Nanoscale sensing of photonic density of states with spins in diamond

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Diamond defects as nanoscale quantum devices

Diamond nitrogen-vacancy (NV) centers can be applied in nanoscale sensing and quantum information processing thanks to their spin properties [1].

Diamond NV centers + Nanophotonic structures

Integrated quantum devices @ 300 K

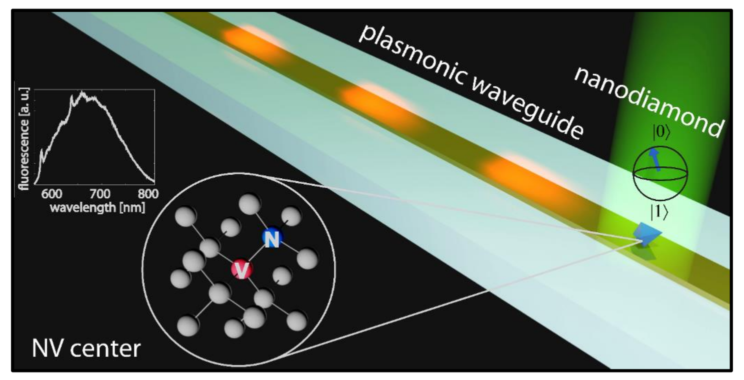
The effect of a broadband Purcell enhancement on the operation of NV spins has so far not been measured. We find that it is possible to use the NV centers as photonic density of states (PDOS) sensors without involving the complicated measurement of their fluorescence decay.

The NV center's optical fluorescence is different for the two spin subsystems.

Spin subsystem	0	1
Decay channels	Radiative only	Radiative + Non-radiative
Fluorescence rate	Higher	Lower

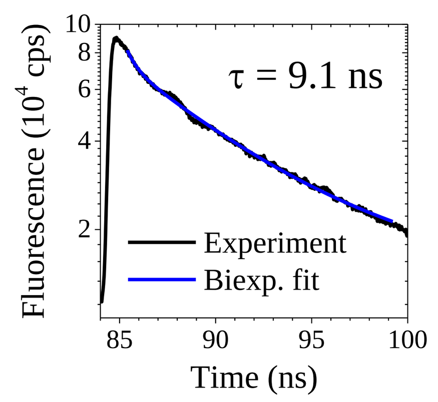
Higher PDOS
⇓
Higher radiative decay
⇓
Less contrast between spin subsystems

