Magnesium-vacancy quantum defects in diamond

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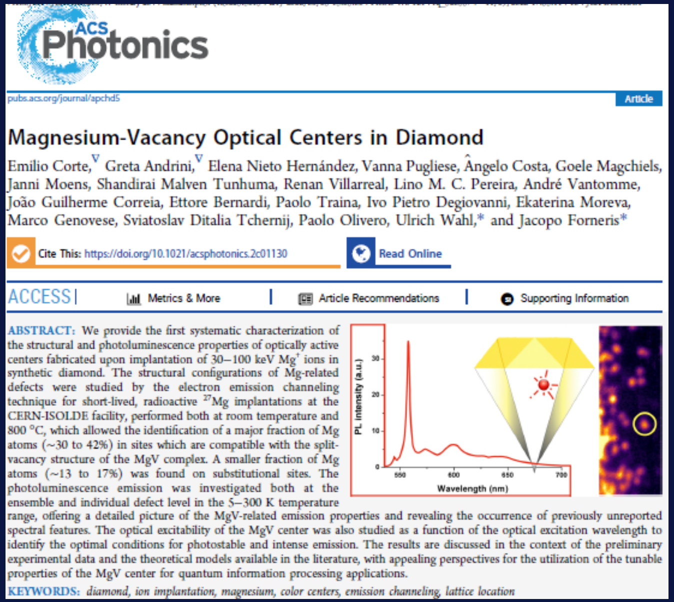
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Magnesium-Vacancy Optical Centers in Diamond

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ABSTRACT: We provide the first systematic characterization of the structural and photoluminescence properties of optically active centers fabricated upon implantation of 30−100 keV Mg^+ ions in synthetic diamond. The structural configurations of Mg-related defects were studied by the electron emission channeling technique for short-lived, radioactive ^{27}Mg implantations at the CERN-ISOLDE facility, performed both at room temperature and 800 °C, which allowed the identification of a major fraction of Mg atoms (~30 to 42%) in sites which are compatible with the split-vacancy structure of the MgV complex. A smaller fraction of Mg atoms (~13 to 17%) was found on substitutional sites. The photoluminescence emission was investigated both at the ensemble and individual defect level in the 5−300 K temperature range, offering a detailed picture of the MgV-related emission properties and revealing the occurrence of previously unreported spectral features. The optical excitability of the MgV center was also studied as a function of the optical excitation wavelength to identify the optimal conditions for photostable and intense emission. The results are discussed in the context of the preliminary experimental data and the theoretical models available in the literature, with appealing perspectives for the utilization of the tunable properties of the MgV center for quantum information processing applications.

KEYWORDS: *diamond, ion implantation, magnesium, color centers, emission channeling, lattice location*

- Mg quantum colour centres in Diamond
- Emission channeling lattice location of ^{27}Mg in diamond

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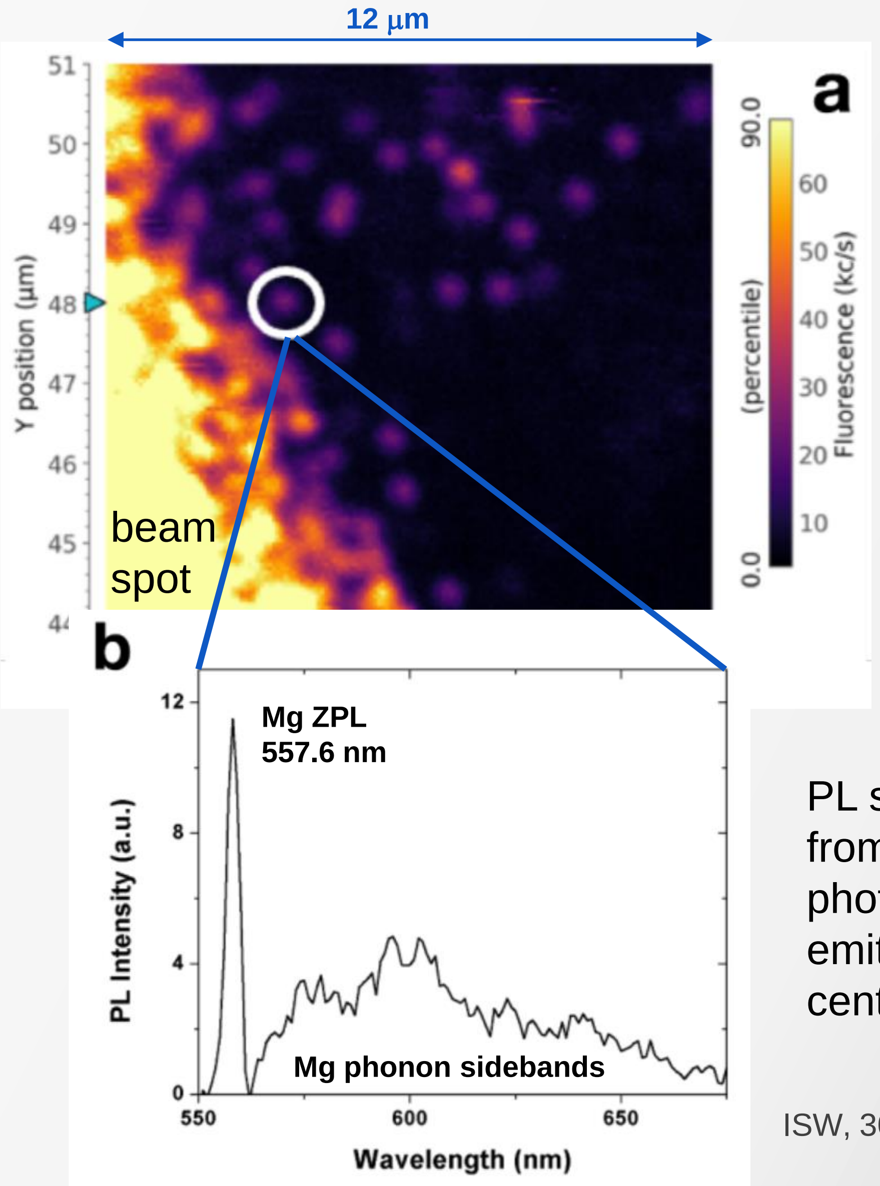
E. Corte, *et al*, “Magnesium-Vacancy Optical Centers in Diamond”, ACS Photonics

