Nanoimplantation and Purcell enhancement of single NV centers in photonic crystal cavities in diamond

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We present the controlled creation of single nitrogen-vacancy (NV) centers via ion implantation at the center of a photonic crystal cavity which is fabricated in an ultrapure, single crystal diamond membrane. High-resolution placement of NV centers is achieved using collimation of a 5 keV-nitrogen ion beam through a pierced tip of an atomic force microscope (AFM). We demonstrate coupling of the implanted NV centers’ broad band fluorescence to a cavity mode and observe Purcell enhancement of the spontaneous emission. The results are in good agreement with a master equation model for the cavity coupling.

The nitrogen-vacancy (NV) center in diamond has been successfully implemented as solid state quantum bit that meets all essential requirements for quantum information processing such as optical initialization, control and readout of the spin state. The challenge remains to extend the quantum system from a small number of qubits to large scale networks. Seminal experiments already demonstrated remote entanglement between individual NV centers via two-photon quantum interference\(^2,3\). The hitherto poor rate of entanglement events\(^2,3\) could be strongly increased by coupling the NV centers to optical microcavities. The effects range from enhancement and spectral reshaping of the NV spectrum over cavity-enhanced spin state readout\(^4\) to cavity mediated entanglement between two NV centers\(^5\). Photonic crystal (PhC) cavities directly fabricated in diamond are ideal for color center-cavity coupling experiments as they exhibit high Q-factors and extremely small mode volumes. For solid state systems, it is however challenging to precisely place the emitter in the maximum of the cavity electric field to achieve optimum coupling.

Past experiments that demonstrated coupling of single NV centers to PhC cavities\(^6,7\) have largely relied on random positioning. Controlled lateral positioning and emitter-cavity coupling has recently been achieved via a tailored fabrication process of a PhC around a single silicon-vacancy center in diamond\(^8\). Here, we pursue the complementary approach based on targeted implantation of NV centers into pre-defined cavities in diamond. In recent years, several techniques for spatially selective formation of single NV centers in bulk diamond have been developed involving focused nitrogen ion beam\(^9\), implantation through pierced AFM-tips\(^10,11\) and through small apertures in e-beam resist\(^12,13\), mica foils\(^14\), and silicon masks\(^15\). Using the silicon mask simultaneously as an etch mask would allow for controlled emitter-cavity placement\(^16\).

In our experiment, we achieve high resolution implantation of NV centers within two-dimensional diamond-based PhC cavities using a combined system of a nitrogen ion beam and an atomic force microscope (Fig. 1(a)) that allows for collimation and lateral positioning of the ion beam\(^10,11\). We verify the successful formation of a small number of NV centers and demonstrate Purcell enhancement of the broad NV emission when coupled to the confined cavity field. The PhCs consist of a trian-
Figure 1(d) shows the room temperature photoluminescence (PL) spectrum of the fabricated M1-cavity revealing three pronounced cavity modes $c_1$, $c_2$ and $c_3$ at 653, 670 and 681 nm, respectively, but no signature of NV emission in the ultrapure diamond material.

For deterministic creation of NV centers within the PhC cavities, we first use an AFM to image the PhC structures (c.f. Figs. 1(c)). A small hole (diameter <30 nm) drilled in the AFM tip serves as an aperture for the ion beam that allows for high resolution implantation of $^{15}$N$^+$ ions with an energy of 5 keV at the cavities’ center at different doses of 0.3 – 4.4 × 10$^{14}$ ions/cm$^2$. The low ion energy is chosen to achieve a high spatial resolution of <15 nm$^3$. According to Monte Carlo simulations (SRIM$^{[19]}$), the average implantation depth of the 5 keV-nitrogen ions is 8 nm with a small ion straggle of 3 nm. After implantation, the diamond sample is annealed at 800°C for 2 h in vacuum such that lattice vacancies diffuse in the diamond host material towards the implanted nitrogen ions to form optically active NV centers. Finally, the sample is cleaned again in a boiling acid mixture for 8 h in order to oxidize any graphite-like residuals and to convert the NV centers to the negative charge state.

