Spin Contrast of Purcell-Enhanced Nitrogen-Vacancy Centers in Diamond

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Abstract: Nitrogen-vacancy centers in diamond allow coherent spin state manipulation and optical readout at room temperature, which has powerful applications in nanoscale sensing. Nanophotonic structures such as plasmonic waveguides, nanoantennae, metamaterials, and metasurfaces can enhance the detected fluorescence rate from such broadband emitters. The fluorescence of the coupled emitter is directed into confined plasmonic modes with high photonic density of states. However, an accurate spin readout requires both high photon counts and a strong contrast between the spin states, both of which can be influenced by the Purcell effect. We introduce a novel method for measuring the spin contrast in large nitrogen-vacancy ensembles. We use this method to study how the photonic density of states must be engineered in order to minimize the uncertainty of spin readout in dense NV ensembles. We describe these results using a kinetic model of the nitrogen-vacancy's internal dynamics.
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1. Introduction

Nitrogen-vacancy (NV) color centers in diamond [1] are fluorescent lattice defects resulting from a vacancy and an adjacent nitrogen substitution. They have proven to be excellent testbeds for realizing nanoscale ultrasensitive electromagnetic field and temperature sensors as well as integrated quantum information registers operating at ambient conditions. These capabilities are due in large part to the unique properties of the NV center electron spin. The NV produces a spin-dependent fluorescence rate, which serves as a means of optical spin state readout. The spin can also be initialized optically and exhibits excellent room temperature coherence.

For sensing applications, dense NV ensembles (NVEs) are used to increase the readout signal. However, even then, the collected fluorescence rates are limited by the NV quantum yield and collection efficiency. To solve this problem, nanodiamonds containing NVEs can be coupled to various integrated optical elements such as waveguides or nanoantennae. In many of such demonstrations, particularly in those employing plasmonic structures, one observes a reduction of NV fluorescence lifetime caused by an increase in the photonic density of states (PDOS) or Purcell effect. This effect can lead to higher NV quantum efficiency and higher single-photon detection rates. The sensing applications, which exploit the exceptional NV center spin coherence, could also benefit from such nanophotonic structures. However, these applications often rely not only on the number of collected photons but also on the fluorescence contrast produced between the spin states (spin contrast). The issue of spin readout in a high PDOS environment has been investigated theoretically [2,3], but no experimental study has been performed so far to quantify or verify the effect of such environment.

2. Experimental results

In our experiment, individual nanodiamonds with sizes around 80 nm, each containing an NVE of 400 NV centers on average, were dispersed on a sapphire substrate. In order to create a wide distribution of fluorescence lifetimes, titanium nitride (TiN) islands were predeposited on the substrate so that NVEs were experiencing different PDOS depending on their location (see Fig. 1(a)). Higher PDOS at the surface of TiN islands is expected due to confined surface plasmon-polariton (SPP) modes. We chose a sapphire area and a TiN area on the sample, randomly selected several nanodiamonds from each area and measured their fluorescence lifetimes and spin contrasts.

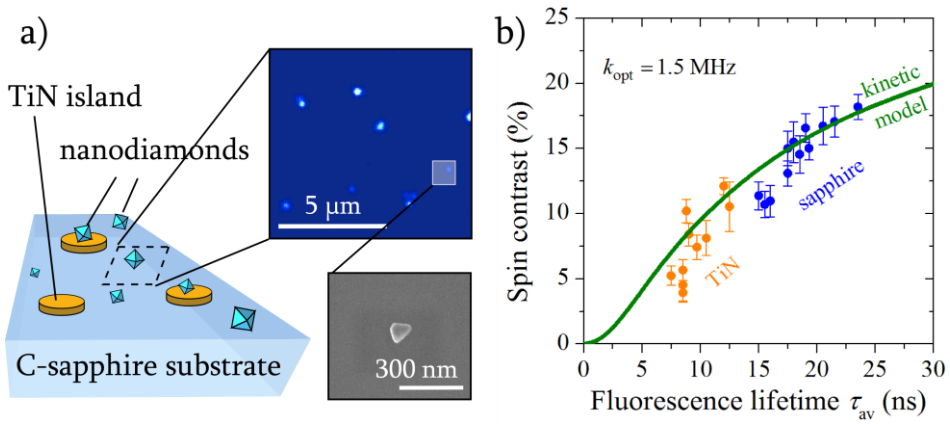

Fig. 1. (a) Layout of the sample, with nanodiamonds dispersed on a sapphire substrates with lithographically predefined TiN islands. Fluorescence and SEM images of the nanodiamonds are shown in the insets. (b) Experimentally measured lifetimes and spin contrasts of NVEs on sapphire and TiN areas. Orange (blue) datapoints correspond to nanodiamonds on TiN (sapphire).

The spin contrast was measured as the difference in fluorescence produced by optically initialized and thermally relaxed spin populations [4]. The spin contrast measured in this way represents 2/3 of the contrast produced by coherently flipping a single spin. However, this method does not require controlled DC and microwave magnetic fields and is not affected by different NV orientations within the crystal. The local PDOS was quantified by fluorescence lifetime measurements performed using the time-correlated single photon counting technique. The range of fluorescence lifetimes τ_{av} for NVEs found on sapphire spanned from 15 to 24 ns. This spread of lifetimes can be attributed to variations in the local density of states as well as varying nonradiative decay rates. The TiN film's SPP modes contribute to the local PDOS and increase the radiative rates of the NVEs.

Because of high NV density, the contrast has to be measured at optical powers well below saturation. This prevents collective NV effects which decrease the spin contrast. We observe a strong dependence of the spin contrast on the fluorescence lifetime (see Fig. 1(b)). The dependence can be schematically explained as follows. The spin contrast is quantitatively defined as $C = 1 - N_1/N_0$, where $N_{1(0)}$ are the numbers of photons detected during the detection time interval for NV centers initially prepared in $m_s = 1(0)$ states. For each photon produced by the $m_s = 0$ state, the $m_s = 1$ state produces $k_{rad}/(k_{rad} + k_{cross})$ photons, where $k_{rad} = \tau_{av}^{-1}$ is the radiative decay rate and k_{cross} is the nonradiative rate to a dark state. We therefore find the simplified monotone dependence of spin contrast on fluorescence lifetime: $C = k_{cross}\tau_{av}/(1 + k_{cross}\tau_{av})$. Careful kinetic modeling of the NV center transient level populations yields a good agreement with the experimental data (see Fig. 1(b)).

We see that while high PDOS aids photon collection, it also leads to a decrease in spin contrast at low optical powers. Therefore, a careful PDOS engineering is required for the efficient spin readout. As an example, we have theoretically considered an NV center located in a small nanodiamond, coupled to a silver plasmonic waveguide. We found that a waveguide with a Purcell factor of 5 to 10 (depending on the intrinsic NV quantum yield), must be designed in order to achieve maximum sensitivity of spin population measurement.

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3. References

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