Mg-related photoluminescence in diamond

- ^{24}Mg implanted into artificial diamond of highest purity (“electronic-grade”, [N]<5 ppb), 100 keV, fluence $2x10^{12}$ cm^{-2}, annealed at 1200°C
- Excitation with 522 nm laser shows Photoluminescence (PL) inside implanted region with a sharp (FWHM 3.3 nm) and intense Zero Phonon Line (ZPL) at 557.6 nm.
- This line was previously assigned to Mg-related colour centers [1,2].
- Confocal PL microscopy with µm resolution reveals isolated bright spots at the edge of the implanted region.
- What is the nature of these bright spots?
- They also show the PL spectrum (ZPL + phonon sidebands) attributed to Mg.



^{24}Mg implantation KU Leuven, confocal PL at U Turin

PL spectrum from single photon emitting center

[1] T. Lühmann, *et al*, “Screening and engineering of colour centres in diamond”, J. Phys. D: Appl. Phys. 51 (2018) 483002
[2] T. Lühmann, *et al*, “Coulomb-driven single defect engineering for scalable qubits and spin sensors in diamond”, Nat. Commun. 10 (2019) 4956

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Mg-related centers as single photon emitters

- Hanbury-Brown – Twiss (HBT) auto-correlation measurements:
- 50% beam splitter, 2 single photon detectors
- Measure the time τ in between the arrival of single photons at the 2 detectors
- Autocorrelation $g^{(2)}(\tau)$ shows drop to 15% in photon-count rate for 0 ns delay in between detected photons.
- Photons are emitted one by one by the same single center.
- Presumably one single Mg atom
- Saturation count rate as function of laser power:
  ~2 Mcts/s (highest known for any single photon emitter in diamond)
- Possible "single photon-on-demand" system, e.g. for secure quantum communication
- **What is the configuration of the underlying Mg-defect?**

