum properties
- Many such systems are produced by ion implantation

Examples

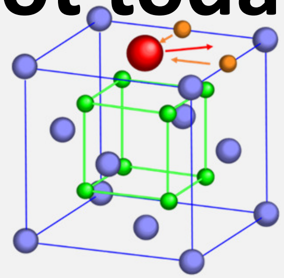
Colour centers in diamond



q-bits and single-photon emitters
⇩
quantum coherence and entanglement
⇩
quantum communication, computation, metrology

^{229m}Th nuclear isomer (~8 eV) in CaF_2 or MgF_2

not today

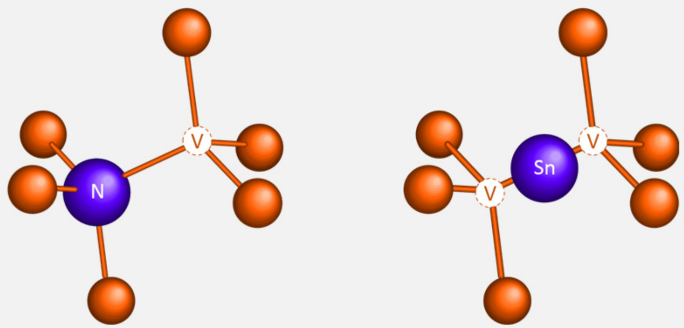


nucleus with lowest isomeric energy: ~8 eV
⇩
stimulated photon emission
⇩
nuclear clock

SnV in diamond as single photon emitter

- NV, SiV, GeV, SnV [1,2] and PbV colour 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 in diamond:

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



D_{3d} "split-*vacancy*" [3,4], assumed for group IV-*vacancy*: **SiV, GeV, SnV, PbV**

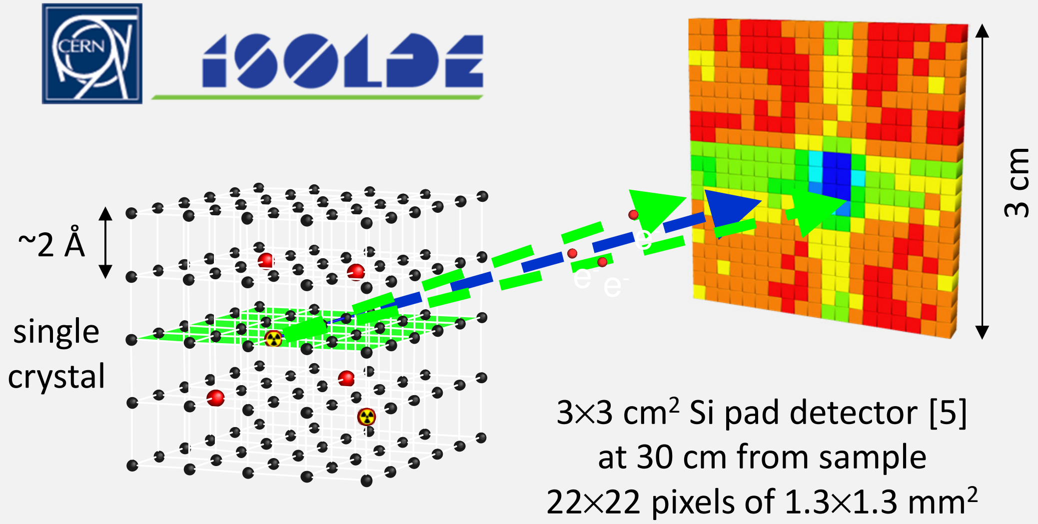
- Superior optical properties of the group-IV-*vacancy* centers are to a large extent a consequence of their D_{3d} inversion (mirror) symmetry.
- Group IV–*vacancy* centers are commonly produced by ion implantation.
- **How to optimize implantation conditions in order to achieve unperturbed split-*vacancy*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

This talk!

[1] D. Tchernij, … J. Forneris, *et al.*, ACS Photonics 4 (2017) 2580
[2] T. Iwasaki, … P. Syushev, *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 (2×10^{12} cm^{-2}) into diamond, measured RT as-implanted and 920°C 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.