Sensing PDOS at the nanoscale

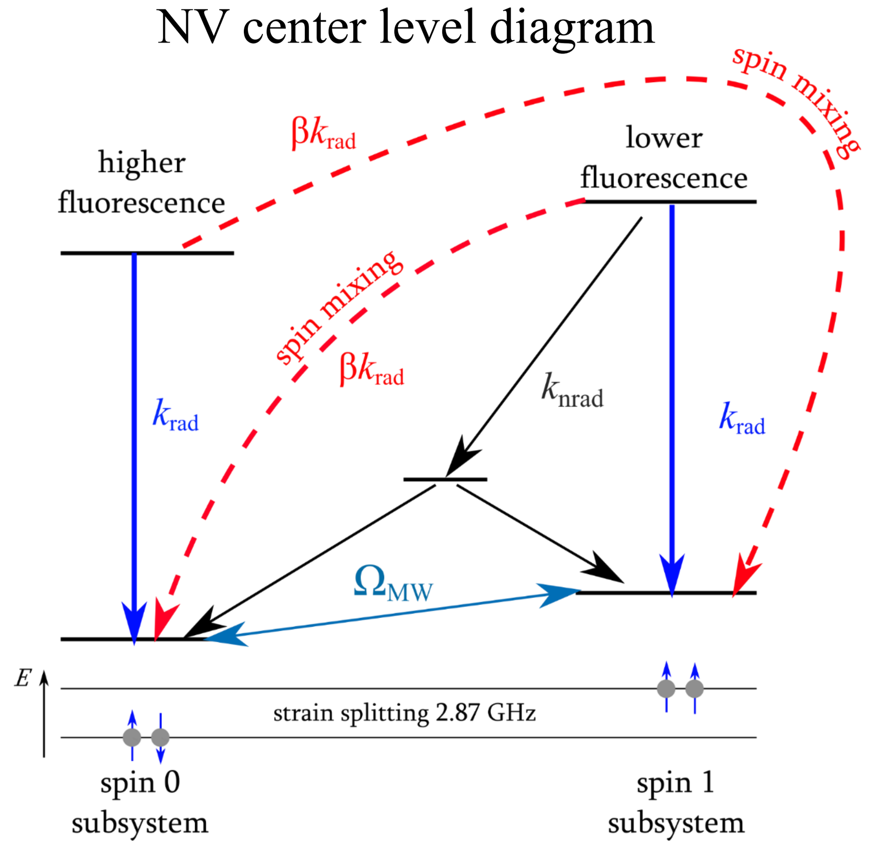


Traditional method:

Time correlated single-photon counting involves recording the time intervals between pump pulses and fluorescence photons



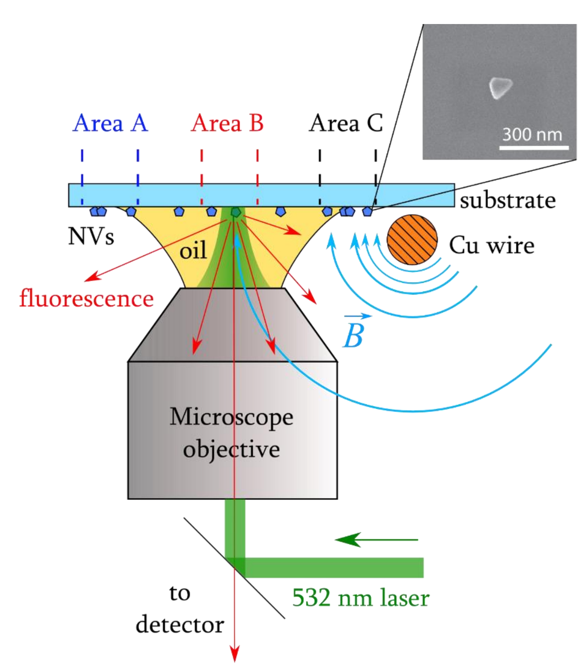
NV center level diagram



Experimental setup

We chose 3 areas 10×10 μm denoted by letters A, B and C and located 230, 130 and 30 μm away from the wire. A, B and C are subject to different levels of microwave field. Fluorescence lifetime τ_{rad} and the spin contrast C were measured for several nanodiamonds in each area

Nanodiamonds	140 nm (~10^3 NV centers)
Substrate	Silica or 200 nm TiN film
Optical excitation	Pulsed 532 nm laser
MW excitation	Cu wire (f = 2.87 GHz)
Objective	Immersion, NA = 1.49



