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Total number of authors:
12

Published in:
arXiv

Publication date:
2020

Document Version
Early version, also known as pre-print

Citation (APA):

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Cavity-Enhanced Photon Emission from a Single Germanium-Vacancy Center in a Diamond Membrane

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(Dated: December 12, 2019)

The nitrogen-vacancy center in diamond has been explored extensively as a light-matter interface for quantum information applications, however it is limited by low coherent photon emission and spectral instability. Here, we present a promising interface based on an alternate defect with superior optical properties (the germanium-vacancy) coupled to a finesse 11,000 fiber cavity, resulting in a 31±15-fold increase in the spectral density of emission. This work sets the stage for cryogenic experiments, where we predict a factor of 20±7 increase in the spontaneous emission rate.

Defect centers in diamond can exhibit long-lived spin states that are accessible via coherent optical transitions [1–3], providing a promising platform for quantum nonlinear optics [4] or quantum networks [5–6]. There have been impressive steps towards creating such a network using the nitrogen-vacancy (NV) center in diamond, with several critical components demonstrated in the last decade [7–15]. Nevertheless, optically-mediated entanglement rates for NVs have been limited to tens of Hz [14] due to the low fraction (3%) of photons emitted into the coherent zero phonon line (ZPL), the difficulty of collecting these photons into a single optical mode, the long excited state lifetime (12 ns), and spectral diffusion of the emitter. This platform could be improved by coupling defects to an optical resonator, thereby increasing the ZPL emission rate into a well-defined mode. However, enhanced spectral diffusion near surfaces has thus far impeded attempts to realize stable NV centers in small-mode-volume optical cavities [15–17].

An alternative approach explores group IV defect centers such as the silicon-vacancy (SiV) [18–19] and germanium-vacancy (GeV) centers [20–21]. While their spins interact strongly with phonons (necessitating experiments at low temperature [3] or in a high-strain regime [22]), their optical properties are superior. Owing to inversion symmetry, these particular defects have larger ZPL fractions (60–70%) and exhibit strongly reduced spectral diffusion compared to the NV center [23–25]. Moreover, there is evidence that the GeV also has high quantum efficiency [24]. Integrating GeV centers into optical resonators is therefore a promising step toward generating highly efficient or even deterministic interactions between spins and photons [26,27].

Over the last decade, two primary approaches for coupling diamond defect centers to optical cavities have emerged: (1) diamond nanophotonics and (2) open-geometry microcavities. Emitters coupled to low-mode-volume nanophotonic resonators can exhibit high Purcell enhancement [15] and can even enter the high-cooperativity regime [26,27]. Despite their inversion symmetry, SiVs in such structures still suffer from increased spectral diffusion and inhomogeneous broadening due to nearby diamond surfaces, lattice damage, and material strain [28]. This is further exacerbated for the GeV, since the heavier ion leads to increased implantation damage. Furthermore, the GeV ZPL occurs at a shorter wavelength (603 nm), making fabrication substantially more challenging as it requires smaller feature sizes and has increased sensitivity to surface roughness.

In contrast to nanofabricated devices, open Fabry-Pérot microcavities offer much narrower resonator linewidths as well as in-situ spectral and spatial tunability. Importantly, these systems can accommodate microns-thick diamond membranes in which even NV centers can exhibit minimal spectral diffusion and bulk-like optical properties [29]. So far it has proved challenging to fabricate membranes with sufficient surface quality as to achieve very high cavity finesses. This is particularly true when the energy of the optical mode is concentrated in the diamond (so-called “diamond-like” modes) since the electric field has an antinode at the air-diamond interface [30]. Indeed, the only demonstration of coupling between a single emitter in a diamond membrane and an open cavity used a finesse $\mathcal{F} = 5,260$ mode localized in...
the potential of the open-cavity approach.

In this work, we observe coupling between a “diamond-like” mode of a Fabry-Pérot microcavity and a single GeV center in a membrane, thereby combining the advantageous optical properties of the GeV with the flexibility of an open-geometry resonator. We build on previous room-temperature experiments with defects in nanodiamonds to observe clear funneling of GeV center emission into the cavity mode. Our experiment represents the first demonstration of GeV-cavity coupling and confirms that both the diamond membrane and cavity are of sufficient quality to support high-finesse (\(F > 10,000\)) “diamond-like” resonances. Furthermore, we observe and study the presence of a dark state in the GeV level structure, elucidating new details regarding the emission dynamics. These results therefore represent an important step toward realizing an efficient spin-photon interface using diamond defects in open cavities.

