

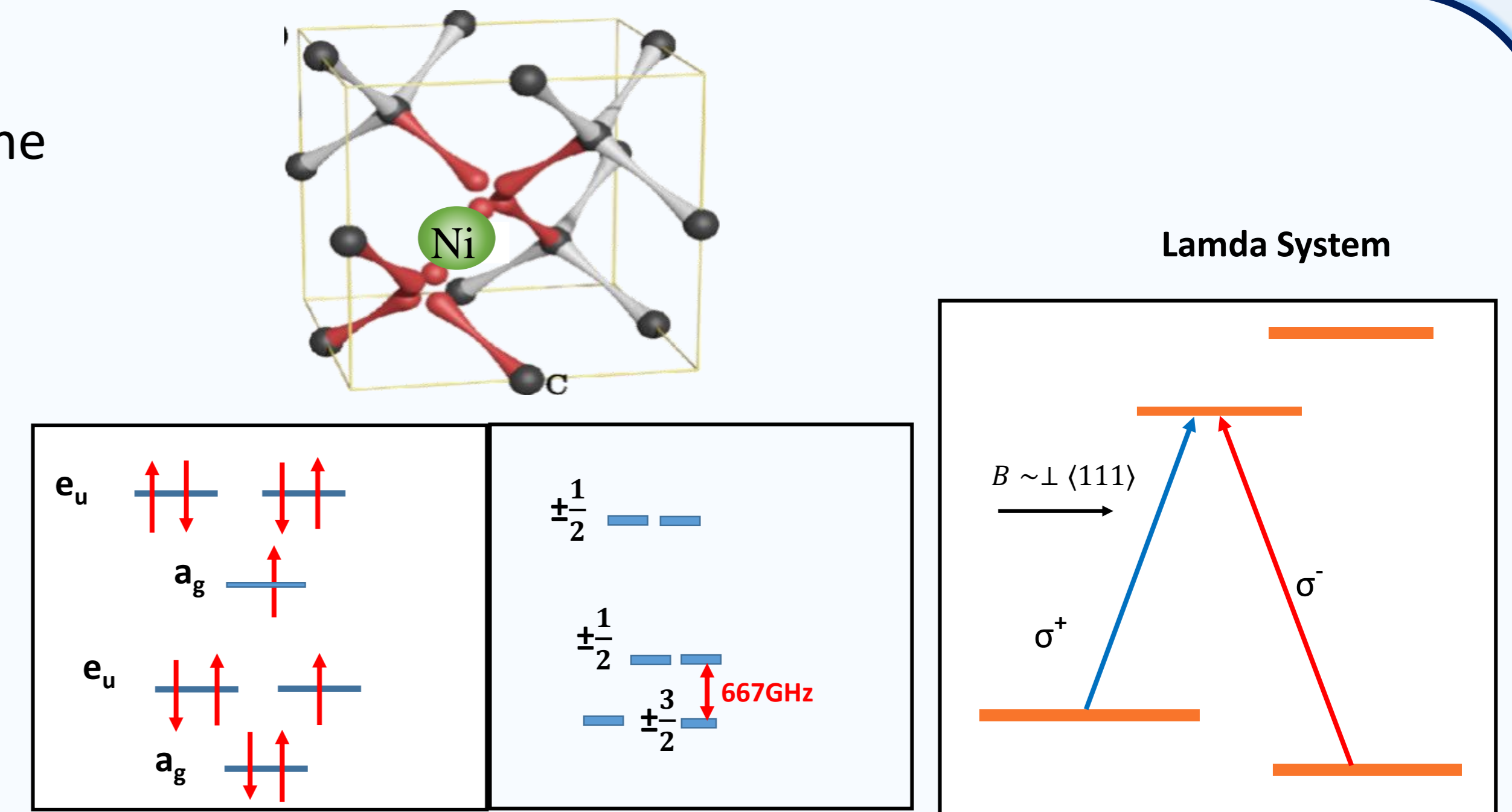


Abstract

We study **nickel-related color centers** in diamond towards their application as a **solid-state light-spin interface**. These centers have strong spin-orbit interaction, thus are potentially suitable for high-temperature operation. Using polarized resonant excitation **we observe signs of both spin and charge optical pumping at 10K**.

