Modeling and characterization of cooperative effects in ensembles of inhomogeneous solid-state emitters

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Abstract: We propose a platform for the study of collective emission in a solid-state system, consisting of silicon-vacancy (SiV) centers implanted in subwavelength ordered arrays. Numerical simulations of emitter-emitter interactions, fabrication, and preliminary characterization are presented.

1. Introduction

Silicon-vacancy centers (SiVs) in diamond provide a highly promising solid-state platform for quantum information processing due to their close-to-lifetime-limited linewidths and relatively stable single-photon emission [1]. Many schemes for coherent transfer of quantum information from quantum systems to long-range information carriers (photons and phonons) involve strong coupling of the emitter with photonic/phononic cavity modes of a host nanostructure and tuning of individual emitter properties to ensure spectral overlap and generation of indistinguishable photons [2]. This approach is often limited by challenges associated with precise placement of defects in nanostructures [3]. In this work, we explore the use of focused ion beam (FIB) implantation to create dense and ordered ensembles of SiVs whose geometries enable strong emitter-emitter coupling and observation of cooperative phenomena such as superradiance, subradiance, and selective radiance. Our experimental efforts are complemented by modeling of the emitter-emitter interactions that consider relevant optical properties and inhomogeneities observed in SiVs.

2. Modeling of collective effects in closely spaced SiVs

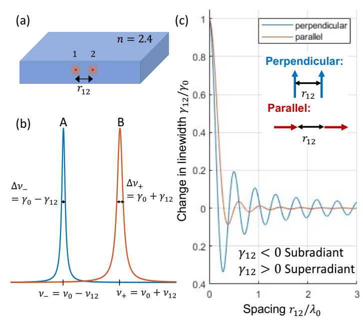

Fig. 1(a) Our model comprises of two or more SiV⁻ separated by small spacing r_{12} in diamond. (b)-(c) For a set of two identical emitters (with $\lambda_0 = 737\ nm$ and transform limited linewidths 135 MHz), the collective emission exhibits mode splitting with the linewidths of the modes (A shown here in (c)) varying based on the emitter-emitter spacing and emitter polarization.

When emitters with two-level systems (TLS) are placed at close enough distances, the emitter decay rates are significantly modified in either the superradiant (enhanced collective decay) or subradiant (suppressed collective decay) regime in a manner that is strongly dependent on the emitter-emitter spacing. To understand the distance-dependent effects on collective emission in ensemble SiVs, we initially considered two identical SiVs in homogeneous medium (diamond, n = 2.4) with central positions $\nu_1 = \nu_2 = 406.7$ THz (corresponding with a transition at 737 nm) and linewidth $\Delta\nu_1 = \Delta\nu_2$ (Fig 1). Dipole-dipole interaction between the emitters leads to mode splitting and for clarity we focus only on the evolution in linewidth for one of the modes ("A" in Fig. 1b). If the distance between two identical emitters is within $r_{12} (< \lambda_0)$, the interaction between emitters is in the superradiant regime, and conversely, distances greater than r_{12} result in the subradiant regime. As r_{12} increases, the effect of the linewidth and frequency change oscillates and gradually decreases, well aligned with previous findings [4]. Based on Fig. 1c, we identified that the coupling effect will be the strongest for emitter-emitter spacing below one-half of the transition wavelength (737 nm), which is within the positioning accuracy of the FIB implantation capabilities. As spectral diffusion and inhomogeneous broadening are well-observed effects for SiVs [1,5], we then accounted for inhomogeneity in our model by varying ν_2, $\Delta\nu_1$, and $\Delta\nu_2$. (Fig 2a) With ν_1 unchanged at 406.7 THz, we implemented a Monte Carlo approach to sample a set of $[\nu_2, \Delta\nu_1, \Delta\nu_2]$ from gaussian distribution while applying linewidth distribution of $\Delta\nu_{1,2} \approx 200 \pm 50\, MHz$ [5] and spectral variance $\nu_2 - \nu_1 \leq 500\, MHz$ [1]. Our model predicts that when accounting for realistic inhomogeneous distribution of frequencies and linewidth broadening in SiVs, collective response is still observable (Fig 1c). This model is being expanded to increase emitter number and to account for heterogeneity in dipole orientation.

