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Nitrogen-vacancy defect emission spectra in the vicinity of an adjustable silver mirror

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Optical emitters of quantum radiation in the solid state are important building blocks for emerging technologies making use of the laws of quantum mechanics. The efficiency of photon extraction from the host material is low for many solid-state systems due to their relatively high index of refraction. In this article we experimentally study the emission spectrum of an ensemble of nitrogen-vacancy defects implanted around 8 nm below the planar diamond surface and in the vicinity of a planar silver mirror. Scanning the distance between diamond and the mirror, we observe an enhancement of the spectral emission power by up to a factor of 3. We construct a model based on classical dipoles and elucidate the observations as being caused by interference in the far field of the emitters.

The nitrogen-vacancy (NV) defect in diamond has received significant attention in the past two decades, mainly due to the excellent coherence properties of the associated electron spin [1], the optical spin polarization and readout techniques [2], and the possibility of emitting single photons at room temperature [3]. With these properties, the NV defect has emerged as a prime candidate for novel quantum technologies with a focus on applications in quantum information processing [4] and sensing of magnetic fields [5–7], electric fields [8] and temperature [9]. Moreover, by mapping the polarization of an electron spin to nearby nuclear spins may eventually facilitate the construction of a quantum simulator [10].

Spin-photon entanglement [11] and quantum interference of photons from two independent NV defects [12, 13] has been observed. Such features are essential for the demonstrations of quantum teleportation [14] and entanglement of spins separated by a large distance [15]. The success rate of these protocols among many other quantities is strictly limited by the collection efficiency of indistinguishable photons within the zero-phonon line (ZPL) of the NV center. At cryogenic temperatures, only about 3% of photons are emitted into the ZPL and typically $\ll 1\%$ of photons are detected with the collection optics of the setup. Enhancing the emission rate or increasing the photon collection efficiency from the far field have been suggested to increase the attainable photon counts on the detector, and demonstrations include broadband plasmonic enhancement [16], plasmon-based cavities [17], an open micro-cavity system [18], or solid immersion lenses milled into the host material [19]. However, these approaches are technically challenging in terms of device processing, nano-fabrication, and control.

In this Letter, we investigate the spectral emission properties of NV defects in a bulk diamond sample and in

proximity to a planar reflecting interface with adjustable distance. Our experiment shows that the collection efficiency in an adjustable spectral range from NV defects implanted approximately 8 nm below the surface can be increased by up to a factor of 3. Our experimental findings are explained by a modification of the NV center far-field emission pattern and collection efficiency into the numerical aperture (NA) of our collection optics.

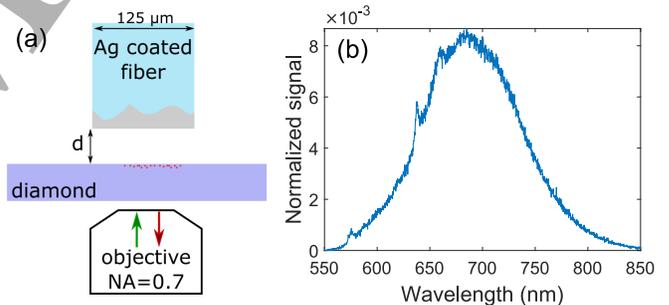


FIG. 1. (a) Schematic illustration of the experimental setup: the diamond sample with a thickness of $300 \mu\text{m}$ and NV centers implanted 8 nm below the surface is mounted in a standard confocal setup equipped with a $\text{NA}=0.7$ microscope objective. A cleaved optical fiber coated with silver is mounted on top of the diamond with a piezo actuator used to adjust the distance d to the diamond sample. (b) Reference NV emission spectrum $S_{ref,0}(\lambda)$ normalized to unit emission power and representative for the diamond sample.

Our experimental setup is illustrated in Fig. 1(a). We use an electronic-grade bulk diamond sample from Element Six with a thickness of $300 \mu\text{m}$. Nitrogen atoms are implanted with an energy of 5 keV and a dose of 10^{13} cm^{-2} approximately 8 nm below the diamond surface. The sample is subsequently annealed at 800°C in high vacuum yielding a uniform distribution of NV defects. The optical investigations are carried out in a standard confocal microscope, equipped with a 532-nm

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continuous-wave laser for the optical excitation of NV defects, an avalanche photo diode for fluorescence detection, and a spectrometer with 500 mm focal length and a grating with 150 lines/mm for spectral analysis. We use an objective lens with a NA of 0.7 and an adjustable collar (Olympus LUCPLFLN 60x) to partially correct for aberrations in the diamond sample. The reflecting interface atop the diamond sample is made of a cleaved optical fiber with a diameter of 125 μm and coated with a more than 300-nm-thick layer of silver deposited using thermal evaporation. The fiber is mounted on a piezo-actuator stage with a fine-tuning range of 20 μm to adjust the distance d between the diamond surface and the fiber mirror with nm precision.