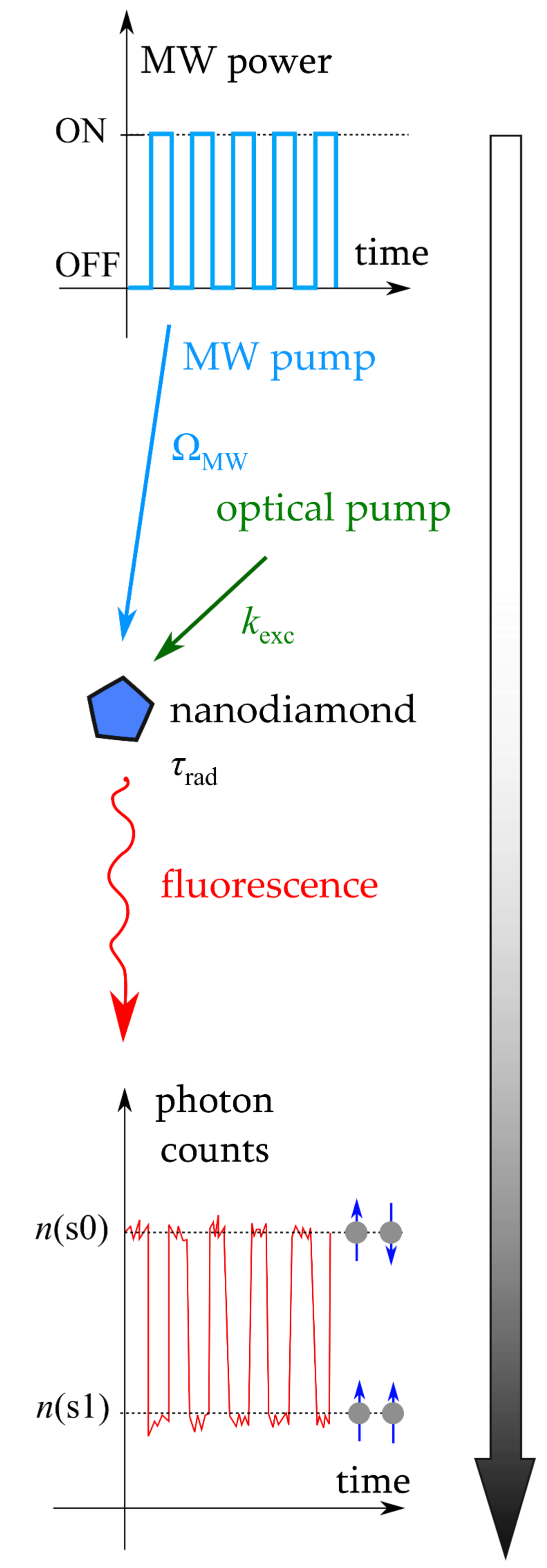
PDOS sensing with NV center spins

Spin contrast C depends on three experimental parameters:

- k_{exc} (opt. exc. rate)
- Ω_{MW} (MW Rabi freq.)
- τ_{rad} (fluo. lifetime)

If the MW power is modulated in time the spin populations are oscillating. The NV center will produce a periodically varying fluorescence rate making it easy to detect the relative contrast between the spin states.

If the optical and microwave excitation rates are known, one can access the NV center lifetime without ever measuring its fluorescence decay. The local PDOS can then be calculated. The results given by this technique are insensitive to the number of NV centers in the ensemble and to the collection efficiency of the detection system.



Spin contrast C can serve as a measure of PDOS

$$C = \frac{n(s0)-n(s1)}{n(s0)} \sim \frac{\tau_{rad}}{\tau_{rad}+\tau_{nrad}}$$

$$\mathrm{PDOS}^{(\mathrm{local})} \propto \tau_{rad}^{-1}$$

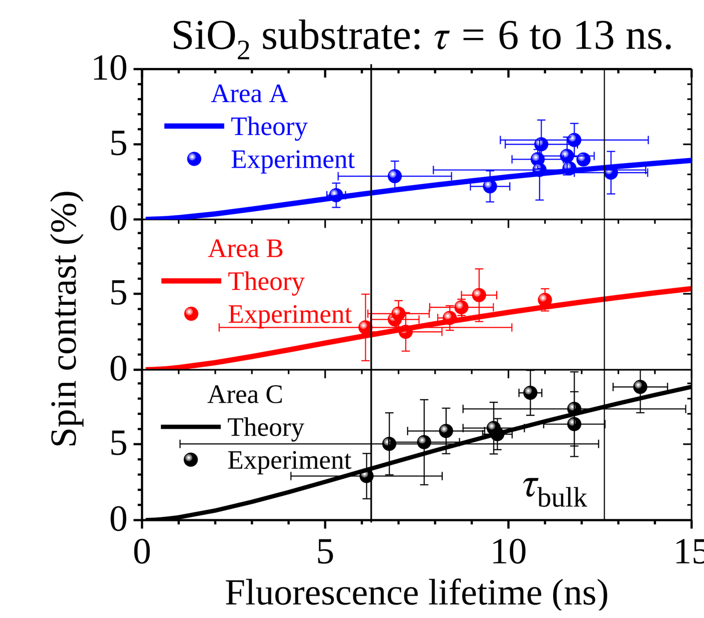
PDOS sensing method	Fluorescence decay	Spin contrast
Source	Pulsed laser	Green LED
Detector	SPAD	Photodiode
Electronics	Ultrafast counter	Lock-in amplifier
Memory	Dedicated module	Real-time measurement

NV center spin contrast dependence on the fluorescence lifetime

Verifying the validity of the theory

For each NV center ensemble we measured the fluorescence lifetime τ and the spin contrast C. The theoretical dependence of C on τ matches the experimental data without free parameters. The spin contrast is higher in areas that are closer to the Cu wire, as these NVEs experience higher magnetic field.

