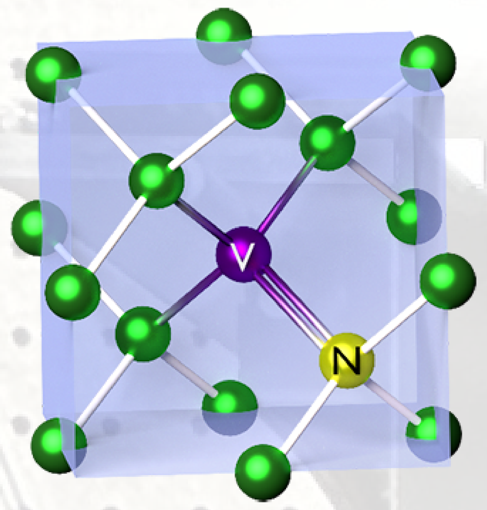

Exploring High Energy States in NV-Centers: Photoluminescence in Single-Point Defects in Diamonds

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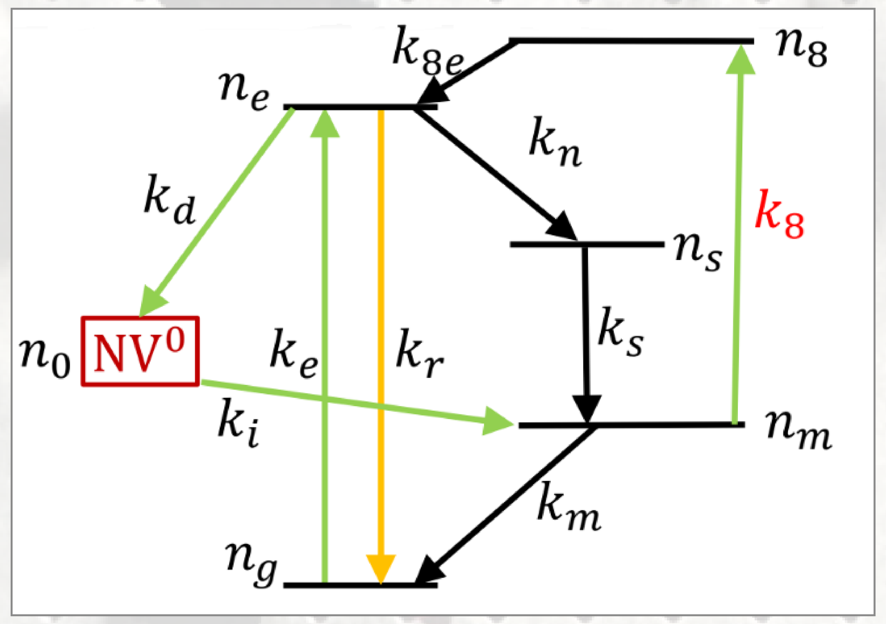
Thesis, Code & Data

Abstract

NV point defects in diamonds have been studied extensively, initially using low laser powers and the four-level model [1]. However, recent investigations by Ádám Gali through ab initio calculations in higher power regimes introduced a fifth level [2], to consider state switching between NV^- and NV^0.