We begin the optical investigations by recording a reference fluorescence spectrum S_{ref} from the NV layer without the fiber mirror. The result is shown in Fig. 1(b). The spectrometer covers the spectral range 540–900 nm, which includes the ZPL of the neutral (NV^0) and the negative (NV^-) charge state of the NV defect and their phonon side-bands, respectively. The optical excitation power was chosen to be about half the saturation pump power of single NV^- defects in our setup in similar diamond with lower NV concentration, and held constant for all investigations reported in this article. We also recorded spectra from 10 different locations away from the reference point and along a line in steps of 1 μm . These spectra are nearly identical to each other, which confirms the uniformity of NV defects in the implantation region. They all show a noticeable ZPL of NV^- at around 637 nm and a broad contribution due to scattering on lattice phonons in the range 640–800 nm. The tiny peak at 575 nm indicates a contribution of NV^0 and hence small fluctuations between the two possible charge states.

Next, we mounted the silver-coated fiber in the confocal setup, centered it on top of the optical excitation spot and minimized the distance to the diamond sample. Since the fiber cleaving process did not yield an absolutely flat surface, irregularities of the fiber surface were seen when touching the diamond at different points and thus the smallest possible distance d_0 we could achieve was in the range from 0.5 μm to 1 μm . Keeping the excitation at a fixed reference point on the diamond and increasing d in steps of 10 nm, we recorded NV center emission spectra $S(d, \lambda)$ for each position of the fiber (here λ is the emission wavelength). The obtained spectra are plotted as a heat map in Fig. 2(a); they produce a complex pattern which is caused by the interference of both the pump laser and NV center fluorescence. Interference of the pump laser solely modulates the pump power at the location of the NV centers, but does not impact the NV emission spectrum $S(d, \lambda)$ for a given distance d . In order to remove the effect caused by laser interference, we normalized the spectra for each distance d to unit spectral counts, $\int S_0(d, \lambda) d\lambda = 1$. Finally, the spectral enhancement E is obtained by comparing the normalized emission spectra to the normalized reference

spectrum in which the mirror is removed from the setup, $E = S_0(d, \lambda)/S_{ref,0}(\lambda)$. The distance-resolved enhancement spectrum E is plotted in Fig. 2(b) and the overall structure resembles the interference pattern of an optical resonator. It is clear that when increasing the distance d , the number of eigenmodes in wavelength space increases. An irregularity in the pattern observed for a mirror position $d \approx 3 \mu\text{m}$ we attribute to a slow thermal drift of the fiber relative to the diamond and it occurs due to the long total measurement time of around 8 hours.

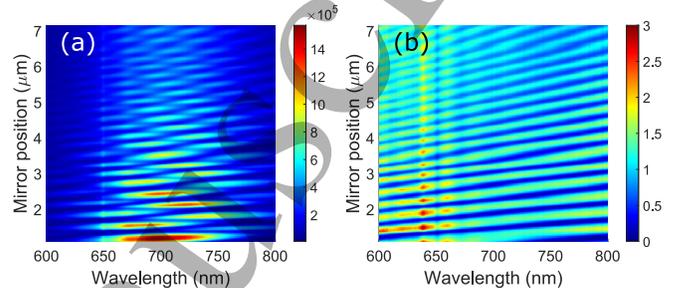


FIG. 2. (a) Absolute NV defect emission spectra and (b) enhancement spectra recorded as a function of mirror position d relative to the diamond sample surface. See the main text for details.

In our model we consider the ensemble of NV defects as non-interacting and classical electric point dipoles with angular frequency $\omega = 2\pi c/\lambda$ located close to an interface with planar stratified media, where c is the speed of light in vacuum. The radiation dynamics [20] and the radiation pattern [21] of electric dipoles in such a configuration have both been described in detail by Lukosz and Kunz. The method consists in decomposing the radiation pattern of a dipolar emitter into plane waves and considering the propagation of each component. Analytical expressions can be obtained for the power-density spectrum in the wave-vector space for dipoles oriented parallel (\parallel) and perpendicular (\perp) to the interface [22, 23]. Here, we apply this model to calculate the power radiated by an ensemble of electric dipoles close to stratified media into the NA of the microscope objective. The stratified media in our case are formed by the diamond with planar surfaces, an air gap of adjustable size d , and the fiber end-facet coated with silver. For a set of d and λ , we first calculate the power detected $P(d, \lambda)$ by integrating the angular power density over the radiation modes within the NA of the objective. The theoretical enhancement spectrum is then obtained by normalizing $P(d, \lambda)$ to the collected power when the mirror is removed from the setup.

