CLOSING REPORT

Quant-ERA: Scalable Electrically Read Diamond Spin Qubit Technology for Single Molecule Imagers

Project period: 03/01/2018 – 10/31/2021

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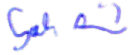
Abstract: Diamond quantum sensors will be one of the first quantum technology devices on the market, with applications in magnetic field sensing and sensors for medicine, biology, and chemistry. The Q-Magine project comes with a disruptive approach for nitrogen-vacancy (NV) spin projection measurements, by developing hybrid photoelectric detection of NV electron spin resonances at room temperature, introduced jointly by the applicants and used for constructing single qubit matrix sensors.

Wigner Research Centre for Physics will contribute to tight control of the photoelectric readout of NV center in diamond by modeling from first principles calculations, identifying surface termination of diamond for NV based quantum sensing, and finding alternative color centers in diamond for quantum sensing.

Date: 12/30/2021

Signature: Dr. Ádám Gali, PI



Introduction

The nitrogen-vacancy (NV) center in diamond is a solid-state defect qubit with favorable coherence time up to room temperature, which could be harnessed in several quantum-enhanced sensor and quantum communication applications, and has a potential in quantum simulation and computing [1]. The quantum control largely depends on the intricate details about the electronic structure and states of the NV center, the radiative and nonradiative rates between these states, and the coupling of these states to external spins, electric, magnetic, and strain fields, and temperature. In particular, the project aimed to harness NV center's favorable properties for quantum sensing applications. From practical point of view, it is desirable to use the photoelectric readout mechanism for creating a compact quantum sensor device which was the target of the project. The photoelectric readout of the NV center is based on the spin-selective photoionization of the defect. The consortium members wrote a chapter in a book about experimental discoveries reported before the start of the project and also during the project entitled "Fundaments of photoelectric readout of spin states in diamond" which deals with the detection of NV center spins in diamond using the photoelectric detection of magnetic resonances (PDMR) method. It provides in particular insights into the physics of electronic transitions of the NV center, leading to the free carrier generation, and discusses methodologies how to implement the photocurrent detection principles in the dynamically evolving field of quantum technologies [2]. Ádám Gali's group at Wigner Research Centre for Physics has the role to understand the optical spinpolarization of the NV center spins and the mechanism of optical and photoelectric readout, in order to setup such a model which can be used then to tightly control the experiments. Before the start of the project, the photoelectric readout for electron spins were already demonstrated [2], but not with a nuclear spin which requires a delicate quantum optics control and understanding the interaction of light, microwave and radiofrequency pulses and external magnetic fields with the NV electron and nuclear spins.

Beside NV center we also investigated other color centers, preferably but not exclusively in diamond, that may substitute NV center in certain quantum technology applications. Here, we were interested in such color centers that have favorable optical emission in terms of the wavelength (closer to near-infrared which is desirable for *in vivo* studies) or stability of the zero-phonon-line (ZPL) emission.

Methods

We applied *ab initio* methods to calculate the electronic structure of color centers which is based on supercell plane wave density functional theory methods (see Ref. [1] and references therein). The color centers were modeled by 512-atom or larger supercell. The spin dynamics of the system was simulated by Lindblad formalism where the spin Hamiltonian parameters were extracted from the *ab initio* methods (see Ref. [3] and references therein). Ádám Gali wrote a comprehensive review paper about the *ab initio* description of solid-state defect qubits [1], which is a reference to methodologies applied in the project.

The calculations were carried out on massively parallelized computer clusters with the use of both local facilities, national (KIFÜ) and international supercomputer centers. The use of local facilities is inevitable for development and assessment of novel algorithms and methods, as well as for execution of high priority calculations. We used the budget to extend our local computer cluster to timely achieve results, and high performance laptops were also purchased to post-process the data from calculations.

Results

Nitrogen-vacancy center

The nitrogen-vacancy (NV) center in diamond is of high importance in quantum information processing applications. The operation of the NV center relies on the efficient optical polarization of its electron spin. However, the full optical spin-polarization process, which involves the intersystem crossing between the shelving singlet state and the ground-state triplet, was not understood. We developed a detailed theory of this process which involves a combination of pseudo- and dynamic Jahn-Teller interactions together with spin-orbit interaction [4]. Our theory provides an explanation for the asymmetry between the observed emission and absorption spectra of the singlet states. We apply density functional theory to calculate the intersystem crossing rates and the optical spectra of the singlets, and we obtain a good agreement with the experimental data. Since the NV center serves as a template for other solid-state-defect quantum bit systems, our theory provides a toolkit to study them that might help optimize their quantum bit operation.

