

Zeeman Induced Ground State Crossing and Resulting Charge State Dynamics

Byoungchan Jang, Ryan Balili

Introduction

Nitrogen-vacancy (NV) centers are point defects in diamonds that have optical and quantum properties that are applicable in nanoscale biological imaging, quantum sensing, and quantum computation. A nitrogen-vacancy center in a diamond consists of a missing carbon in the lattice adjacent to a nitrogen impurity.

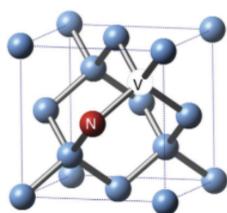


Figure 1. Diamond structure with a missing carbon (V) and an adjacent substitutional nitrogen (N). (Image: OrsayPhysics)

The NV defects often come in two possible charge states: the neutral charge state NV^0 and the negatively charged NV^- . When excited by green light, the NV defects in diamonds emit characteristic spectra shown in Figure 2. By applying the appropriate magnetic field, the energy levels of the electron states could be shifted via the Zeeman effect such that one level becomes preferentially populated over another.

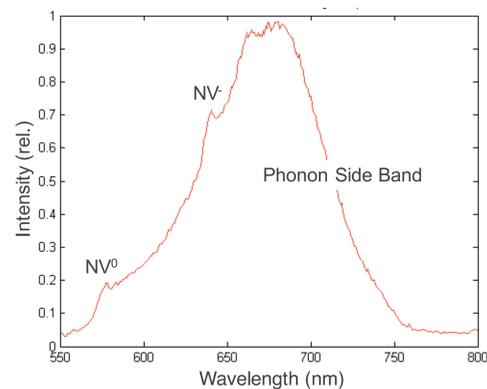


Figure 2. Spectra of the NV center in nanodiamond with its characteristics labeled.

Goals

1. Explore magnetic dependence of the transition between the two charge states of nanodiamond NV centers, the negatively charged NV^- and the neutrally charged NV^0 .
2. Determine which spin state electron dominates in the charge transition.

Methods

A combination of confocal microscopy and spectrometer was used to measure the spectra of the NV center with varying laser intensity and varying magnetic field. We used magnetic field to force the $m_s = -1$ spin state at the NV^- to cross with its $m_s = 0$ state. For this setup, two calibrations were needed: one for the spectrometer and another for the magnetic field. In order to calibrate the spectrometer, a halogen white light with a known intensity per wavelength was used. A Gaussmeter was used to calibrate the magnetic field of the magnet as a function of the distance from our sample diamond to the magnet. Once the calibration was done we measured spectra with varying green laser (532nm) power and magnetic field.

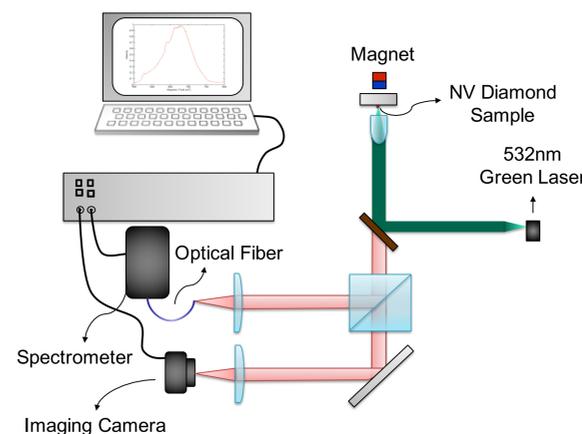


Figure 3. Confocal microscopy setup for fluorescence measurements.

Results

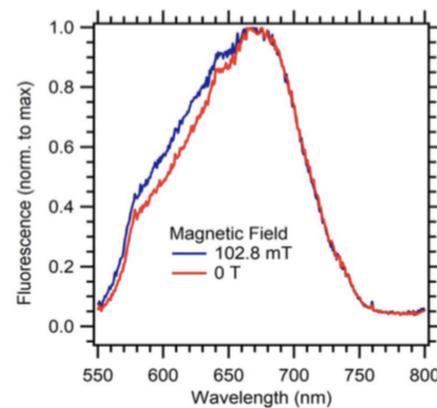


Figure 4. Normalized spectra with 0 mT and 102.8 mT magnetic field, comparing the shape of the spectra.