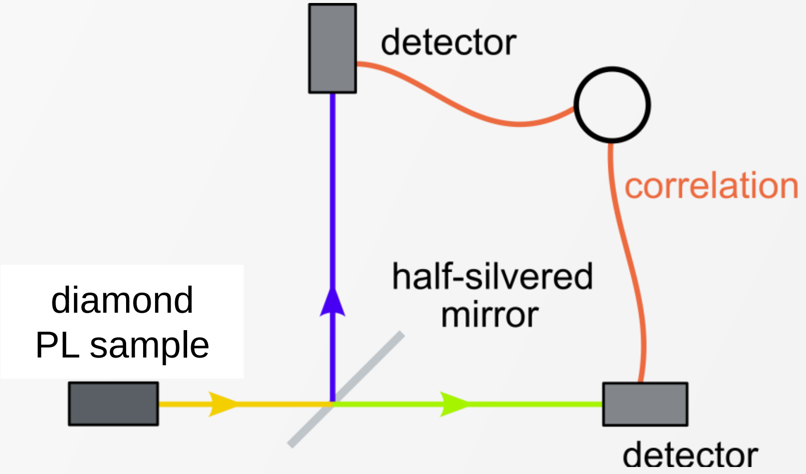
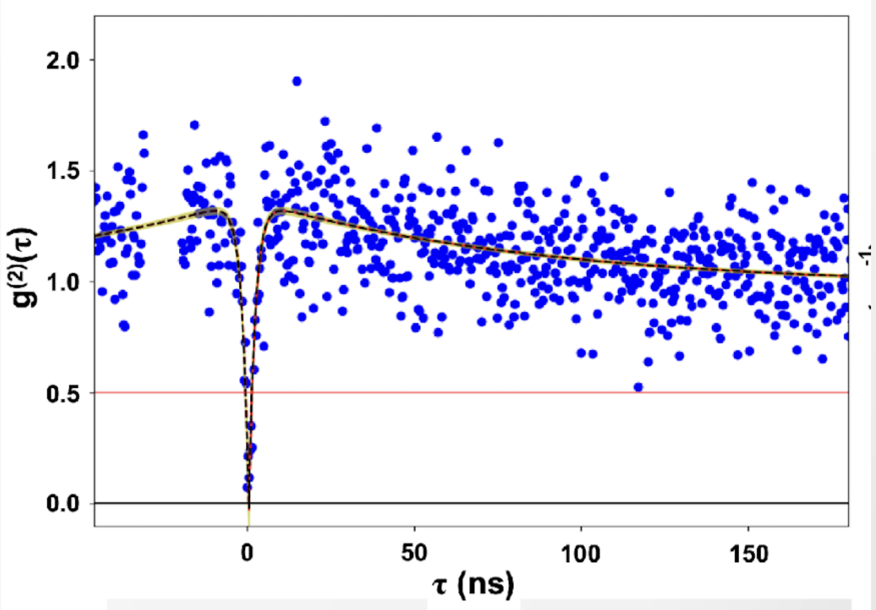
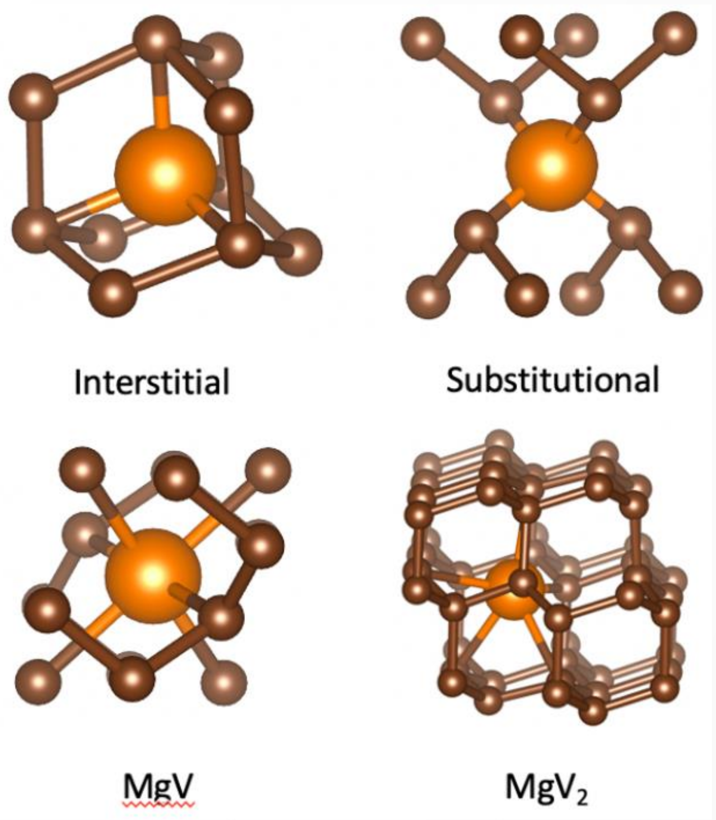


Diagram from: HBT method on Wikipedia

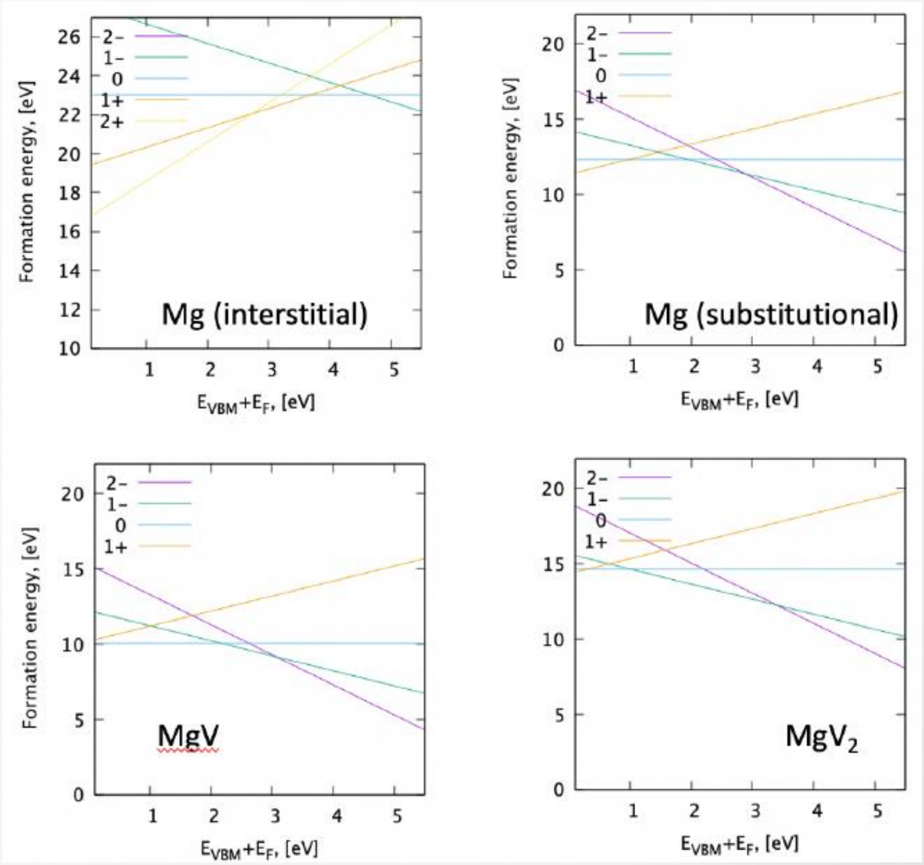


Predicted structures of Mg defects in diamond



- Theoretically investigated structures of Mg-related complexes in diamond [3]:
- 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]



