Quantum colour centers in diamond studied by emission channeling with short-lived isotopes (EC-SLI) and radiotracer photoluminescence (IS668)

L.M.C. Pereira[1], U. Wahl[2], J.G. Correia[2], B. Biesmans[1], A.R.G. Costa[1], M.R. da Silva[3], E. David Bosne[4], A. Lamelas[3], G. Magchiels[1], J. Moens[1], S.M. Tunhuma[1], R. Villarreal[1], A. Vantomme[1] (EC-SLI collaboration)

V. Amaral[3], E. Corte[5], S. Ditalia Tchernij[5], J. Forneris[5], B. Green[6], K. Johnston[7], E. Nieto Hernandez[5], P. Olivero[5], V. Pugliese[5]

1 KU Leuven, Quantum Solid State Physics, Belgium

2 Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Portugal

3 CICECO- Instituto de Materiais de Aveiro, Universidade de Aveiro, Portugal

4 Institute of Experimental and Applied Physics, Technical University of Prague, Czech Republic

5 Physics Department, University of Torino, and Instituto Nazionale di Fisica Nucleare (INFN), Torino, Italy

6 University of Warwick, UK

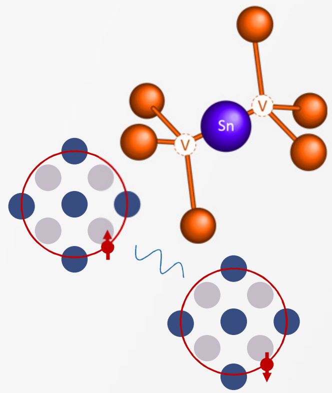
7 CERN-EP ISOLDE, Geneva, Switzerland

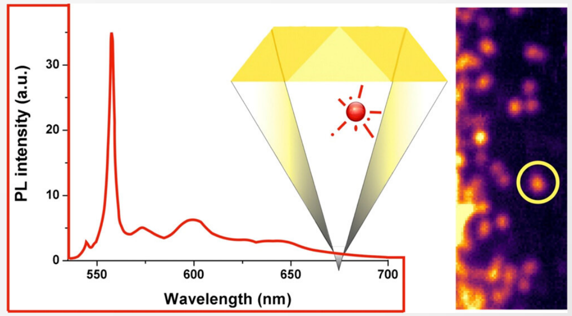
Spokespersons: L.M.C. Pereira, U. Wahl

Local contact & technical coordinator: J.G. Correia

Quantum colour centers in diamond

- Research interest of IS668: impurities in diamond which exhibit quantum properties useful for future applications: "quantum centers"
- General characteristics:
  Dilute impurity atoms embedded in diamond matrix (historically termed "colour centers")
- Useful quantum properties are related to spin interactions, (stimulated) photon emission, coherence, entanglement, polarization of photons…
- Quantum properties emerge from the electronic interaction of the impurity with the diamond host
- Microscopic structure of centers determines their quantum properties





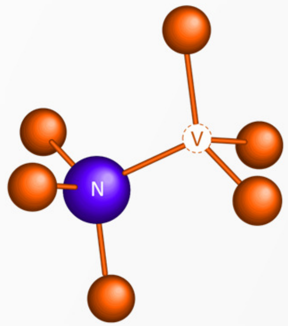
q-bits and single-photon emitters
⇩
quantum coherence and entanglement
⇩
quantum computation, encryption, metrology

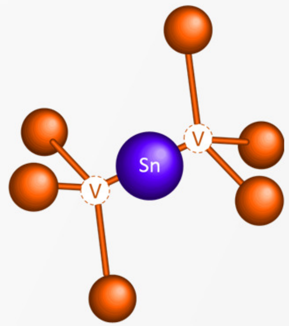
C²TN | TÉCNICO LISBOA | KU LEUVEN

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 larger impurity atoms pair with a *vacancy V*.
- Two possible configurations for impurity-*vacancy* centers in diamond:

