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Magneto-Optical Properties of Quantum Emitters in Diamond

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14. ABSTRACT An active search for new emitters in large bandgap materials such as diamond or hexagonal Boron-Nitride has taken place over the last years with the purpose of enabling novel quantum applications. One of the methods for creating defects in hexagonal Boron Nitride consists of ion implantation with an ion gun or indirect ion implantation, technique in which an accelerated ion hits an atom in rest, which in turn is implanted into the matrix. Towards this goal the project had mounted an experimental setup for implanting defects into large bandgap materials. A master's in physics student accomplished this task. Once a defect is created, the characterization of its optical and magnetic properties is required. These properties are highly modulated by the interaction between the defect and the vibrations of the lattice in which it is embedded. In this regard, we have modeled the interaction between phonons and the electronic spin 1/2 associated to defects founding a remarkable electron spin resonance suppression, an effect hypothesized in the past but never estimated. In addition, we have modeled how nearby nuclear spins to the electronic spin associated to the nitrogen-vacancy color center can be polarized using several methods and most importantly how these polarization schemes are affected by non-radiative transitions.			
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Abstract:	<p>An active search for new emitters in large bandgap materials such as diamond or hexagonal Boron-Nitride has taken place over the last years with the purpose of enabling novel quantum applications. One of the methods for creating defects in hexagonal Boron Nitride consists of ion implantation with an ion gun or indirect ion implantation, technique in which an accelerated ion hits an atom in rest, which in turn is implanted into the matrix. Towards this goal the project had mounted an experimental setup for implanting defects into large bandgap materials. A master's in physics student accomplished this task. Once a defect is created, the characterization of its optical and magnetic properties is required. These properties are highly modulated by the interaction between the defect and the vibrations of the lattice in which it is embedded. In this regard, we have modeled the interaction between phonons and the electronic spin $\frac{1}{2}$ associated to defects founding a remarkable electron spin resonance suppression, an effect hypothesized in the past but never estimated. In addition, we have modeled how nearby nuclear spins to the electronic spin associated to the nitrogen-vacancy color center can be polarized using several methods and most importantly how these polarization schemes are affected by non-radiative transitions. The project has involved 10 international and national collaborators co-authoring these works, PhD and Master and Bachelor students. The project is framed in an interdisciplinary research for combining single emitters with topological materials with research on the experimental and theoretical fronts. Future single emitters that will be considered are carbon substitutional and carbon trimmers in hexagonal Boron-Nitride as this material can be readily placed in close proximity to several topological insulators, constituting an ideal platform for studying the optoelectronics coupling between them.</p>
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Archival Publications:	<p>Third period publications:</p> <ul style="list-style-type: none"> ■ Sensing Electrochemical Signals Using a Nitrogen-Vacancy Center in Diamond, Hossein T. Dinani, Enrique Muñoz and Jeronimo R. Maze, <i>Nanomaterials</i> 11, 358 (2021). https://doi.org/10.3390/nano11020358 ■ M. Block, B. Kobrin, A. Jarmola, S. Hsieh, C. Zu, N.L. Figueroa, V.M. Acosta, J. Minguzzi, J.R. Maze, D. Budker, and N.Y. Yao, Optically Enhanced Electric Field Sensing Using Nitrogen-Vacancy Ensembles, <i>Physical Review Applied</i> 16 (2021) url ■ Effect of intersystem crossing rates and optical illumination on the polarization of nuclear spins close to nitrogen-vacancy centers, <i>Physical Review B</i> 103, 195443 (2021), http://doi.org/10.1103/PhysRevB.103.195443 <p>Second period publications:</p> <ul style="list-style-type: none"> ■ Ariel Norambuena, Jerónimo R. Maze, Peter Rabl, and Raúl Coto, Quantifying phonon-induced non-Markovianity in color centers in diamond, <i>Physical Review A</i> 101, 022110 (2020). https://10.1103/PhysRevA.101.022110 ■ Ariel Norambuena , Alejandro Jimenez, Christoph Becher and Jerónimo R Maze, Effect of phonons on the electron spin resonance absorption spectrum, <i>New J. Phys.</i> 22 (2020) 073068, https://doi.org/10.1088/1367-2630/ab9da0 <p>First period publications:</p> <ul style="list-style-type: none"> ■ Bayesian estimation for quantum sensing in the absence of single-shot detection, Hossein T. Dinani, Dominic W. Berry, Raul Gonzalez, Jeronimo R. Maze, and Cristian Bonato, <i>Physical Review B</i> 99 (2019) url ■ Functionalization of stable fluorescent nanodiamonds towards reliable detection of biomarkers for Alzheimers disease, Francisco Morales-Zavala et al., <i>Journal of Nanobiotechnology</i> 16 (2018), https://doi.org/10.1186/s12951-018-0385-7