Background

- Diamond is rich in point defects that are able to change the optical and electrical properties of the host material, often called color centers.
- One of the prominent color centers in diamond is the nitrogen –vacancy (NV) defect.
- Nickel is a typical contaminant in high pressure high temperature (HPHT) diamonds.
- The 883/885 nm (1.4eV) center, is attributed to NiV⁻ (S=1/2)
- + 1/2 and -1/2 spin state couple through a single **polarized lambda system**.
- The spin-orbit interaction in the ground state is **~700 GHz** – equivalent to a temperature of 35K



Atomic and electronic structure of the nickel-vacancy defect in diamond

Rational

Goals:

- Spin pumping by polarized resonant excitation
- Stabilizing -1 charge state by off-resonant excitation

Research question:

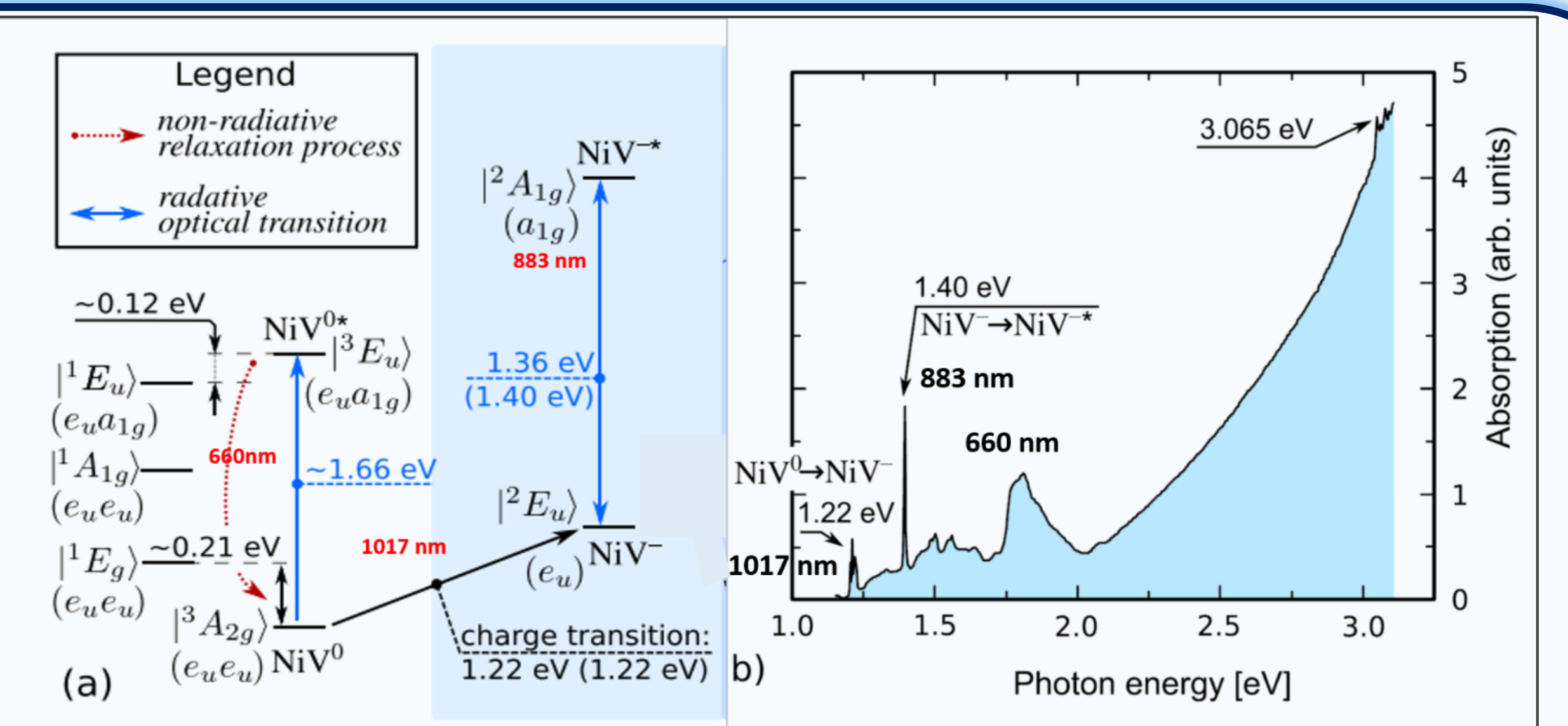
What wavelength and power are required for optimal charge stabilization?

