Minh Anh Phan Nguyen

# Creation and Manipulation of Quantum Emitters in Solid-State Materials

Thesis submitted in fulfilment of the requirements for the degree of Doctor of Philosophy

*Faculty of Science*
School of Mathematical and Physical Sciences
*Primary Supervisor:*
Prof. Igor Aharonovich
*Co-supervisors:*
Prof. Milos Toth
Dr. Sejeong Kim
Dr. Carlo Bradac
Dr. Mehran Kianinia

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# Certificate of Original Authorship

I, Minh Anh Phan Nguyen, declare that this thesis, is submitted in fulfilment of the requirements for the award of Doctor of Philosophy, in the School of Mathematical and Physical Sciences at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise referenced or acknowledged. In addition, I certify that all information sources and literature used are indiciated in the thesis.

This document has not been submitted for qualifications at any other academic institution.

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Signature of Student:

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# Contributing publications

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# Abbreviations

| | |
|---|---|
| 2D | Two-Dimensional |
| AFM | Atomic Force Microscope |
| APD | Avalanche Photodiode |
| BFP | Back-Focal Plane |
| CL | Cathodoluminescence |
| CVD | Chemical Vapour Deposition |
| CW | Continuous Wave |
| DFT | Density Function Theory |
| DW | Debye-Waller |
| ELOG | Epitaxially-Laterally Overgrown |
| EMCCD | Electron-Multiplied Charged Coupled Device |
| eV | Electron Volts |
| FDTD | Finite-Difference Time-Domain |
| FWHM | Full-Width Half-Maximum |
| FT | Fourier-Transform |
| GeV | Germanium Vacancy |
| GSD | Ground-State Depletion |
| HAADF | High-Angle Annular Dark-Field |
| hBN | Hexagonal Boron Nitride |
| HDD | High Density of Dislocations |
| HPHT | High-Pressure High-Temperature |
| HVPE | Hydride Vapour Phase Epitaxy |
| IPA | Isopropyl Alcohol |
| IRF | Instrument Response Function |
| LDD | Low Density of Dislocations |
| LDOS | Local Density of States |

| | |
|---|---|
| LSP | Localised Surface Plasmon |
| MBE | Molecular Beam Epitaxy |
| MOCVD | Metal-Organic Chemical Vapour Deposition |
| MOVPE | Metal-Organic Vapour Phase Epitaxy |
| NA | Numerical Aperture |
| ND | Nanodiamond |
| NV | Nitrogen Vacancy |
| $N_VB_N$ | Anti-site Nitrogen Vacancy in hBN |
| PAMBE | Plasma-Assisted Molecular Beam Epitaxy |
| PSB | Phonon-Sideband |
| QE | Quantum Efficiency |
| SiC | Silicon Carbide |
| SMLM | Single-Molecule Localisation Microscopy |
| SNR | Signal-to-noise |
| SPE | Single-Photon Emitter |
| SPP | Surface Plasmon Polariton |
| SPS | Single-Photon Source |
| TCSPC | Time-Correlated Single Photon Counting |
| TD | Threading Dislocation |
| TEM | Transmission Electron Microscope |
| Ti:SAP | Titanium Sapphire |
| TMDC | Transition-Metal Dichalcogenides |
| UV | Ultra-violet |
| VLS | Vapour-Liquid-Solid |
| ZPL | Zero-Phonon Line |

# Abstract

Single-photon emitters are considered as a fundamental building block upon which many quantum-based applications are established. Of the many solid-state quantum emitters discovered, there exists three which garnered an increasing interest over the past few years; gallium nitride (GaN), germanium vacancy (GeV) colour centres in diamond, and quantum emitters in hexagonal boron nitride (hBN). Each of these solid-state emitter systems have unique advantages, making them intriguing candidates for quantum applications. However, there is still much to be understood regarding their optical properties and origin. Thus, the focus of this thesis is then established—to understand the origins of these solid-state sources through systematic studies of their growth and fabrication, followed by how they interact with the surrounding environment, and finally the modification of these interactions upon the addition of nanophotonic architectures.

Two separate studies were done on quantum emitters in GaN. First, the effects of microstructure and growth mechanics on the formation of emitters in GaN were investigated through multi-spectroscopic analysis in a systematic study of various material properties. No observable correlation was recorded, suggesting the origin of emitters was of an extrinsic nature, rather than intrinsic. The second study detailed the characterisation of the optical properties of GaN SPEs through resonant excitation, approaching Fourier-transform-limited linewidths of ~250 MHz—the narrowest reported for these emitters.

Next, a determination of the quantum efficiency (QE) of GeV colour centres in nanodiamond was performed by measuring and comparing radiative emission rates in a changing dielectric environment. Combined with Fourier-plane imaging of the resulting emission

patterns, a quantum efficiency of 22% was calculated from ensembles, several times higher than the SiV colour centre.

Finally, two separate studies on hBN SPEs were performed—the first study demonstrating the creation of emitters with high-energy electron irradiation. In this study, different hBN multilayer and monolayer flakes were irradiated with electrons in the megaelectronvolt regime, resulting in emitter creation within the flat regions of the hBN flakes, areas not seen in prior methods. The second study details the hybridisation of hBN emitters with plasmonic nanospheres, assembled via an atomic force microscope. An enhancement resulted in a maximum count rate of approximately 5.8 M counts/second, with the linear transition dipole exploited to maximise coupling to the nanospheres.

All of these studies serve to highlight the unique properties of their respective material systems, and further their development towards reliable integration with fundamental nanophotonic devices for applications in quantum information science.