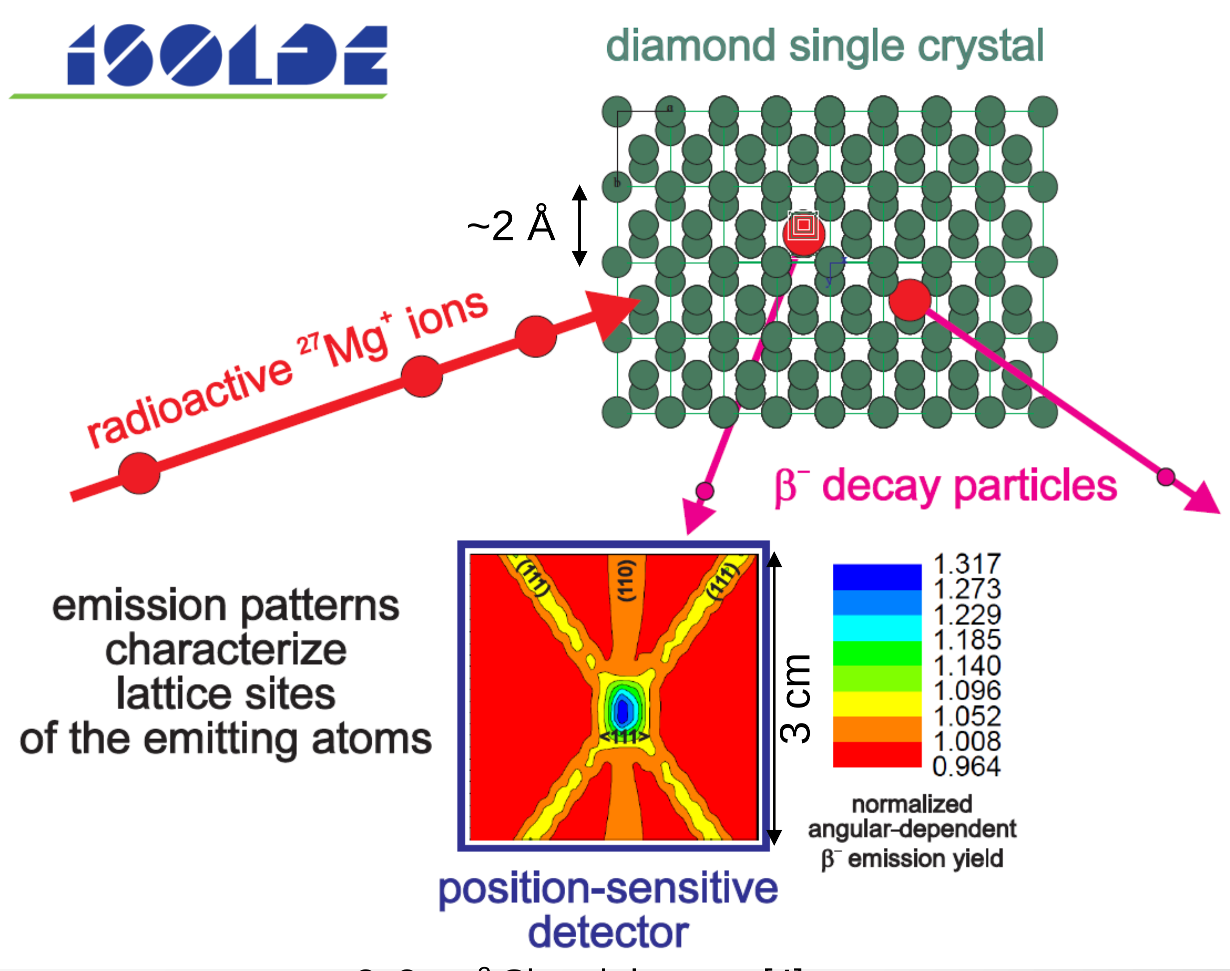
[3] A. Pershin, *et al*, "Highly tunable magneto-optical response from Mg*V* color centers in diamond", npj Quantum Information 7 (2021) 99

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

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

- Radioactive ^{27}Mg ($t_{1/2}$=9. 5 min) probe atoms are produced and ion implanted into single crystals at ISOLDE, 30 keV, 10^{12}-10^{13} cm^{-2}
- Thermal processing: due to short $t_{1/2}$ we can only vary implantation temperature T_i.
- Position- and energy sensitive detector [4] is used to detect emission channeling [5] effects of β^- decay particles in the vicinity of major crystallographic directions.

