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We experimentally demonstrate a simple and robust optical fiber based method to achieve simultaneously efficient excitation and fluorescence collection from Nitrogen-Vacancy (NV) defects containing micro-crystalline diamond. We fabricate a suitable micro-concave mirror that focuses scattered excitation laser light into the diamond located at the focal point of the mirror. At the same instance, the mirror also couples the fluorescence light exiting out of the diamond crystal in the opposite direction of the optical fiber back into the optical fiber within its light acceptance cone. This part of fluorescence would have been otherwise lost from reaching the detector. Our proof-of-principle demonstration achieves a 25 times improvement in fluorescence collection compared to the case of not using any mirrors. The increase in light collection favors getting high signal-to-noise ratio optically detected magnetic resonance signals and hence offers a practical advantage in fiber-based NV quantum sensors. Additionally, we compacted the NV sensor system by replacing some bulky optical elements in the optical path with a 1 × 2 fiber optical coupler in our optical system. This reduces the complexity of the system and provides portability and robustness needed for applications like magnetic endoscopy and remote-magnetic sensing. Published by AIP Publishing. https://doi.org/10.1063/1.5037807

The negatively charged Nitrogen-Vacancy (NV) color center in diamond is a promising candidate for quantum information processing and spin-based quantum sensing of magnetic fields,1–3 electric fields,4,5 and temperatures.6–8 To efficiently use the NV center in these applications, enhancing the excitation and collection of the color-center’s fluorescence is demanded. For instance, a high fluorescence collection efficiency would benefit precision metrology. Some methods, for example, circular gratings9,10 or solid immersion lenses (SILs),11–13 when fabricated on diamond substrates, extract much fluorescence from a single NV center out of the high refractive index (~2.4) material. While Micro/nanoresonator cavities and tapered optical fiber have been shown to enhance both the single NV center’s fluorescence emission and coupling-out efficiencies,14–16 optical antennas and plasmonic structures have been used to enhance the local light field, thus improving the fluorescence efficiency through the Purcell effect, and enhance emission from a single NV defect in diamond.17,18

Here, we demonstrate a simple method to enhance the excitation efficiency and also fluorescence collection from a micrometer-sized NV color-center rich diamond attached to the end of a sphere optical fiber. This single-port sensor configuration offers flexibility needed for some applications that require precision field-measurements in a non-standard location (e.g., magnetic endoscopy, subcutaneous measurements, and remote and inaccessible locations). Our enhancement technique is based on using a matched micro-concave (MC) mirror to a sphere optical-fiber end.

A diamond micro-crystal is fixed on the sphere tip optical fiber and excited by 532 nm laser light guided through the fiber. The diamond scatters incident light except for a fraction that is absorbed by the NV defects. The MC mirror reflects and focuses the scattered laser light into the diamond (on the sphere fiber tip) that is positioned at the focal point of the MC mirror. At the same time, the MC mirror also serves a dual purpose to reflect and focus the fluorescence light that is scattered out of the diamond into the optical fiber within its acceptance angle, thereby increasing the effective numerical aperture of the light collection system. In a proof-of-principle demonstration and validation experiment, we were able to collect over 25 times higher fluorescence intensity from the diamond located at the focal point of an MC mirror compared to the intensity collected without any mirrors. Although we only demonstrated the MC mirror for
enhancing excitation and fluorescence collection of NV ensembles in micrometer-sized diamond, we could foresee that this method can improve laser excitation and fluorescence collection of single NV centers and other solid-state quantum emitters such as Silicon-Vacancy centers in diamond and silicon-vacancies in Silicon carbide and rare-earth ions. In addition, we simplified the optical fiber-based NV sensor system by using a fiber optical coupler to replace the bulk lens and dichroic-beam-splitter based optical path splitter. This replacement reduces the complexity, enhance the portability, and improve the robustness of this optical-fiber-based NV system.