- Candidates: 660 nm and 1017 nm (two sharp absorption resonances)

Experimental methods:

- Simultaneous and sequential two-color excitation
- Polarization pulsed excitation

Proposed model: Gergő Thiering, and Adam Gali. "Magneto-optical spectra of the split nickel-vacancy defect in diamond." *Physical Review Research* 3, 043052 (2021).



Sample and Experimental Setup

Sample

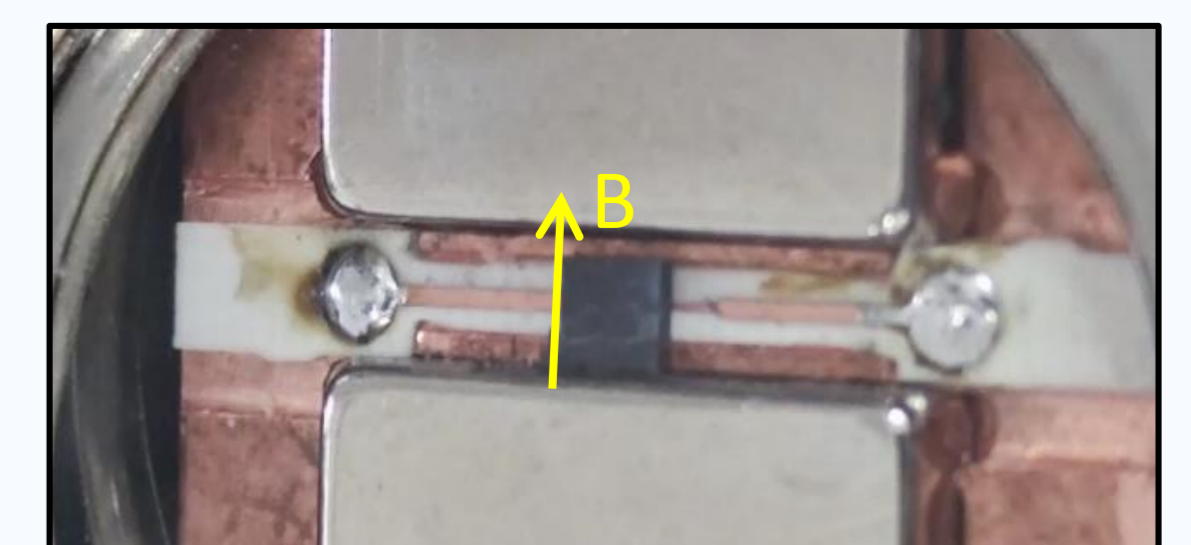
- The substrate is a <111> single-sector type IIb HPHT-grown artificial diamond (New Diamond Technology)
- Nickel was ion-implanted at 2 MeV at a dose of $5 \cdot 10^{12} \text{ cm}^{-3}$.
- The sample was then HPHT treated at 2000C and 8 Gpa for 2 hours.

