Optica Quantum 2.0 Conference 2025 © 2026 Optica Publishing Group

Two-Photon Excitation Fluorescence 3D Imaging of NV- Color Centers in Bulk Diamond Samples

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Abstract: Linearly polarized output of a18.9 MHz repetition rate Yb-fiber laser delivering 0.4 ps pulses at around 1030 nm is used for two-photon excitation fluorescence 3D imaging of CVD grown single-crystal diamond samples containing 4 PPM nitrogen-vacancy (NV) centers.

1. Introduction

Single-crystal diamond samples that contain controlled levels of nitrogen-vacancy (NV) centers are of great scientific interest since these color centers offer a unique solid-state platform with spin qubits that can be initialized and read out with long qubit lifetimes at room temperature. Combined with a 3D optical microscope imaging technique, such as confocal fluorescence microscopy and applying a moderate DC magnetic field, the spin state (m_s = 0, m_s = +/- 1) of excited state electrons in the 3D volume can be determined by techniques like optically detected magnetic resonance (ODMR) [1] or fluorescence lifetime imaging (FLIM) [2]. In both cases, a microwave antenna is placed next to the bulk diamond sample and a microwave field of varying frequencies is applied around the Ω = 2870 MHz resonant frequency, which corresponds to the energy difference between the m_s = 0 and +/-1 spin states in case of zero magnetic field. In the former case, the fluorescence intensity $I(x,y,z)$ is measured as the function of microwave frequency (Ω), while in the latter case histograms corresponding to the different fluorescence lifetime components are recorded for each pixel using TCSPC (time correlated single photon counting) technique. In the case of ODMR, fluorescence intensity drops at the resonance frequency (or frequencies in the presence of non-zero DC magnetic/electric field), while in case of FLIM the weight factors for the different time constants for the m_s = 0 spin electrons (τ_1 ~ 12 ns) and for the m_s = +/-1 spin electrons (τ_1 ~ 8 ns) provide a contrast: if a large portion of the excited state electrons are prepared to the m_s = 0 spin state, then the weight factor (a_1) for the τ_1 ~ 12 ns lifetime component is large compared to that (a_2) of the τ_2 ~ 8 ns lifetime component. When a microwave at the resonance frequency is provided to the sample, however, the a_2 component will increase at the expense of a_1, hence the a_2/a_1 ratio will reflect the relative number of excited electrons in the m_s = +/- 1 state and those in the m_s = 0 state [2].

In principle, there is an alternative for 3D magnetometry on the microscopic scale, which is based on multi-photon excitation fluorescence of NV- color centers in diamond. In a recent work of Peng. Ji et al. [3], two-photon (2P) excitation emission spectra of NV0 and NV- were investigated in diamond samples. Based on their results we can say that using an ultrashort pulse laser with central wavelength above 1 micron, fluorescence emission from NV- is dominant over that of NV0, which is also an important issue for ODMR or FLIM magnetomerty.

2. Experimental setup, spectroscopic and imaging results

In order to implement 2P FLIM, one requires a pulsed laser source with a variable repetion rate [4] that matches the fluorescence lifetime values of the NV0 and NV- color centers mentioned above. To this end, we optimized our variable repetiton rate, sub-ps Yb-fiber laser for 2P 3D fluorescence imaging experiments: we ended up with a 18.9 MHz repetition rate laser operating at center wavelength of 1030 nm with a spectral bandwidth of ~12 nm. The typical pulse duration of the system after a grating compressor is ~0.4 ps. For our 2P 3D imaging and magnetometry experiments, we modified an LSM 7MP scanning two-photon microscope system (product of Calr Zeiss, Jena, Germany): bandpass filters matcing the emission spactra of NV- and NV0 were placed in front of the NDD1 (EM: 680-720 nm) and NDD2 (EM: 565-610 nm) non-descanned detectors, while the dichroic long pass beamsplitter above the objective was also replaced by an LP750 filter. In order to protect the NDD detectors from stattered and back reflected laser light, a short pass laser blocking filter (LBF750) was also used in front of the GaAsP APD detectors, which allow fluorescence detection up to 750 nm. In order to measure the 2P excitation fluorescence spectra of our samples, we also placed similar dichroic beamsplitters in front of our scanner mirrors in a descanned configuration, and coupled the emitted fluorescence signal to a large core diameter (~ 0.5 mm) multimode optical fiber. Using a 10 µm entrance slit in an Ocean Optics QEPro spectometer, we could record the two photon excitation