

Fluorescence lifetime analysis and mapping of a H rich diamond

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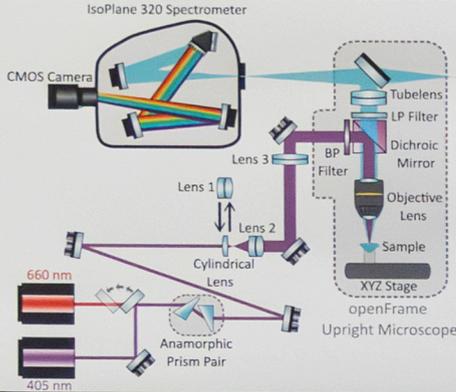
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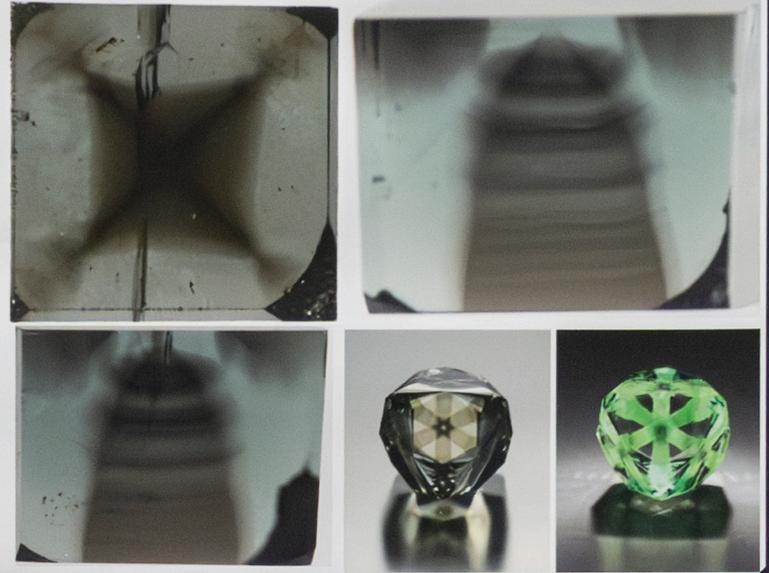
1. Instrument and motivation

- A custom-built instrument at GIA was developed to measure the lifetime of color centers in diamond on picosecond (ps), to millisecond (ms) times scales. Lifetime is the measure of how long the electron will stay in its excited state before it decays and releasing a photon of light.
- This instrument can perform lifetime mapping, for observing the spatial distribution of lifetime, and perform lifetime analysis as a function of spectrum. This allows us to analyze how the lifetime changes with different color centers which emit with different wavelengths (colors).