The pseudo- and dynamic Jahn-Teller interactions are special forms of strong electron-phonon coupling. The Jahn-Teller formalism helps to provide a simple but powerful model for electron-phonon coupling which we can use to calculate the polaronic wavefunctions for large, 512-atom, systems by calculating the adiabatic potential energy surface of the electronic states by density functional theory. In particular, a breakthrough has been achieved in understanding the decay from the shelving singlet state to the ground state triplet which closes the optical spinpolarization loop [4]. We found that the pseudo-Jahn-Teller effect creates such polaronic wavefunctions in the singlet manifold which can effectively connect the shelving singlet and the ground state triplet states which is otherwise forbidden by considering only the pure electronic solution. In other words, the Herzberg-Teller theorem, which was used to explain second-order optical transition between polaronic wavefunctions, was extended towards the intersystem crossing transition. We believe that this finding has an importance beyond NV center and generally applied to other point defects (see also below and Refs. [5,6]).

Ádám Gali is a co-author in a recent general review paper on solid state defect quantum bits in which this finding is highlighted [7]. Defects with associated electron and nuclear spins in solid-state materials have a long history relevant to quantum information science that goes back to the first spin echo experiments with silicon dopants in the 1950s. Since the turn of the century, the field has rapidly spread to a vast array of defects and host crystals applicable to quantum communication, sensing and computing. In this review paper, we expanded upon all the key components of solid-state spin defects, with an emphasis on the properties of defects and of the host material, on engineering opportunities and on other pathways for improvement with providing broad guidelines for the field of solid-state spin defects for quantum information.

It is emphasized that the key results achieved in Ref. [4] established the rate equations between different states of NV defect needed for optimization of electric readout. We further employed a combination of spectrally resolved optical pump-probe spectroscopy and excited-state *ab initio* molecular dynamics (ESAIMD) simulations to study the ultrafast vibrational relaxation dynamics of the ^{3}E excited state of NV center [8]. The experimental results revealed vibrational relaxation in the phonon sideband with a time constant of approximately 50 fs, in excellent agreement with the ~40-fs structural equilibration timescale predicted by ESAIMD simulations. The observed ultrafast vibrational energy relaxation implies that

dynamical processes triggered by photoexcitation into the phonon sideband of the NV center occur primarily in the lowest vibronic level of the ^{3}E state [8]. This finding further helped us to setup a reliable model for the rate equations of the optical and intersystem crossing transitions.

We also investigated the NV center in diamond at magnetic fields corresponding to the ground-state level anticrossing (GSLAC) region gives rise to rich photoluminescence (PL) signals due to the vanishing energy gap between the electron spin states, which enables for a broad variety of environmental couplings to have an effect on the NV center's luminescence (see Ref. [3] and references therein). Previous works have addressed several aspects of the GSLAC photoluminescence, however, a comprehensive analysis of the GSLAC signature of NV ensembles in different spin environments at various external fields was missing. We employed a combination of experiments and recently developed numerical methods to investigate in detail the effects of transverse electric and magnetic fields, strain, P1 centers (N-donors), NV centers, and the ^{13}C nuclear spins on the GSLAC photoluminescence. Our comprehensive analysis provided a solid ground for advancing various microwave-free applications at the GSLAC, including but not limited to magnetometry, spectroscopy, dynamic nuclear polarization (DNP), and nuclear magnetic resonance (NMR) detection. We demonstrate that not only the most abundant ^{14}NV center but the ^{15}NV can also be utilized in such applications.

By combining the results in Refs. [3,4,8], we addressed the photoelectric readout of a single nuclear spin in diamond both in theory and experiment. Nuclear spins are of great interest because they are leading candidates for future quantum technologies, including quantum computation, communication, and sensing. Nuclear spins in diamond are particularly attractive due to their long coherence time. With the NV center, such nuclear qubits benefit from an auxiliary electronic qubit, which, at cryogenic temperatures, enables probabilistic entanglement mediated optically by photonic links. We demonstrated a concept of a microelectronic quantum device at ambient conditions using diamond as wide bandgap semiconductor. The basic quantum processor unit – a single ^{14}N nuclear spin coupled to the NV electron – was read photoelectrically and thus operates in a manner compatible with nanoscale electronics [9]. The underlying theory by Ádám Gali's group provided the key ingredients for photoelectric quantum gate operations and readout of nuclear qubit registers. This demonstration is, therefore, a step towards diamond quantum devices with a readout area limited by inter-electrode distance rather than by the diffraction limit. Such scalability could enable the development of electronic quantum processors based on the dipolar interaction of spin-qubits placed at nanoscopic proximity.