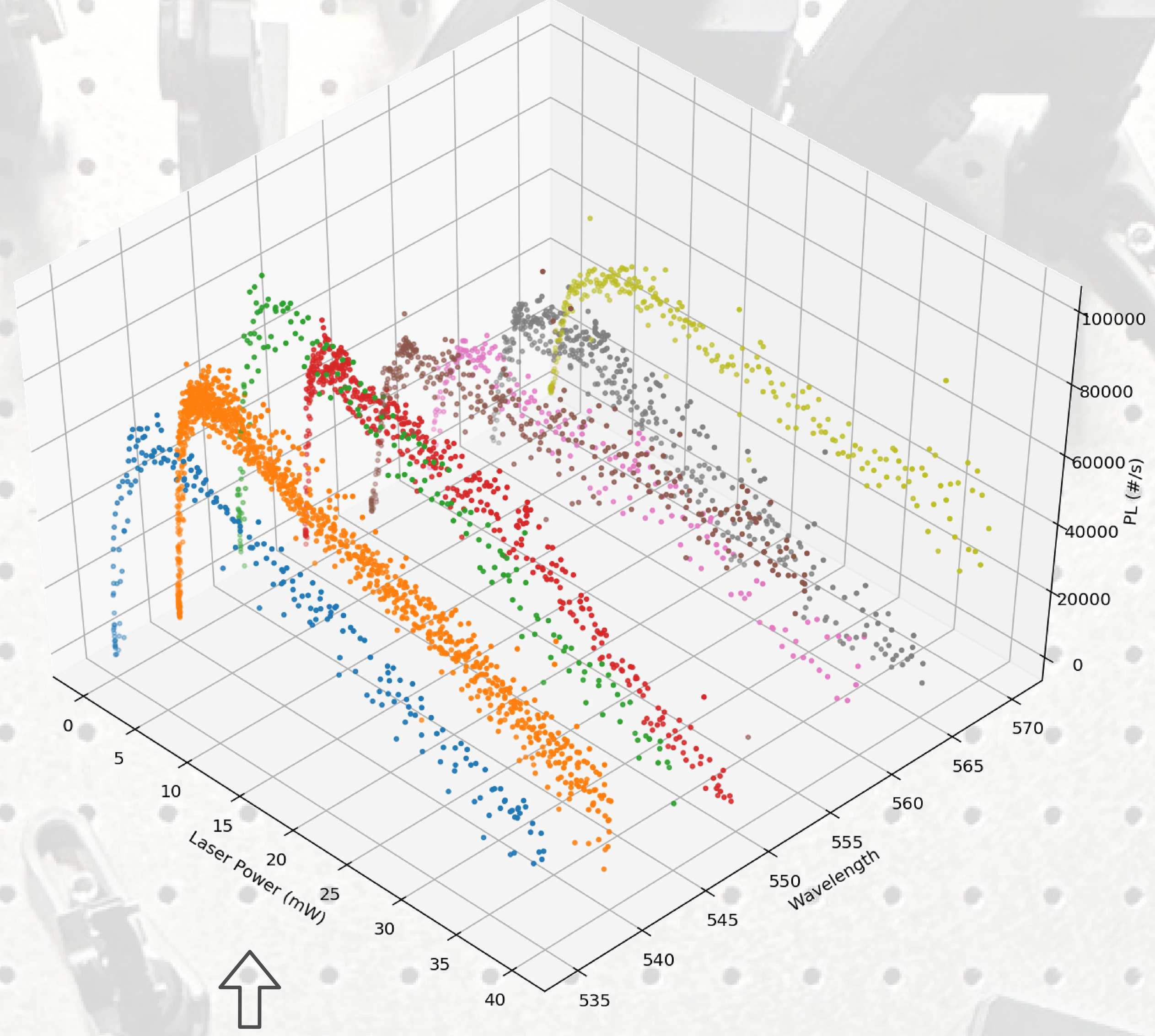
The theoretical model initially predicted zero photoluminescence (PL) at high laser powers, but ongoing experiments have indicated a possible high energy n_8-state, close to the conduction band, which would lead to non-zero asymptotic PL at high laser power. To shed light on this discrepancy, our research group modeled a NV-center with a six-level state, and derived a subsequent formula to calculate the population of the excited state at higher laser powers. Saturation curve measurements are obtained to study the validity and presence of this new high energy state. Setup refinements are required before making conclusions about the existence of state n_8.

