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Systematic search for new color centers in diamond for quantum technologies

De Leon, Nathalie
TRUSTEES OF PRINCETON UNIVERSITY
1 NASSAU HALL
PRINCETON, NJ,
US

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Nathalie De Leon

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TRUSTEES OF PRINCETON UNIVERSITY
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14. ABSTRACT
The goal of this YIP was to systematically search for new color centers in diamond that will act as bits for storing and processing quantum information. Color centers in diamond are atomic scale defects in diamond with energetically deep, confined electronic states that can have exceptional spin and optical coherence. In particular, the nitrogen vacancy (NV) center in diamond has been widely deployed in many recent experiments in quantum science, and is actively explored for quantum sensing and network technologies. However, known color centers suffer from either poor optical coherence or poor spin coherence, motivating a search for alternative color centers that are less sensitive to their environment. Prior work on new defects in diamond has relied on serendipitous discovery. Our approach, by contrast, is to systematically search for new color centers in diamond by introducing heteroatoms through ion implantation into ultrahigh purity, CVD diamonds, using thermal annealing to remove ion implantation damage, and to characterize the resulting color centers with bulk spin and optical spectroscopy techniques that are optimized for interrogating thin layers of implanted defects. We then perform detailed single center spectroscopy on defects that display promising properties.

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Final Report, AFOSR YIP grant # FA9550-17-1-0158
Systematic Search for New Color Centers in Diamond for Quantum Technologies
PI: Nathalie de Leon
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The goal of this YIP was to systematically search for new color centers in diamond that will act as bits for storing and processing quantum information. Color centers in diamond are atomic scale defects in diamond with energetically deep, confined electronic states that can have exceptional spin and optical coherence. In particular, the nitrogen vacancy (NV) center in diamond has been widely deployed in many recent experiments in quantum science, and is actively explored for quantum sensing and network technologies [1], [2]. However, known color centers suffer from either poor optical coherence [3] or poor spin coherence [4], motivating a search for alternative color centers that are less sensitive to their environment. Prior work on new defects in diamond has relied on serendipitous discovery. Our approach, by contrast, is to systematically search for new color centers in diamond by introducing heteroatoms through ion implantation into ultrahigh purity, CVD diamonds, using thermal annealing to remove ion implantation damage, and to characterize the resulting color centers with bulk spin and optical spectroscopy techniques that are optimized for interrogating thin layers of implanted defects. We then perform detailed single center spectroscopy on defects that display promising properties.

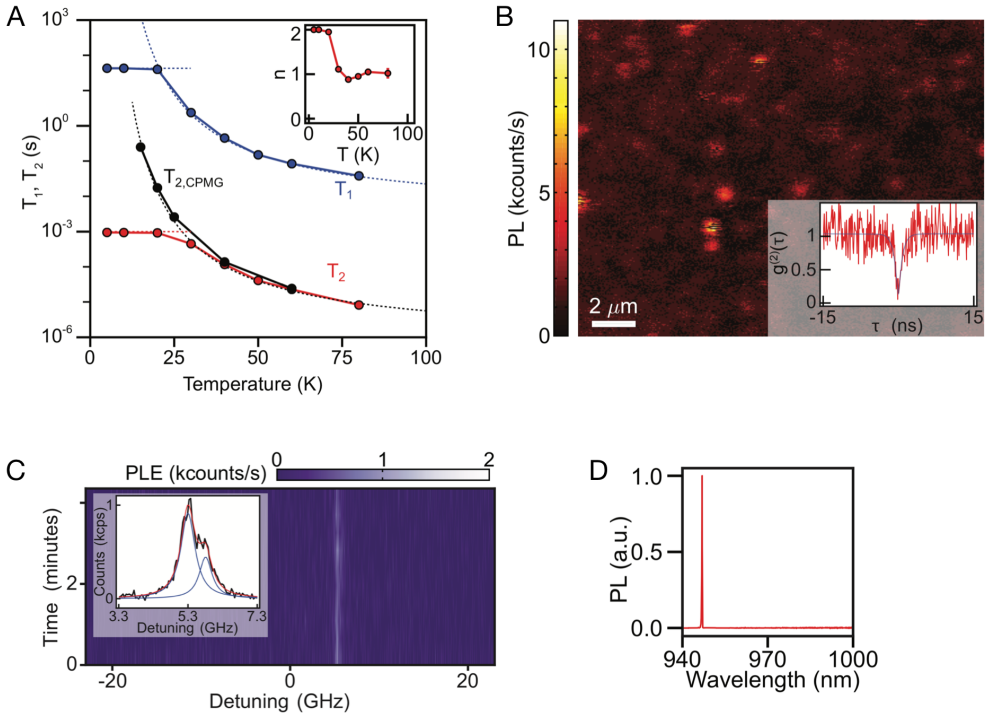

Figure 1: Coherence properties of SiV^0 (reproduced from [5]). (A) Spin T_1, $T_{2,Hahn\ echo}$, and $T_{2,CPMG}$ showing long relaxation and coherence times at low temperature. (inset) The stretched exponent of $T_{2,Hahn\ echo}$ changes from n=2 to n=1 around 25 K, indicating that T_2 is limited by spectral diffusion of ^{13}C at low temperatures, but is dictated by the Orbach process at higher temperatures, as explored in reference [6]. (B) Single SiV^0 centers isolated by scanning confocal microscopy. (Inset) Second order correlation statistics from a single center show antibunching. (C) Photoluminescence excitation (PLE) spectroscopy shows a near-transform-limited optical transition that is stable over time. (D) Photoluminescence (PL) spectroscopy shows that over 90% of the optical emission is in the zero-phonon line.

