Introduction

Quantum mechanics was first developed at the beginning of the 20th century to explain physical observations such as the blackbody radiation [1] or the photoelectric effect [2]. The quantum theory was subsequently developed in the first half of the 20th century and showed great success, in particular in giving a physical framework for chemistry [3] and leading to a microscopic understanding of matter [4]. In the second half of the 20th century, technological development based on the new understanding of light and matter have led to what was retroactively called the first quantum revolution [5]. Technologies developed in the 50s and 60s include the semiconductor transistor [6], the laser [7], and nuclear magnetic resonance [8] which have since revolutionized the fields of information science, global communications and medical imaging.

A new field of quantum technology has emerged in the last two decades. This "second quantum revolution" differs from the first one in the usage of individual quantum systems, such as single photons [9], atoms [10] or electrons [11], which have properties that can not be emulated by larger classical systems. These properties include quantum entanglement, superposition or measurement and form the basis of the newly formed quantum information science [12–14].

Central to quantum information science is the idea of quantum bits or qubits [15], which are the building blocks of quantum information in analogy to the bits of classical information theory. Physical implementation of a qubit could be any quantum system with two well defined quantum states, as long as these states can be initialized, manipulated and readout [16]. Popular qubit candidates include superconducting circuits [17, 18], photons [19, 20], quantum dots [21, 22], trapped ions [23, 24] and single crystal defects [25–27].

Defects in large band gap crystals are sometimes referred to as artificial atoms [28], as the localized wave function of the electrons surrounding the impurity give rise to discrete energy levels similar to that of an atom. Of all the possible crystals and defects, the most studied defect for quantum applications is the negatively charged nitrogen-vacancy center in diamond [29, 30], often abbreviated to "NV center". Although this defect had been known and correctly identified almost 50 years ago [31], interest in the NV center really sparked after it was first isolated in 1997 [32]. The detection of single NV centers proved that NV centers fluoresce brightly [32], are photostable [33], and that their electronic spin can be polarized and readout optically [34], making them an promising solid state qubit.

Since then, single NV centers have been successfully used to create quantum entanglement over several km [35], to store quantum information at room temperature for more than one second [36], to detect single proteins via nuclear magnetic resonance [37], and for many other applications in quantum communication [38], quantum computing [30] or quantum sensing [39].

As the interest in single NV center grew, diamond synthesis improved significantly [40–42] which benefited not only single NV centers but also NV center ensemble. While single NV center are required for most quantum information processes [43], several other applications benefit from a greater concentration of defects and are therefore more suited to operate with NV centers ensemble. Four of these applications are pictured in Fig. 1.

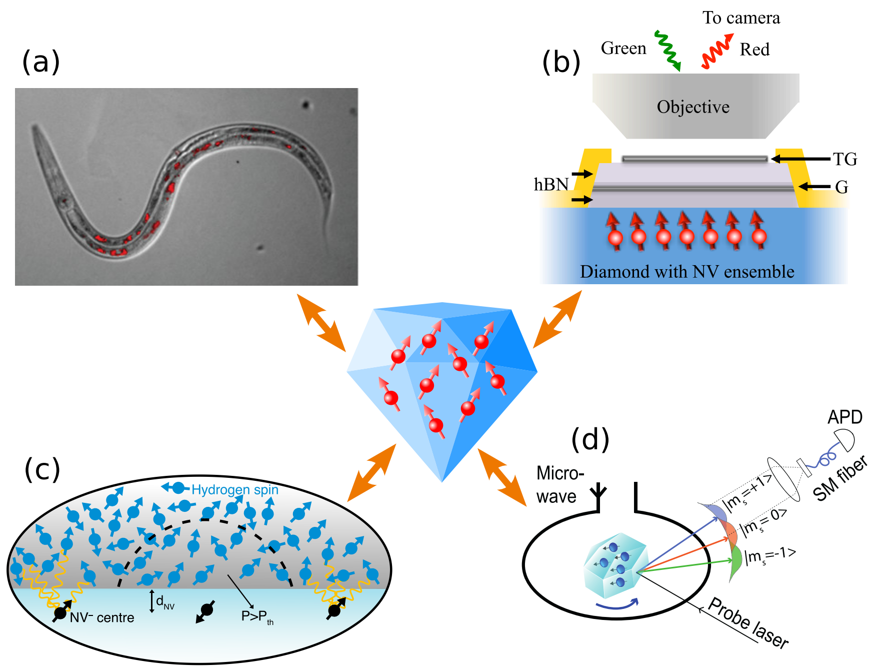

Figure 1: Example of applications with dense ensemble of NV centers. (a) In vivo nanodiomond biomarkers (from [44]). (b) Quantum sensing with NV ensemble (from [45]). (c) Dynamic nuclear polarization of an external spin bath (from [46]). (d) Spin mechanics with trapped micro diamonds (from [47]).

