Effect of Synthesis Techniques on the Creation of Nitrogen-Vacancy Centers in Diamond

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Abstract:

Nitrogen-vacancy (NV) centers are defects in diamond's lattice structure whose properties enable its application to quantum sensing. NV diamond quantum sensing has numerous applications including wide-field magnetic imaging, biosensing, geosensing, and quantum information systems. To make robust NV diamond sensors, they must contain a relatively high concentration of NV-centers. This project studies the effects of NV diamond synthesis and treatment techniques, specifically post annealing temperatures, on NV diamond composition with a goal of optimizing growth parameters to create better quantum sensors.

Summary of Research:

NV centers form when a substitutional nitrogen is adjacent to a vacancy. This substitutional nitrogen (Ns) donates an electron to a neutral NV (NV0) center, creating a negative NV center (NV-) (Eq 1) [1]. A high concentration of NV-centers in diamond is necessary to make robust quantum sensors. The concentration of NV-centers and other nitrogen-related impurities can be controlled during synthesis and treatment processes. In this research, NV diamonds were synthesized using chemical vapor deposition (CVD). CVD uses hydrocarbon plasma and nitrogen gas to grow NV diamonds on a high-pressure and high-temperature NV diamond substrate [2]. CVD grown diamonds have a small concentration of NV centers compared to total nitrogen concentration; therefore, a treatment process of electron-beam (EB) irradiation and annealing is required. EB irradiation creates vacancies with a negative or neutral charge state. This concentration of vacancies is approximately equivalent to the diamond's nitrogen concentration [1]. Annealing allows for these vacancies to diffuse, causing them to be captured by Ns to form NV centers. Previous studies have shown that vacancies begin to move at 700°C and nitrogen begin to move at 1600°C; therefore, a range of 800-1400°C is common for annealing [1]. The optimal annealing temperature is yet to be understood, which is the motivation of this research. A summary of NV diamond synthesis and treatment can be seen in Figure 1.

$$\text{Eq 1. } N_s^0 + NV^0 \leftrightarrow N_s^+ + NV^- \quad [1].$$

Methods. Three NV diamond samples of varying nitrogen concentration (8ppm, 4ppm, and 2ppm) were synthesized using CVD. Next, they underwent EB irradiation with an EB fluence of 5.0×10^{17} e/cm^2. Finally, the samples were annealed at three different temperatures: 700°C, 1000°C, and 1400°C to determine the optimal annealing temperature. The samples were characterized after each synthesis step using optical images, photoluminescence (PL), and Fourier transform infrared (FTIR) spectroscopy. Optical images indicated external changes of the diamonds after each step, PL measured the NV-and NV0 intensity, and FTIR spectroscopy measured the Ns0 and Ns+ concentrations. After measurements, Ns0/(Ns0+Ns+) and NV-/(NV0+NV-). To confirm PL and FTIR results, a new sample with a nitrogen concentration of 8 ppm was characterized. This characterization was done using electron paramagnetic resonance (EPR) spectroscopy to measure the concentration of Ns0, negative vacancy (V-), and NV-at each synthesis step.

Results. The optical images of the samples indicated that the diamond became pink after 1000°C annealing (Figure 2). This likely occurred because vacancies diffused toward Ns to create NV centers. Additionally, it can be seen that the sample with higher nitrogen concentration (8ppm) was darker pink than the sample with lower nitrogen concentration (2ppm). The FTIR and PL results of the samples after each synthesis step can be seen in Figure 3. The FTIR results indicate that after EB irradiation there was a decrease in Ns0 concentration, and after annealing Ns0 concentration slightly increased. Based on Figure 1 and Eq 1, this is expected because Ns0 is used to create NV centers. Furthermore, an increase in nitrogen concentration in the sample

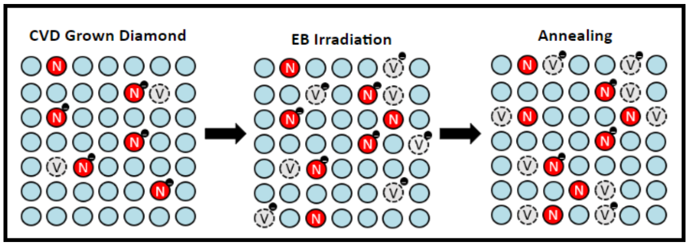

Figure 1: NV diamond synthesis and treatment procedure. Red is nitrogen and gray is a vacancy. After EB irradiation vacancies are formed, and after annealing the vacancies are diffused near Ns to create NV centers.

