Monolithic diamond optics for single photon detection

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In this work, we experimentally demonstrate a novel and simple approach that uses off-the-shelf optical elements to enhance the collection efficiency from a single emitter. The key component is a solid immersion lens made of diamond, the host material for single color centers. We improve the excitation and detection of single emitters by one order of magnitude, as predicted by theory.

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Over the past decade, solid-state optically active defects have attracted attention owing to the applications in quantum information science. Among the numerous studied color centers in diamond, the negatively charged nitrogen-vacancy (NV) defect is particularly interesting. For the NV center, a variety of applications has been demonstrated, such as diamond-based single photon sources,^{1,2} magnetic field sensors,^{3,4} and quantum information protocols.⁵ Most of these emerging technologies rely on the spin state of NV defects, which can be coherently manipulated with high fidelity,⁶ owing to a long coherence time.⁷ Moreover, the spin of NV defects can be efficiently read out⁸ and coupled to photons through spin-dependent transitions,⁹ a key ingredient toward long-distance quantum communications using photons as flying qubits.

For all of the above mentioned experiments, the photon collection efficiency is crucial. For single NV defects, this is reduced by the high refractive index of diamond ($n_d=2.4$), leading to total internal reflection at the diamond-air interface. The collection efficiency can be enhanced by the guiding emission in photonic nanostructures such as diamond nanowires¹⁰ or by using NV defects hosted in nanocrystals with a size smaller than the emission wavelength.¹¹ In this case, refraction becomes irrelevant, and the defect can be considered as a point source emitting in the surrounding medium. However, the spin properties of NV defects hosted in nanocrystals are often affected by surface defects leading to a short coherence time.¹² To overcome these limitations, we engineer a solid immersion lens (SIL) directly into the diamond matrix.^{13,14}

To estimate theoretically the collection efficiency, we assume a single emitter in diamond and use geometrical optics and Fresnel's reflection laws. For a single emitting dipole, the normalized photoluminescence (PL) intensity I_s (I_p) having s (p) polarization reads¹⁵

$$I_s = \frac{3}{8\pi} [1 - \sin^2(\theta) \cos^2(\phi)] \sin^2(\phi), \quad (1)$$

$$I_p = \frac{3}{8\pi} [1 - \sin^2(\theta) \cos^2(\phi)] \cos^2(\phi), \quad (2)$$

where θ is the emission angle and ϕ is the azimuthal angle measured from the dipole axis.

We first consider a dipole parallel to a planar diamond surface [Fig. 1(a)] and a collection optic characterized by its numerical aperture (NA). At the diamond-air interface, the total internal reflection is achieved as soon as $\theta \geq \theta_{\text{TIR}} = \arcsin(1/n_d) \approx 24.6^\circ$. The collection efficiencies η_s and η_p are then given by

$$\eta_{s,p} = \int_0^{\theta_m} \int_0^{2\pi} I_{s,p} T_{s,p} \sin(\theta) d\phi d\theta, \quad (3)$$

where $\theta_m = \arcsin(\text{NA}/n_d)$ and $T_{s,p}$ are the transmission coefficients of the s and p polarization components through the interface.

We now consider a dipole located at the center of a hemispherical diamond SIL used as a primary collection optic [Fig. 1(b)]. In that case, all light rays exit the SIL in a direction normal to the surface, thus avoiding any refraction at the interface. As a result, the collection efficiencies η_s and η_p are defined by Eq. (3), while using $\theta_m = \arcsin(\text{NA})$ and $T_s = T_p = 4n_c n_d / (n_c + n_d)^2$.

In the general case, the collection efficiency depends on the dipole orientation inside the diamond matrix. For NV defects, the PL is associated with two orthogonal dipoles D_x

FIG. 1. (Color online) [(a) and (b)] Ray propagation (a) for a single emitting dipole oriented parallel to the diamond flat surface and (b) for a dipole located at the center of a hemispherical diamond SIL. (c) Computed collection efficiencies of NV defect PL as a function of the NA of the collection optics. The simulation is performed for NV defects in a [100]-oriented sample with and without SIL. The inset shows the atomic structure of the NV defect in diamond.

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and D_y located in a plane perpendicular to the NV defect symmetry axis,¹⁶ which is a $\langle 111 \rangle$ crystalline axis [Fig. 1(c)]. In addition, the absolute orientation of the two transition dipoles is determined by the direction of nonaxial local strain in the diamond matrix. Since local strain is randomly distributed over a diamond sample, the orientation of the two orthogonal dipoles is random in a plane perpendicular to the NV defect symmetry axis.

For a $[100]$ -oriented diamond sample, the absolute angle between the NV defect axis and the diamond flat surface is the same for the four possible NV defect orientations.¹⁷ To evaluate the collection efficiencies for this case, we take into account the orientation of the two dipoles with respect to the flat diamond surface and apply the required rotations to Eq. (3). An average over the two dipoles and over all possible strain-induced orientations is taken. The results of the calculation are shown on Fig. 1(c). Using a 0.85-NA microscope objective, a 7.5-fold enhancement in collection efficiency is expected while using a diamond SIL. Note that this theoretical value requires a diamond SIL with a surface roughness of approximately $\lambda/10$ (Ref. 18).