Our main result from this systematic search has been the discovery of a new color center in diamond, the neutral charge state of the silicon vacancy center (SiV^0), the first color center that has been demonstrated to have long spin coherence times ($T_1 \sim 1$ min, $T_2 \sim 1$ s) and near-transform-limited optical

linewidths (Figure 1) [5]. Specifically, we achieved the stabilization and characterization of SiV^0 by deliberate engineering of the diamond host.

After establishing the optical and spin coherence properties of SiV^0 at low temperatures, we then turned to detailed characterization of the ground state spin dynamics at high temperatures [6]. Above 20 K, T_1 decreases rapidly with a temperature-dependence characteristic of an Orbach process and, surprisingly, is strongly anisotropic with respect to magnetic-field orientation. As the angle of the magnetic field is rotated relative to the symmetry axis of the defect, T_1 is reduced by over three orders of magnitude. The electron spin coherence time (T_2) follows the same temperature dependence but is drastically shorter than T_1. We proposed that these observations result from phonon-mediated transitions to a low-lying excited state that are spin conserving when the magnetic field is aligned with the defect axis.

In the final year of the project, we established optically detected magnetic resonance (ODMR) in this new defect [7]. Specifically, using electron spin resonance detection with resonant optical excitation, we were able to identify an additional, previously unknown, excited state in SiV^0 at 952 nm, as well as several other higher lying transitions at 890, 850, and 835 nm that we have identified as Rydberg-like bound exciton transitions. These higher lying states allow for efficient bulk spin polarization, comparable to what is observed for resonant excitation at the zero-phonon line. This highly efficient spin polarization enabled the observation of ODMR for the first time. This work has resulted in three manuscripts and a patent (references [5]-[8]).

Building on this result, we are currently pursuing several directions to develop SiV^0 for quantum networks. First, we aim to establish ODMR in single centers and map the excited state fine structure and establish a spin-photon interface. For single center measurements, we have primarily been using two-photon spectroscopy (Figure 2), in which we tune two lasers or an EOM sideband to two optical transitions and look at time-dependent fluorescence, enabling interrogation of both spin-conserving and non-conserving transitions. These experiments have allowed us to identify different optical transitions that are split by ~GHz scales, and we can observe optical pumping between them, as well as evolution of the ground states in the dark. We would also like to extend these measurements by applying strain and high magnetic fields to elucidate selection rules and to understand the interactions of SiV^0 with its environment.

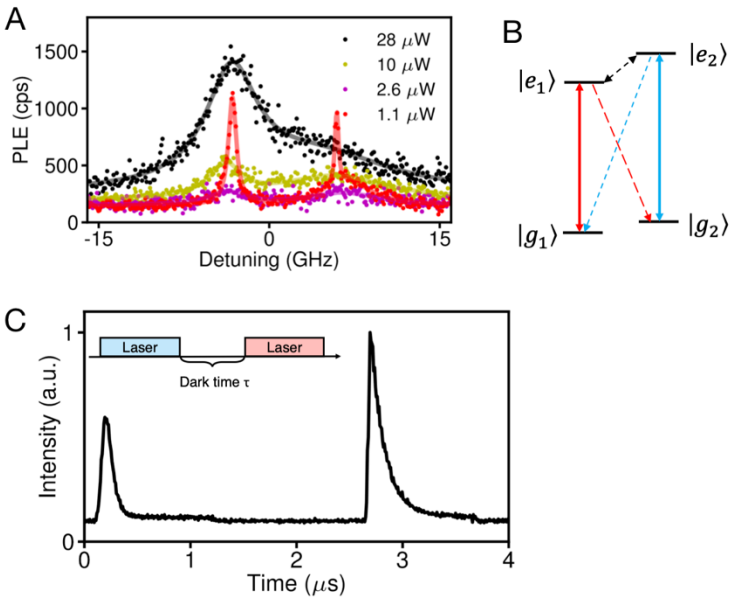

Figure 2 Two-photon spectroscopy of single SiV^0 centers. (A) At high excitation power, two broad features are apparent in PLE, which disappear at low power (black, yellow, purple traces). When two lasers are incident with the center frequencies of the broad peaks, the PLE is recovered at low power (red trace), indicating that there are two optical transitions that can be probed by optical repumping, as shown in (B). (C) Time-resolved PL resulting from pulsed excitation at the two colors shows optical pumping into the dark state at each frequency.