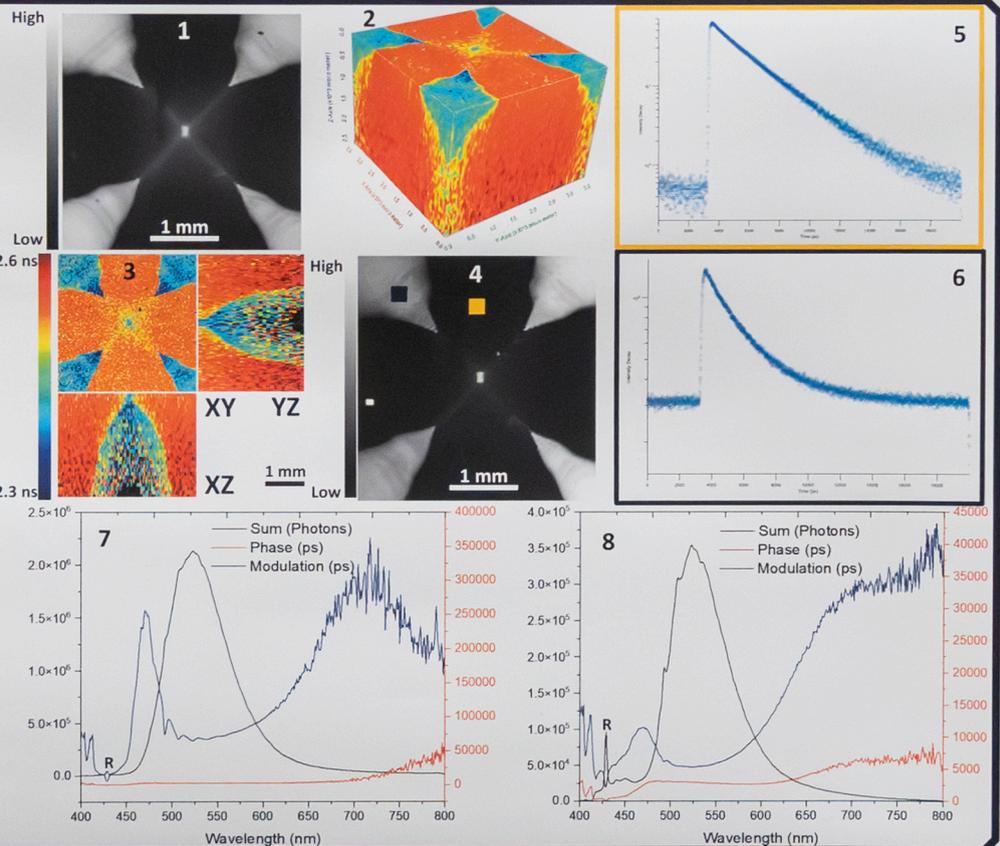
2. Sample information and history

- The sample studied is a 0.92ct diamond cut into a cube for study. It is a natural diamond crystal cut from a rough diamond of Zimbabwe origin subsequently heated at 1500°C for 5 minutes and irradiated at 3MeV for 10 minutes
- The diamond is a type Ia hydrogen rich diamond with a stellate hydrogen cloud in the center of the cube seen below in the mapping results. Often these type of diamonds are polished octahedra or faceted to showcase these clouds (see example, 1).



