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Improving the Optical Coherence of Diamond Tin-Vacancy Centers by Heralded Initialization

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Abstract: We demonstrate heralded initialization of charge state and optical transition frequency of diamond tin-vacancy centers, using (off-)resonant lasers, photon detection and real-time logic. Using this, we show frequency tunability > 100 MHz and strongly improved optical coherence. © 2024 The Author(s)

1. Introduction

Group-IV color centers in diamond are an emerging favorable alternative due to their high Debye-Waller factor [1] and their inversion symmetry [2], which allows for integration in nanophotonic devices [3]. The negatively charged Tin-Vacancy (SnV^{-}) is of particular interest due to its high quantum efficiency and large spin-orbit coupling, resulting in elevated operation temperatures compared to other Group-IV color centers [4].

Upon resonant excitation (619 nm) of the SnV^- center, the emitter can go into a dark state and it can be brought back into the bright state by off-resonant excitation (515 nm) [5]. However, the off-resonant repumping can cause a frequency shift [6]. This is presented in Fig. 1(a), where the resonant frequency is swept over the optical transition of the SnV^- . A repump pulse is applied when no resonant frequency is measured in the preceding scan. This frequency shift poses two challenges for quantum networking applications based on SnV-centers: i) initialization errors due to the probabilistic repump process, ii) frequency shifts hinder efficient optical spin initialization and readout. This causes reduced photon indistinguishability, which negatively impacts remote entanglement generation which rely on photon interference. Here, we overcome these challenges by heralded initialization of the charge state and the transition frequency using a combination of (off)-resonant lasers, photon detection and read-time logic.



Fig. 1. (a) The fluorescence of 1000 PLE scans taken in \sim 1.3 GHz/s over the optical transition. The state of the emitter is estimated from the counts detected during each scan. (b) The pulse sequence of the experiment data shown in (c) and (d). (c) 2D histogram of C_2 , as a function of C_1 .

2. Results

Motivated by the observed spectral stability before ionization, we explore the possibility of using photon counts during a resonant probe pulse as a heralding signal for the successful preparation of the SnV⁻ center charge state with its optical transition at a pre-set frequency, see pulse sequence in Fig. 1(b). An off-resonant 515 nm 'repump' pulse of 50 μ s and 100 μ W is followed by two identical resonant 619 nm laser pulses of 50 μ s and 100 nW, named

here 'Pulse 1' and 'Pulse 2'. In Fig. 1(c) we plot in log-scale the distributions of the photons detected during Pulse 2, C_2 , as a function of the number of photons detected during the preceding Pulse 1, C_1 . A clear correlation between C_1 and C_2 is visible. The horizontal and vertical bands of uncorrelated counts is attributed to ionization of the emitter. The horizontal band mainly corresponds to cases where ionization occurred during Pulse 1. Considering heralded initialization, those cases could lead to an incorrect heralding signal and should thus be minimized. In Fig. 1(d) we plot C_2 conditioned on having more or less than 150 photon counts detected during Pulse 1. It can be seen that if we condition on $C_1 > 150$, the photon count distribution of Pulse 2 follows a Poisson distribution centered around 170 counts, while the reverse conditioning shows a broader, lower-counts distribution mixed with a peak near zero counts. The thresholding approach demonstrated here allows to filter the desired bright state condition of the color center out of the statistical distribution of possible charge-resonance states.



Fig. 2. (a) CRC pulse sequence protocol. (b) CRC preceded PLE scans, for a low (high) C_{pass} in the top (middle) panel. Bottom panel: CRC preceded PLE scans with changing frequency set point.

For quantum protocols it is key that the initialization is heralded and that the selecting of the correct charge state and transition frequency is done real-time [7]. We present a Charge-Resonance-Check (CRC) protocol, see Fig. 2(a), which relies on photon detection during a resonant probe pulse and two count threshold, C_{repump} and C_{pass} that resemble a check on the charge state and the frequency detuning respectively. We employ the CRC in conjecture with resonant frequency scans in Fig. 2(b). In the top and middle panel we show the difference between a low and high C_{pass} , where we see the emitter make more frequency jumps but of lower magnitude. In the bottom panel of Fig. 2(b), we employ this measurement on another emitter follows the frequency of the probe pulse, allowing us to tune the emitter >100 MHz. In addition, we performed a Ramsey interferometry experiment preceded by a CRC. We obtain a T_2^* for C_{pass} of 100, (10) of (6.3 ± 0.4) ns, ((4.3 ± 1.7) ns). This demonstrates that implementing a CRC can mitigate the effects of spectral detuning leading to an increase in optical coherence time, which is key to improve photon interference experiments [8].

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