In the calculation, the emission of a NV defect is represented with two classical dipoles lying in a plane perpendicular to the NV symmetry axis [24]. Here, we assume that the orientation of the dipoles is homogeneously distributed in the plane. Averaging over the four possible orientations of NV defects in the diamond crystal lattice results in the relative dipole strength parallel $a_{\parallel} \approx 0.659$

and perpendicular $a_{\perp} \approx 0.341$ to the planar diamond-air interface [25], which nearly is a homogeneous distribution of dipoles. The refractive index of silver over the entire spectral range $n_{Ag}(\lambda)$ is taken from Johnson and Christy [26], while the refractive index of diamond is assumed to be constant with $n_d = 2.41$.

The calculated enhancement spectrum for the same range of mirror position and λ as in the experiment is presented in Fig. 3(a). We obtained qualitatively the best match with the experiment when limiting the NA to 0.35 in the calculation. This observation we attribute to aberrations due to the high index of refraction and dispersion of diamond, which was not accounted for by the objective. Comparing the number of oscillations within the mirror scanning range (for a fixed λ) between experiment and simulation as well as the slope of the first mode when the mirror is closest to the diamond, we can estimate the smallest mirror distance d_0 in the experiment to be around $0.5 \mu\text{m}$.

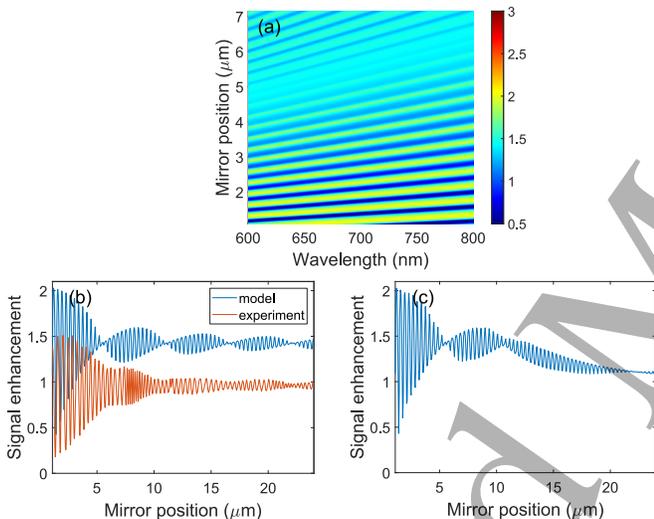


FIG. 3. (a) Calculated enhancement spectrum and (b) line cuts comparing the calculated and measured enhancement spectrum at $\lambda = 700 \text{ nm}$. (c) Evaluation of signal enhancement at $\lambda = 700 \text{ nm}$ using a modified model assuming a laterally limited silver mirror (see main text for details). Under this assumption, the enhancement tends to the same value as in the experiment when the mirror is brought to infinity.

Line cuts of the enhancement spectrum for $\lambda = 700 \text{ nm}$ taken from the experiment and simulation are presented in Fig. 3(b). One can see that the general trend including high- and low-frequency oscillations is well reproduced numerically. The model suggests a maximum enhancement factor of around 2, while a slightly smaller factor of up to around 1.5 is observed for this wavelength in the experiment for a mirror position $d < 5 \mu\text{m}$. This minor discrepancy is likely due to imperfections of the silver mirror yielding a lower reflectivity than considered in the model.

More importantly, numerical modelling yields an upper limit for the signal enhancement at infinite distance than

what we obtain experimentally. It is therefore important to note that we use a planar geometry in modelling, i.e. the mirror has an infinite size and hence maintains its effect even at infinite distance. In the experiment and due to the limited lateral mirror size, final tilt, and surface roughness, retracting the mirror to infinity effectively excludes it from the experiment. To verify that the discrepancy in the enhancement limit is indeed imposed by the (decreasing with distance) limited NA of the mirror, we show the result of a very crude model that attempts to take that effect into account [Fig. 3(c)]. In this adjusted model we assumed the mirror size to be $8 \mu\text{m}$ and set the mirror reflectivity to zero for wavenumbers exceeding the NA of the mirror. Clearly, at large distances, the intensity enhancement now tends to values obtained experimentally even though the exact shape of the profile is not well reproduced.