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

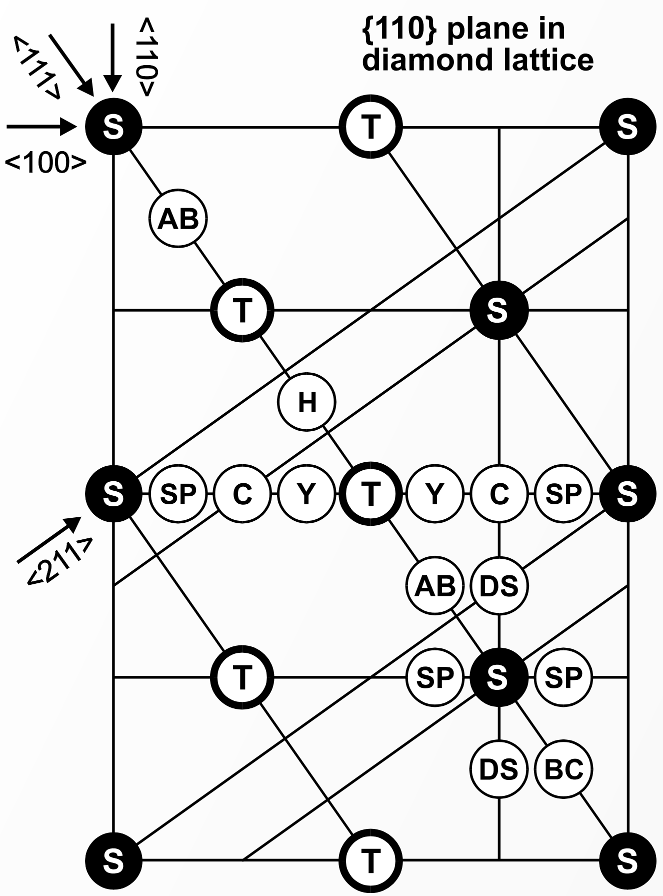


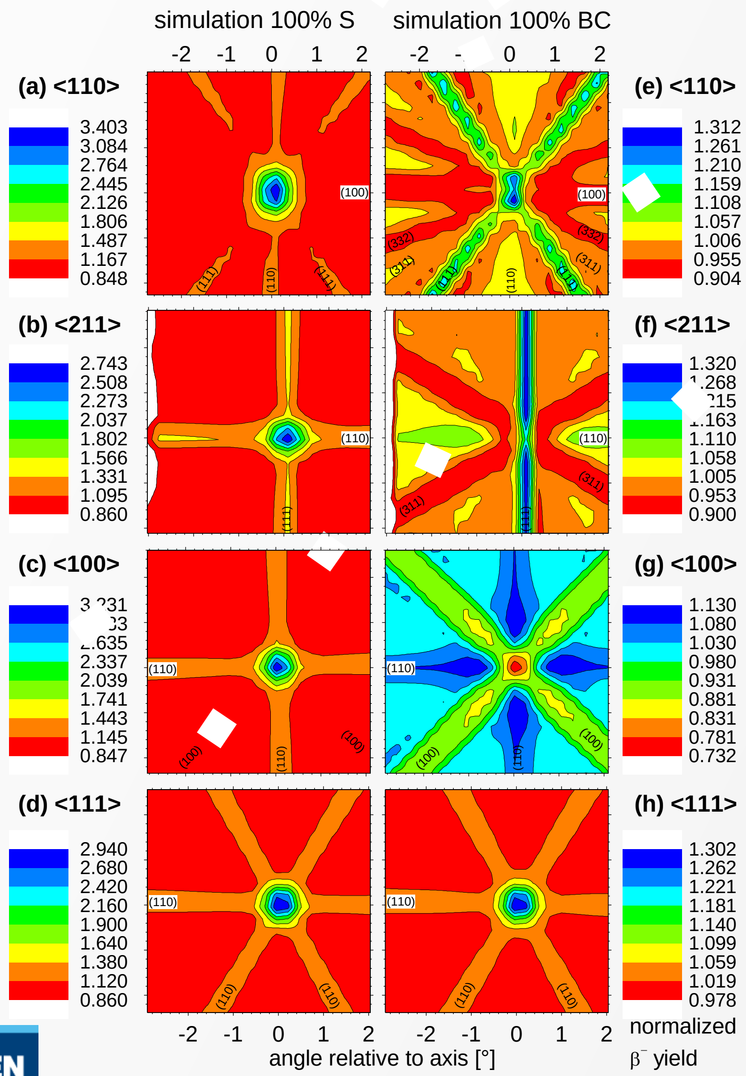
3×3 cm^2 Si pad detector [4]
at 30 cm from sample
22×22 pixels of 1.3×1.3 mm^2

“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” [5,6] 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.