C_{3v} "full-*vacancy*",
for **N*V***,
e.g. applications in
nano-magnetometry



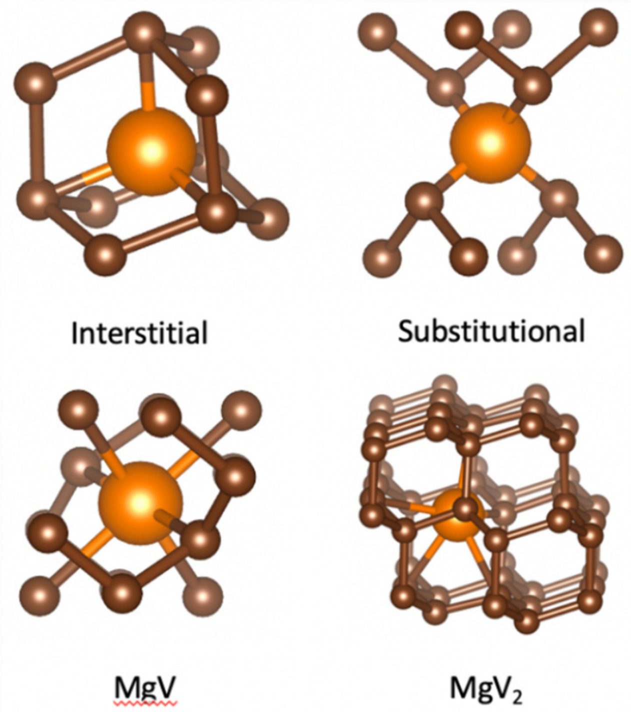


D_{3d} "split-*vacancy*",
for group IV-*vacancy*: **Si*V* [1], Ge*V* [2], Sn*V* [3,4], Pb*V* [5],** but also for **Mg*V* [6]** and others

- 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.

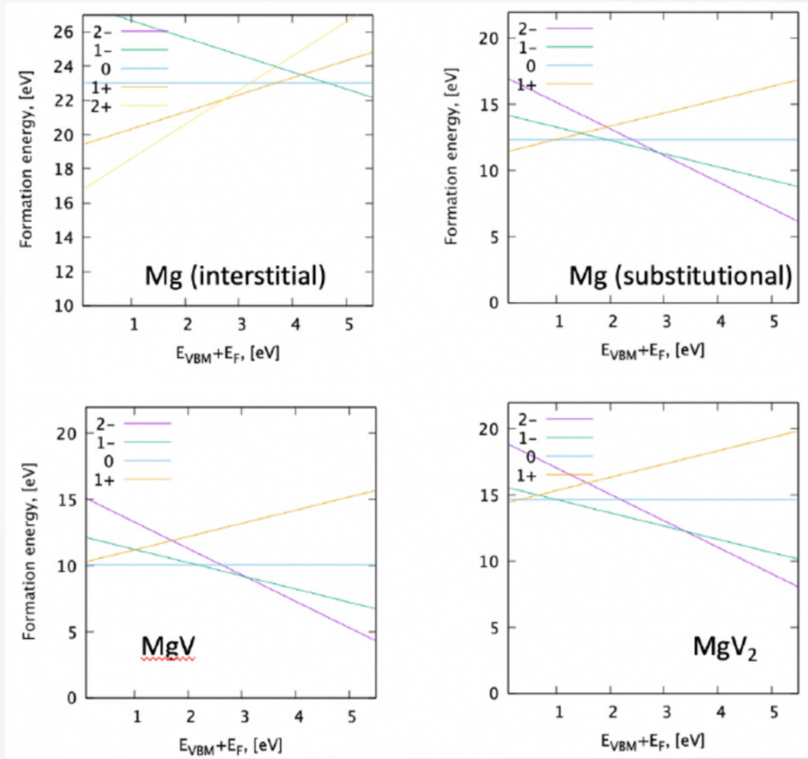
[1] J.P. Goss *et al*, Phys. Rev. Lett. 77 (1996) 3041
[2] T. Iwasaki *et al*, Sci. Rep. 5 (2015) 12882
[3] S.D. Tchernij, … J. Forneris, *et al*, ACS Photonics 4 (2017) 2580
[4] T. Iwasaki, *et al*, Phys. Rev. Lett. 119 (2017) 253601
[5] S.D. Tchernij, … J. Forneris, *et al*, ACS Photonics 5 (2018) 4864
[6] E. Corte, …L.M.C. Pereira, U. Wahl, J. Forneris, *et al*, ACS Photonics 10 (2023) 101

Example: Predicted structures of Mg defects in diamond



- Theoretically investigated structures of Mg-related complexes in diamond [7]:
- 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>) predicted with ZPL=563 nm.
- MgV_2: (C_1 symmetry <100>)

Formation energy *vs* Fermi-level [3]