The first application for NV center ensemble is the use of nano or micro diamonds as fluorescent biomarkers [44, 48]. NV centers are good candidates for biomarkers as they emit a bright and stable photoluminesecence, and because diamond nanocrystals present low toxicity [49]. Some applications also use the spin degree of freedom of the NV centers, either to perform background free fluorescence microscopy [50] or to monitor the angular motion of the nanodiamonds [51, 52]. For most of these applications, NV ensemble provide a better visibility than single NV centers and are therefore preferred.

The second NV ensemble application is for quantum sensing [39]. NV centers are predominantly used as magnetic field sensors [41, 53], although they can also detect electric fields [54, 55], temperature [56, 57] ,crystal strain [58, 59] or rotation [60, 61]. Both single and ensemble of NV centers can be used for sensing: single centers offer unmatched spatial resolution [62] down to a few nm [37], while ensemble provide better sensitivities [41, 63].

The third NV ensemble application is nuclear magnetic resonance (NMR) dynamical nuclear polarization [64]. Dynamic nuclear polarization consists in increasing the nuclear polarization of a sample by transferring electron polarization to the nuclei bath [65]. Because the NV electronic spin can be optically polarized at room temperature, micro and nano diamonds containing NV centers are promising candidates as hyperpolarization agents [66]. Nuclear polarization of spins inside the diamond lattice was demonstrated with single NV centers [67, 68] and NV ensemble [69–71]. Polarization transfer to nuclear spins outside of the diamond is an ongoing challenge [46, 64], but dense NV ensemble are believed to be necessary to achieve high external polarization [66, 72].

Finally, the fourth NV ensemble application is the recent field of spin mechanics [73]. Similarly to optomechanics [74], the goal of spin mechanics is to couple a quantum system to the mechanical degree of freedom of a macroscopic object. Strong coupling between single NV center and an external mechanical oscillators has been observed [75, 76], but coupling between NV centers and the motion of the diamond has so far only been achieved with dense NV ensemble [47, 77, 78]. Coupling several NV centers to the same mechanical oscillator could also lead to interesting magnetic phase transitions [79, 80].

Most of the NV ensemble experiments discussed previously work under the assumption that the ensemble is constituted of N independent single NV centers. However, as often in physics, *more is different* [81]. Interaction between the NV center themselves or with their environment can lead to significantly different behavior between ensemble and single spins. These difference include a modification of the charge state dynamics [82] and of the spin dynamics [83–86], as well as the apparition of exotic phenomena such as time crystal phase [87], Anderson localization [88] or superradiance [89, 90].

The work presented in this manuscript focuses on dipolarly coupled dense ensemble of NV centers. More specifically, we look at spin polarization exchange between the NV centers themselves and with their environment. Polarization exchange (flip-flop, spin diffusion, cross-relaxation, ...) in dense ensemble of spins has been abundantly studied in the context of NMR [65] and spintronics [91]. Because NV centers tend to be relatively dilute, the question of spin exchange between between NV themselves [86] or with their environment [92] has only recently been addressed. However, as samples with higher NV concentration become available [93–95], these aspects are becoming more and more relevant.

The goal of this present work is both to better understand the physics of dense NV ensemble, which is crucial for many of the NV ensemble applications cited previously, and to exploit some of these many-body properties for new applications.

This manuscript is organized in the following way:

The first chapter introduces the theoretical and experimental concepts used in this manuscript. This includes a brief overview of diamond and NV centers fabrication processes, the physics of single NV centers and a presentation of magnetic dipole-dipole interaction and cross-relaxation.

The second chapter covers the observation of cross-relaxation between NV centers and other spin impurities in CVD-grown diamond. We use measurements on the NV centers to identify these impurities and to quantify some of their attributes. Most of the results of this chapter have been published in [96] and [97].

The third chapter focuses on the dipole-dipole mediated spin relaxation within dense ensemble of NV centers. Following the work presented in [86], we introduce the concept of NV-flutcuator to explain this phenomenon and present experimental and theoretical results solidifying this hypothesis. The results of this chapter have been partly published in [98] and [77].