Changes in Research objectives:	
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Change in AFOSR Program Manager, if any:	
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Collaborators:	<p>Researchers (co-authoring published works):</p> <p>Hossein Dinani, Centro de Investigación DAiTA Lab, Facultad de Estudios Interdisciplinarios, Universidad Mayor, Santiago, Chile. Role: Modelled dynamics of electronic spins and nuclear spins in diamond.</p> <p>Vincent Jacques, Laboratoire Charles Coulomb, Université de Montpellier and CNRS, Montpellier, France. Role: scientific collaboration on the dynamics of electronic and nuclear spins in diamond</p> <p>Dominic Berry, Department of Physics and Astronomy, Macquarie University, Sydney, Australia. Role: scientific discussion on improving sensing of defects in diamond</p> <p>Cristian Bonato, Institute of Photonics and Quantum Sciences, SUPA, Heriot-Watt University, United Kingdom. Role: Scientific discussion on improving sensing of defects in diamond</p> <p>Norman Yao, Department of Physics, University of California, Berkeley, USA. Role: scientific discussion on describing optical dynamics of defects in diamond.</p> <p>Dmitry Budker, Helmholtz Institut Mainz, Johannes Gutenberg Universitat Mainz, Germany. Role: scientific discussion on describing optical dynamics of defects in diamond.</p> <p>Andrey Jarmola, U.S. Army Research Laboratory, Adelphi, Maryland, USA. Role: Scientific discussion on describing optical dynamics of defects in diamond.</p> <p>Ariel Norambuena, Centro de Óptica e Información Cuántica, Facultad de Ciencias, Universidad Mayor, Santiago, Chile. Role: scientific discussion on modelling interaction of defects in diamond and phonons.</p> <p>Raul Coto, Centro de Investigación DAiTA Lab, Facultad de Estudios Interdisciplinarios, Universidad Mayor, Santiago, Chile. Role: scientific discussion on modelling interaction of defects in diamond and phonons.</p>
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	<p>Peter Rabl, Vienna Center for Quantum Science and Technology, Atominstitut, TU Wien, 1040 Vienna, Austria. Role: scientific discussion on modelling the interaction of defects with phonons.</p> <p>Christoph Becher, Universität des Saarlandes, Fachrichtung Physik, Saarbrücken, Germany. Role: scientific discussion on modelling interaction of defects with phonons.</p> <p>Students:</p> <p>Enrique Rodriguez, PhD Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: characterize single emitters experimentally.</p> <p>Alejandro Jimenez, Master in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: modelled interaction of single emitters with phonons.</p> <p>Arturo Reyes, Master in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: built setup for implanting ions in diamond.</p> <p>Vicente Santibañez, Master in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: optical characterized showllow single emitters.</p> <p>Loreto Martinez, Master in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: constructed optical setup for analyzing single emitters.</p> <p>Simone Crozier, Bachelor in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: built setup for studying the charge dynamics of single emitters.</p> <p>Raul Gonzalez, Master in Physics student, Institute of Physics, Pontificia Universidad Catolica de Chile, Santiago, Chile. Role: built optical setup for characterizing the electronic spin resonance of defects in diamond</p>
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AFOSR program officer:	Geoffrey Andersen, Technical Director
Extensions granted or milestones slipped, if any:	

Accomplishments

- **Research Objectives:**
The main research goal of the project is to find and characterize the magnetic or spin degrees of freedom as well as the optical properties of new color centers primarily but not exclusively in diamond and other materials with large bandgaps for applications in quantum information and quantum metrology assessing the potential use of electronic and nuclear spin degrees of freedom.