Our microcavities comprise a macroscopic flat mirror (substrate from Coastline Optics) and a microscopic spherical mirror deposited on the tip of a single-mode optical fiber (Thorlabs G30HP). The spherical dimple of the fiber mirror is machined using a laser ablation technique and has an effective radius of curvature \(R = 43.1 \pm 0.6 \text{ µm}\). Both the flat and fiber substrates are coated with dielectric Bragg stack mirrors (LASEROPTIK) with alternating high and low index layers of Ta2O5 and SiO2, respectively. The flat mirror is low-index terminated such that the addition of the diamond membrane increases the mirror reflectivity; it also produces an electric field-antinode at the mirror-diamond interface. In contrast, the fiber mirror is high-index terminated (designed for use in air) and has an electric-field node at the air-mirror interface. The layer designs are numerically optimized to have matched transmissions (into diamond and air respectively) of \(T_{\text{flat}} = T_{\text{fiber}} = 70 \text{ ppm}\) at 603 nm. At the pump wavelength (532 nm) the mirrors have \(T_{\text{flat}} = 99.8\%\) and \(T_{\text{fiber}} = 0.7\%\) to prevent laser leakage into the fiber. We incorporate an emitter into the cavity by Van der Waals bonding a diamond membrane containing GeVs centers (fabrication details in [34]) to the flat mirror, which is mounted on a three-axis stage with piezoelectric control (see Fig. 1). A long working distance objective (Olympus LUCPLFLN 40×, 0.6 NA) faces the backside of the flat mirror, and is used to excite the emitter and collect the cavity mode; a separate stage facing the sample is used for mounting either an objective (Mitutoyo 100× Plan Apo, 0.7 NA) for confocal characterization, or the fiber mirror for cavity characterization. An additional tip-tilt stage is used to align the fiber mirror normal to the sample, with a piezoelectric scanner along the optical axis for fine tuning the cavity length. We align the objectives and fiber mirror to the excitation laser, and scan the sample relative to the excitation spot to study different emitters.

We first characterize the membrane in the confocal configuration to map out the position of GeVs. Confocal images are obtained by exciting emitters through the back of the flat mirror, while the emission is collected through the 100× objective and filtered with a passband of 600 – 605 nm before coupling into a single-mode fiber (Fig. 2a). Here, bright spots correspond to single GeVs and contours are drawn at 4,000 counts/s to qualitatively show emitter localization. We perform a detailed study of the well-isolated GeV center marked with an arrow (henceforth referred to as the studied emitter). We confirmed that this was a single defect (measured intensity autocorrelation of \(g^2(0) = 0.25\pm0.16 [34]\) with a lifetime of \(\tau = 6.0 \pm 0.1\) ns.

The modest fluorescence count rates observed can be attributed to a combination of low transmission in the collection path, narrow spectral filtering, and much of the emission either exiting through the flat mirror (which is designed for normal incidence) or being confined to the membrane in the form of guided modes [34]. Counts are further reduced by the presence of an additional dark state, which may be a different charge state of the GeV center [35]. To quantify the impact of such a state, we probed the population dynamics of the studied emitter by measuring \(g^{(2)}\) as a function of excitation power and found that the dynamics were well-described by a three-level model including power-dependent shelving [34]. From this analysis, we predict a dark-state equilibrium population of 96 ± 18% at infinite pump power, corresponding to a bright-state photon emission rate of \((6.7 \pm 0.9) \times 10^6\) photons/s. Reassuringly, there has been some evidence that the dark-state population can be reduced via laser repumping/gating [27, 35]. Moreover, non-radiative decay paths may further limit photon counts, as varying estimates of quantum efficiency have been reported recently [24, 35, 36]. Finally, photon emission was further limited by our inability to saturate most emitters due to the low absorption cross-section at 532 nm [37] and destructive interference of the pump laser within the diamond membrane.

We can replace the 100× objective with the fiber mirror to observe cavity-coupled GeV emission as a function of cavity length, where Fig. 2b shows such a measurement at the position of the studied GeV. There is a clear peak in photon counts where the cavity length is resonant with the thermally broadened ZPL; optical filtering results in a profile that deviates from the expected Lorentzian. We repeat this measurement at various x-y positions on the membrane and plot the difference between maximum and minimum counts in Fig. 2c; where the lateral resolution is set by the 532 nm excitation spot size (\(\approx 1 \text{ µm}\)) since the cavity waist diameter is much larger (\(\approx 3 \text{ µm}\)). The contours obtained from the confocal measurement are overlaid for comparison, revealing
FIG. 2. a) A confocal image of membrane fluorescence (600 – 605 nm) taken at $P = 19$ mW. The white contours indicate emitter localization and the studied GeV center is indicated with an arrow. b) Cavity-coupled emission as a function of cavity length at the position of the studied GeV center ($\mathcal{F} = 11.200 \pm 170$ at 603 nm). The non-Lorentzian shape is determined by the filters in the collection path. c) A map of cavity-coupled emission amplitude taken at $P = 19$ mW over the same area as in a). The confocal contours are overlaid and the studied defect is indicated. d) Broadband cavity transmission as a function of cavity length. The dashed lines correspond to numerical fitting of the fundamental and first order transverse modes of the $m = 15$ and 16 longitudinal modes.