Finally, the fourth chapter presents a study on NV-NV dipolar relaxation under low magnetic field. This chapter covers both the rich physics of NV centers under low magnetic field, and a potential application of the low field depolarization as a new magnetometry protocol, as well as comparisons with other NV magnetometry protocols. The results of this chapter have in large part been published in [98].

Bibliography

[1] Max Planck. "Zur theorie des gesetzes der energieverteilung im normalspektrum". In: *Berlin* (1900), pp. 237–245.

[2] Albert Einstein. "Über einen die Erzeugung und Verwandlung des Lichtes betreffenden heuristischen Gesichtspunkt". In: (1905).

[3] Linus Pauling. "The nature of the chemical bond. II. The one-electron bond and the three-electron bond". In: *Journal of the American Chemical Society* 53.9 (1931), pp. 3225–3237.

[4] Charles Kittel, Paul McEuen, and Paul McEuen. *Introduction to solid state physics*. Vol. 8. Wiley New York, 1996.

[5] Rob Thew, Thomas Jennewein, and Masahide Sasaki. "Focus on quantum science and technology initiatives around the world". In: *Quantum Sci. Technol* 5.1 (2019), p. 010201.

[6] John Bardeen and Walter Hauser Brattain. "The transistor, a semiconductor triode". In: *Physical Review* 74.2 (1948), p. 230.

[7] Theodore H Maiman et al. "Stimulated optical radiation in ruby". In: (1960).

[8] Anatole Abragam. *The principles of nuclear magnetism*. 32. Oxford university press, 1961.

[9] Sebastien Gleyzes et al. "Quantum jumps of light recording the birth and death of a photon in a cavity". In: *Nature* 446.7133 (2007), pp. 297–300.

[10] W Neuhauser et al. "Localized visible Ba+ mono-ion oscillator". In: *Physical Review A* 22.3 (1980), p. 1137.

[11] Steve Peil and Gerald Gabrielse. "Observing the quantum limit of an electron cyclotron: QND measurements of quantum jumps between Fock states". In: *Physical Review Letters* 83.7 (1999), p. 1287.

[12] Michael A Nielsen and Isaac Chuang. *Quantum computation and quantum information*. 2002.

[13] Vlatko Vedral et al. *Introduction to quantum information science*. Oxford University Press on Demand, 2006.

[14] Masahito Hayashi. *Quantum information.* Springer, 2006.

[15] Benjamin Schumacher. "Sending entanglement through noisy quantum channels". In: *Physical Review A* 54.4 (1996), p. 2614.

[16] David P DiVincenzo. "The physical implementation of quantum computation". In: *Fortschritte der Physik: Progress of Physics* 48.9-11 (2000), pp. 771–783.

[17] Yasunobu Nakamura, Yu A Pashkin, and JS Tsai. "Coherent control of macroscopic quantum states in a single-Cooper-pair box". In: *nature* 398.6730 (1999), pp. 786–788.

[18] TP Orlando et al. "Superconducting persistent-current qubit". In: *Physical Review B* 60.22 (1999), p. 15398.

[19] Charles H Bennett. "Quantum cryptography using any two nonorthogonal states". In: *Physical review letters* 68.21 (1992), p. 3121.

[20] Han-Sen Zhong et al. "Quantum computational advantage using photons". In: *Science* 370.6523 (2020), pp. 1460–1463.

[21] M Veldhorst et al. "An addressable quantum dot qubit with fault-tolerant control-fidelity". In: *Nature nanotechnology* 9.12 (2014), pp. 981–985.

[22] David M Zajac et al. "Resonantly driven CNOT gate for electron spins". In: *Science* 359.6374 (2018), pp. 439–442.

[23] Nicolai Friis et al. "Observation of entangled states of a fully controlled 20-qubit system". In: *Physical Review X* 8.2 (2018), p. 021012.

[24] Kenneth Wright et al. "Benchmarking an 11-qubit quantum computer". In: *Nature communications* 10.1 (2019), pp. 1–6.

[25] Fedor Jelezko and Jörg Wrachtrup. "Single defect centres in diamond: A review". In: *physica status solidi (a)* 203.13 (2006), pp. 3207–3225.

[26] Pavel G Baranov et al. "Silicon vacancy in SiC as a promising quantum system for single-defect and single-photon spectroscopy". In: *Physical Review B* 83.12 (2011), p. 125203.