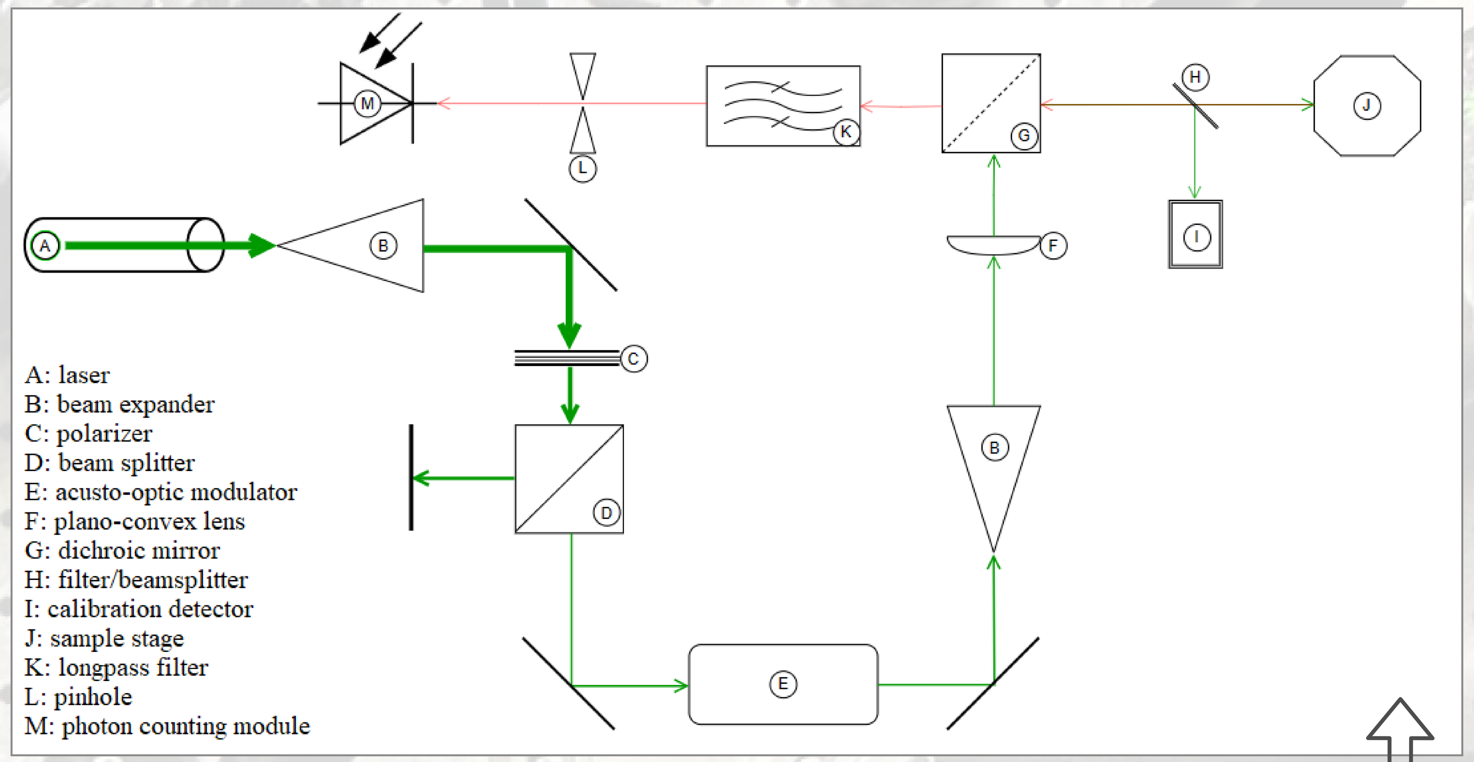
During measurements, PL changes were observed due to changes in applied voltage on the sample. The PL dependence on potential has been investigated extensively using Optically Detected Magnetic Resonance (ODMR) and Rabi Oscillations, which confirmed initialization issues when cycling from non-zero to