FIG. 3. a) GeV center spectrum taken in the confocal configuration. The pure dephasing rate $\gamma^*/(2\pi) = 5.22 \pm 0.05$ THz is indicated. b) GeV center fluorescence as a function of cavity length. The colored region represents the integration window. c) Integrated spectrum taken in the cavity configuration. The light trace shows the raw data, and the dark trace has been averaged over 40 points.

a clear spatial correspondence between emitters in the confocal scan and high emission into the cavity. This measurement corresponds to emission into the $m = 15$ longitudinal cavity mode, as determined by numerical fitting of the broadband cavity transmission measurements (Fig. 2d) [34].

Beyond spatial correlations between Figs. 2a and c, further evidence that we are probing a cavity-coupled GeV center can be obtained by comparing emitter spectra from both setups. Figure 3b shows a confocal spectrum for the studied emitter, which exhibits a strong ZPL emission around 603 nm with a FWHM linewidth of $\gamma^*/(2\pi) = 5.22 \pm 0.05$ THz [34]. A corresponding cavity measurement can be obtained by exciting the emitter while scanning the cavity length and acquiring a spectrum at each position (Fig. 3c). Integrating the emission along one mode using a 2 nm window about the resonance results in the light trace shown in Fig. 3c, where the noise can be attributed to mechanical instability of the cavity. To show the underlying spectral shape we also average the data using a 40 point window (dark trace).

Both measurements were taken using a 605$\pm$15 nm bandpass filter (Semrock), which did not limit the observed spectra. The peaks around 597 and 609 nm in Fig. 3c
are due to mechanical disturbances causing the cavity to pass through the same resonance twice. The qualitative similarities between the confocal and cavity spectra further confirm that the collected emission is coming from a GeV center.

In principle, comparison of cavity-coupled and free-space GeV fluorescence rates allows us to quantify cavity funneling. However, we must carefully account for different excitation intensities arising from interference effects in the diamond membrane and cavity. We thus compare the saturating fluorescence counts at infinite pump power ($I(\infty)$) obtained by fitting saturation curves with the model $I(P) = I(\infty) \frac{P}{P_{\text{sat}}} + c_0 g$, where $I$ is the observed count rate, $P$ is the excitation power, $P_{\text{sat}}$ is the saturation power, and $c_0 g$ accounts for a linear background. In the confocal setup, we measure a saturation power of $P_{\text{sat}} = 3.9 \pm 0.3$ mW and extract a saturating fluorescence count rate of $I_{\text{free, meas}} = 4,000 \pm 400$ counts/s. We note that this is lower than the count rates observed in Fig. 2a and we attribute this discrepancy to variation in optical alignment. There is consequently some additional error on the free-space emission rate which is not captured by our estimate of the collection efficiency for the confocal path ($\eta_{\text{free}} = (3.5^{+0.9}_{-1.5}) \times 10^{-3}$ counts/photon), which yields $I_{\text{free}} = I_{\text{free, meas}} / \eta_{\text{free}} = (1.2^{+0.3}_{-0.5}) \times 10^6$ photons/s, the total emission rate of the defect. Comparing this value to the maximum photon emission rate predicted at infinite pump power results in a quantum efficiency of $17^{+5}_{-8}$% for the studied GeV center, which falls within the range of reported values [24, 35, 36]. Furthermore, we collect photon emission between 600 - 605 nm, corresponding to a photon spectral density of 90 - 40 photons/(s GHz) (see [34] for details).

We then study the saturation behavior of the emitter coupled to the $m = 15, 16, 17$ longitudinal modes of the cavity. For each excitation power, we measure fluorescence as a function of cavity length (Fig. 4b). The amplitudes of these resonances are estimated by fitting with Gaussians, and are plotted as a function of excitation power in Fig. 4b. As expected, the cavity-coupled fluorescence decreases with mode number as the cavity mode volume increases. Fitting the $m = 15$ data to the saturation model yields $P_{\text{sat}} = 3.1 \pm 0.5$ mW and $I_{\text{cav, meas}} = 380 \pm 50$ counts/s. We estimate a collection efficiency of $\eta_{\text{cav}} = (8.2 \pm 1.2) \times 10^{-2}$ counts/photon for the cavity setup, resulting in a corrected saturating fluorescence count rate of $I_{\text{cav}} = I_{\text{cav, meas}} / \eta_{\text{cav}} = 4,700 \pm 900$ photons/s [34]. Moreover, we estimate a cavity linewidth of $\kappa/(2\pi) = 1.08 \pm 0.17$ GHz, leading to a cavity-enhanced peak spectral density of $2,800 \pm 700$ photons/(s GHz), a factor of $31^{+12}_{-5}$ greater than what was obtained with the confocal measurements. The experimentally determined efficiency of emission into the $m = 15$ mode is then $\beta_{\text{exp}} = I_{\text{cav}} / (I_{\text{free}} + I_{\text{cav}}) = 0.40^{+0.14}_{-0.19}$% (details in [34]).