[27] Manjin Zhong et al. "Optically addressable nuclear spins in a solid with a six-hour coherence time". In: *Nature* 517.7533 (2015), pp. 177–180.

[28] Iulia Buluta, Sahel Ashhab, and Franco Nori. "Natural and artificial atoms for quantum computation". In: *Reports on Progress in Physics* 74.10 (2011), p. 104401.

[29] Igor Aharonovich, Dirk Englund, and Milos Toth. "Solid-state single-photon emitters". In: *Nature Photonics* 10.10 (2016), pp. 631–641.

[30] Nathalie P de Leon et al. "Materials challenges and opportunities for quantum computing hardware". In: *Science* 372.6539 (2021), eabb2823.

[31] Gordon Davies and MF Hamer. “Optical studies of the 1.945 eV vibronic band in diamond”. In: *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences* 348.1653 (1976), pp. 285–298.

[32] A Gruber et al. “Scanning confocal optical microscopy and magnetic resonance on single defect centers”. In: *Science* 276.5321 (1997), pp. 2012–2014.

[33] Christian Kurtsiefer et al. “Stable solid-state source of single photons”. In: *Physical review letters* 85.2 (2000), p. 290.

[34] Fedor Jelezko et al. “Observation of coherent oscillations in a single electron spin”. In: *Physical review letters* 92.7 (2004), p. 076401.

[35] Bas Hensen et al. “Loophole-free Bell inequality violation using electron spins separated by 1.3 kilometres”. In: *Nature* 526.7575 (2015), pp. 682–686.

[36] Peter Christian Maurer et al. “Room-temperature quantum bit memory exceeding one second”. In: *Science* 336.6086 (2012), pp. 1283–1286.

[37] Igor Lovchinsky et al. “Nuclear magnetic resonance detection and spectroscopy of single proteins using quantum logic”. In: *Science* 351.6275 (2016), pp. 836–841.

[38] Stephanie Wehner, David Elkouss, and Ronald Hanson. “Quantum internet: A vision for the road ahead”. In: *Science* 362.6412 (2018), eaam9288.

[39] Christian L Degen, Friedemann Reinhard, and Paola Cappellaro. “Quantum sensing”. In: *Reviews of modern physics* 89.3 (2017), p. 035002.

[40] J Achard, Vincent Jacques, and A Tallaire. “Chemical vapour deposition diamond single crystals with nitrogen-vacancy centres: a review of material synthesis and technology for quantum sensing applications”. In: *Journal of Physics D: Applied Physics* 53.31 (2020), p. 313001.

[41] John F Barry et al. “Sensitivity optimization for NV-diamond magnetometry”. In: *Reviews of Modern Physics* 92.1 (2020), p. 015004.

[42] Andrew M Edmonds et al. “Generation of nitrogen-vacancy ensembles in diamond for quantum sensors: Optimization and scalability of CVD processes”. In: *arXiv preprint arXiv:2004.01746* (2020).

[43] Thaddeus D Ladd et al. “Quantum computers”. In: *nature* 464.7285 (2010), pp. 45–53.

[44] Nitin Mohan et al. “In vivo imaging and toxicity assessments of fluorescent nanodiamonds in Caenorhabditis elegans”. In: *Nano letters* 10.9 (2010), pp. 3692–3699.

[45] Mark JH Ku et al. “Imaging viscous flow of the Dirac fluid in graphene”. In: *Nature* 583.7817 (2020), pp. 537–541.

[46] AJ Healey et al. “Polarization transfer to external nuclear spins using ensembles of nitrogen-vacancy centers”. In: *Physical Review Applied* 15.5 (2021), p. 054052.

[47] Tom Delord et al. “Spin-cooling of the motion of a trapped diamond”. In: *Nature* 580.7801 (2020), pp. 56–59.

[48] Chi-Cheng Fu et al. “Characterization and application of single fluorescent nanodiamonds as cellular biomarkers”. In: *Proceedings of the National Academy of Sciences* 104.3 (2007), pp. 727–732.

[49] Romana Schirhagl et al. “Nitrogen-vacancy centers in diamond: nanoscale sensors for physics and biology”. In: *Annu. Rev. Phys. Chem* 65.1 (2014), pp. 83–105.

[50] Robert Chapman and Taras Plakhoitnik. “Background-free imaging of luminescent nanodiamonds using external magnetic field for contrast enhancement”. In: *Optics Letters* 38.11 (2013), pp. 1847–1849.