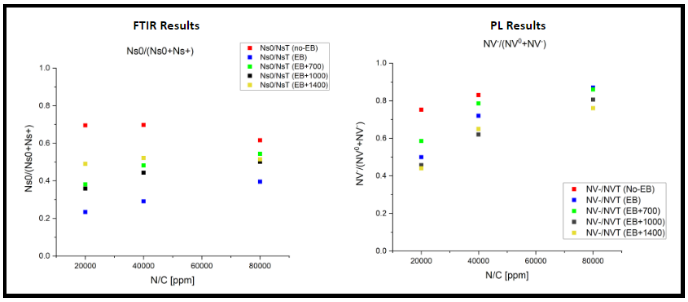

Figure 3: FTIR and PL results of each NV diamond sample after each annealing step. The unexpected data of the samples after CVD growth (No-EB) is likely due to temperature affecting measurements and a different analysis procedure used. No-EB measurements taken by Ms. Shoko Manako.

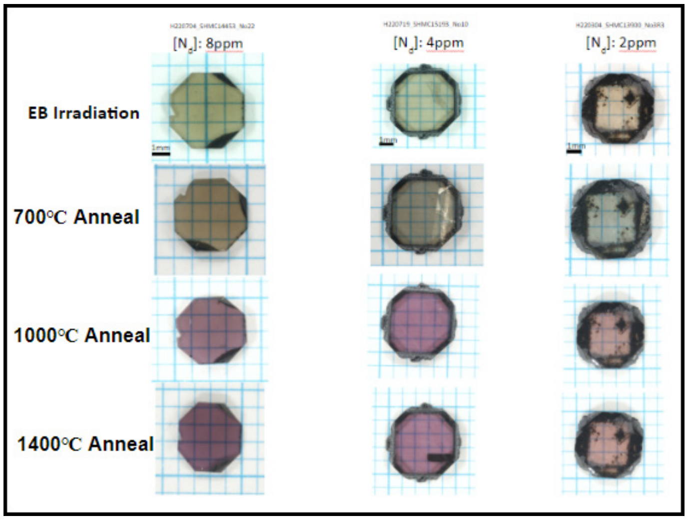

Figure 2: Optical images of NV diamond samples at each synthesis step. After annealing at 1000°C, the samples change from a yellow/brown color to pink.

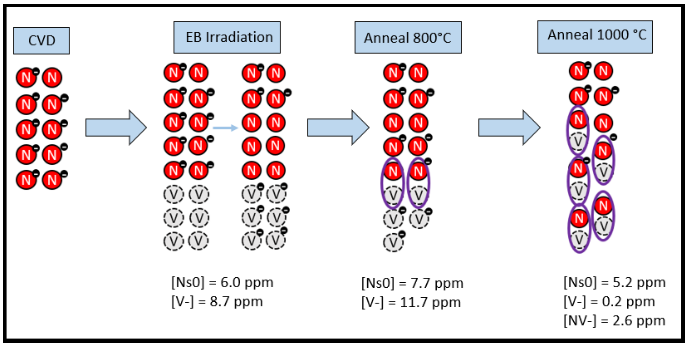

Figure 4: EPR results of an 8ppm diamond sample confirmed that the optimal annealing temperature is 1000°C. Red is nitrogen, gray is a vacancy, and purple is an NV center. Measurements taken by Dr. Chikara Shinei.

resulted in a small change of Ns0 to total Ns after each synthesis step. The PL results indicated that after each step NV0 and NV- intensity increased. To make a robust NV diamond sensor, a high concentration of NV-compared to NV0 concentration is required. Based on this requirement, the PL data concluded that an annealing temperature of 1000°C is optimal. The PL data of the No-EB sample has a very high intensity, which is unexpected. This was likely due to the data being taken prior to this project when a different analysis procedure was used. Finally, EPR data of a new 8 ppm NV diamond sample was used to confirm results of PL and FTIR (Figure 4). After EB irradiation there was a high concentration of Ns0 and V-, annealing at 800°C increased the concentration of V-compared to Ns0, and finally annealing at 1000°C resulted in a low concentration of V-and a detectable concentration of NV-. This confirmed the FTIR and PL results that 1000°C is the optimal annealing temperature.

Conclusions and Future Work:

The results from this research determined that the optimal annealing temperature for NV diamond synthesis is 1000°C. Additionally, the FTIR and PL results show that a higher concentration of nitrogen results in a higher concentration of NV centers. Using this data, the synthesis of NV diamonds can be tailored based on their desired application, leading to the creation of better quantum sensors. To improve this research, a new set of samples grown at the same concentration should be studied to confirm these results. Furthermore, studying a new set of samples will help confirm the unexpected PL data that was measured after CVD growth. Finally, to better understand the properties of the samples and how they can be used as optimal quantum sensors, their coherence properties should be measured using optically detected magnetic resonance.

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References:

[1] Shinei, C., et al. (2021). Equilibrium charge state of NV centers in diamond. Applied Physics Letters, 119(25), 254001. https://doi.org/10.1063/5.0079687.

[2] Barry, J., et al. (2020). Sensitivity optimization for NV-diamond magnetometry. Reviews of Modern Physics, 92(1), 015004. https://doi.org/10.1103/RevModPhys.92.015004.