Experimental setup

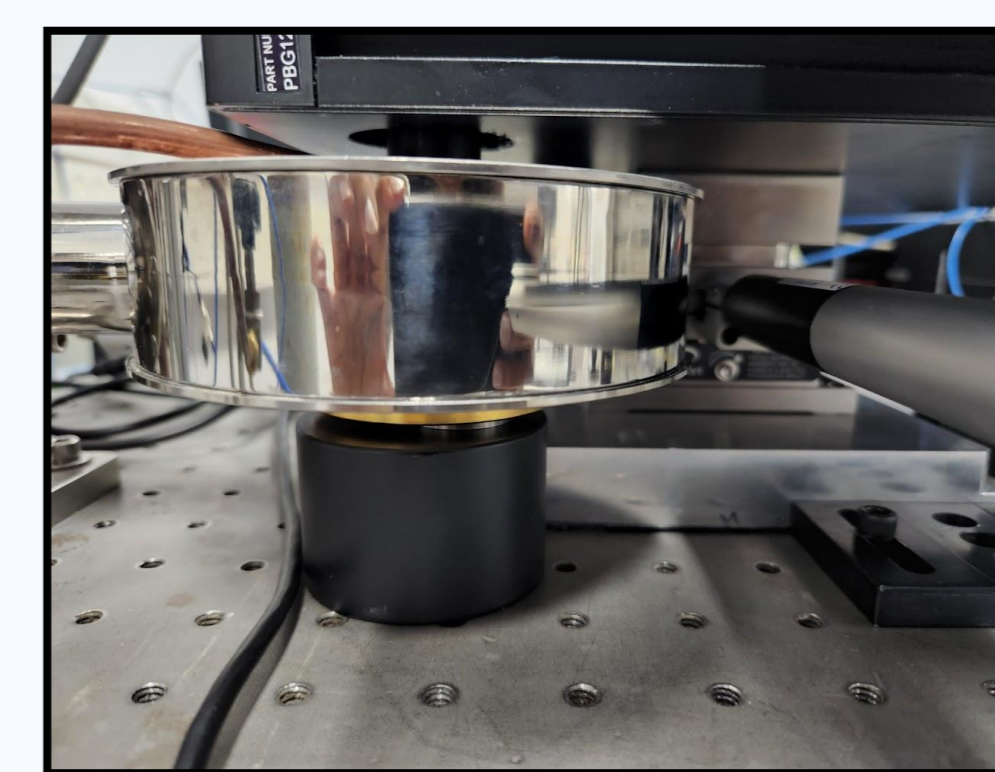
- 10K closed-cycle cryostat (ARS). Permanent magnets inside.
- Confocal microscope with three laser sources at 660 nm, 883 nm, and 1017 nm.
- 0.75 m spectrometer with a cooled ccd camera (Teledyne-PI)
- Avalanche photo detector and time-tagger electronics (Swabian Instruments)