[51] Liam P McGuinness et al. “Quantum measurement and orientation tracking of fluorescent nanodiamonds inside living cells”. In: *Nature nanotechnology* 6.6 (2011), pp. 358–363.

[52] Xi Feng et al. “Association of nanodiamond rotation dynamics with cell activities by translation-rotation tracking”. In: *Nano letters* 21.8 (2021), pp. 3393–3400.

[53] Loïc Rondin et al. “Magnetometry with nitrogen-vacancy defects in diamond”. In: *Reports on progress in physics* 77.5 (2014), p. 056503.

[54] Florian Dolde et al. “Electric-field sensing using single diamond spins”. In: *Nature Physics* 7.6 (2011), pp. 459–463.

[55] Julia Michl et al. “Robust and accurate electric field sensing with solid state spin ensembles”. In: *Nano letters* 19.8 (2019), pp. 4904–4910.

[56] Victor M Acosta et al. “Temperature dependence of the nitrogen-vacancy magnetic resonance in diamond”. In: *Physical review letters* 104.7 (2010), p. 070801.

[57] Georg Kucsko et al. “Nanometre-scale thermometry in a living cell”. In: *Nature* 500.7460 (2013), pp. 54–58.

[58] Preeti Ovartchaiyapong et al. “Dynamic strain-mediated coupling of a single diamond spin to a mechanical resonator”. In: *Nature communications* 5.1 (2014), pp. 1–6.

[59] Marcus W Doherty et al. “Electronic properties and metrology applications of the diamond NV- center under pressure”. In: *Physical review letters* 112.4 (2014), p. 047601.

[60] MP Ledbetter et al. "Gyroscopes based on nitrogen-vacancy centers in diamond". In: *Physical Review A* 86.5 (2012), p. 052116.

[61] Ashok Ajoy and Paola Cappellaro. "Stable three-axis nuclear-spin gyroscope in diamond". In: *Physical Review A* 86.6 (2012), p. 062104.

[62] Morgan W Mitchell and Silvana Palacios Alvarez. "Colloquium: Quantum limits to the energy resolution of magnetic field sensors". In: *Reviews of Modern Physics* 92.2 (2020), p. 021001.

[63] Thomas Wolf et al. "Subpicotesla diamond magnetometry". In: *Physical Review X* 5.4 (2015), p. 041001.

[64] James Eills et al. "Spin hyperpolarization in modern magnetic resonance". In: (2022).

[65] Anatole Abragam and Maurice Goldman. "Principles of dynamic nuclear polarisation". In: *Reports on Progress in Physics* 41.3 (1978), p. 395.

[66] J-P Tetienne et al. "Prospects for nuclear spin hyperpolarization of molecular samples using nitrogen-vacancy centers in diamond". In: *Physical Review B* 103.1 (2021), p. 014434.

[67] V Jacques et al. "Dynamic polarization of single nuclear spins by optical pumping of nitrogen-vacancy color centers in diamond at room temperature". In: *Physical review letters* 102.5 (2009), p. 057403.

[68] Benjamin Smeltzer, Jean McIntyre, and Lilian Childress. "Robust control of individual nuclear spins in diamond". In: *Physical Review A* 80.5 (2009), p. 050302.

[69] Jonathan P King et al. "Room-temperature in situ nuclear spin hyperpolarization from optically pumped nitrogen vacancy centres in diamond". In: *Nature communications* 6.1 (2015), pp. 1–5.

[70] Jochen Scheuer et al. "Optically induced dynamic nuclear spin polarisation in diamond". In: *New journal of Physics* 18.1 (2016), p. 013040.

[71] Ilai Schwartz et al. "Robust optical polarization of nuclear spin baths using Hamiltonian engineering of nitrogen-vacancy center quantum dynamics". In: *Science advances* 4.8 (2018), eaat8978.

[72] R Rizzato et al. "Polarization Transfer from Optically Pumped Ensembles of N-V Centers to Multinuclear Spin Baths". In: *Physical Review Applied* 17.2 (2022), p. 024067.

[73] Maxime Perdriat et al. "Spin-mechanics with nitrogen-vacancy centers and trapped particles". In: *Micromachines* 12.6 (2021), p. 651.

[74] Markus Aspelmeyer, Tobias J Kippenberg, and Florian Marquardt. "Cavity optomechanics". In: *Reviews of Modern Physics* 86.4 (2014), p. 1391.