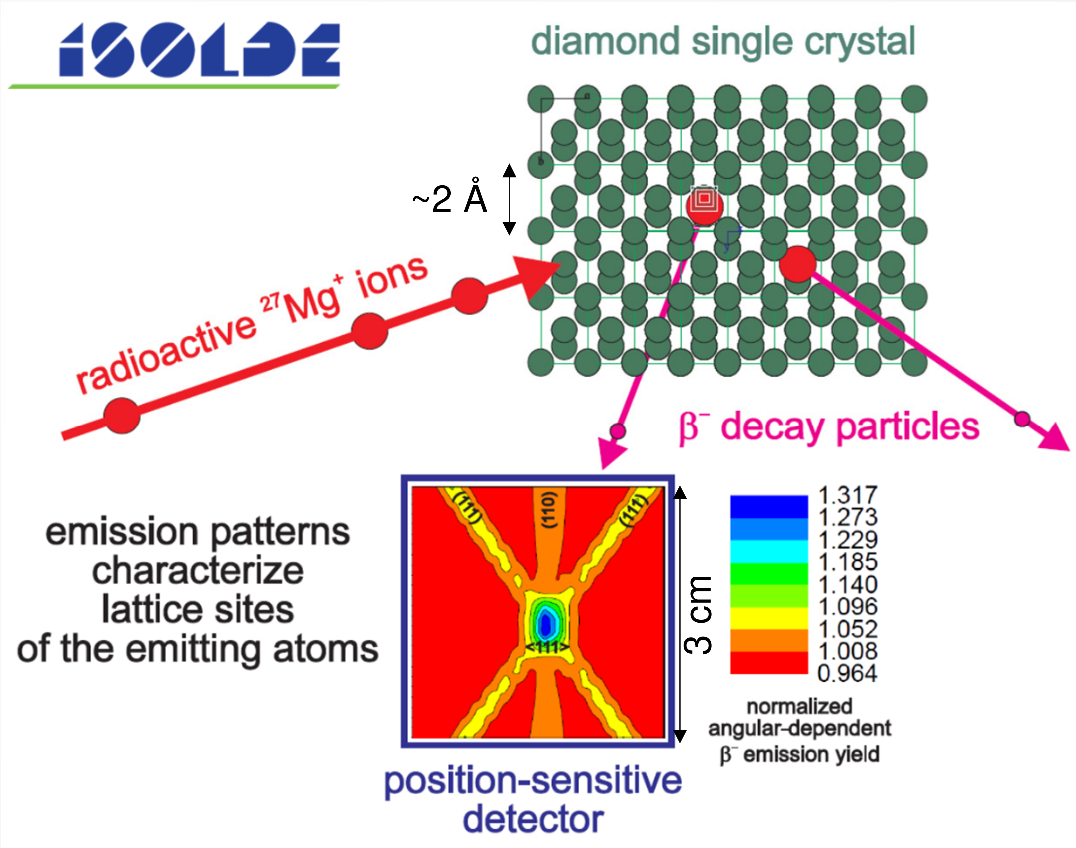


[7] A. Pershin, *et al*, “Highly tunable magneto-optical response from Mg*V* color centers in diamond”, npj Quantum Information 7 (2021) 99

Formation energies favour Mg*V*, Mg(S), possibly MgV_2, rule out Mg_i

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

- Radioactive probe atoms are produced and ion implanted into single crystals at ISOLDE, 30-50 keV, 10^{11}-10^{13} cm^{-2}
- Thermal processing: post-implant annealing at T_a or vary implantation temperature T_i
- Position- and energy sensitive detector [8] is used to detect emission channeling [9] effects of β^- decay particles in the vicinity of major crystallographic directions.