We would like to emphasize that our observations are not due to a Purcell enhancement. In fact, for our configuration we calculate a moderate Purcell suppression factor (compared to an emitter in bulk diamond) in the range of 0.68–0.75 when the mirror is more than 100 nm away from the diamond. At shorter distances, there is Purcell enhancement, but there is no spectral enhancement due to quenching of radiation near metallic surface of the mirror. Instead, our experimental observations are well explained by interference and a modification of the emitter far-field radiation pattern depending on the mirror position d . In Fig. 4, we compare the calculated angular power density (at $\lambda = 700 \text{ nm}$) for various distances corresponding to local maxima [Figs. 4(a,c)] and minima [Figs. 4(b,d)] in the oscillating signal enhancement. Both the displayed radiation pattern as well as the power radiated within the NA of our setup (indicated by red solid lines) differ significantly. It is interference in the far field of the classical dipoles that causes the change and therefore explains the observation.

By analyzing angular power spectra (not shown) that were used to obtain the enhancement map presented in Fig. 3(a), we conclude that the high-frequency oscillations in Fig. 3(b) stem from periodic alterations between an odd and an even number of nodes in the angular spectrum within the NA of the objective. The low-frequency beating corresponds to an increase by one in the number of alternating nodes, i.e. a change from alternating nodes zero and one to alternating nodes one and two, then a change to nodes two and three, and so forth. Since in real objectives, the NA is not sharply defined due to diffraction at the edges of the objective entrance pupil, such transitions to a higher number of nodes is also blurred. This explains why the low-frequency beating is less pronounced in the experimental plot in Fig. 3(b).

Finally, we note that in the experiment and for $d \approx 5 \mu\text{m}$, the enhancement at $\lambda = 637 \text{ nm}$ and $\lambda = 659 \text{ nm}$ is significantly more pronounced than for other wavelengths – an enhancement factor of up to 3 is obtained [cf. Fig. 2(b)]. These features are observed at the NV⁻ ZPL and the first-order phonon scattering peak red shifted

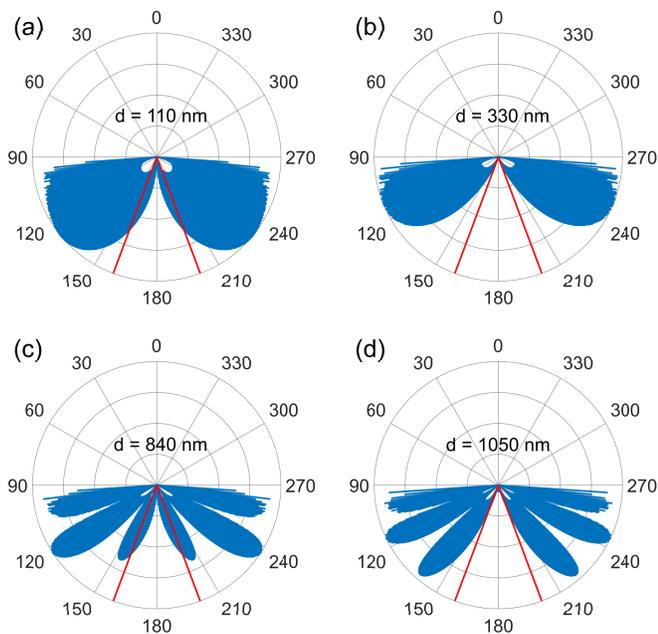


FIG. 4. Calculated angular radiation pattern for (a) $d = 110$ nm, (b) $d = 330$ nm, (c) $d = 840$ nm, and (d) $d = 1050$ nm, while $\lambda = 700$ nm. Panels (a) and (c) correspond to local maxima and (b) and (d) to local minima in the oscillating signal enhancement in Fig. 3(b). Red lines indicate the range of angles within the effective NA = 0.35 of the setup.

by ≈ 65 meV, which might indicate complex emitter dynamics. Increase of the ZPL brightness is of great impor-

tance in solid-state quantum optics and is conventionally attempted using resonant high-finesse Fabry-Perot cavities [27, 28]. The quality of reflecting surfaces in our experiment (silver mirror and diamond-air interface) is, contrary, relatively low, therefore it will be interesting to further investigate the physical nature and the potential use of the observed phenomenon. Our model is based on classical dipoles with an emission λ and clearly does not explain such features in the observed spectrum. A more sophisticated model including quantized energy levels, coherences between them, and scattering on lattice phonons might be required to quantitatively describe the observation.

In conclusion, we experimentally studied the emission spectrum of NV defects implanted in a bulk diamond sample near its surface and in the vicinity of a planar silver-coated mirror. We experimentally observe a spectral enhancement by up to a factor of 3, which can be tuned in spectrum by adjusting the distance between the diamond and the mirror. We constructed a simple model with classical dipoles and attribute our observations to interference in the far field of the emitter radiation. Enhancement features at the NV⁻ ZPL and the first-order phonon scattering peak are not explained by this model and require further investigations. Our experimental approach is easy to implement and may be applied to other solid-state systems such as emitters in GaAs.

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