Cathodoluminescence and Spin-Dephasing Time of Nitrogen Vacancy Centers in Diamond

IRTE Intern: Anais El Akkad

Affiliation(s): School of Physics, Georgia Institute of Technology

Program: Global Quantum Leap International Research Training Experience (IRTE)

Funding: NSF AccelNet Program via OISE-2020174

Project Host: Dr. Tokuyuki Teraji, Group Leader, Semiconductor Defect Design Group, National Institute for Materials Science, Tsukuba, Japan

Contact: aelakkad@gatech.edu, teraji.tokuyuki@nims.go.jp

Abstract:

Nitrogen-vacancy (NV) centers are defects in diamond with unique properties well-suited for robust quantum sensing. Imperfections in the host diamond, however, limit the sensitivity of NV sensors. This project characterizes the relationship between the material properties of nitrogen-doped CVD grown diamonds and the spin-dephasing time of negatively charged NV centers within the samples.

Summary of Research:

NV centers are defects in the diamond crystal lattice involving a substitutional nitrogen atom adjacent to a vacancy. NV centers in diamond have attracted significant interest as a platform for quantum sensing, as they have numerous desirable properties including long coherence times, the possibility of room temperature operation, and robustness in extreme conditions, including high pressures (up to 60 GPa) and high temperatures (up to 600 K) [1]. However, diamond material properties such as the presence of other defects, inhomogeneous strain distribution, and lattice dislocations pose challenges to NV-based quantum sensing [2].

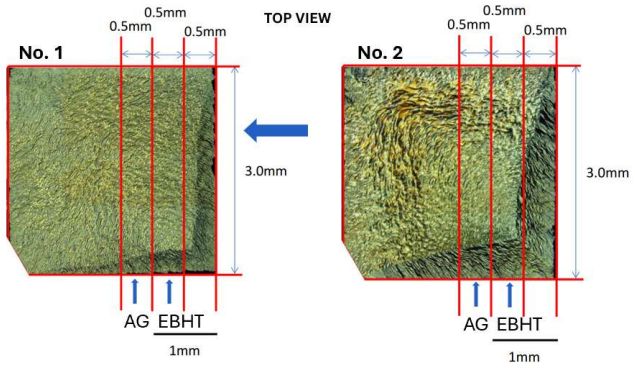

Figure 1: Top surface view of diamonds no. 1 and no. 2. Two samples were characterized from each diamond, with one left in its original as-grown state (AG), and one irradiated with a high energy electron beam and annealed (EBHT).

While NV centers can occur naturally, they form at relatively low concentrations in natural diamond [3]. Thus, diamonds for NV-sensing applications are typically nitrogen-doped and synthesized using either high-pressure high-temperature (HPHT) synthesis, or chemical vapor deposition (CVD) diamond growth. While HPHT synthesis results in diamonds with less overall strain, nitrogen concentration control is limited. Conversely, CVD diamonds grown to a thick (> 10 μm) layer typically exhibit higher strain, however lower nitrogen doping concentrations can be achieved [4].

Four samples from two diamonds grown via CVD on an HPHT substrate were characterized. The diamonds were grown with differing nitrogen concentrations (~0.8 ppm and ~0.2 ppm for samples no. 1 and no. 2, respectively) and isotopically enriched with 12C in order to limit undesirable nuclear spin interactions. The diamonds were then cut into three 0.5 mm wide samples. One sample from each diamond was subsequently irradiated with a high energy (2 MeV) electron beam and annealed at 1350°C for 2 hours. The samples having undergone this procedure are referred to as EBHT (electron beam irradiated and high temperature annealed), and they were compared to another sample from each diamond left in its as-grown state (AG). Characterization was performed from the right-hand cross-section, as indicated by the arrow in Figure 1.

Cathodoluminescence (CL) spectroscopy and imaging was performed on the samples in order to characterize key material properties, such as the presence of other defects and strain. CL is particularly interesting because it is not as well-studied as other spectroscopic techniques, offers a high spatial resolution, and can shed light on NV center charge state dynamics. Performing CL with a very fine grating resulted in the detection of negatively charged NV centers (NV-) for both EBHT samples, as indicated in Figure 2, which is rarely reported in literature [3]. This detection furthers our understanding of what conditions, both in measurement and in diamond material properties, are necessary to observe NV- under electronic excitation.