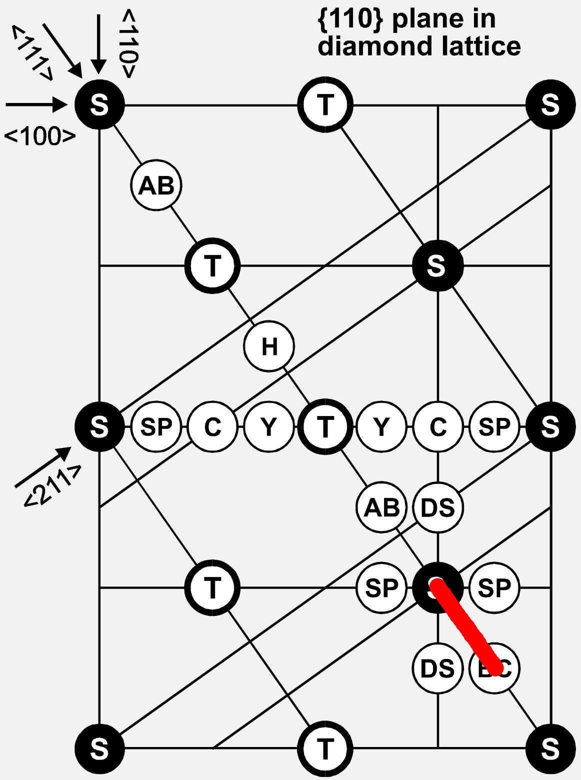


Angular dependent β^- emission patterns characterize the lattice site distribution of the ^{121}Sn emitter atoms.

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

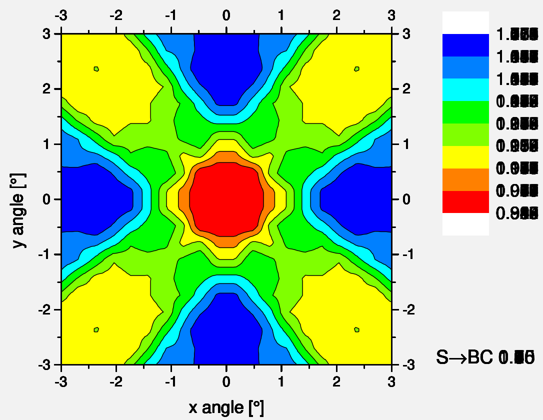
Many-beam" calculation of β⁻ emission yields

high- symmetric sites in diamond



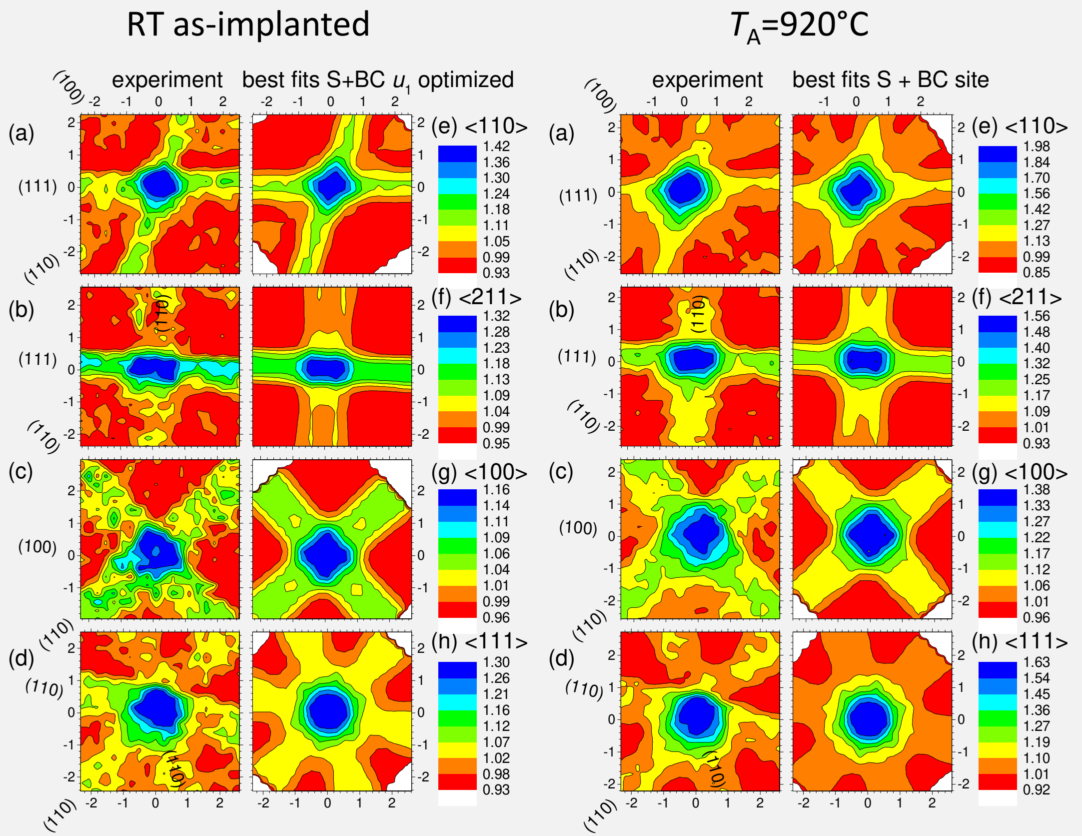
- β⁻ angular emission yield patterns from ^{121}Sn are calculated for ~250 lattice sites in the diamond unit cell using the "many-beam" [6,7] approach.