The main technical approach was to build a setup able to implant ions into large bandgap materials such as diamond, and hBN in order to create defects. In parallel, the project had a theoretical component in charge of modelling the interaction of defects with phonons and in particular the interaction between electronic spins associated to the defects and nearby nuclear spins. The creation and use of the implantation system constitute a major milestone of the project at the end of the first year. This milestone was 100% complete on time but used few times before the pandemic started. Right now, it is fully operative, and a new Master student will operate on December 2021st. Another milestone of the project was the creation of a setup able to control the magnetic field orientation up to 700 Gauss for the third and last year of the project. The completion is 80% and it will be completed on January 2022nd. On the theoretical front several works were completed as we describe below.

- **Details of accomplishments:**
 - Scientific publications: 7 publications in peer reviewed journals.
 - These completed: 4 master students and one doctorate student.
 - Theses under supervision: 3 doctorate students.
 - Funded AFOSR collaborative project between RMIT (Melbourne Australia) and PUC (Santiago, Chile).
- **Dissemination activities:**
 - Dissemination of results in a Window of Science funded by AFOSR (Geoffrey Andersen) to visit RMIT, Melbourne, Australia in February 2020. This visit led to a new collaborative project entitled “Diamond scanning probe for characterization of topological insulators” granted by AFOSR (FA2386-21-1-4125) on which participates the researchers Brant Gibson, Andrew Greentree from RMIT, and Jerónimo Maze from PUC (Chile).

Impacts

Development of the principal discipline(s) of the project

The successful development of quantum applications relies on the identification of

systems at the nanoscale that can be highly isolated or unperturbed from its nearby environment to preserve coherence: the ability to maintain the phase of a quantum state, for example, a given superposition of spin up and spin down of an electronic spin. However, accessing such state for later use necessarily requires interacting with the system that hosts it. Therefore, a complex trade of between accessibility and isolation is inevitably present. In this regard, defects in large bandgap materials offer an on demand optical accessibility and can be thought as trapped ions or molecules in solids, almost textbook systems trapped by nature. Almost, as a major price must be paid, which is their interaction with phonons of the hosting material, or vibrations that are constantly heating the nearby environment of the system and slowly degrading its quantum properties. However, even this undesired interaction must obey to the rules set by the host material and the system such as the symmetry of the trapped molecule. A detailed understanding on the interaction of these systems with vibrations might allow clever ways to design and engineer trapped molecules to preserve coherence. In this project we have understood how the symmetry of the system contributes to maintain coherence and how the interaction with vibrations can affect the internal dynamics such as non-radiative transitions and its consequence on the manipulation of quantum state associated to electronic and nuclear spins of these systems.

Other disciplines:

The research field on this project required a close collaboration with national researchers in the field of chemistry.

Impact on the development of human resources:

The project offered new research opportunities for several students who developed their theses among the research lines of the proposal. The following is a list of completed and under development theses.

• Theses completed:

1. Arturo Reyes, Master in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Creación de defectos ópticos poco profundos en sólidos por implantación de iones” (“Creation of shallow optical defects in solids by ion implantation”), March 2020.
2. Santibañez Cisternas, Vicente Andres. Master in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Chemical Reaction Sensor Based on NV-Centers”, 2021.
3. Martinez Prieto, Loreto Alejandra. Master in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Dynamics of optically trapped magnetic particles“, 2021.
4. Jiménez Contreras, Alejandro Adrián. Master in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Analysis of the effect of strain in negatively charged nitrogen-vacancy centers through effective Hamiltonians“, 2021.

5. Duarte Portilla, Hector Andres. Doctorate in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Polarización de Espines Nucleares en diamante con Nitrógeno-Vacante“, 2021.
- Theses under supervision:
 6. Escalante De Posada, Richard Albert. Doctorate in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Single Molecule Magnets for Spintronics and Quantum Information“.
 7. Rodríguez Jiménez, Enrique Arturo. Doctorate in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Nano-magnetometry with a Nitrogen vacancy centre in Diamond”
 8. Waselowski Butikofer, Victor Hugo Daniel. Doctorate in Physics, Institute of Physics, Pontificia Universidad Católica de Chile. Title: “Applications in biology and materials science of single-shot read-out using defects in diamond“.