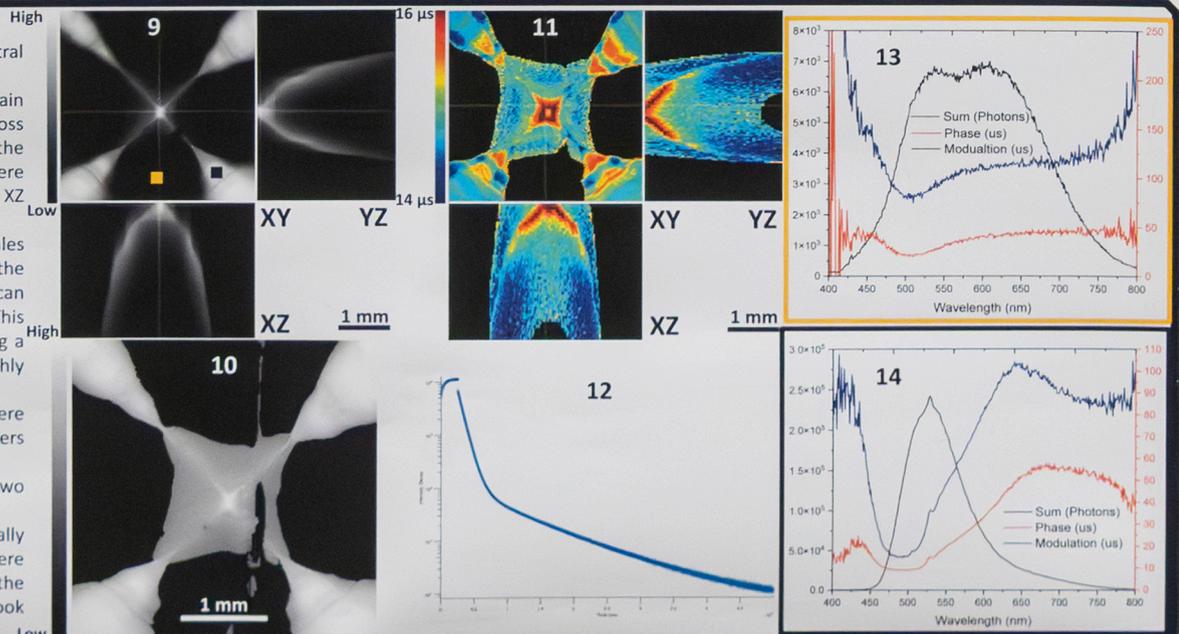
3. Fluorescence time scales

- On fluorescent times scale, on the order nano seconds (ns) we can measure the decay of light intensity, which allows us to calculate the lifetime of these color centers. This is accomplished by using a monochromator to remove all light apart from a thin spectral section, typically <1 nm in width and measure the decay. If multiple color centers are emitting, and the lifetime of each is known, we can calculate the fraction of each. In this case, we do not know their lifetime, and so only an average is calculated.
- Spectrally resolved decays to extract lifetime can be measured, as shown in the graphs, or we can take a single spectral band and map over the sample, collecting a decay at each point.
- In Fig 1, we show the total intensity of emission at 500 nm, indicating that the bulk of the light comes from the stellate clouds. This is the surface of the sample.
- In Fig 2, we show the 3D representation of the full cube as measured with lifetime, where the color represents the mean lifetime. In Fig 3 we show 3 images as cross-sections of the data cube, showing that the range of mean lifetime is from 2.3 to 2.6 ns, suggesting multiple emitters or quenching. The darker regions of the sample have a more consistent lifetime, suggesting 1 emitter, or uniform quenching, whilst the diagonal regions have a reduced lifetime, suggesting multiple emitters or variations in quenching.
- Graphs 5 and 6 show example decays taken from the orange and blue squares in Fig 4, where it is clear most of the photons in bright diagonal cross (6) come from a large fixed value background, suggesting emission from a longer-lived color center. In graph 5 we see a mostly monoexponential decay, suggesting a color center with a lifetime near 2.6 ns. There is very little background, suggesting no long-lived color centers. This is in agreement with the polished octahedral data shown in box 2, where the bulk of the emission comes from the clouds.
- The spectrally resolved lifetime graphs in 7 and 8 show the intensity spectra, and the average lifetime using two different methods. One is the phase lifetime which is very sensitive to small changes in lifetime around the ns range, whilst the modulation lifetime is less sensitive to ns changes, but more sensitive to larger changes in the lifetime 100's ns. These reveal a broad spectra hump in the green with a longer lifetime (matching emission from the octahedral stone), and sharp peaks with a shorter lifetime.



4. Microsecond time scales

- On microsecond time scales we start to see different spectral shapes and further contrast in the stellate clouds.
- In Fig 9 we see the intensity at 525 nm emission, showing again that the bulk of the emission originates from the diagonal cross stellate clouds. The funnel shape shows more emission from the outside due to the cloud absorption light, as the images were collected from the top down XY view, which is vertical in the XZ view, so the cloud obscures.
- In Fig 11 we see the mean lifetime on microsecond time scales which goes from 14 to 16 μs, with a complex distribution on the top surface. By looking at the cross-sections in XZ and YZ we can see that the lifetime is higher towards to top of the sample. This is where the green color from radiation is highest suggesting a color center with a longer component is more highly concentrated there.
- In Fig 10 we see the background emission, suggesting that there are subtle differences in the even longer-lived color centers around the stellate cloud.
- In graph 12 we see an example decay showing distinctly two components with very different lifetimes.
- In graph 13 and 14 we again see intensity spectra and spectrally resolved phase and modulation lifetime. This reveals that there is complex emission occurring both in the dark regions of the sample and in the stellate clouds. We note that the spectral look much different to those on ns time scales.



Conclusions

- This hydrogen rich diamond revealed many interesting contrasting in 3D lifetime analysis, showing that despite looking uniform to the naked eye under specific excitation, there are actually multiple different color centers and potentially quenching occurring. Whilst the origin of these centers remains elusive, this demonstrates the power of lifetime imaging in 2D, 3D, and spectrally resolved.

Acknowledgements and References

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1 Faceted DIAMOND Octahedron With a Remarkable Stellate Cloud Inclusion
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