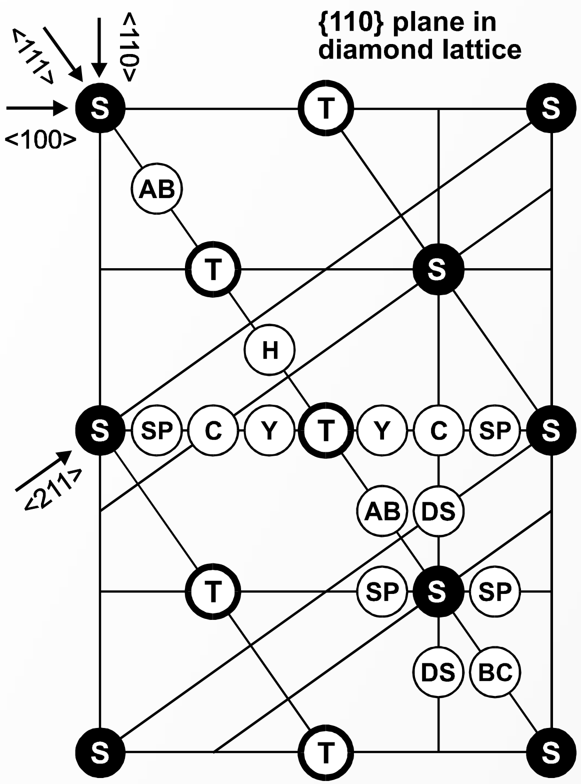
3×3 cm^2 Si pad detector [8]
at 30 cm from sample
22×22 pixels of 1.3×1.3 mm^2

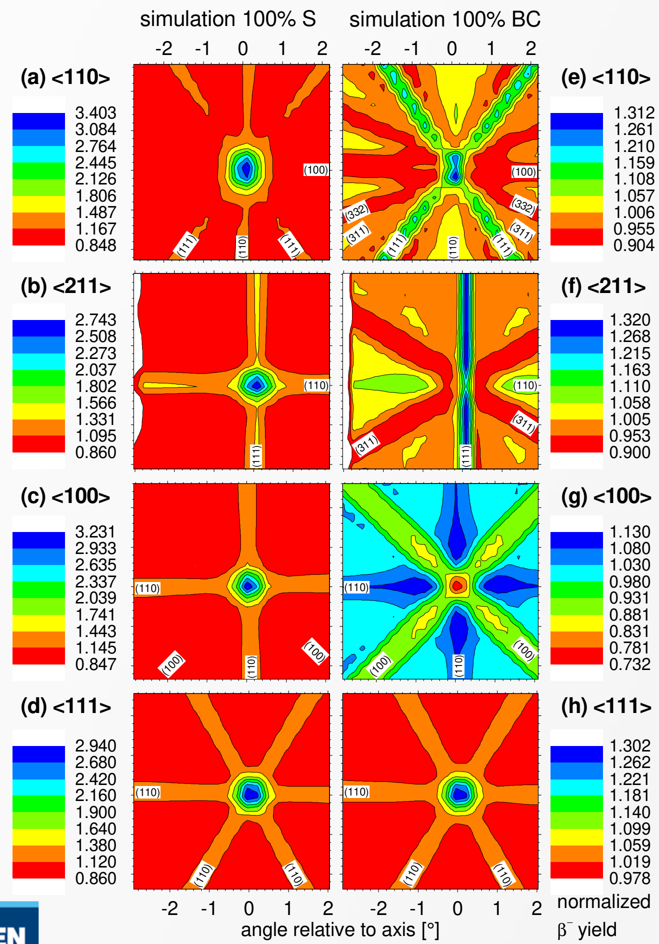
[8] U. Wahl *et al*, Nucl. Instr. Meth. A 524 (2004) 245
[9] H. Hofsäss, G. Lindner, Phys. Rep. 201 (1991) 121

“Many-beam” calculation of β⁻ emission yields

Occupied lattice sites identified by comparison of experimental results to simulated yields