Contrast depends on lifetime as expected



Sensing PDOS near surface of TiN film

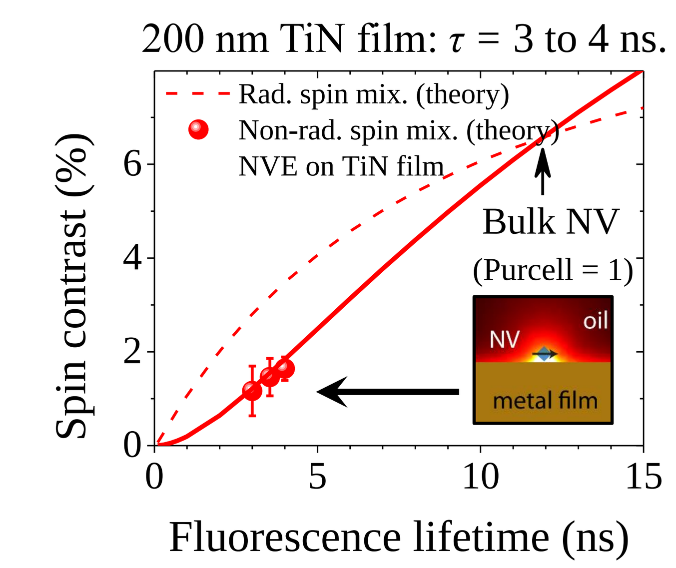
TiN film provides surface-plasmon-polariton (SPP) modes that contribute to the local PDOS. NV centers on TiN must have shorter fluorescence lifetime

- Expected Purcell factor: 2.9 ± 0.3 (depends on nanodiamond size)
- Spin-contrast measurement results on 3 nanodiamonds: 3, 3.5, 4

The nature of NV spin-mixing transitions

Spin contrast of NV centers on TiN fitted with two different models

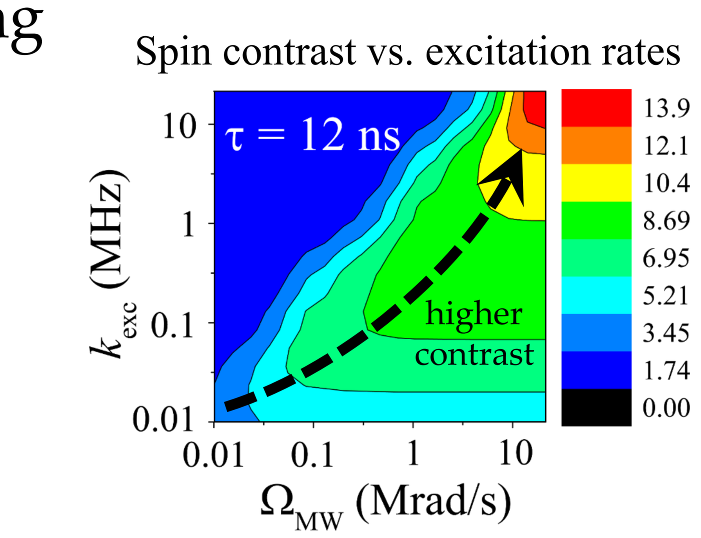
- Radiative spin-mixing transitions model: Good fit
- Non-radiative spin-mixing transitions: Bad fit



Optimal conditions for PDOS sensing

How does spin contrast C depend on the optical (k_{exc}) and microwave (Ω_{MW}) excitation rates?

- The contrast is highest when both k_{exc} and Ω_{MW} are high.
- Estimated sensitivity of the PDOS detector: $\eta_\tau = \frac{\sigma_C\sqrt{\Delta t}}{\partial C/\partial \tau} \sim 5\ \mathrm{ns}\sqrt{\mathrm{Hz}}$



Conclusions

- We measured the spin contrast in NV centers as a function of the fluorescence lifetime matches theoretical description
- Spin contrast could be used to measure local PDOS on the nanoscale using simplest optoelectronic devices
- Spin mixing transitions in the NV center appear to be radiative in nature

References

[1] Doherty et al., Phys. Rep. 528, 1 (2013);

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Acknowledgements

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