Ensemble NV emission spectra taken at different reference spots implanted at high dose (5 × 10$^{14}$ ions/cm$^2$) aside the photonic structures reveal that up to 70% of the NV centers are converted to their negative charge state after all oxidation steps. In our analysis, we take into account the PL intensities integrated in a spectral range of 20 nm around the NV$^0$ ($\lambda = 575$ nm) and NV$^-$ zero-phonon line (ZPL) ($\lambda = 637$ nm) as well as varying detection efficiencies of our spectrometer, different absorption$^{[20]}$ and quantum$^{[21]}$ efficiencies of the two charge
We determine the number of NV centers created within each M1-, M3- and M7-cavity that were implanted at various ion doses by integrating the background corrected PL signal in the spectral range of 637 ± 8 nm and normalize it to the average ZPL intensity of a single NV center. As displayed in Fig. 2(c), the number of produced NV centers monotonically increases as a function of the applied implantation dose. By dividing the number of NV centers by the amount of implanted nitrogen ions, we obtain the NV creation yield shown in Fig. 2(d). For the ion energy of 5 keV, we find a creation yield of 0.8 ± 0.2% that is constant over the whole range of implantation dose. The small creation yield is within the range of experimental observations of yields < 0.1%23 to ∼25%24 which strongly depend on annealing and surface conditions. The creation yield is limited by loss of vacancies to the surface upon shallow implantation and surface effects possibly shifting the NV center charge state to NV⁺. From Fig. 2(c), we deduce an optimal dose of 1 × 10¹³ ions/cm² at an ion energy of 5 keV for deterministic creation of one single optically active NV center.

In the following, we analyze the intensity enhancement of the NV emission at the resonant wavelength of the M1-cavity mode c₁. Thereby, we take into account that the mode c₁ preferentially overlaps with the NV phonon side band (PSB) and not with the ZPL. As the resonance wavelengths and linewidths are known from the M1-cavity spectrum (Fig. 1) prior implantation, we can estimate the bare NV emission (orange line in Fig. 3(a)) without cavity modes. By comparing the integrated intensity of the cavity-enhanced emission I⁺ (orange + gray area in Fig. 3(b)) to the bare spectrum Ioff (orange area), we find an experimental enhancement factor of I⁺/Ioff = 1.24. Here, we solely focus on the intensity increase by the dominant c₁ mode and disregard other modes c₂ and c₃. In addition the emission efficiency into the mode c₁ is β = I⁺/Ioff = 0.31, where I⁺ is the emission channeled into mode c₁ (pink area in Fig. 3(c)) and Ioff is the overall intensity (pink + gray area).

To relate the resonant intensity enhancement of the NV PSB to a generalized Purcell factor F⁺, we adopt the master equation model55,28 for broad-band emitter-
the cavity coupling. In analogy to cavity coupling, the NV emission is modeled as a multi-level system (inset in Fig. 3(a)). The model input parameters are obtained from Lorentzian fits to the uncoupled NV spectrum (Fig. 3(a)). Solving the master equation model, we compute the generalized Purcell factor $F^\text{P} = F^\ast/(1 + F^\ast)$ (Fig. 3(d)) into the cavity when the resonant mode $\epsilon_1$ with $Q_\text{eff} = 160$ and $V_\epsilon = 1.1(\lambda/n)^3$ is tuned across the modeled NV spectrum. The individual contributions of the ZPL and PSBs to the total emitter-cavity coupling are shown by the filled curves in Figs. 3(d,e). For simplicity, our analysis assumes unity quantum efficiency and perfect spatial and orientational overlap of the two NV dipoles with the cavity field. At the resonant wavelength $\lambda_{\text{cavity}} = 653\text{ nm}$ of the M1-cavity mode (c.f. Fig. 3(d)), we find a theoretical Purcell enhancement of $1 + F^\ast = 1.7$ and an emission efficiency $\beta = 0.42$ that result in a theoretical intensity increase $I_{\text{on}}/I_{\text{off}} = ((1 + F^\ast)\epsilon_1+1 - (1 - \epsilon_1)/\gamma = 1.2$, considering that the $\epsilon_1$ mode preferentially overlaps with the first and second NV PSB with a relative contribution to the total NV emission of $\epsilon_1 = 0.29$. These theoretical values are in excellent agreement with our experiment.

In conclusion, we have demonstrated high resolution creation of a small number of NV centers at the center of diamond-based PhC cavities using collimated nanoimplantation of nitrogen ions through a pierced AFM-tip. For an ion energy of 5 keV, we found a constant NV creation yield of $0.8\%$, independent of implantation dose and cavity size. The lowest ion dose of $3 \times 10^{13}\text{ ions/cm}^2$ yielded $3 \pm 1$ NV centers placed at the center of a M1-cavity. The emitter-cavity coupling leads to an intensity enhancement of $I_{\text{on}}/I_{\text{off}} = 1.24$ when the cavity mode is in resonance with the NV PSB and an emission efficiency into the cavity mode of $\beta = 0.31$ which is in very good agreement with theoretical predictions. From our experiment, we deduce an optimal dose of $1 \times 10^{13}\text{ ions/cm}^2$ for the targeted creation of one single NV center within a PhC cavity. The here demonstrated high resolution implantation of single NV centers within PhC cavities is an essential step towards scalable solid-state quantum networks²⁷ or quantum repeaters²⁸ based on NV nanocavity systems.

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