Impact on teaching and educational experiences:

The project implied to train student in the field of group theory for describing the dynamics of the defects or systems that were created. Toward this goal a new section entitled “Group Theoretical Approach for describing molecules” was inserted in the program of the course FIM8340 Atomic and Molecular physics.

Impact on physical, institutional, and information resources that form infrastructure:

The experimental part of the project and the actual pandemic condition forced us to develop online access to our experiments. This knowledge was used to implement online and remote access to experimental setups for a new experimental course entitled “Experimental Physics III”. The course included experiences on modern physics and wave phenomena such as electric spin resonance and measuring the speed of light via heterodyne detection.

Impact on society beyond science and technology:

Nothing to report.

Changes

Changes in approach

Nothing to report.

Problems or delays

Due to the actual pandemic situation, the project experienced problems to progress on the

experimental side as students and PI could not access to the laboratories over several months. As a workaround to this problem, the group developed tools to control the setups remotely and minimize the number of times that was required to be present at the lab. Although this was useful, did not apply to all our setups and just half of them could take advantage of this modality.

Expenditure Impacts

Nothing to report.

Significant changes in the use or care of human subjects, vertebrate animals and/or biohazards

Nothing to report.

Changes to the primary place of performance from that originally proposed

Nothing to report.

Technical Updates

- Current Progress on creating single emitters

An experimental setup was prepared for implanting Nitrogen ions on large bandgap materials. First implantations were performed early on 2020 (January and February) on diamond using a home-made setup shown on the figure below.

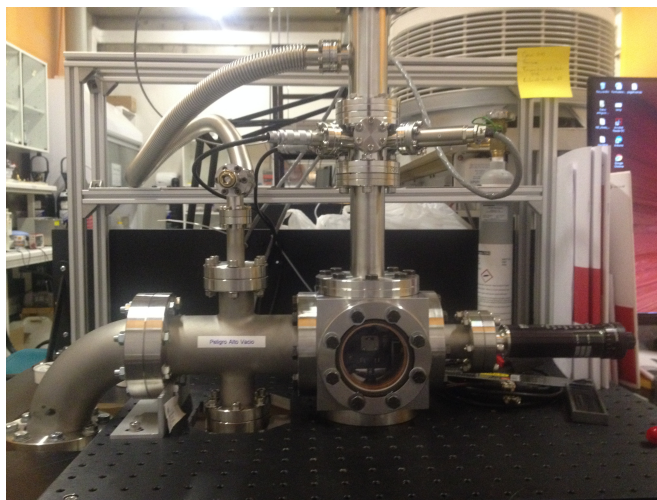
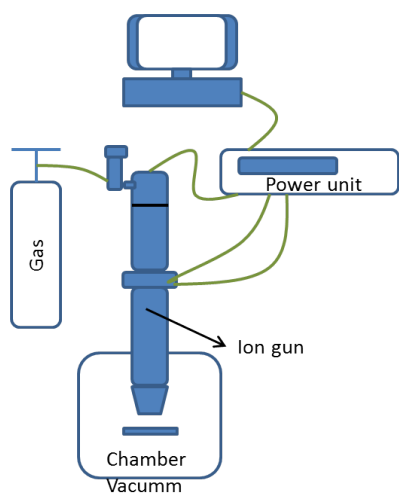


Figure 1. Left: Schematic of an ion gun for implanting ions with low energy. It can implant ions with energy between 0 and 5000 eV under vacuum (10^{-6} mbar). Right: a picture of the implemented setup.

The sample was annealed at 800 Celsius for two hours and then analyzed in a confocal microscope connected to an optical spectrometer. The figure below summarized the work performed by Arturo Reyes, a Master in Physics student graduated in March 2020.