- Anisotropy and contours of patterns change with position of ^{121}Sn in the lattice,
  e.g. the <100> pattern when moving from S to BC sites:



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

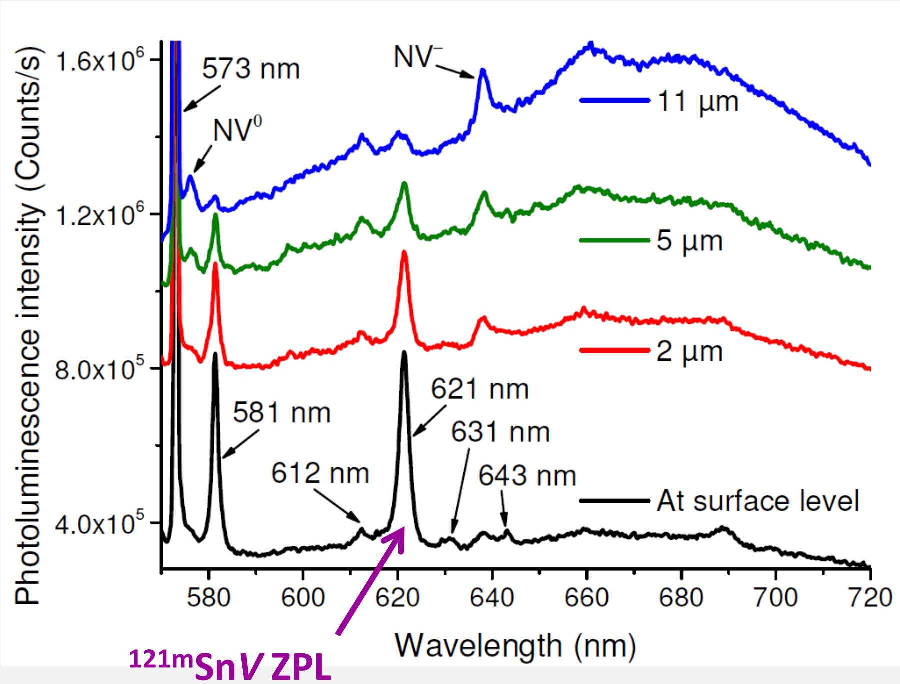
Emission channeling ^{121}Sn in diamond



- Strong channeling effects along all axial and planar directions indicate that ^{121}Sn substitutional sites must be involved.
- 920°C annealing ~doubles the maximum yield (β^- anisotropy) of all patterns.
  A considerable fraction is found on BC sites (split-*vacancy* configuration).