The working principle, schematic diagrams, and ray optic fluorescence excitation and collection simulations of the MC mirror-micro-crystal-optical fiber system are shown in Fig. 1. Although the micro-crystals have high density of NV defects, because of low optical absorption cross-section of the NV defects, except for a small portion of the laser light being absorbed by the dense NV centers, most of the laser light exiting the optical fiber will pass through the diamond. In this case, an MC mirror located with its focal point at the diamond center will reflect and focus the unabsorbed laser light back into the diamond to excite the NV centers, and this enhances the excitation efficiency [Figs. 1(a) and 1(c)]. Moreover, because of the unstructured surface of the diamond, the excitation laser undergoes partial light-trapping due to total-internal refection at the surface. Hence, the excitation laser light undergoes many rounds of reflection within the diamond crystal before being excited from the diamond; thankfully, the presence of the MC mirror further couples the laser light into the diamond, thus promoting further excitation of NV centers. Simultaneously, the MC mirror will also reflect and focus most of the fluorescence that could exit out of the diamond in the opposite direction of the optical fiber back into the optical fiber within its light acceptance cone. In this way, the MC-fiber system enhances the fluorescence collection from a NV micro-crystal [Figs. 1(b) and 1(d)].

The MC mirrors are fabricated by slightly pressing the center of a small section (<2 mm) of thin aluminum (Al) foil (~15 μm in thickness; purchased from a local supermarket) with an optical fiber with its tip that is shaped as a hemi-spherical end on an ultra-precision fiber optic alignment stage (M-561D-XYZ, Newport, USA). The Al foil is first glued onto a small plastic sheet with a hole at the center. The spherical tipped optical fiber is fabricated by using arc discharge of a tapered optical fiber tip. The desired taper and the spherering are obtained by heating and simultaneously stretching in an electric fusion splicer (Fitel S153A, Japan). The shape and focal length of the mirror can be adjusted through modifying the fabrication parameters such as the stretching force and the arc discharging current. In the initial phases, we made a set (array) of 5–10 MCs and individually tested the enhancements. The enhanced collection, normalized to the maximum of the fluorescence signal, deviates less than 1%–5% in total. Hence, for subsequent studies, we just fabricate one MC and that works well as expected. In other words, the good-micromirror turn-out ratio is near unity taking into consideration the 1%–5% tolerances. The MC mirror’s shape and focal length are quite robust and will not change once the tip-sphered optical fiber is chosen. The fabricated MC mirror is quite stable against drifts and other conditions: even after months, it still preserves its shape and gives the same efficiency of enhancements. Figures 2(a)–2(c) show scanning electron microscopy images of two tip-sphered optical fibers and one fabricated Al MC mirror. Figure 2(d) shows one fabricated Al MC mirror focusing the illumination light and forming a bright spot in an optical microscopy image.

We used the experimental setup depicted in Fig. 3 to study and quantify the enhancement of the MC mirror-system’s NV fluorescence excitation and collection efficiency. The excitation light (solid arrows in Fig. 3) from a 532 nm laser source is collimated and passed through a clean-up filter and then coupled into the 10% port of a 90:10
1 × 2 fiber optical coupler (Beijing XingYuan AoTe Technology Co., Ltd., China) using an objective (10×, NA = 0.1). About ~10% of the laser power is guided to the ~8-μm diamond glued using an UV curing glue at the center of the sphered end of the graded index multi-mode fiber (GIF625, Thorlab). The fluorescence (dashed arrows in Fig. 3) is collected by the same fiber and guided back into the input port of the fiber optical coupler; ~90%. The collected fluorescence light passes through a long-pass (LP) filter (cut-off wavelength 615 nm) and is detected using an optical spectrometer (BTC655, B&W tek, USA) or photo-diode (APD430A/M, Thorlab) (OS/PD in Fig. 3). The micro-size diamond is crushed from NV centers enriched in an HPHT (high pressure, high-temperature) type-Ib single crystal which was electron-irradiated and subsequently annealed. It should be noted that the laser light exiting out of our optical fiber is unpolarized, and so, the orientation of the diamond crystal will have little effect on the NV excitation efficiency.19 The Al MC mirror is mounted on a precision adjustable three-dimensional translation stage facing the diamond on the sphered fiber end (inset of Fig. 3). For experiments that require signal acquisition using a lock-in amplifier or using a data acquisition (DAQ) card, the connections depicted by the dashed line and double-solid lines in Fig. 3 were swapped.