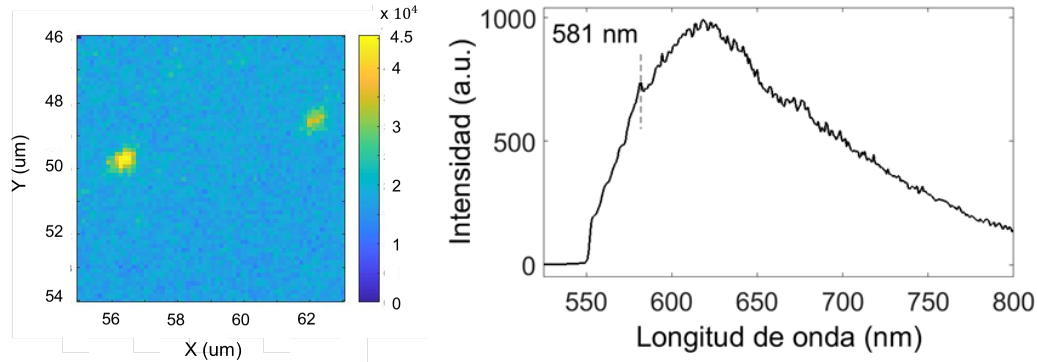


Figure 2. Left: Confocal Image showing two bright spots belonging to the same focal plane. No other bright spots were found at different focal planes. Right: Emission spectrum under green excitation of a defect found in diamond after implantation and annealing. The signature at 581 nm corresponds to a yet unclassified center in diamond.

- Undergoing collaborations

1. Brant Gibson, Andrew Greentree, RMIT, Melbourne, Australia, on studying the magnetic sensitivity of ensemble samples and the coupling between single emitters and topological materials.
2. Patrick Maletinsky, Basel University, Switzerland, on describing the excited state structure of NV centers at high magnetic fields and high strain.
3. Shimon Kolkowitz, Wisconsin University, on studying the charge migration under optical illumination in diamond with ensemble of nitrogen-vacancy defects and nitrogen impurities.
4. Enrique Muñoz (PUC), Carolina Parra (UTFSM), Loik Gence (PUC), Francisco Muñoz (UCH), Rodrigo Soto (PUC), on Light-Matter Interactions in Topological Nanomaterials towards Low-Consumption Energy Devices. Grant: ANID-ACT192023. From 2019 to 2022.

A current collaboration with Brant Gibson and Andrew Greentree (RMIT) aims to characterize topological materials with color centers. Topological materials have unique boundary conditions which mixes electric and magnetic fields. The following figure (left panel) shows the magnetic field produced by a topological material because of applying an electric field with a conductive sphere. Right panel shows the optically detected magnetic resonance of nitrogen-vacancy centers indicating a highly sensitive operation point on which a small external magnetic field can be detected. This collaboration aims to explore the electromagnetic fields at the surface of novel topological insulators.

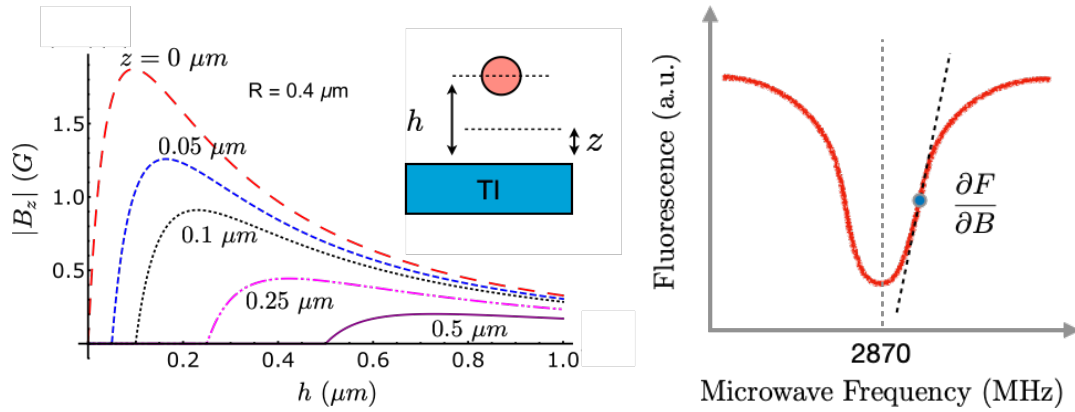


Figure 3. Left: Magnetic field emanated by a topological insulator as a response to the electric field produced by a conductive sphere. Image adapted from Ref: A. Martin-Ruiz et al, *Physical Review A*, 100, **042124** (2019). Right: Optically detected magnetic resonance of nitrogen-vacancy centers showing the operation point for detecting the magnetic field shown on the right panel.