For comparison, we simulate the expected $\beta$ value (Fig. 5) for an emitter resonantly coupled to the $m = 15$ cavity mode as a function of diamond thickness [34]. Here, the solid line shows the result for the target implantation depth (125 nm) and the shaded regions reveal the spread for one- or two-times the standard deviation in depth ($\sigma = 20$ nm), resulting in $\beta$ between 0.08% and 2.0% at $t_d = 862$ nm. The peaks in efficiency as a function of diamond thickness correspond to “diamond-like” modes, where the electric field in the diamond is maximized. We note that the observed $\beta_{\text{exp}} = 0.40^{+0.14}_{-0.19}$% lies at the lower range of expected values, which we tentatively attribute to non-ideal emitter location along the cavity axis due to implantation straggle.

Cooling such a high-quality membrane to cryogenic temperatures should result in a narrowing of the ZPL optical transitions, approaching the radiative limit of $\gamma/(2\pi) = 27$ MHz for a 6.0 ns excited-state lifetime. In this regime the cavity linewidth should far exceed that of the emitter, allowing for a Purcell enhancement of resonantly coupled optical transitions. The expected enhancement can be calculated using the extracted room-temperature system parameters as $F_p \approx \frac{\xi^2}{\beta_{\text{exp}}} \beta_{\text{exp}}$, where $\xi = 0.6$ is the GeV Debye-Waller factor [38]. This would result in a ZPL enhancement of $F_p = 32^{+12}_{-16}$ (assuming unity quantum efficiency and full depopulation of the dark state), corresponding to a $20^{+10}_{-10}$-times reduction in the excited state lifetime and > 95% of photons emit-
FIG. 5. Simulated $\beta$ for an emitter resonantly coupled to the $m = 15$ mode as a function of diamond thickness, with the experimentally derived value of $\beta_{exp} = 0.40^{+0.14}_{-0.19}\%$ plotted at the extracted membrane thickness $t_d = 862^{+4}_{-1}$ nm.

In summary, we demonstrate coupling of a single GeV center in a diamond membrane to a “diamond-like” mode of an open-cavity system, resulting in an $31^{+12}_{-15}$ times increase in the spectral density of single-photon emission. Moving forward, it should be possible to lock the cavity length (and therefore frequency) to a ZPL transition at cryogenic temperatures [39]; such resonant coupling would lead to a projected Purcell enhancement similar to what has been achieved for NV centers in open-cavities [17] with a much larger reduction in the excited state lifetime. This could be further improved by reducing the cavity mode volume via the mirror radius of curvature, where a shallow mirror with a $R \approx 5$ μm would increase the enhancement by a factor of $5$ [40]. In parallel, the suitability of the GeV as an efficient spin-photon interface should be confirmed through further characterization of the observed dark state and explicit measurement of the quantum efficiency (which could be done using such a tuneable cavity). In the future, this platform could be easily adapted for coupling to novel emitters based on heavier group IV elements such as the tin-vacancy (SnV) [12] and the lead-vacancy (PbV) centers [43], which should exhibit longer spin coherence times. This work therefore encompasses several important steps toward realizing an efficient spin-photon interface for defect centers coupled to open microcavities.

We would like to thank Marie-Josée Gour for assistance with membrane fabrication. We acknowledges support from Canada Foundation for Innovation and Canada Research Chairs project 229003 and 231949, Fonds de Recherche - Nature et Technologies FQRNT PR-253399, National Sciences and Engineering Research Council of Canada NSERC RGPIN 435554-13, l’Institut Transdisciplinaire d’Information Quantique (INTRIQ), the Danish Research Council through the Sapere Aude project DIMS, and the Danish National Research Foundation through the Center for Macroscopic Quantum States (bigQ, DNRF142). L. Childress is a CIFAR Fellow in the Quantum Information Science Program. Y. Fontana acknowledges support from SNSF Early Mobility Fellowship; CDRR was supported by a CONACYT fellowship; Y. Hi was supported by a MITACS Globalink internship.

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