For systematically studying the fluorescence enhancements, we recorded the spectrum of the diamond placed on the sphered fiber end in three different configurations. Case I, Diamond is placed far away from the MC mirror (not using any mirror). Case II, The diamond faces a plane mirror (focus at infinity). Case III, the diamond is approximately located at the focal point of the MC mirror. The results are shown in Fig. 4(a), and the respective configurations are labeled as cases I, II, and III. The incidence laser power (into the fiber) used for these experiments is ~0.49 mW. From the results of Fig. 4(a), we could observe that case III (with MC) shows a considerable increase in the collected fluorescence. To quantify the results, we calculate the integrated area from the obtained spectrum (from case III) and compare that with that of not using any mirror (case I). We get a modest value of over 25 times increase in the fluorescence collection when the MC mirror is placed at the focus (case III) [Fig. 4(a)].

For studying the advantages of enhanced fluorescence collection on magnetometry applications, we performed the optically detected magnetic resonance (ODMR) on the micro-crystals, fiber, and MC system. As an addition to the previous setup, we placed a permanent magnet and a micro-wave coil (RF coil) near the diamond. The NV defects in a diamond micro-crystal occupy four crystal orientations; hence, for these studies, we oriented the field arbitrary to the crystal orientation such that we obtain eight ODMR transitions (two for each of the crystal orientations). We could also orient the magnetic field to the NV quantization axis of a crystal orientation to obtain fewer lines and a better

**FIG. 2.** (a) and (b) Scanning electron microscopy (SEM) image of two prepared tip-sphered optical fibers with different sphere shapes. (c) SEM image of a fabricated MC mirror on aluminum (Al) foil; (d) optical microscopy image of a fabricated aluminum MC mirror focusing the illumination light and forming a bright spot in the middle and a bright circle encircling it.

**FIG. 3.** Experimental setup for assessing the MC mirror’s NV fluorescence excitation and collection enhancement. The inset depicts the set of the diamond, the Al MC mirror (Al foil), and the microwave conducting wire (RF coil). In the optically detected magnetic resonance (ODMR) scanning experiments: when no lock-in amplifier is used, the dashed line was connected instead of the three double-solid lines, and the photo-diode was read by the data acquisition (DAQ) card; when the lock-in amplifier is used, the three double-solid lines were connected instead of the two dashed lines, and the photo-diode was read directly by the lock-in amplifier.

**FIG. 4.** (a) The measured fluorescence spectra of an ~8-μm diamond glued on a spherical optical fiber end in three different configurations: Case I (not using any mirror), case II (facing an Al flat mirror), and case III (diamond at the focus of the Al MC mirror). ODMR spectra of the diamond without any averaging (b) case III and (c) case I. The laser power (~0.49 mW) and magnetic field (~2.74 mT) are kept the same.
The concave etched fiber is then coated with a thin film of a smooth concave surface aligned to the core of the fiber. This is done by first etching the graded index fiber GIF625 flat end face with 40% hydrofluoric acid for about 1 min. This forms an MC mirror directly on an optical fiber. The fabrication is done by following the ODMR transition frequencies and ensuring the lines ($m_s = 0 \rightarrow \pm 1$) split to maximum and also symmetrically to that of zero-field-splitting (2870 MHz). For the ODMR experiments, a permanent magnetic field is placed perpendicular to the optical fiber and fine adjustments are made to align the B-field. We recorded the ODMR spectrum shown in Figs. 4(b) and 4(c) which show the results obtained by sweeping the microwave frequency and synchronously recording the fluorescence intensity for cases I and III. It is evident that in case III, we can easily obtain the ODMR spectrum in a single-scan (without averaging) [Fig. 4(b)], while the results from case I [Fig. 4(c)] are very noisy in a single-scan experiment. For a proper comparison, we have fixed the excitation laser power to be the same for both the cases shown in Figs. 4(b) and 4(c).

To obtain a high resolution ODMR spectrum, we have used a lock-in amplifier configuration. The microwave field that drives the NV electron spins is amplitude modulated (at a modulation frequency of 25 KHz and a depth of 100%), and the signal is recorded using a photo-diode. When the lock-in technique is applied, we get ODMR signals with a significantly high signal-to-noise ratio (SNR). The Nitrogen hyperfine lines are clearly visible (inset in Fig. 5). Comparing the ODMR results of case III [Fig. 5(a)] and case I [Fig. 5(b)], we could certainly ascertain that much better SNR of the ODMR signal is obtained when the diamond is positioned at the focal point of the MC.

