

Photophysics of Nitrogen Vacancy centres in Nanodiamonds

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Abstract

NVs are cool because they do cool science stuff. All cool stuff comes from NV⁻. People use 532 nm excitation to increase charge polarisation so they can optimise cool stuff and neglect NV⁰. They looked at it with only one excitation wavelength at a time. Clearly charge polarisation depends on wavelength. So clearly quenching must occur with second wavelength. Must understand all of this stuff to integrate NVs into other systems that use lasers.

INTRODUCTION

The Nitrogen Vacancy (NV) centre is a point defect consisting of a nitrogen-vacancy lattice pair embedded along the [111] axis of a diamond (ref1). The NV centre has two stable charge states, the neutral charge state (NV⁰) and the negatively charged state (NV⁻), with photo-induced interconversion between these two states (ref2). The NV⁻ charge state is an intensively studied material that has shown a wide range of applications in both Physics and Biology due to its high stability and interesting optical properties. Biologists have used them extensively for labelling and imaging of internal biological structures (ref3 and 4). Meanwhile, Physicists have been investigating their

use in a wide range of nanoscale sensing and quantum sensing applications(ref5). By exploiting the quantum mechanical interactions of the defects internal spin state, room temperature quantum effects can be observed in the NV⁻ centre providing a platform to study a wide variety of quantum manipulation protocols (ref6). However, these desirable effects rely solely on the properties arising from the NV⁻ charge state and in most applications the excitation wavelength is chosen to be around 510-540nm, as this region was shown to have the highest charge state polarisation (ref7). By using a single optimised excitation wavelength the impact of the neutral charge state NV⁰ could be neglected despite the optimal charge state polarisation limited at 75%.

The NV^- consists of a ground triplet state 3A_2 and an excited triplet state 3E , as well as two metastable singlet states 1E_1 and 1A_1 ref11???. Within the spin triplet states the $m_s = 0$ and $m_s = \pm 1$ spin states are split in energy at zero magnetic field by $D = 2.87$ GHz for the ground triplet and $D = 1.42$ GHz for the excited triplet (Ref12??). The spin transition rate between the ground $m_s = 0$ and $m_s = \pm 1$ spin states is given by the spin-lattice relaxation time T_1 and has been measured to be ≈ 6 ms at room temperature and zero magnetic field (ref13??). This spin mixing rate can be dramatically increased by applying a large magnetic field in the vicinity of the NV centre ref14??.

★ I AM BEING VAGUE ABOUT THE SPIN MIXING RATE BECAUSE BECAUSE I BELIEVE THE CURRENT EXPLANATION OF ZEEMAN SPLITTING THE SPIN SUBLEVELS HAS HOLES IN IT. ASK ME FOR MORE INFO OR LOOK AT COMMENT IN TEX FILE

The principle zero-phonon line between the 3A_2 and 3E is centred at 637 nm and can be efficiently excited with spin conservation at most wavelengths below 640 nm (ref15??). The radiative lifetime of the excited state is 13 ns for NV centres in bulk diamond (ref16??) and approximately 25 ns for NV centres in nanodiamonds (ref17??). Only a few percent of the fluorescence is emitted at the ZPL, most fluorescence appears in the

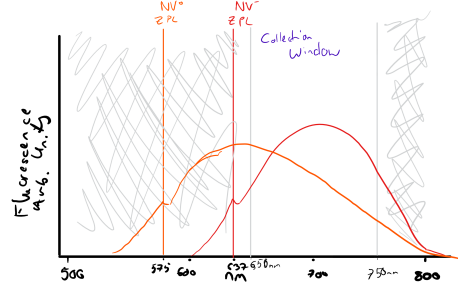


FIG. 2: Fluorescence profiles of NV charge states

phonon side bands between 600 and 800 nm as shown in Fig. 2.

The excited triplet states can also decay to the excited singlet state, the rate from the $m_s = \pm 1$ excited triplet state is $2\pi \times 9.4 \times 10^6$ GHz = 16.9 ns, whereas the rate from the $m_s = 0$ excited triplet state is almost an order of magnitude smaller at $2\pi \times 1.8 \times 10^6$ GHz = 88.4 ns (ref18??). Whilst it is not completely understood why there is a large discrepancy between these decay channels it is noteworthy that this discrepancy is what leads to many of the interesting optical properties of the NV centre. It causes a difference in fluorescence intensity between the two excited spin states which in turn leads to a mechanism for an all optical readout of the centres internal spin state. The excited singlet state has a lifetime of ≈ 1 ns (ref19??) and populates the longer lived ground singlet state and it has been shown to emit fluorescence at a ZPL of 1042 nm ref20??. The longer lived ground

AND THE LIFETIME I FOUND WAS IN BULK QUENCHING MODELS

DIAMOND. EITHER NEED TO FIND VALUE FOR ND'S OR ARGUE THAT IT SHOULD ALSO INCREASE THE SAME WAY AS NV-DID...

Ionisation & Recombination

To convert between the two charge states we need to examine both the Ionisation process from NV^- to NV^0 and the Recombination process from NV^0 to NV^- . The desirable effects of the NV centre rely solely on the properties arising from the NV^- charge state and as a result the standard excitation laser used in NV centre applications is chosen so that it produces the highest charge state polarisation in an effort to optimise the effect and allow them to neglect any effects due to the NV^0 charge state(ref7??).

Ionisation Process

Describe NV^- to NV^0 Process

Recombination Process

Describe NV^0 to NV^- Process

EXPERIMENTAL DETAILS

Describe the Quenching Experiment.

In order to determine the intrinsic photo-physics of our nanodiamonds we developed an 8 level rate equation model that incorporates both the ionisation and recombination mechanisms as well as the STED like mechanisms. The free parameters of the model were varied in order to determine the most likely dynamics of the system. Four approaches were investigated and the most likely model was identified by the Akaike information criteria blah blah

Insert simple diagram of the model and then for each model include separate rates.

Underlying assumptions and unknowns

Blah

'STED' model

Blah

Simple Ionisation and Recombination Model

Where the Ionisation and recombination rates are linearly dependant on laser power independent of wavelength.

Wavelength Dependant I&R

Where the Ionisation and recombination rates are linearly dependant on laser power and dependant on wavelength.

Spin Depenatant I&R

Where the Ionisation and recombination rates are linearly dependant on laser power and dependant on wavelength. In addition there are separate ionisation rates from the $ms = \pm 1$ and $ms = 0$ of the excited NV- charge state to the ground state. However the ratio between these two ionisation channels is held constant for each laser wavelength.

AKAIKE INFORMATION CRITERIA

Blah Blah

Highlight the optimal model, Maybe put table here or in appendix.

DISCUSSION

The optimal model is blah?

Model Parameters

The the values make sense? What are the ionisation rates like? are the close to 10ms/mW. Spin dependancy may occur from different dipolar cross section between the

two excited states. Similar with the recombination rates? Link it to singlet nitrogen. Recombination occurs into which ground state of the NV- Singlet recombination between 0.66 and 1 which is consistent.

Charge state

Blah

Spin State

Blah

COMPARISON WITH OTHER WORK

Blah

CONCLUSION

Say that this indicates that this is the channels that are liekly to occur. If you want to use NV with other lasers than only the 532nm these dynamics must be understood. and then provide array of ways that could investigate these affects.

Acknowledgements

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