

# Spectral Emission Dependence of Tin-Vacancy Centers in Diamond from Thermal Processing and Chemical Functionalization

Emilio Corte, Selene Sachero, Sviatoslav Ditalia Tchernij,\* Tobias Lühmann, Sébastien Pezzagna, Paolo Traina, Ivo Pietro Degiovanni, Ekaterina Moreva, Paolo Olivero, Jan Meijer, Marco Genovese, and Jacopo Forneris\*

A systematic photoluminescence (PL) investigation of the spectral emission properties of individual optical defects fabricated in diamond upon ion implantation and annealing is reported. Three spectral lines at 620, 631, and 647 nm are identified and attributed to the SnV center due to their occurrence in the PL spectra of the very same single-photon emitting defects. It is shown that the relative occurrence of the three spectral features can be modified by oxidizing the sample surface following thermal annealing. The relevant emission properties of each class of individual emitters, including the excited state emission lifetime and the emission intensity saturation parameters are reported.

## 1. Introduction

Diamond-based color centers are appealing candidates as solid-state single-photon sources for applications in emerging fields of quantum technologies, such as quantum computing, quantum information, and quantum sensing.<sup>[1–5]</sup> A class of group-IV-

related quantum emitters, fabricated upon Sn ion implantation, thermal annealing, and surface oxidation was recently discovered.<sup>[6,7]</sup> These color centers exhibit intriguing opto-physical properties, such as high emission rate, narrow spectral width, and emission concentrated in the zero-phonon line (ZPL), thus sparking the interest of the scientific community toward practical applications in the field of quantum technologies,<sup>[4]</sup> including quantum sensing,<sup>[8]</sup> nanophotonics,<sup>[9]</sup> and spintronics.<sup>[10,11]</sup>

The characteristic spectral feature of the optical activity of the SnV center consists of

a strong room-temperature ZPL emission at 620 nm, which is widely attributed to the negative charge state of the defect (SnV<sup>-</sup>) based on the convincing support of independent theoretical works based on *ab initio* simulations.<sup>[6,11,12]</sup> In addition, multiple works have independently observed additional emission lines at 593 nm,<sup>[6,7,13,14]</sup> 631 nm,<sup>[7,14,15]</sup> 647 nm,<sup>[6–8,13–15]</sup> and 663 nm.<sup>[12,14,15]</sup>

While the 663-nm line has been convincingly interpreted as a radiation-induced defect,<sup>[12,15]</sup> the attribution of the remaining photoluminescence (PL) peaks is still uncertain and requires further disambiguation. DFT simulations have highlighted the possibility to observe the SnV center in different charge states,<sup>[11,16]</sup> however a conclusive model of the experimental findings according to this interpretation has not been achieved yet. Recent reports on this subject suggest that the aforementioned emission lines are originating from different Sn-containing lattice complexes.<sup>[6,8,9]</sup> In these works the defects are created by ion irradiation, and different experimental conditions (ion implantation energy, annealing temperature and duration, and chemical termination of the latter) are explored. In particular, it is worth noting that the ion irradiation parameters result in different depth distributions of the defects, as well as their distance from the surface. Remarkably, high-pressure/high-temperature annealing processing performed without specific additional surface treatments resulted in the observation of the sole 620 nm emission line.<sup>[6]</sup> Moreover, the excitability of the 647 nm line under 633 nm laser excitation provided a strong experimental indication that such spectral feature cannot be attributed to the negative charge state of the SnV defect (620 nm ZPL).<sup>[15]</sup>

Finally, the electrical control of the Fermi level of an O-terminated p-i-p diamond junction has shown that all the

E. Corte, S. Ditalia Tchernij, P. Olivero, J. Forneris  
Dipartimento di Fisica, Università degli Studi di Torino, Istituto Nazionale di Fisica Nucleare (INFN), Sez. Torino  
Istituto Nazionale di Ricerca Metrologica (INRiM)  
via P. Giuria 1, 10125 Torino, Italy  
E-mail: sviatoslav.ditaliatchernij@unito.it; jacopo.forneris@unito.it

S. Sachero  
Dipartimento di Fisica  
Università degli Studi di Torino  
via P. Giuria 1, 10125 Torino, Italy

T. Lühmann, S. Pezzagna, J. Meijer  
Applied Quantum Systems, Felix-Bloch Institute for Solid-State Physics  
Universität Leipzig  
Linnéstraße 5, 04103 Leipzig, Germany

P. Traina, I. P. Degiovanni, E. Moreva, M. Genovese  
Division of Quantum Metrology and Nanotechnologies  
Istituto Nazionale di Ricerca Metrologica (INRiM)  
Strada delle Cacce 91, 10135 Torino, Italy

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adpr.202100148>.

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