The fabrication of MC mirror using an Al film is simple and straightforward, but its relatively large size reduces the compactness of the optical fiber-based NV sensor systems. To efficiently solve this issue in a simple way, we integrated an MC mirror directly on an optical fiber. The fabrication is done by first etching the graded index fiber GIF625 flat end face with 40% hydrofluoric acid for about 1 min. This forms a smooth concave surface aligned to the core of the fiber. The concave etched fiber is then coated with a thin film of gold obtained by thermal deposition. Figure 6(a) shows the scanning electron microscopy (SEM) image of one fabricated on-fiber MC mirror.

One piece of diamond micro-crystal is taken from NV enriched HTHP synthetic mono-crystalline diamond powder and placed at the center of a flat optical fiber end facing an on-fiber integrated MC mirror. We obtained an efficiency enhancement of about 10 times in fluorescence excitation and collection for such a configuration from a $\sim10 \mu m$ diamond. Although the integrated MC system shows a modest 10 times increase together with the compactness of the system, the enhancement obtained in this case is certainly not on par with those achieved with the Al foil MC system. This is because the chemical etching has not formed a perfectly matched MC mirror that enables focusing the laser into the diamond and focusing the fluorescence back into the light acceptance cone of the optical fiber. We believe that through optimizing the etching parameters or using other controllable micro-fabrication techniques such as CO$_2$ laser ablation$^{20}$ and ion etching,$^{21}$ to form optimized shaped on-fiber MC mirrors such as the parabolic reflector on the fiber end, this compact design can also achieve a high-efficiency enhancement.

In this method, the MC mirror can only help in collecting the fluorescence that has exited out of the diamond. As the diamond has a high refractive index ($\sim 2.4$), fluorescence from the NV defects undergoes multiple total internal reflections within the diamond micro-crystal. This trapped light in the diamond cannot be collected by the MC mirror. However, methods such as the solid immersion lens (SIL)$^{11–13}$ and the circular Bragg grating$^{9,10}$ can be used for extracting fluorescence out of the diamond. Combining the MC mirror with this SIL or circular Bragg grating on the diamond itself will effectively enhance the fluorescence collection efficiency. For example, Fig. 6(b) shows the working principle schematic diagram of combining a simple parabolic shaped MC mirror and a fluorescence extracting SIL to further enhance the diamond NV center’s fluorescence excitation and collection. Figure 6(c) depict how our MC mirror based configuration can be potentially used in liquid sample sensing or integrated with a microfluidic system. In addition, the metal MC mirror is also found to be able to act as a microwave near-field antenna sufficient to perform NV spin manipulation when it is connected to a microwave synthesizer and properly impedance matched.

![FIG. 5. 20 times averaged lock-in amplifier outputs of the ODMR spectra of an $\sim 8 \mu m$ diamond glued on a spherical optical fiber end (a) located at the focal point of the MC mirror (case III) and (b) facing no mirror (case I). The insets show enlarged spectra of Nitrogen hyperfine splitting of the NV centers. The laser power ($\sim 0.49$ mW) and magnetic field ($\sim 1.52$ mT) values are the same.](image)

![FIG. 6. (a) SEM image of a fabricated MC mirror. (b) Principle schematic diagram of combining the solid immersion lens (SIL) with the parabolic shape MC mirror to further enhance the fluorescence collection efficiency of NV centers. (c) A possible usage setup: the MC mirror is used as a test sample keeper; simultaneously, it is connected to the microwave synthesizer for delivering microwaves to the NV centers.](image)
We have demonstrated a simple method based on an MC mirror to enhance the laser excitation and also fluorescence collection in an optical fiber-based NV sensor system. Using this technique, we achieve 25 times more fluorescence collection from an NV enriched micrometer-sized diamond attached to an optical fiber end when compared to that not using the MC mirror. This enhanced detection efficiency also results in ODMR signals with high SNR. This MC mirror + Fiber system method can also be applied to other solid-state quantum emitters such as silicon-vacancy centers in diamond, quantum dots, and rare-earth ion-based color centers. Additionally, we made the system very compact by using a 1 × 2 fiber optical coupler to replace the bulky optical element based optical path splitter in the fiber-based NV quantum sensor system. This single-ended sensor configuration together with the improved SNR could be of immense use in portable and robust magnetic sensor applications.

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