Second, we aim to carefully characterize the optical quantum efficiency by performing resonance fluorescence in bulk samples, and by tailoring the electromagnetic density of states of the defect. Single center saturation powers and PL count rates are consistent with unity quantum efficiency. To perform resonance fluorescence, we are currently building a microscope with a Fabry-Perot cavity for spectral filtering, as well as careful polarization control to filter the incident laser. Separately,

we are developing a GaAs-on-diamond nanophotonic platform for Purcell enhancement of SiV^0 emission. These measurements should allow us to directly quantify the quantum efficiency.

In parallel, we plan to continue our systematic search for other defects in diamond through our pipeline of materials engineering and spectroscopy. Of particular interest is chalcogen and halogen based defects, which can have small ionic radii, allowing for efficient incorporation into diamond. For example, there are some preliminary reports of oxygen-based defects in diamond [8], but these assignments are not conclusive, and until now their quantum coherence properties have not been investigated. We have recently implanted oxygen into diamond, and are starting to perform bulk characterization now.

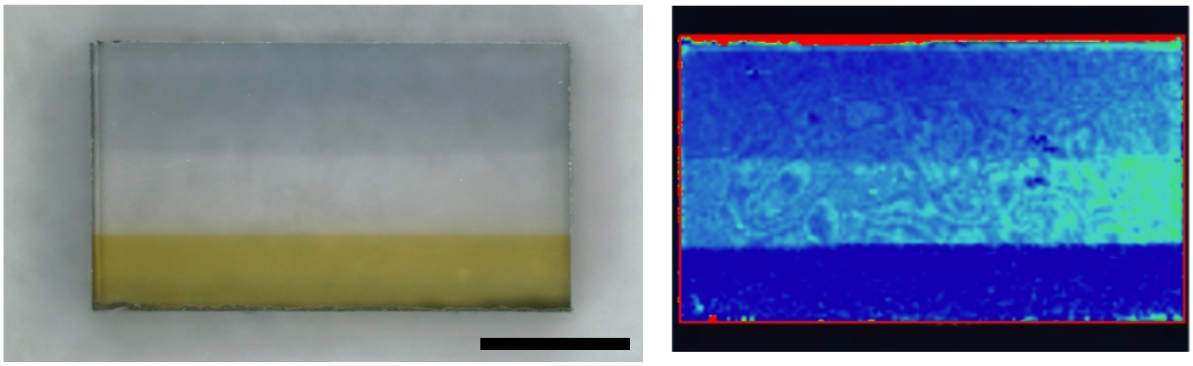

Figure 3 Diamond sample with different doping concentrations of boron, implanted with Ge ions. The bottom yellow bar is a low purity HPHT layer, the center is a high purity layer, the top blue bar is a lightly boron doped layer. Scale bar represents 1mm. (Right) Photoluminescence mapping at 602 nm shows that GeV^- emission is suppressed in the boron doped layer.

Finally, in collaboration with Matthew Markham at Element Six, we have been working to optimize the synthesis of high purity boron-doped diamond. The main challenge is that boron doping tends to lead to aggregation of other impurities, and the use of boron precursors contaminates a microwave CVD chamber, precluding other growth chemistries. Markham has commissioned a new, dedicated reactor for this purpose, and we are currently implanting a range of samples with Si to characterize the properties of SiV^0 centers in these substrates. With optimized high purity boron-doped diamond, we will employ a similar strategy to our work with SiV^0 to systematically explore new charge states of other known color centers, such as other group-IV vacancy complexes. Preliminarily, we have implanted Ge ions to try to study the neutral GeV center. So far we observed suppression of the negatively charged GeV (Figure 3), but we were unable to identify any new spin or optical transitions related to the new charge state. We hope to study this further with new, optimized boron-doped substrates.

Answers to specific reporting questions

New discoveries, inventions, or patent disclosures:

Do you have any discoveries, inventions, or patent disclosures to report for this period?

N. P. de Leon, B. C. Rose, D. Huang, Z-H. Zhang, A. M. Tyryshkin, S. Sangtawesin, S. Srinivasan, L. Loudin, M. L. Markham, A. M. Edmonds, D. J. Twitchen, and S. A. Lyon, "Synthetic engineered diamond materials with spin impurities and methods of making the same," WO2019055975, 2019.

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B. C. Rose, D. Huang, Z.-H. Zhang, P. Stevenson, A. M. Tyryshkin, S. Sangtawesin, S. Srinivasan, L. Loudin, M. L. Markham, A. M. Edmonds, D. J. Twitchen, S. A. Lyon, and N. P. de Leon, "Observation

of an environmentally insensitive solid-state spin defect in diamond," *Science*, vol. 361, no. 6397, pp. 60–63, Jul. 2018.

B. C. Rose, G. Thiering, A. M. Tyryshkin, A. M. Edmonds, M. L. Markham, A. Gali, S. A. Lyon, and N. P. de Leon, "Strongly anisotropic spin relaxation in the neutral silicon vacancy center in diamond," *Phys. Rev. B*, vol. 98, no. 23, p. 235140, Dec. 2018.

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