high- symmetric sites in diamond



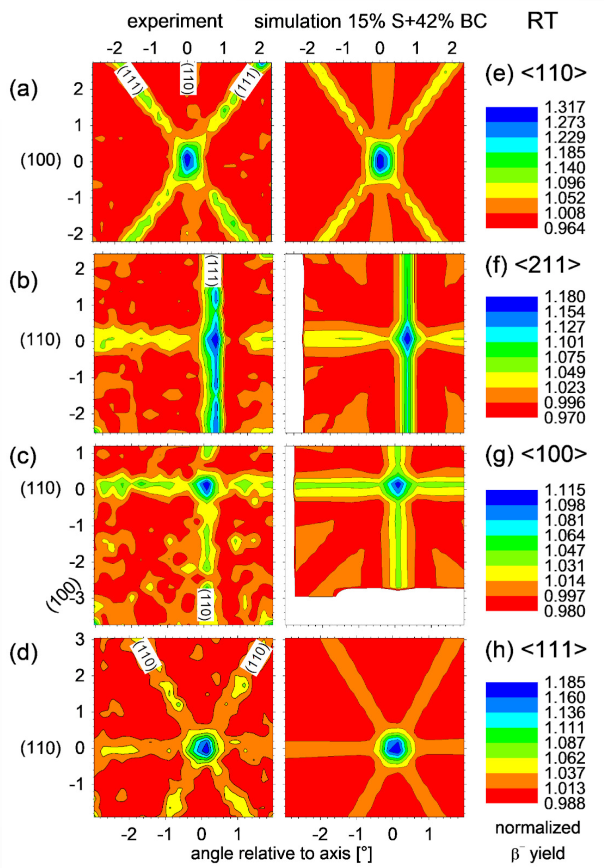


- β⁻ angular emission yield patterns are calculated for ~250 lattice sites in the diamond unit cell using the “many-beam” [9,10] approach.
- Anisotropy and contours of patterns change with position of emitter in the lattice, as shown for the <110>, <211>, <100>, and <111> patterns from ^{27}Mg on S and BC sites.
- The occupation of several sites results in a linear superposition of patterns.

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

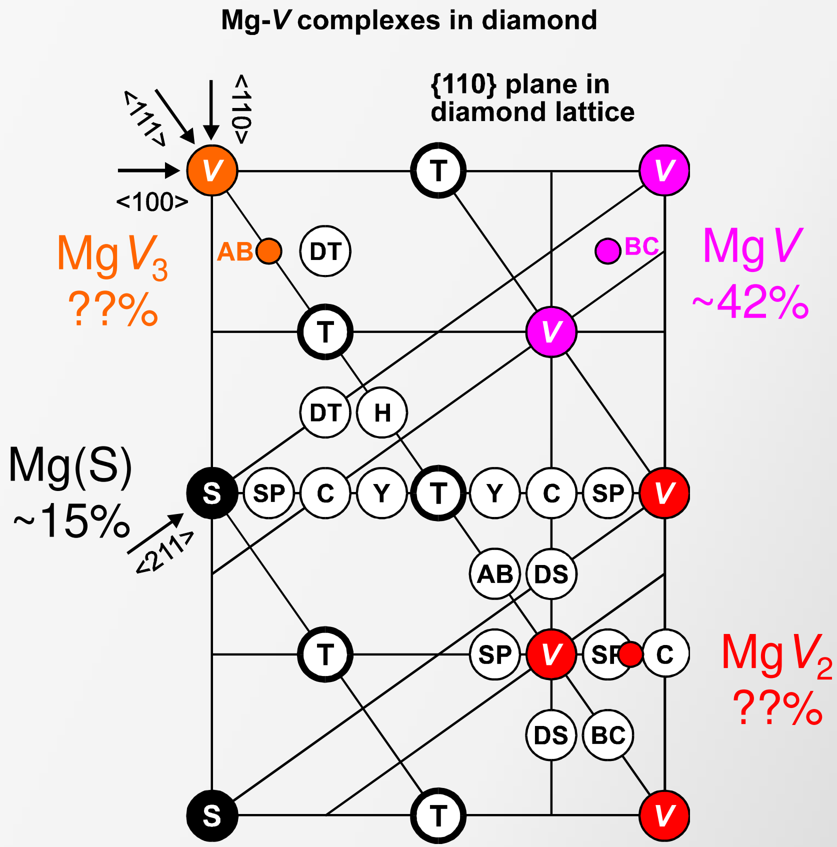
C2TN TÉCNICO LISBOA KU LEUVEN

EC characterization of ^{27}Mg colour centers in diamond (RT)