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

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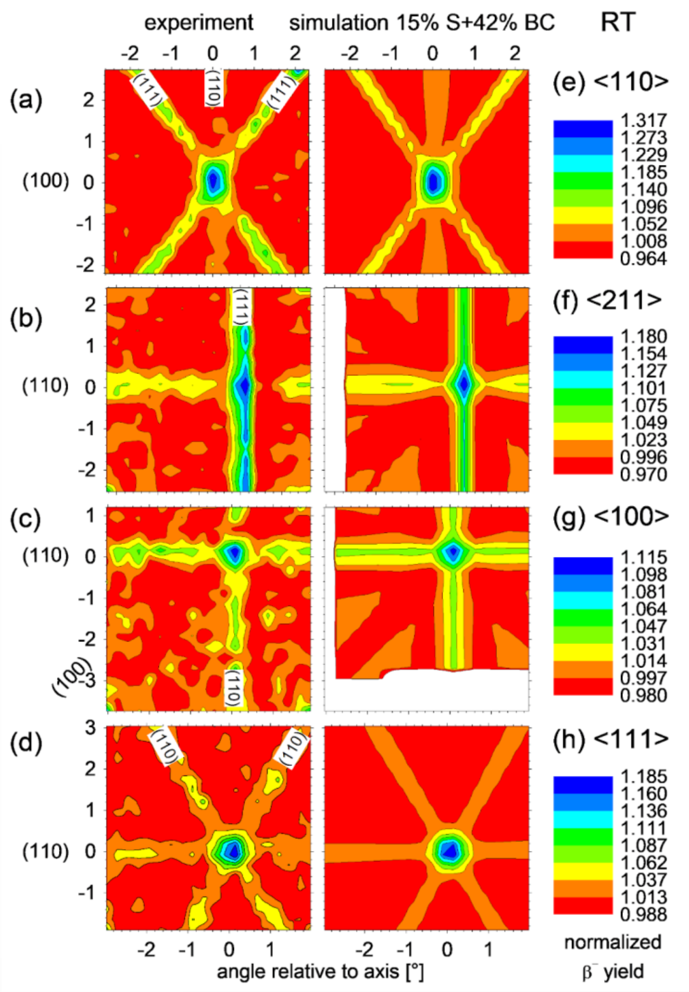
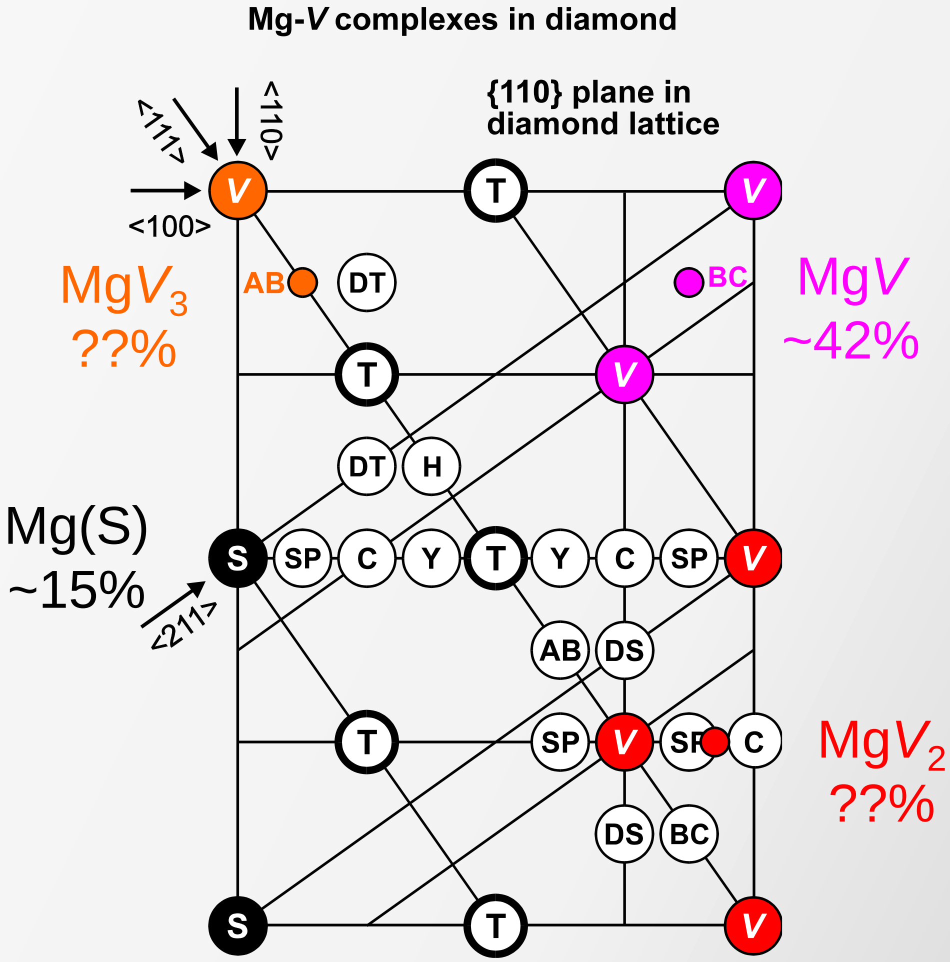
EC characterization of ^{27}Mg colour centers in diamond (RT)