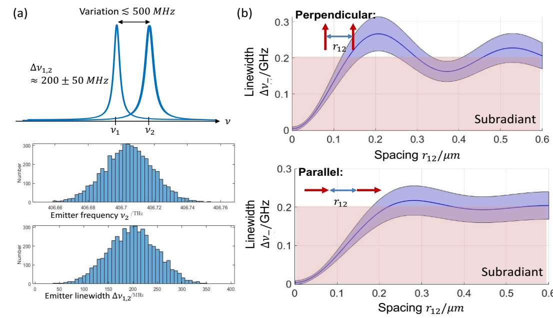

Fig. 2(a) Inhomogeneities in the SiV$^-$ emission are modeled using a Monte Carlo approach, where we simulate many iterations of emitter-emitter interactions with random sets of emitter frequencies and linewidths selected from Gaussian distributions of emitter frequency difference 0 ± 500 MHz and linewidth 200 ± 50 MHz . (b) Simulated linewidth of the collective emission by two SiVs as a function of emitter-emitter spacing. The center blue curves represent the mean response of all iterations, while the shaded blue regions are bounded by the standard deviations.

3. Fabrication and Preliminary Characterization

An electronic grade diamond ($< \mathbf{1}\, \boldsymbol{ppb}$ Nitrogen) was processed following standard procedures, details are shown in [6]. Next, a FIB implantation system with expected accuracy of **~60 nm** implanted SiVs with accelerating voltage of 35 keV and 70 keV onto different patterns, resulting in depths of **~20 nm** and **50 nm**. The SiVs were implanted onto a total of 30 regions, each with varying spatial distance. Room- and cryogenic scanning confocal images (Fig. 3a-b) were taken at 532 nm and 636 nm excitation (with the latter avoiding excitation of nearby native NV centers). While it is not possible to resolve interatomic spacing within each dense array, imaging of each cluster of closely spaced emitters shows positioning accuracy well below 200 nm (diffraction limit of our microscope). Extensive characterization of the spectral properties of FIB-implanted SiVs at 4 K is in progress (see photoluminescence spectrum from an SiV ensemble in Fig. 3c), with the spectral linewidths and transition frequencies of SiV arrays to be evaluated as function of emitter density and spacing under resonant excitation.

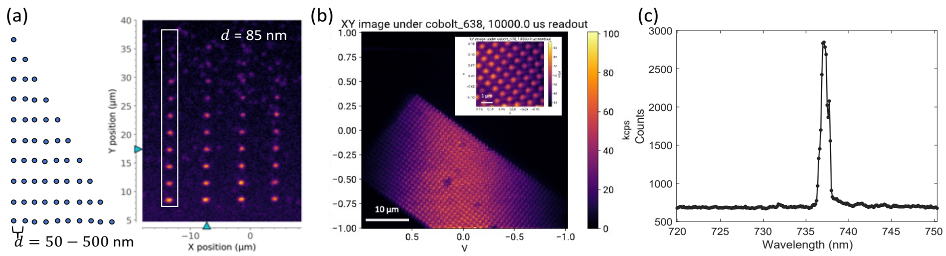

Fig. 3(a) Left: 1D chains of SiV$^-$ arrays that will be explored in this work. Right: Confocal microscopy (with 532-nm excitation) of a set of 1D chains. (b) Cryogenic confocal images taken with 638-nm excitation. (c) Emission spectrum (with above-band excitation) of a region of dense SiVs at 4 K, indicating narrowing of emission and suppression of phonon sidebands at low temperature.

4. Conclusion and outlook

We simulated, fabricated, and have begun characterization on the ordered arrays of SiVs to study to collective phenomenon in solid-state emitters. Our modeling and fabrication results suggest the FIB technique provides sufficient positioning accuracy to observe collective phenomena, even when accounting for emitter inhomogeneities. Future work will focus on resonant excitation spectroscopy to quantify spectral changes due to collective emission.

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6. References

[1] Sipahigil, A., Jahnke, K. D., Rogers, L. J., Teraji, T., Isoya, J., Zibrov, A. S., Jelezko, F. & Lukin, M. D. Indistinguishable Photons from Separated Silicon-Vacancy Centers in Diamond. Phys. Rev. Lett. 113, 113602 (2014).

[2] Van der Sar, T. *et al. Applied Physics Letters* **98**, (2011)

[3] Santori, C. *et al. Nanotechnology* **21**, 274008 (2010).

[4] Z. Ficek and S. Swain, *Quantum Interference and Coherence: Theory and Experiments*. in Springer Series in Optical Sciences. Springer, 2005.

[5] Evans, R. E., Sipahigil, A., Sukachev, D. D., Zibrov, A. S. & Lukin, M. D. Narrow-Linewidth Homogeneous Optical Emitters in Diamond Nanostructures via Silicon Ion Implantation. Phys. Rev. Applied 5, 044010 (2016).

[6] Titze, M., Byeon, H., Flores, A., Henshaw, J., Harris, C. T., Mounce, A. M. & Bielejec, E. S. In situ ion counting for improved implanted ion error rate and silicon vacancy yield uncertainty. Nano Letters 22, 3212–3218 (2022).