Ni-implanted <111> diamond samples



IIb sample between the magnets inside the cryostat

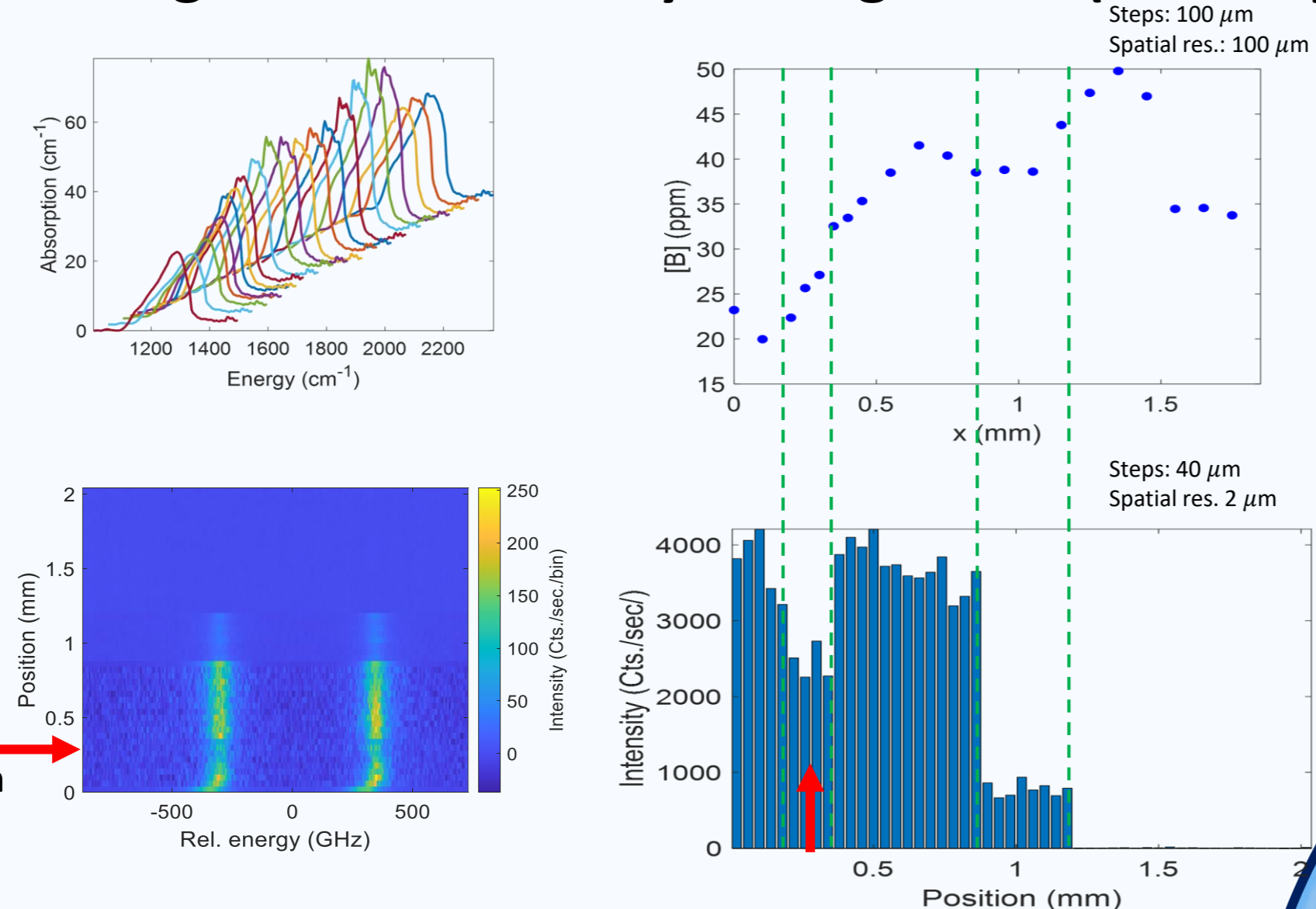


Microscope objective above sample chamber and magnet below it

Results

I. Controlling the charge environment by adding boron (an acceptor)

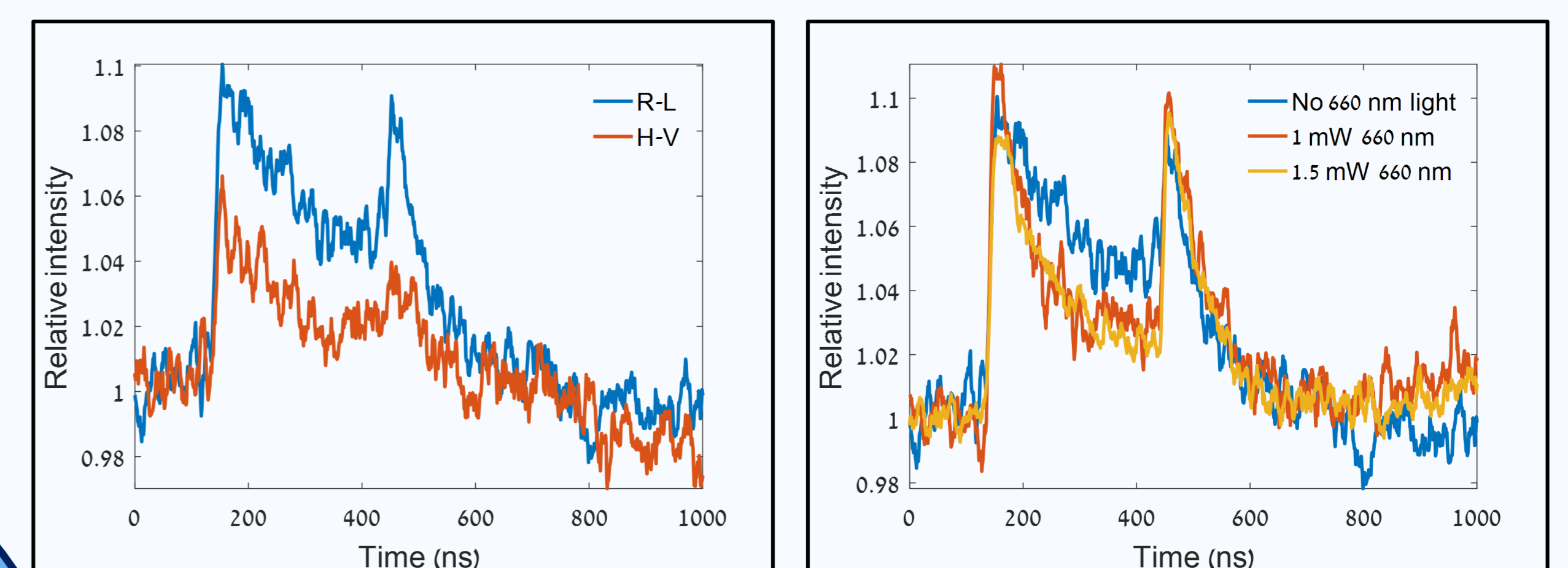
FTIR absorption: [B] varies across the sample: E_f lowers with [B]: the environment gets more positive.