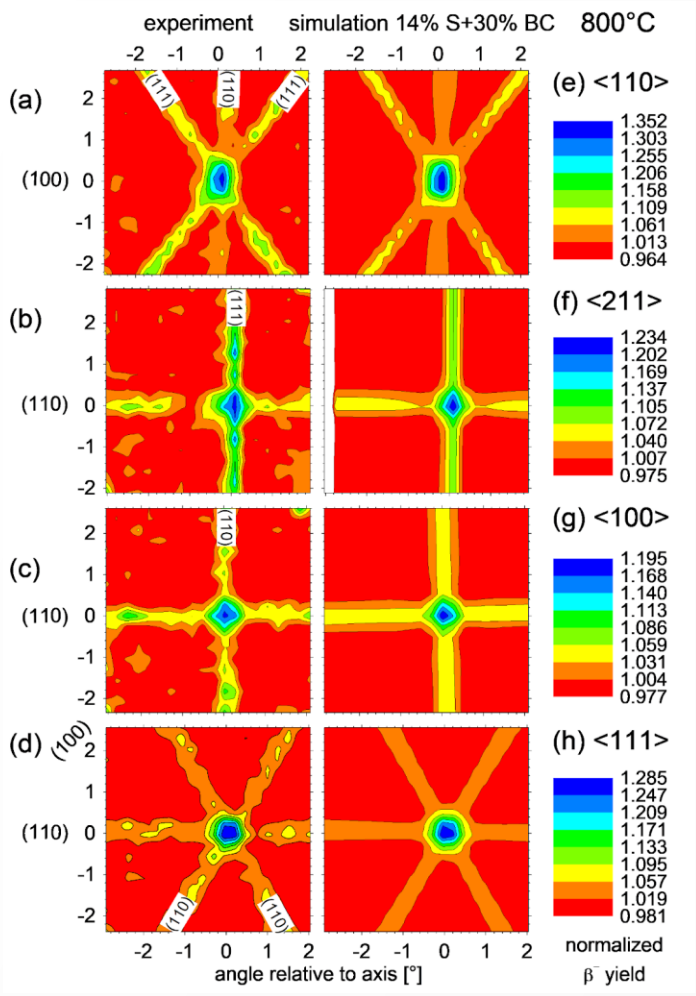


T_i=20°C

- EC from RT implanted ^{27}Mg show 15% on S and 42% on bond-center (BC) sites
- 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 MgV_2 and MgV_3 complexes:
  lower symmetry
  ⇔ quite weak channeling

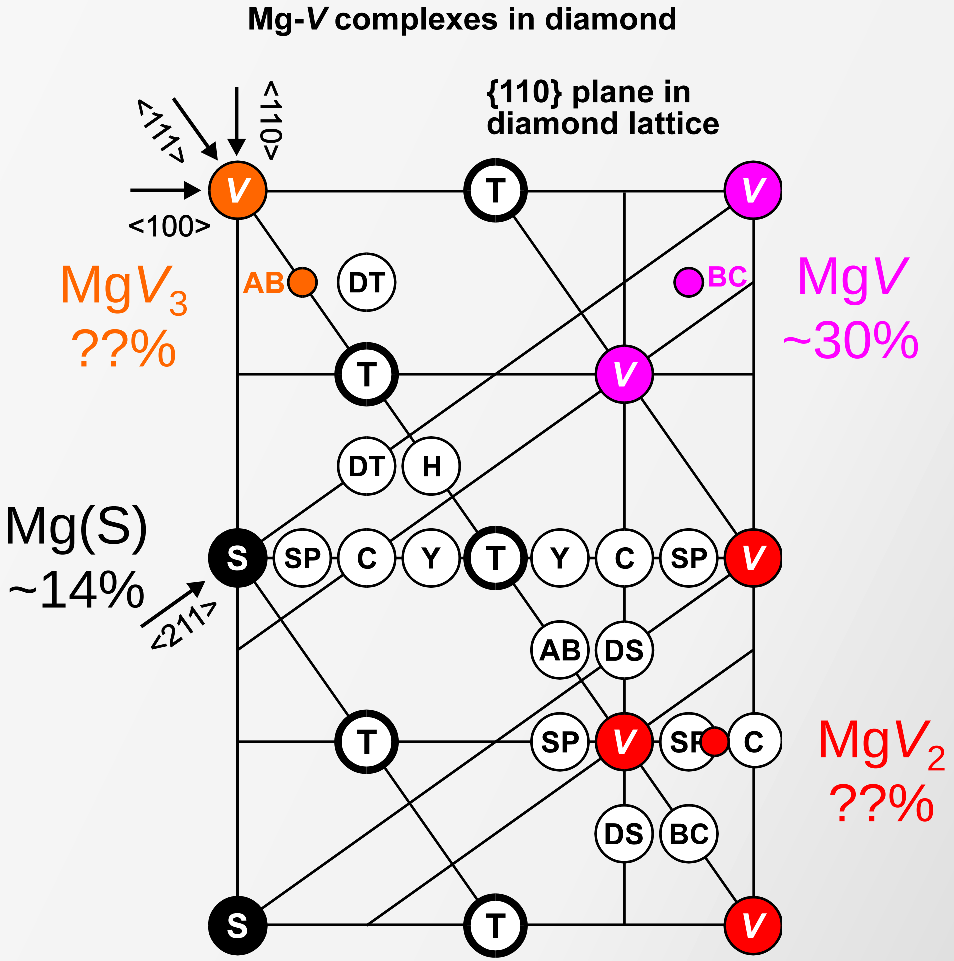


EC characterization of ^{27}Mg colour centers in diamond (800°C)