Finally, we mention our results on the surface termination of diamond which is important in the quantum sensor application. There is an immediate quest to find suitable diamond surfaces for NV sensors. The surface terminators of (113) diamond to host shallow NV centers were studied by means of first principles calculations [10]. Results indicated that complete oxygen termination of (113) diamond creates positive electron affinity with neither strain on the surface nor in-gap levels. This is a very surprising result as the commonly employed oxygenated (001) diamond surface is often defective due to the disorder created by the strain of ether groups at the surface that seriously undermine the coherence properties of the shallow NV centers. The special atomic configurations on (113) diamond surface are favorable for oxygen bonding, in contrast to (001) and (111) diamond surfaces. These simulations implied that oxygenated diamond (113) surface can be produced by conventional diamond chemical vapor deposition growth. Combining this with the $\sim$73% preferential alignment of as-grown NV centers in (113) oriented diamond, oxygenated (113) diamond is presently supposed to be the most prospective host for NV quantum sensors [10].

Alternative color centers

Nickel-vacancy center in diamond

There is a continuous search for solid state spin qubits operating at room temperature with excitation in the infrared communication bandwidth. Recently, we have introduced the photoelectric detection of magnetic resonance (PDMR) to read the electron spin state of NV centers in diamond, a technique which is promising for applications in quantum information technology. By measuring the photoionization spectra on a diamond crystal, we found two ionization thresholds of unknown origin. On the same sample we also observed absorption and photoluminescence signatures that were identified in the literature as Ni-associated defects. We performed *ab initio* calculations of the photoionization cross section of the nickel split-vacancy complex (NiV) and N-related defects in their relevant charge states and fitted the concentration of these defects to the measured photocurrent spectrum, which led to a surprising match between experimental and calculated spectra. This study enabled us to identify the two unknown ionization thresholds with the two acceptor levels of NiV. Because the excitation of NiV is in the infrared, the photocurrent detected from the paramagnetic NiV color centers is a promising way towards the design of electrically readout qubits [11].

We note that nickel is a common impurity in high-pressure high-temperature diamond and may contaminate chemical vapor deposited diamond used for high-power electronics or quantum technology applications. Magneto-optical fingerprints of nickel have been known since decades, however, no consensus has been reached about the microscopic origins of nickel-related electron paramagnetic resonance, photoluminescence, and optically detected magnetic resonance spectra. The unknown nickel-related defect structures in diamond make it difficult to control them or harness them for a given application. As a consequence, nickel is considered as an impurity in diamond that should be avoided or its concentration should be minimized. Recent advances in the development of *ab initio* magneto-optical spectroscopy have significantly increased its accuracy and predictive power that can be employed for identification and in-depth characterization of paramagnetic color centers in diamond. We extended the accuracy of the *ab initio* magneto-optical spectroscopy tools towards self-consistent calculation of second-order spin-orbit coupling for paramagnetic color centers in solids. We applied the full arsenal of the *ab initio* magneto-optical spectroscopy tools to characterize the split nickel-vacancy defect in diamond which is one of the most stable nickel-related defect configurations. As a result, electron paramagnetic resonance and optical centers are positively identified in various charge states of the nickel-vacancy defect in diamond. In particular, the 1.40-eV optical center and the NIRIM-2 electron paramagnetic resonance center are identified as the single negative charge state of the split NiV center. The defect possesses S=1/2 spin state with an orbital doublet ground state. We find that the coherence time of the ground-state spin is about 0.1 ms at cryogenic temperatures which can be optically initialized and readout by a Λ-scheme protocol. Since the defect has inversion symmetry the optical signal is insensitive to the stray electric fields, which is an advantage for creating indistinguishable solid-state single-photon sources. We predict that the negatively charged NiV defect has similar optical properties to those of the well-known silicon-vacancy (SiV) defect in diamond but is superior in terms of electron spin coherence times. Our study resolved a few decades controversy about the nickel-related spectroscopy centers in diamond and turns nickel from an impurity to a resource in quantum technology applications [12].