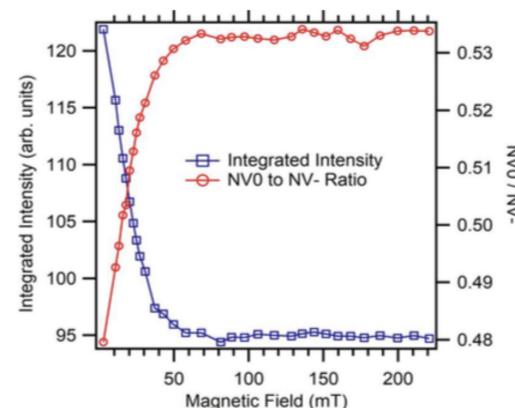


Figure 5. Integrated fluorescence intensity and the NV^0 to NV^- ratio of NV fluorescence as a function of magnetic field at 10 mW, continuous 532 nm laser excitation.

To figure out which charge state, $m_s = 0$ or $m_s = \pm 1$, contributes more to the NV^0 transition, we looked at the shape of the NV fluorescence with and without the magnetic field as shown in Figure 4. However, it is important to note that as soon as electrons are diverted to populate the $m_s = \pm 1$ states, a marked decrease in fluorescence is observed because large portions of the electrons will go through the nonradiative decay path. We used magnetic fields to force NV electron spin levels to mix so that, instead of preferentially populating a single spin direction, we force the electron spins to point in two different directions. As soon as the electron population was mixed, a marked decrease in fluorescence was observed as shown with the blue line in Figure 5. In addition, there was a clear change in ratio between the NV^0 , integrated intensity between 550-620 nm, versus the NV^- fluorescence, integrated intensity between 640-800 nm, which has never been observed before as shown with the red line in Figure 5.

Analysis

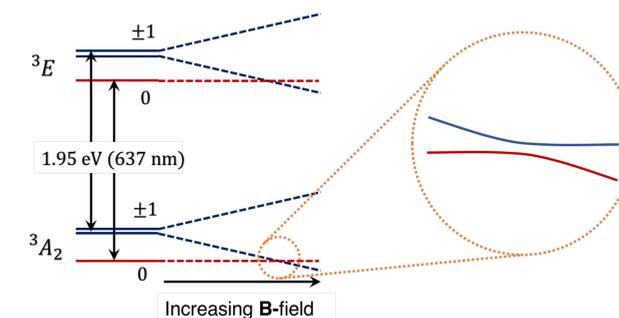


Figure 6. Splitting of states due to magnetic field. Avoided crossing showing coupling between $m_s = 0$ and $m_s = -1$ is shown in the magnified view.

When applying an appropriate magnetic field, the states mix at the energy level crossing allowing for a relatively uniform population between the $m_s = 0$ and $m_s = -1$ states instead of the regular spin polarization towards the state $m_s = 0$. The state preferentially occupied when pumping with green laser at zero magnetic field.

To populate $m_s = \pm 1$ ground state, the basic idea is to apply a magnetic field until the spin 1 projection crosses the $m_s = 0$ ground state. Due to some interaction at the crossing, the two states become coupled (which should theoretically happen at 1028 Gauss). So instead of optically initializing the $m_s = 0$ spin state, the electron is split between the $m_s = 0$ and $m_s = -1$ states leading to a decrease in fluorescence.

Conclusions

The results were preliminary, but this gives us an indication of which spin population contributes more to the charge state transfer from NV^- to NV^0 . For now, evidence points to a still unknown process that makes electrons populating the $m_s = -1$ state contribute more to the charge state transfer from NV^- to NV^0 .

Acknowledgements

We are grateful for the generosity of our collaborators M.V. Gurudev Dutt and Peng Ji from Univ. of Pittsburgh for sharing with us the nanodiamond samples used in the experiments. This work is supported by Calvin College and the Calvin Alumni Association.