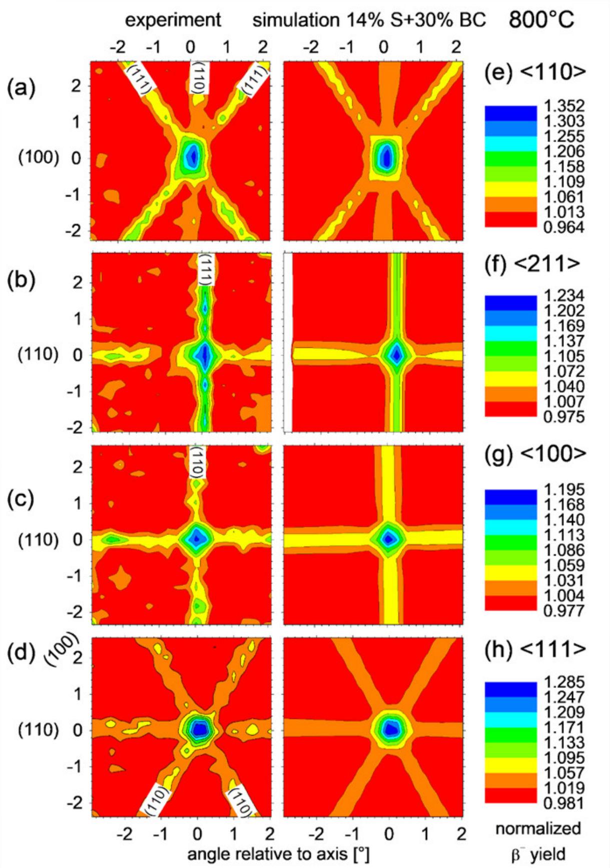
T_i=20°C

- EC from RT implanted ^{27}Mg shows 15% on S and 42% on bond-center (BC) sites [6]
- The occupation of BC sites is due to Mg *V* in the split-vacancy configuration.
- High yield of formation (42%) of the Mg *V* defect
- However, ~43% of emitters are in “random” sites: could be within Mg V_2 and Mg V_3 complexes: lower symmetry ⇔ quite weak channeling



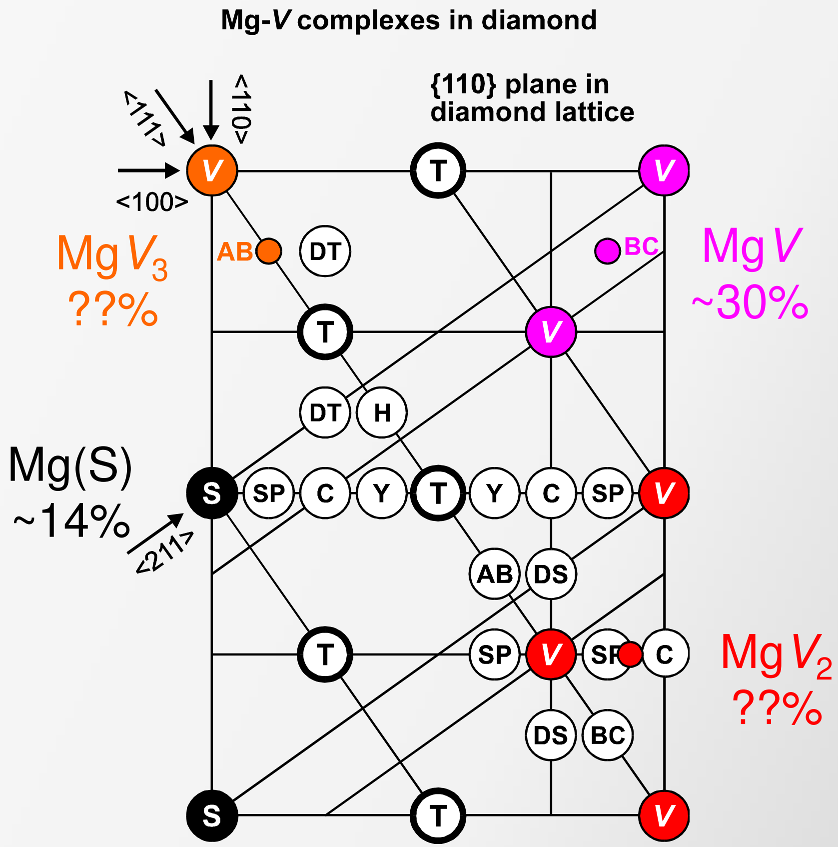
[6] E. Corte, …L.M.C. Pereira, U. Wahl, J. Forneris, *et al*, ACS Photonics 10 (2023) 101

EC characterization of ^{27}Mg colour centers in diamond (800°C)



T_i=800°C

- EC from 800°C implanted ^{27}Mg show 14% on S and 30% on bond-center (BC) sites [5]
- The occupation of BC sites is due to Mg*V* in the split-vacancy configuration.
- High yield of formation (30%) of the Mg*V* defect
- However, ~56% of emitters are in "random" sites: could be within MgV_2 and MgV_3 complexes:
  lower symmetry
  ⇔ quite weak channeling