The potential of NiV defect quantum bit is to be explored in the near future. In the next papers, we worked tightly with experimentalists to harness the prepared color centers in diamond and silicon carbide.

Neutral silicon-vacancy center in diamond

Neutral silicon-vacancy (SiV0) centers in diamond are promising candidates for quantum networks because of their excellent optical properties and long spin coherence times. However, spin-dependent fluorescence in such defects has been elusive due to poor understanding of the excited state fine structure and limited off-resonant spin polarization. We reported the realization of optically detected magnetic resonance and coherent control of SiV0 centers at cryogenic temperatures, enabled by efficient optical spin polarization via previously unreported higher-lying excited states [13]. We assigned these states as bound exciton states using group theory and density functional theory. These bound exciton states enable new control schemes for SiV0 as well as other emerging defect systems. We note here that the group theory of inversion symmetric bound exciton states is completely novel to our knowledge which could explain first order and vibration assisted optical spinpolarization resonances. Furthermore, we developed a theory for scaling the bound exciton states and energy levels within supercell formalism which are described in the Supplementary Materials of Ref. [13].

Tin-vacancy center in diamond

The recently discovered negatively charged tin-vacancy (SnV) center in diamond is a promising candidate for applications in quantum information processing (QIP). We presented a detailed spectroscopic study encompassing single photon emission and polarization properties, the temperature dependence of emission spectra as well as a detailed analysis of the phonon sideband and Debye–Waller factor by using first principles calculations [14]. Using photoluminescence excitation spectroscopy we probe an energetically higher lying excited state and prove fully lifetime limited linewidths of single emitters at cryogenic temperatures. For these emitters we also investigate the stability of the charge state under resonant excitation. These results provide a detailed insight into the spectroscopic properties of the SnV center and lay the foundation for further studies regarding its suitability in QIP.

Magnesium-vacancy center in diamond

Defect quantum bits (qubits) constitute an important emerging technology. However, it is necessary to explore new types of defects to enable large-scale applications. We examined the potential of magnesium-vacancy (MgV) in diamond to operate as a qubit by computing the key electronic- and spin properties with robust theoretical methods beyond density functional theory such as quantum chemistry wave function methods and density matrix renormalization group algorithms. We found that the electronic structure of MgV permits the coexistence of two loosely separated spin-states, where both can emerge as a ground state and be interconverted depending on the temperature and external strain. These results demonstrated a route to control the magneto-optical response of a qubit by modulating the operational conditions [15].

In particular, we performed an extensive theoretical investigation to elucidate the role of Mg in the formation of color centers in diamond. Our calculations revealed that MgV is the most stable defect configuration among those considered, with a photostable −1 charge state, achieved in experiment by photoionization with a green laser (see references in Ref. [15]). Furthermore, we showed that this defect feature possesses an inversion symmetry and emits light from the third bright excited state, at odds with Kasha's rule. It also possesses quasi-degenerate quartet and doublet ground states, each of them exhibiting distinct photophysical properties. Although we assigned the experimental photoluminescence

signal to the $^2E_u^{(2)}$–^2E_g transition, the quartet state (which appeared to be in thermal equilibrium with the doublet ^2E_g) has ZPL of ~1.1 eV, lying in the near-infrared window in biological tissue.

Si-vacancy quantum bits in 4H silicon carbide

We mention here two important results for color centers in silicon carbide. Silicon carbide material is akin to diamond in terms of relatively large band gap (3.3 eV) and high Debye-temperature. In particular, the application of Jahn-Teller theory in NV center within C_{3v} symmetry [4] could be extended to the tetrahedral symmetry distorted to C_{3v} symmetry for the Si-vacancy in 4H silicon carbide.

Silicon-vacancy qubits in silicon carbide (SiC) are emerging tools in quantum-technology applications due to their excellent optical and spin properties. We explored the effect of temperature and strain on these properties by focusing on the two silicon-vacancy qubits, V1 and V2, in 4H SiC. We applied density-functional theory beyond the Born-Oppenheimer approximation to describe the temperature-dependent mixing of electronic excited states assisted by phonons [6]. We obtained a polaronic gap of around 5 and 22 meV for the V1 and V2 centers, respectively, which results in a significant difference in the temperature-dependent dephasing and zero-field splitting of the excited states, which explains recent experimental findings. We also computed how crystal deformations affect the ZPL of these emitters. Our predictions are important ingredients in any quantum applications of these qubits sensitive to these effects.