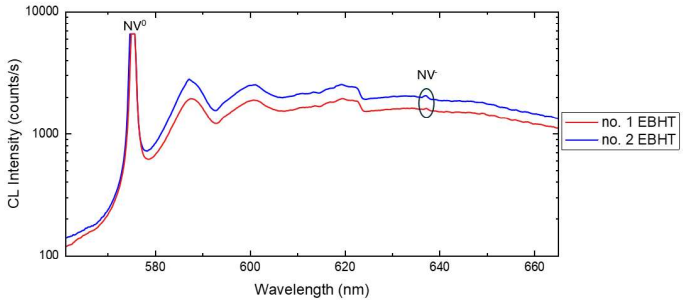

Figure 2: CL spectrum of samples no. 1 EBHT and no. 2 EBHT, obtained with an accelerating voltage of 10 kV, a probe current of 7.2 nA, x1000 magnification, and a grating of 300 g/mm.

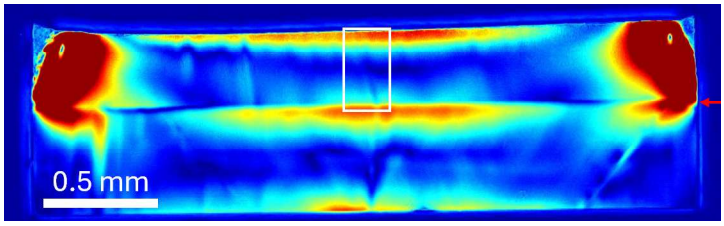

Figure 3: Birefringence image of sample no. 1 AG.

Birefringence imaging shed further light on the strain distribution within the samples. A birefringence image of sample no. 1 AG (Figure 3) shows dislocations originating from the substrate, which were also observed in CL imaging. Unexpectedly, this image also revealed a region of high strain near the CVD-substrate interface in the HPHT substrate, yet low strain in the CVD diamond layer nearest the interface. A potential explanation of this phenomenon may be due to the mismatch in lattice parameter between the substrate and CVD-grown diamond, caused by the much higher concentration of nitrogen in the substrate than in the CVD layer.

Finally, pulsed optically detected magnetic resonance (ODMR) was used to measure the spin-dephasing time (T2*), which limits the sensitivity of NV- to magnetic fields. T2* mapping performed across sample no. 1 EBHT revealed a correlation between regions of relatively lower strain and higher spin-dephasing times. As shown in Figure 4, the regions with the highest T2* tended to be the regions with the lowest strain. Interestingly, such regions seemed to be constrained to within 100 µm of the CVD-substrate interface. There is a decline in crystal quality as the thickness of CVD-grown diamond is increased, which may explain this phenomenon.

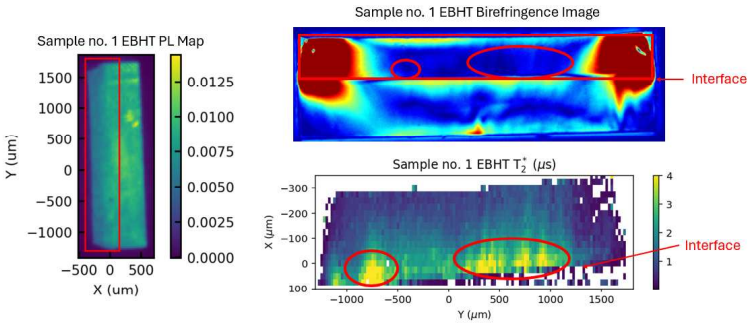

Figure 4: Photoluminescence (PL) map, birefringence image, and T2 map of sample no. 1 EBHT. PL mapping was first performed in order to differentiate the CVD layer from the substrate.

Conclusions and Future Steps:

In summary, the detection of NV- has been reported under electronic excitation, and lower strain and higher T2* was observed in CVD diamond near the CVD-substrate interface. Future work includes measuring T2* in the lower nitrogen concentration sample, as well as attempting CVD diamond growth on a substrate with a lower nitrogen concentration. Such work supports efforts towards diamond growth with optimal properties for NV- magnetometry.

References:

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