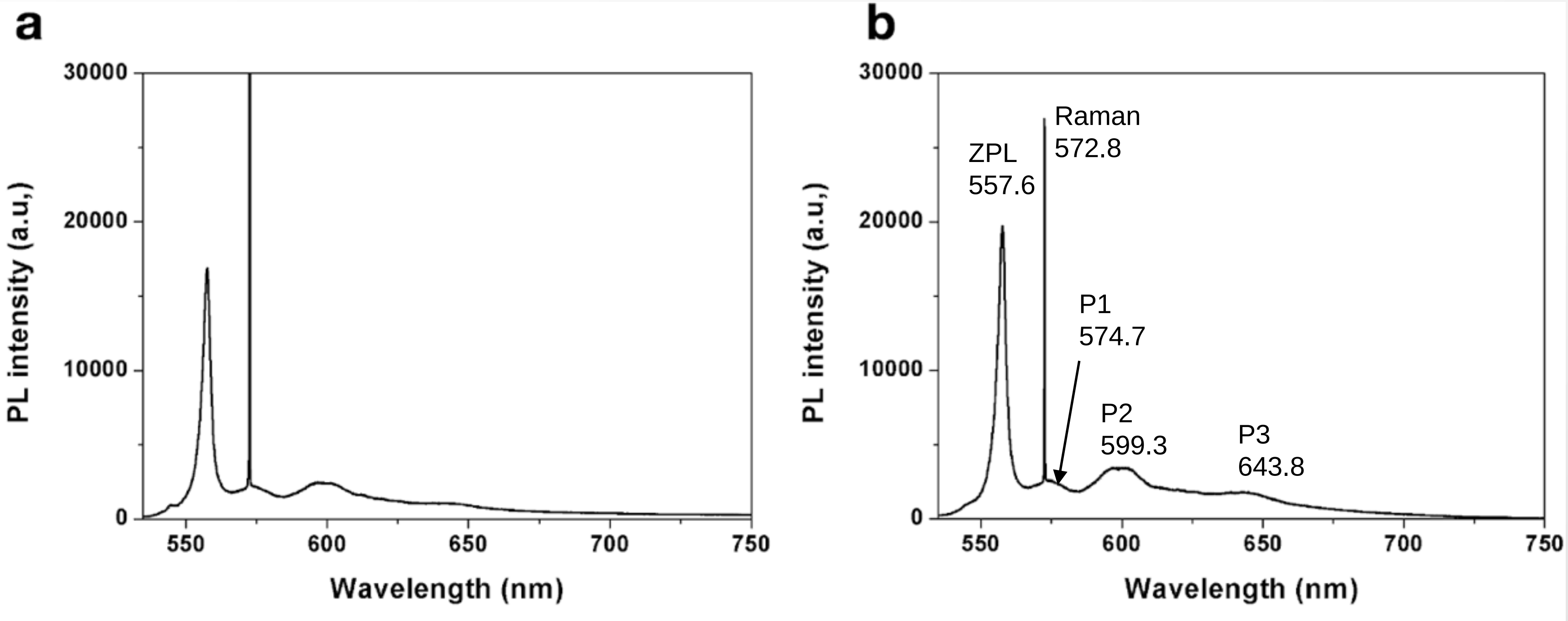
T_i=800°C

- EC from 800°C implanted ^{27}Mg show 14% on S and 30% on bond-center (BC) sites
- 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



PL from ^{26}Mg implanted in diamond at ISOLDE

- Stable ^{24}Mg implanted into "electronic-grade" diamond ([N]<5 ppb) at ISOLDE, 30 keV, 1×10^{12} cm^{-2} (takes 30 s ☺)
- Excitation with 532 nm laser shows ZPL from ensemble of Mg*V* centers at 557.6 nm, as well as characteristic phonon side bands P1, P2, P3 (measured at U Turin).
- ZPL is observed with narrow FWHM (3.4 nm) 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) [7].



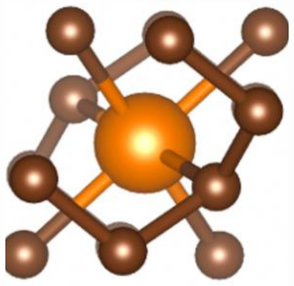
Implanted at T_i = RT, annealed 20 min at T_a=800°C

Implanted at T_i=800°C

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

FWHM of ZPL is a measure of structural quality of Mg*V* centers

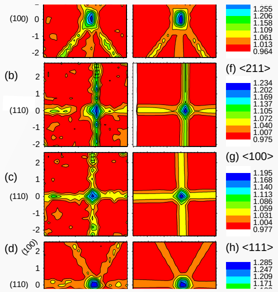
Conclusions



Structural formation yield

Implanted ^{27}Mg shows a surprisingly high structural yield of formation of Mg*V* complexes: 30-40%

However, the major part of Mg is most likely in MgV_2 and MgV_3 complexes of more complicated structure.

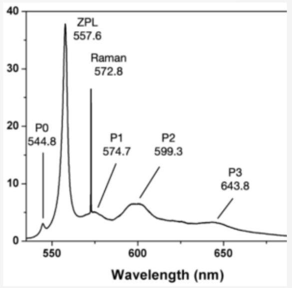


Formation mechanism

Formation of Mg*V* takes place immediately following implantation during cool-down of implantation cascade.

No annealing is needed in order to drive vacancies towards Mg!

(in stark contrast to the formation mechanism of N*V*).



Role of annealing

Thermal annealing serves…

- to restore the optical properties of implanted diamond,

- to ***remove*** implantation defects from the neighbourhood of the Mg*V* complexes.

Very simple production process