Si-vacancies in SiC can be used to realize quantum systems combining indistinguishable photon generation and spin-based quantum information processing that are essential for remote quantum applications and networking. We investigated the silicon vacancy center in SiC and demonstrated controlled emission of indistinguishable and distinguishable photons via coherent spin manipulation. Using strong off-resonant excitation and collecting zero-phonon line photons, we showed a two-photon interference contrast close to 90% in Hong-Ou-Mandel type experiments [5]. Further, we exploited the system's intimate spin-photon relation to spin-control the color and indistinguishability of consecutively emitted photons. Here *ab initio* calculations with the use of modern theory of polarization could quantify the coupling of electric stray fields to the NV center in diamond and Si-vacancy in 4H SiC, and found that the coupling strength is much weaker for the latter which makes it a good candidate to create indistinguishable single photon sources. Our results provide a deep insight into the system's spin-phonon-photon physics and underline the potential of the industrially compatible silicon carbide platform for measurement-based entanglement distribution and photonic cluster state generation. Additional coupling to quantum registers based on individual nuclear spins would further allow for high-level network-relevant quantum information processing, such as error correction and entanglement purification.

Outlook

Despite on the pandemic crisis in the year of 2020, which forced us to elongate the project with eight months, significant results could be achieved. The results were published in prestigious journals, e.g. Nature Communications, Nature Partner Journal Quantum Information or Physical Review Letters and also summarized in two review papers (one single author paper and one paper in Nature Materials Reviews). The published papers already received considerable number of citations (>100) and attracted great attention in the community of quantum technology researchers. The results were also presented in contributed and invited talks at workshops and conferences. Here we list the invited talks below. We note that because of the pandemic crisis the conference invitations were naturally ceased from 2020:

- *Novel color centers in diamond for communication and sensing*, European Materials Research Society Fall Meeting 2019, Warsaw (Poland), September 16-19, 2019

- *Theory of silicon-vacancy and related colour centres in diamond*, 30th International Conference on Diamond and Carbon Materials, Sevilla (Spain), September 10-14, 2019

- *The progress in ab initio description of solid state defect qubits*, 30th International Conference on Defects in Semiconductors, Seattle (United States), July 21-26, 2019

- *Ab initio study of defect qubits for hyperpolarization and quantum sensing*, Gordon Research Conference on Quantum Sensing, Hong Kong (China), June 2-6, 2019

- *Ab initio study of defect qubits for hyperpolarization, quantum sensing and communication*, QDiamond Workshop, Tel Aviv (Israel), May 13-16, 2019

- *Theory of electrical readout of deep defect qubits in solids*, APS March Meeting, Boston (USA), March 4-8, 2019

- *Ab initio theory of intersystem crossing in NV center in diamond*, OSA Incubator Meeting, Defects by Design: Quantum Nanophotonics in Emerging Materials, Washington, DC (USA), October 28-30, 2018

- *Toward full ab initio description of qubits in solids*, CECAM-Workshop on Crystal defects for qubits, single photon emitters and nanosensors, Bremen (Germany), July 9-13, 2018

- *Theory on the optical spin-polarization loop of the nitrogen-vacancy center in diamond*, 25th Central European Workshop on Quantum Optics, Mallorca (Spain), May 21-25, 2018

- *Closing the loop - ab initio theory on NV center in diamond*, QDiamond Workshop 2018, Tel Aviv (Israel), April 23-27, 2018

Young researchers were trained and worked in the project. One of them, Péter Udvarhelyi, started to work as a PhD candidate on the project, and he could successfully defend his PhD thesis in 2020 (partially based on the project results) and continued to work on the project as postdoc researcher.

As a consequence of the results achieved by Ádám Gali's group and the overall consortium, new joint projects have been started in which one of the projects have been granted by the European Commission within the EIC Pathfinder program (project QuMicro about sensing of microwave fields via array of NV centers in diamond) and another one will be financed by NKFIH via Quant-ERA program for exploring a room temperature quantum processor based on the photoelectric readout of arrays of NV centers in diamond (project Maestro). Further development is foreseen in the charge control of NV center and application of nickel defects for quantum sensor applications.

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