zero voltages, which is something researchers should be taking into account in future research.



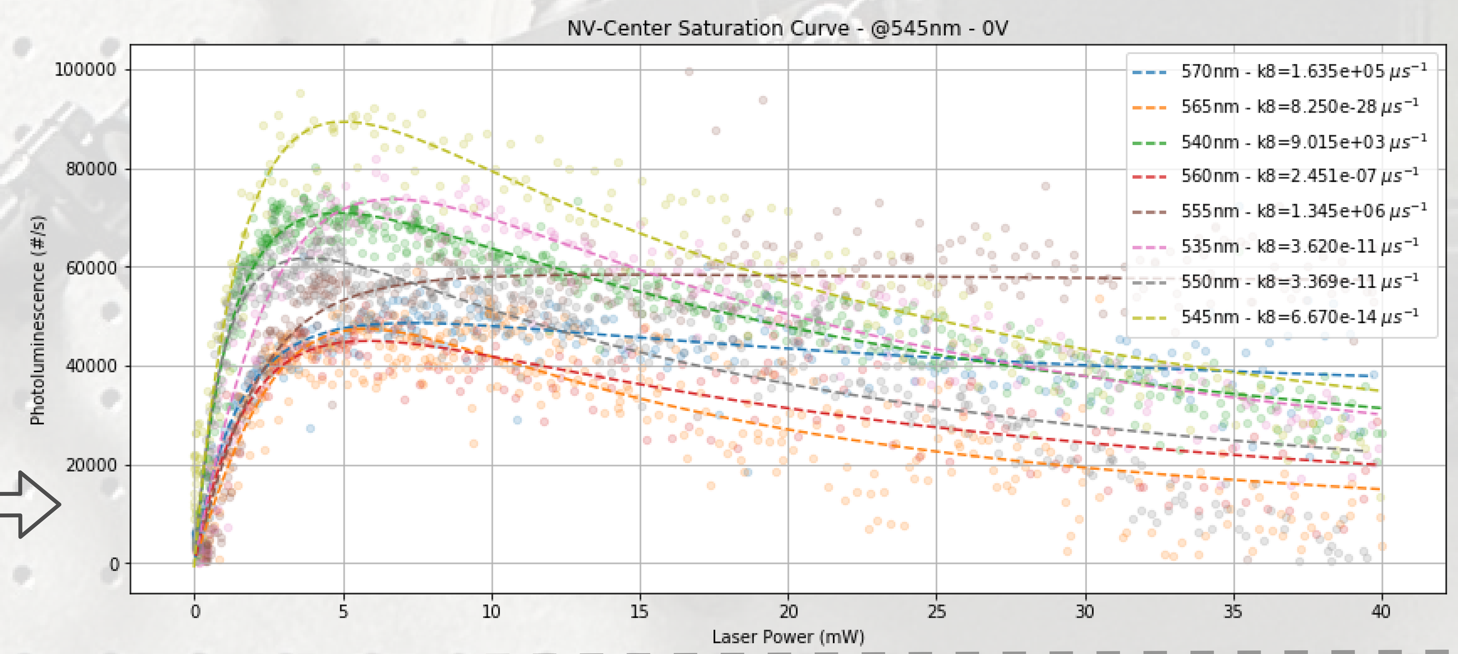
Using **density of states**, below equation has been derived to model PL in presence of a new high energy state, and used to obtain fits and values for k_8. Green arrows indicate laser-driven transitions.