[75] Peter Rabl et al. “Strong magnetic coupling between an electronic spin qubit and a mechanical resonator”. In: *Physical Review B* 79.4 (2009), p. 041302.

[76] Shimon Kolkowitz et al. “Coherent sensing of a mechanical resonator with a single-spin qubit”. In: *Science* 335.6076 (2012), pp. 1603–1606.

[77] C Pellet-Mary et al. “Magnetic torque enhanced by tunable dipolar interactions”. In: *Physical Review B* 104.10 (2021), p. L100411.

[78] M Perdriat et al. “Angle locking of a levitating diamond using spin diamagnetism”. In: *Physical Review Letters* 128.11 (2022), p. 117203.

[79] Bo-Bo Wei et al. “Magnetic ordering of nitrogen-vacancy centers in diamond via resonator-mediated coupling”. In: *EPJ Quantum Technology* 2.1 (2015), pp. 1–7.

[80] Yue Ma et al. “Proposal for quantum many-body simulation and torsional matter-wave interferometry with a levitated nanodiamond”. In: *Physical Review A* 96.2 (2017), p. 023827.

[81] Philip W Anderson. “More is different: broken symmetry and the nature of the hierarchical structure of science.” In: *Science* 177.4047 (1972), pp. 393–396.

[82] R Giri et al. “Coupled charge and spin dynamics in high-density ensembles of nitrogen-vacancy centers in diamond”. In: *Physical Review B* 98.4 (2018), p. 045401.

[83] VV Dobrovitski et al. “Decoherence dynamics of a single spin versus spin ensemble”. In: *Physical Review B* 77.24 (2008), p. 245212.

[84] A Jarmola et al. “Temperature-and magnetic-field-dependent longitudinal spin relaxation in nitrogen-vacancy ensembles in diamond”. In: *Physical review letters* 108.19 (2012), p. 197601.

[85] Mariusz Mrózek et al. “Longitudinal spin relaxation in nitrogen-vacancy ensembles in diamond”. In: *EPJ Quantum Technology* 2 (2015), pp. 1–11.

[86] Joonhee Choi et al. “Depolarization dynamics in a strongly interacting solid-state spin ensemble”. In: *Physical review letters* 118.9 (2017), p. 093601.

[87] Soonwon Choi et al. “Observation of discrete time-crystalline order in a disordered dipolar many-body system”. In: *Nature* 543.7644 (2017), pp. 221–225.

[88] Georg Kucsko et al. “Critical thermalization of a disordered dipolar spin system in diamond”. In: *Physical review letters* 121.2 (2018), p. 023601.

[89] Carlo Bradac et al. “Room-temperature spontaneous superradiance from single diamond nanocrystals”. In: *Nature communications* 8.1 (2017), pp. 1–6.

[90] Andreas Angerer et al. “Superradiant emission from colour centres in diamond”. In: *Nature Physics* 14.12 (2018), pp. 1168–1172.

[91] Igor Žutić, Jaroslav Fabian, and S Das Sarma. “Spintronics: Fundamentals and applications”. In: *Reviews of modern physics* 76.2 (2004), p. 323.

[92] LT Hall et al. “Detection of nanoscale electron spin resonance spectra demonstrated using nitrogen-vacancy centre probes in diamond”. In: *Nature communications* 7.1 (2016), pp. 1–9.

[93] Victor M Acosta et al. “Diamonds with a high density of nitrogen-vacancy centers for magnetometry applications”. In: *Physical Review B* 80.11 (2009), p. 115202.

[94] Alexandre Tallaire et al. “High NV density in a pink CVD diamond grown with N2O addition”. In: *Carbon* 170 (2020), pp. 421–429.

[95] Olga A Shenderova et al. “Synthesis, properties, and applications of fluorescent diamond particles”. In: *Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena* 37.3 (2019), p. 030802.

[96] Clément Pellet-Mary et al. “Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition”. In: *Physical Review B* 103.10 (2021), p. L100411.

[97] Midrel Wilfried Ngandeu Ngambou et al. “Improving NV centre density during diamond growth by CVD process using N2O gas”. In: *Diamond and Related Materials* 123 (2022), p. 108884.

[98] C Pellet-Mary et al. “Spin-Relaxation of Dipolar-Coupled Nitrogen-Vacancy Centers: The role of Double-flip Processes”. In: *arXiv preprint arXiv:2207.13899* (2022).