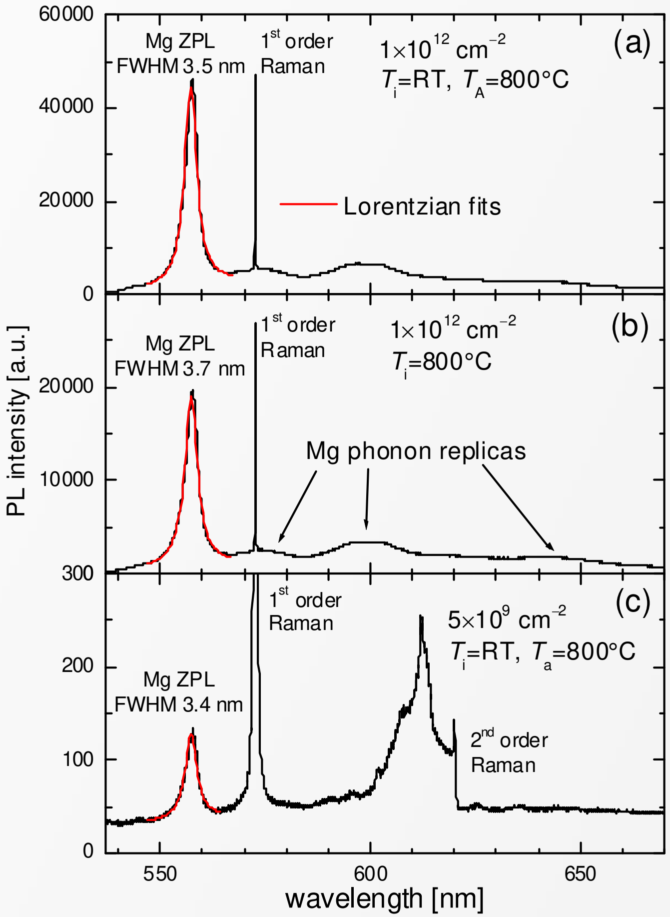


[6] E. Corte, …L.M.C. Pereira, U. Wahl, J. Forneris, *et al*, ACS Photonics 10 (2023) 101

PL from ^{24}Mg implanted in diamond at ISOLDE

- Stable ^{24}Mg implanted into "electronic-grade" diamond ([N]<5 ppb) at ISOLDE, 30 keV
- 532 nm laser excitation shows Zero Phonon Line (ZPL) from Mg*V* centers at 557.6 nm, as well as characteristic phonon replicas (measured at U Turin).
- ZPL with narrow FWHM (3.4-3.7 nm, measure of structural quality of Mg*V* centers) already after RT implantation and annealing at 800°C, or 800°C implantation.
- Same FWHM as in literature after 1600°C annealing (3.5 nm) [11].

[11] E. Osmic, *et al*, "Unusual temperature dependence of the photoluminescence emission of Mg*V* centers in diamond", Appl. Phys. Lett. 121 (2022) 084101



1×10^{12} cm^{-2}
implanted at T_i = RT,
annealed 20 min at T_a=800°C

1×10^{12} cm^{-2}
implanted at T_i=800°C

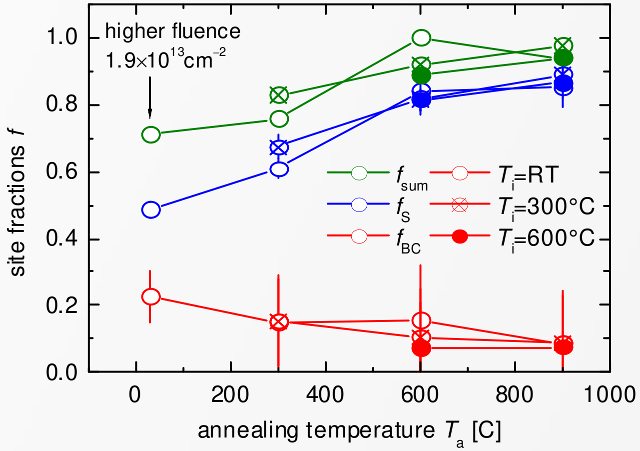
5×10^{9} cm^{-2}
implanted at T_i = RT,
annealed 20 min at T_a=800°C