- RT as-implanted: best fits obtained for
  63% S with u_1=0.18 Å
  41% BC with u_1=0.11 Å
- T_A=920°C: best fits obtained for
  79% ideal S with u_1=0.034 Å
  32% ideal BC with u_1=0.034 Å

Confocal depth-resolved photoluminescence ^{121m}Sn in diamond



- Same sample as used for EC also contains the long-lived isomer ^{121m}Sn ($t_{1/2}$=55 y)
- Photo Luminescence (PL) excitation by 532 nm laser 1 mW

- 920°C annealed: sharp (FWHM 2.3 nm) zero phonon line (ZPL) from Sn*V*⁻ at 621 nm [1,2] near the surface
- Our sample showed the lowest ensemble FWHM at RT so far reported in the literature.
- Recently published: U. Wahl *et al.*, Phys. Rev. Lett. 125 (2020) 045301/1-7

[1] D. Tchernij, … J. Forneris, *et al.*, ACS Photonics 4 (2017) 2580

[2] T. Iwasaki, … P. Syushev, *et al.*, Phys. Rev. Lett. 119 (2017) 253601

Our current experiment IS668 at ISOLDE/CERN

Isotope	$t_{1/2}$	yield [ions/μC]	target + ion source
^{31}Si	157 min	^{31}Al: 2.5×10^{5}	UC_x + Al RILIS
^{75}Ge	82.8 min	^{75}Ga: 3×10^{7}	UC_x + Ga RILIS
^{121}Sn	27.1 h	1×10^{8}	UC_x + Sn RILIS
^{209}Pb	3.25 h	?	UC_x + Pb RILIS + LIST?
^{27}Mg	9.46 min	1×10^{7}	Ti + Mg RILIS
^{45}Ca	164 d	^{45}K: 1×10^{7}	UC_x - W
^{89}Sr	50.5 d	^{89}Rb: 5×10^{9}	UC_x - W
^{6}He	807 ms	5×10^{7}	UC_x or BeO
^{23}Ne	37.2 s	1.6×10^{6}	UC_x plasma
^{41}Ar	109 min	3.2×10^{7}	TiO_2 or UC_x
^{87}Kr	76.3 min	2×10^{8}-2×10^{9}	UC_x or PbBi
^{133}Xe	5.24 d	6×10^{7}	PbBi plasma
^{135}Xe	9.14 h	1.5×10^{8}	ThC plasma

- IS668 = "Quantum colour centers in diamond studied by emission channeling with short-lived isotopes (EC-SLI) and radiotracer photoluminescence"
- 20×8h shifts approved, first successful run on ^{27}Mg, ^{45}Ca, ^{89}Sr in diamond 6.-9.7.2021

- Isotopes in red are also suitable for radiotracer PL.

- Participating institutes:
- C²TN, IST (Portugal)
- QSP, KU Leuven (Belgium)
- Universidade de Aveiro (Portugal)
- University of Torino (Italy)
- University of Warwick (UK)
- CERN

Collaborations

- Diamond:
- Lino Pereira, André Vantomme (Quantum Solid State Physics , KU Leuven, Belgium)
- Vítor Amaral (CICECO, Universidade de Aveiro, Portugal)
- Miloš Nesládek (Hasselt University, Belgium)
- Karl Johnston (CERN, Geneva, Switzerland)
- Ben Green (University of Warwick, UK)
- Petr Syushev (group of F. Jelezko, University of Ulm, Germany)
- Jacopo Forneris (University of Turin, Italy)
- Ádám Gali (Wigner Research Center, Budapest, Hungary)
- Gerald Auböck (Silicon Austria Labs)

Quantera application on Quantum Repeaters based on Sn*V*, Pb*V*

- ^{229m}Th:
- Lino Pereira, André Vantomme (Quantum Solid State Physics, KU Leuven, Belgium)
- Piet van Duppen (Instituut voor Kern- en Stralingsfysica, KU Leuven, Belgium)
- Thorsten Schumm (Atominstitut, TU Wien, Austria)

Conclusions

- Emission channeling offers unique opportunities for structural studies of quantum colour center formation in diamond.
- First direct structural evidence and quantification for implanted ^{121}Sn in the Sn*V* "split-*vacancy*" configuration (surprisingly high yield of ~30%)
- PL signature of ^{121m}SnV^{-} detected: sharp ZPL at 621 nm
- Besides ^{121}Sn, beam time is also granted for further studies of other colour centers in diamond, using emission channeling and radiotracer PL.