$$n_e = \frac{1}{1+\frac{k_r}{k_e}+\frac{k_d}{k_m+k_8}+\frac{k_n}{k_m+k_8}}$$

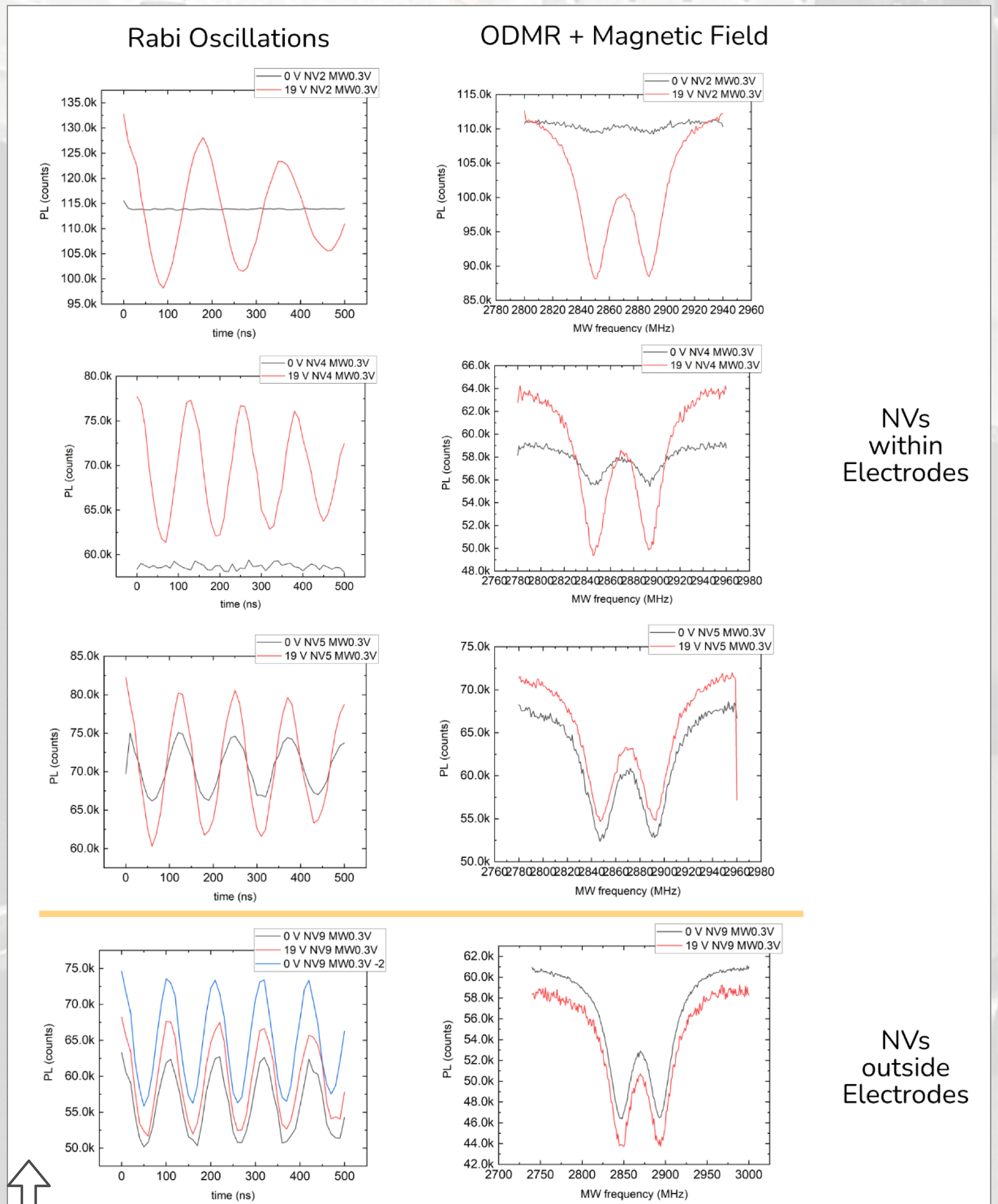




Confocal setup used to measure PL of NV centers.

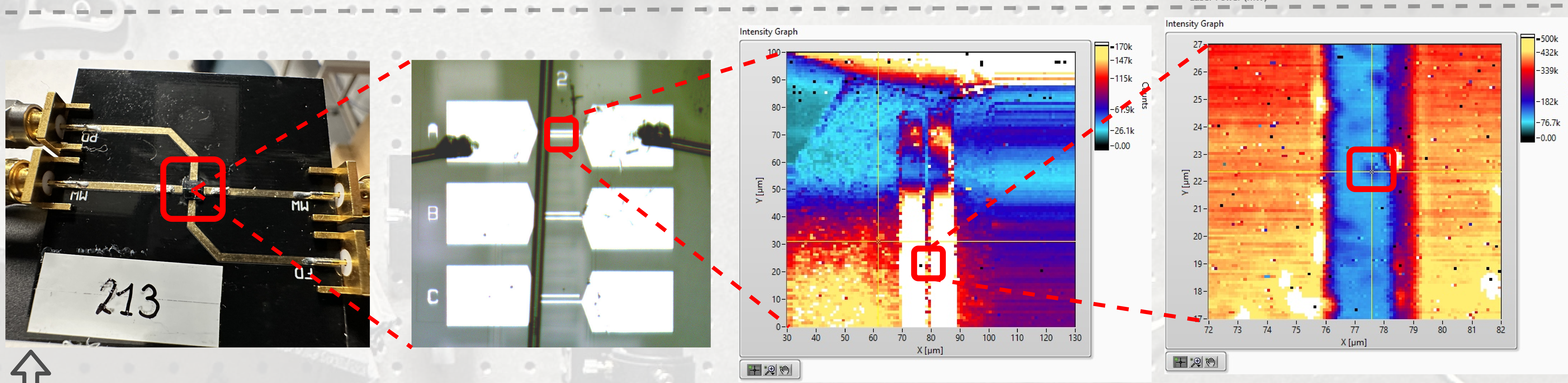


Saturation curves at different wavelengths (above) together with the obtained fits (right). Fitted values for k_8 are shown, which govern non-zero asymptotic behaviour of a saturation curve. Values for k_m, k_n and k_r are fixed and obtained from literature [3]. We note that our fitted k_8's tend to zero, just like the saturation curves at high laser power, which suggests an absence of the hypothesized n_8 state. Curves with a non-zero k_8 lack a maximum at low laser powers are attributed to measurement issues.