A diamond SIL was fabricated as follows. Homoepitaxial chemical vapor deposition (CVD) diamond growth was performed using a selected high pressure, high temperature grown substrate, which had been processed prior to growth in order to minimize the dislocation density in the grown CVD diamond. A two stage chemical process was subsequently used to produce a grown CVD layer with a thickness of about 2.3 mm made up with a first layer containing ~ 0.2 ppm of single substitutional nitrogen. The final layer of ~ 0.6 mm was engineered with a significant reduced nitrogen concentration (< 5 ppb) to reduce the number of native NV defects. Two different nitrogen conditions were used, as the first higher doped nitrogen layer was believed to reduce the level of strain in the CVD layer before the second thinner but higher purity layer was grown. The SIL was subsequently processed from this CVD diamond sample, the flat surface being composed by the high purity layer, oriented along a $[100]$ crystalline axis. Using a combination of laser and mechanical processing stages,¹⁹ a 1 mm diameter SIL was produced with flatness better than 10 nm (rms), measured by atomic force microscopy.

Native single NV defects in the diamond SIL were imaged using conventional confocal microscopy, as depicted in Fig. 2(a). A frequency doubled Nd:yttrium aluminum garnet laser ($\lambda = 532$ nm) was tightly focused onto the flat surface of the diamond SIL using a 0.85-NA microscope air objective. The PL was collected by the same objective using the SIL as primary collection optics and directed either to a spectrometer or to a Hanbury-Brown and Twiss (HBT) interferometer for photon correlation measurements. A PL raster scan of a region close to the center of the diamond SIL is depicted in Fig. 1(c), showing an isolated photoluminescent spot. The spectrum recorded at this point exhibits a broadband emission with a characteristic zero-phonon line at the wavelength $\lambda_{ZPL} = 637$ nm [Fig. 2(d)], which is associated with a NV defect. The single-atom nature of the emitter was then verified by recording the second-order autocorrelation function $g^{(2)}(\tau)$ using the HBT interferometer.² As shown in Fig. 2(e), a subpoissonian photon statistics [$g^{(2)}(0) \approx 0.1$] is observed, proving a single NV defect. We note that optical aberrations can be clearly observed in the confocal raster

FIG. 2. (Color online) (a) Experimental setup. BS: quartz plate; O: microscope air-objective (NA=0.85); PH: 50 μ m diameter pinhole; F: 580 nm long-pass filter; FM: flip mirror directing the collected PL either to an imaging spectrometer or to a Hanbury-Brown and Twiss interferometer consisting of two silicon avalanche photodiodes placed on the output ports of a 50/50 beamsplitter. (b) Picture of a diamond SIL placed on a cover glass substrate. (c) Typical raster scan of a region close to the center of the SIL for a laser power of 215 μ W. The bright spot indicates a single NV defect, imaged with a signal to background ratio of 45:1. (d) PL spectrum showing a broad emission with a ZPL at $\lambda_{ZPL} = 637$ nm [(*) symbol]. (e) Second-order autocorrelation function $g^{(2)}(\tau)$ recorded for a laser power of 215 μ W.

scan, since the shape of the bright spot is not perfectly circular. Such aberrations are attributed to a lateral offset of 10 μ m between the NV defect location and the center of the diamond SIL.¹⁸

With the aim of quantifying the enhancement in collection efficiency, the PL rate R was measured as a function of the laser intensity $\mathcal{I}$ (Fig. 3). Experimental data were then fitted using a two-level model with the relation $R = R_{\infty} [\mathcal{I} / (\mathcal{I} + \mathcal{I}_{\text{sat}})]$, where $\mathcal{I}_{\text{sat}}$ is the saturation laser intensity for the given focus size and R_{∞} is the emission rate at saturation. For

FIG. 3. (Color online) Background-corrected PL as a function of the laser intensity. Points correspond respectively to a single NV defect imaged through the diamond SIL and through a planar diamond surface. Solid lines are data fitting using a two-level model. The inset shows the signal to background ratio as a function of the laser intensity.

the studied single NV defect imaged through the diamond SIL, we find $R_{\infty}^{\text{SIL}} = 493 \pm 5 \times 10^3$ cps and $\mathcal{I}_{\text{sat}}^{\text{SIL}} = 61 \pm 1 \mu\text{W}$. The same experiment was then performed for a single NV defect imaged from the backside of the SIL, i.e., imaged through a planar diamond surface. In that configuration, the saturation parameters are $R_{\infty}^{\text{noSIL}} = 80 \pm 2 \times 10^3$ cps and $\mathcal{I}_{\text{sat}}^{\text{noSIL}} = 1330 \pm 30 \mu\text{W}$. A more than sixfold enhancement of the collection efficiency is therefore achieved in high saturation conditions. Such a value is slightly smaller than the one predicted by pure geometrical optic arguments [Fig. 1(d)]. This discrepancy is attributed to the optical aberrations arising from the unperfect positioning of the NV defect in the center of the diamond SIL.¹⁸ We note that similar collection efficiency enhancements were obtained for two others single NV defects located around 10 μm away from the center of the SIL. However, if this distance exceeds 30 μm , the enhancement vanishes.

Beyond the collection efficiency enhancement, we note that exciting through the diamond SIL yields to a smaller excitation volume. Indeed, the saturation laser intensity $\mathcal{I}_{\text{sat}}$ is decreased by roughly one order of magnitude. This effect might have interesting applications in the context of stimulated emission depletion (STED) microscopy since the resolving power of STED is proportional to $\sqrt{\mathcal{I}_{\text{sat}}/\mathcal{I}}$ (Ref. 20). Furthermore, the reduced focus of the excitation laser funnels more efficiently photons toward the single emitter. This allows for the detection of a single defect in transmission through an efficient extinction of the excitation laser beam.²¹

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¹⁷The NV defect can be oriented along $[111]$, $[\bar{1}\bar{1}1]$, $[1\bar{1}1]$, or $[\bar{1}11]$ direction.

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