1.4 eV emission decreases across the sample: it comes from a charge state that is unstable in a too-positive environment

III. Polarization pulse

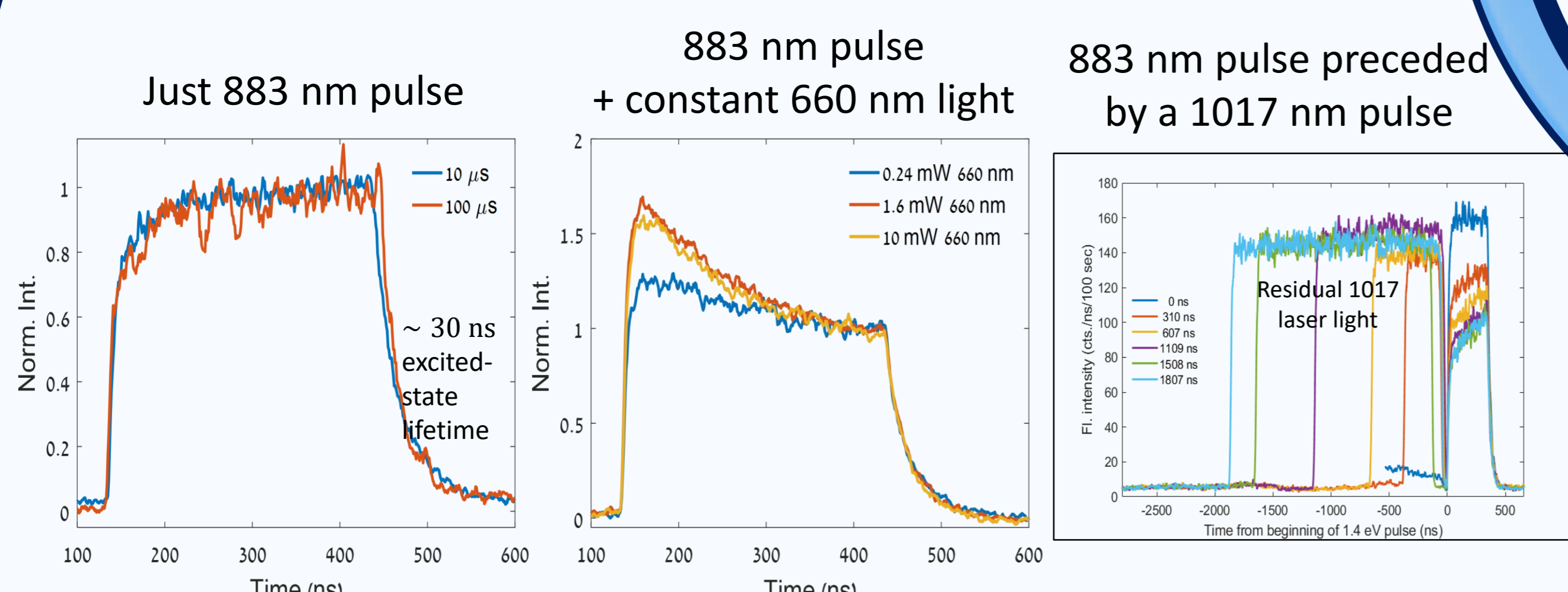
Change only the polarization of the 883 nm light



Polarization cannot affect charge state

→ The spikes are an evidence for spin pumping induced by circularly-polarized resonant excitation

II. Controlling fluorescence dynamics using an additional color



883 nm partially pumps out of the emitting state into a long-lived (>100 us) dark state. 660 nm light shortens the dark state's lifetime by pumping back into bright state. 1017 nm light pumps more population out of the bright state dark state.

Combined charge and spin dynamics

Conclusions and future work

Three options:

- Dark state is an opposite **spin state**, with very long lifetime. 660 scrambles the spin state.
- Dark state is a different **charge state**. 1017 charges to that state, and 660 charges back.
- There are **two dark states**, one of a different spin and the other of a different charge. 660 resets both.

How to discriminate?

- Introduce a 660 nm 'reset' pulse, just before the 883 pulse.
- Look for polarization dependence of the fluorescence dynamics.
 - If there's none, it's only charge pumping.
 - Otherwise, spin pumping is present as well, and their ratio can be extracted