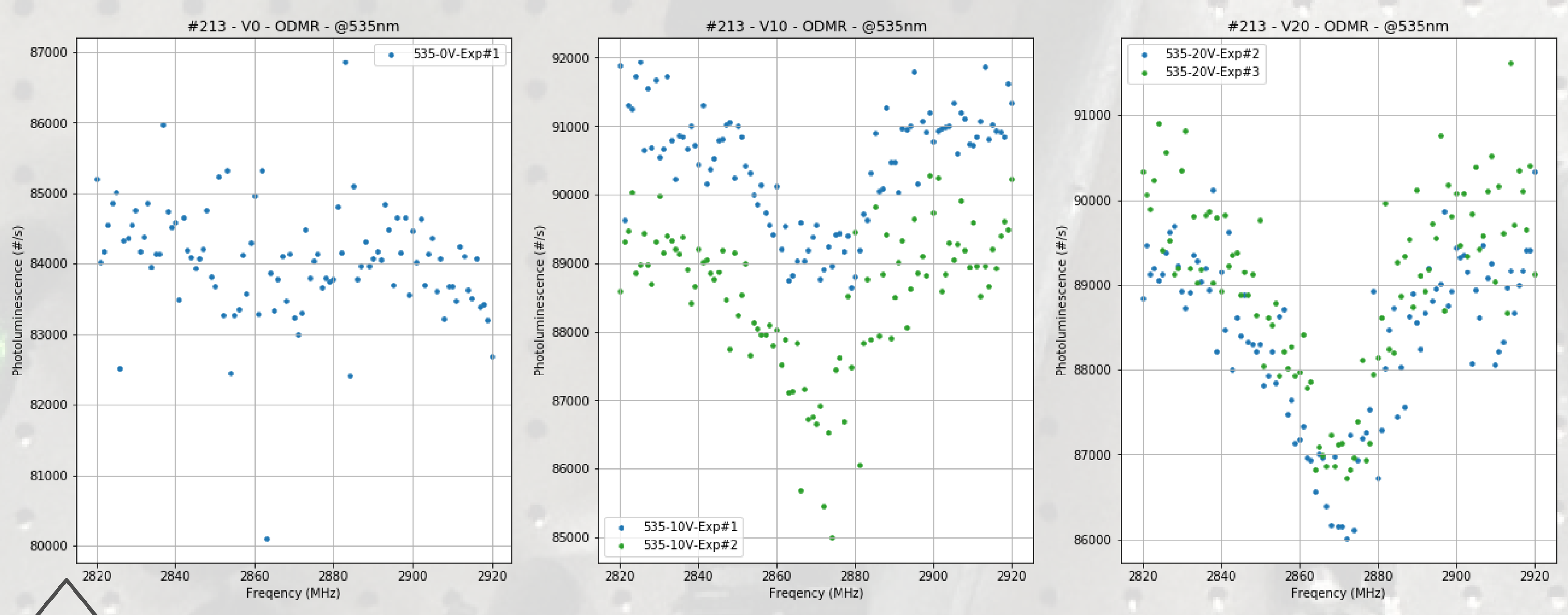


Rabi Oscillations and ODMR Measurements with applied magnetic fields are conducted, at 0V and 19V, both between and outside the electrodes. Outside the electrodes, the measurements' contrast remain constant throughout voltage cycling, whereas within the electrodes the PL contrasts show up to be inconsistent.

This highlights that different voltages change the environment of the NV center, and demonstrates initialization errors of the spin states when measuring PL between the electrodes.



To showcase the scale of operation, shown is a sequential zooming of sample #213 (5cm x 5cm). Bright pillars on the right represent electrodes. Measurements have been conducted between and outside of the electrodes.



ODMR measurements across different applied voltages (0V-10V-20V) have been conducted. Characteristic of an NV center is the resonance frequency at which PL dips (2871 MHz). When shifting potential from |5V| to 0V, we noticed that ODMR contrast is effectively eliminated, which is unexpected. For clarity: the contrast on the most left graphic should look like that on the other two graphs. This is the first indication of initalization errors of the ground state of an NV-center, when cycling between non-zero and zero potentials.

Conclusion

Our results indicate that k_8 exhibits a nearly zero value at most wavelengths, except for a few specific ones, which brings the existence of the n_8 state into question. This is further confirmed by analysing obtained saturation curves at high laser power: we see a clear saturation towards zero PL at most wavelengths. Deviation from theory and proposed high energy states in literature, could stem from setup errors (manual rotation of polarizer), limitations of the the suggested model, or inherent complexities within the NV center which aren't yet well understood. Further research is needed to address these uncertainties.

Moreover, our data demonstrates the influence of voltage on environmental conditions between electrodes, captured by ODMR and Rabi oscillations, which suggest specifically that the initialization of quantum state populations need to be carefully considered.

References

[1]Rondin, L., Tetienne, et al (2013). *Magnetometry with nitrogen-vacancy defects in diamond.* Reports on Progress in Physics, 77(5)
[2]Gali, Ádám (2019), *Ab initio theory of the nitrogen-vacancy center in diamond*, Nanophotonics, vol. 8, no. 11
[3] D. Wirtitsch, G. Wachter et al. (2023) *Exploiting ionization dynamics in the nitrogen vacancy center for rapid, high-contrast spin, and charge state initialization*, Physical Review Research