Mg*V* of good structural quality can be efficiently produced

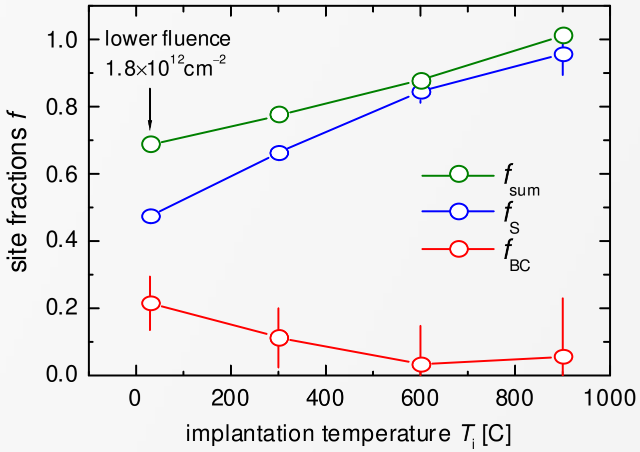
Preliminary EC results on ^{75}Ge

- RT implantation results in ~20% on BC sites and ~50% on S sites.
- BC fraction decreases to a few % for implantation *or* annealing at higher temperatures.
- No general differences visible in between implantation at higher temperatures or post-implant annealing.
- Variation of fluence by factor of 5 had no visible effect.
- Significantly lower BC fractions than for other elements, but still much higher than reported optical activation of 0.4-0.7% [12]

[12] Y. Zhou, *et al*, “Direct writing of single germanium vacancy center arrays in diamond,” New J. Phys. 20 (2018) 125004

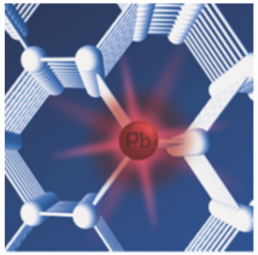


As function of annealing temperature T_i for implantations at RT, 300°C and 600°C, higher fluence

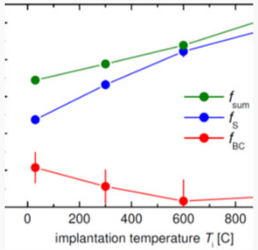


As function of implantation temperature T_i up to 900°C, lower fluence

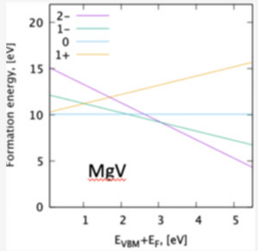
Proposed experiments: 3 key areas



Elements not available during last 2 years



Low temperature implantation



Implantation into pre-doped diamonds

Here the focus will be on ^{209}Pb, which is of particular relevance since Pb*V* centers in split-vacancy configuration have not yet been structurally verified.

Also, it was not yet possible to study ^{31}Si, ^{6}He, ^{23}Ne, ^{41}Ar, ^{87}Kr.

So far, we only studied implantation at RT or elevated temperatures. Especially in the case of Ge, there are indications that low-*T* implantation could improve Ge*V* formation.

Literature has reported improved optical activation of Sn and Mg in *n*-doped diamond, however, the mechanism is unclear: change of charge state of Sn$V^0 \rightarrow$ SnV^-, Mg$V^0 \rightarrow$ MgV^- ***or*** increased production of split-vacancy configuration?

Beam request

isotope	half-life	yield (atoms/μC)	target - ion source	Shifts (8h)
^{209}Pb	3.25 h	no yield in data base	UC_x-Nb - RILIS Pb or LIST Pb	4
^{75}Ga→^{75}Ge	126 s→82.8 min	3×10^7	UC_x-W - RILIS Ga	3
^{121}Sn	27.06 h	1×10^8	UC_x-W - RILIS Sn	4
^{27}Mg	9.5 min	1×10^7	Ti-W - RILIS Mg	2.5
^{28}Mg	21 h	6×10^6	Ti-W or UC_x-W - RILIS Mg	0.5
^{6}He	807ms	7×10^7	UC_x or BeO cold plasma	3.0
^{23}Ne	37.2 s	1.6×10^6	UC_x plasma	1.0
^{41}Ar	109 min	1.6×10^6	UC_x or TiO_2 plasma	0.5
^{87}Kr	76.3 min	2×10^8	UC_x or PbBi plasma	0.5
^{31}Al→^{31}Si	644 ms→157 min	2.5×10^5	UC_x-W - RILIS Al	1

Total requested shifts: 20 (split into ~8 runs over 2 years

Most runs or targets are to be shared with other users