Enhancing fluorescence excitation and collection from the nitrogen-vacancy center in diamond through a micro-concave mirror

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Efficiently exciting the Nitrogen-Vacancy color-center in a diamond with a green light and collecting the emitted near-infrared fluorescence is critical for spin-based quantum sensing at room temperatures. Here, we experimentally demonstrate a simple and robust fiber-optics based method to achieve simultaneously efficient excitation and fluorescence collection. Fiber optic systems usually have limited Numerical Aperture (NA) while our method mitigates this “bottleneck”. We use a simple technique to fabricate a suitable micro-concave mirror that focuses the scattered excitation laser beam back into the diamond located at the focal point of the mirror. At the same instance, the mirror also couples back the fluorescence light exiting out of the diamond opposite the direction of the optical fiber into the optical fiber within its light acceptance cone, otherwise, it would have been lost. Our proof-of-principle demonstration achieves a 25 times improvement in fluorescence collection compared to not using any mirror. Additionally, we made the NV sensor system compact by replaced some bulky optical elements in the optical path with a 1 × 2 fiber optical coupler in our optical system. In addition to reducing the complexity of the system, this also provides portability and robustness, these added features potentially promote unique application fields those were previously inaccessible.

Negatively charged Nitrogen-Vacancy (NV) color center in diamond is a promising candidate for both quantum information processing1,2 and spin-based quantum sensing of magnetic field1,2,3,4,5,6,7,8,9,10 electric field10,11 and temperatures10,11. To advance the use of NV center in both applications, efficiently exciting the color center and collecting its fluorescence are demanded, especially a high fluorescence collection efficiency would benefit precision sensing. Research efforts have been taken to meet these demands and some achievements have been reported12. For example, a circular grating or a solid-immersion-lenses have been fabricated on the single crystal diamond substrate itself to direct and extract the fluorescence out of the diamond, thus the fluorescence can be collected more efficiently13,14,15,16; micro/nanosonator cavity and tapered optical fiber have been used to enhance both the NV center’s fluorescence emission and coupling out efficiency17,18,19,20. Optical antennas and plasmonic structures have been also used to enhance the local light field density thus improving the fluorescence emission efficiency, (the Purcell effect) to enhance fluorescence collection from the NV center in diamond17,18,19,20.

Here, we demonstrate a simple method to enhance the excitation efficiency and also fluorescence collection from an NV center rich micrometer-sized diamond attached to an optical fiber. This single-port sensor offers flexibility for applications that require field-measurements in non-standard locations (e.g., magnetic endoscopy, subcutaneous measurements). Our technique is based on using a micro-concave mirror to reflect and focus the excitation laser light into the diamond located at the focal point of the micro-concave mirror. At the same time, the micro-mirror also reflect and focus the fluorescence 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 experiment, we collected over 25 times higher fluorescence intensity from the diamond located at the focal point of an Aluminum (Al) foil micro-concave mirror than that from not using any mirror. Although we only demonstrated the micro-concave mirror for enhancing excitation and fluorescence collection of NV ensembles in micrometer-sized diamond, we believe this method can also improve laser excitation and fluorescence collection of single NV centers and other solid-state quantum emitters such as Silicon-Vacancy centers in diamond, quantum dots, 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 filter based optical path splitter. This replacement can reduce the complexity, enhance the portability, and improve the robustness of this optical-fiber-based NV system; which will potentially boost its application fields.

The micro-concave mirror fluorescence exiting and collection working principle schematic diagrams and its ray optics simulations are shown in Fig. 1(a) and (c).
show that after exiting the optical fiber, except for a small portion of the laser light being absorbed by the NV centers, most of the laser light will pass the diamond; a micro-concave mirror (locates with its focal point at the diamond center), will reflect and focuses the pass laser light back into the diamond to excite the NV centers again, thus enhancing the excitation efficiency. Simultaneously, reflect and focuses most fluorescence hit it back into the optical fiber within its acceptance cone, thus enhancing the fluorescence collection efficiency (Fig.1 (b) and (d)). The micro-concave mirrors are fabricated by using a tip-sphered optical fiber lightly pressing the center of a thin aluminum foil (Al foil, ~15 µm in thickness, purchased from a local supermarket) that is glued on a center-empty thick plastic sheet. Tip-sphered optical fiber is fabricated by arc discharging heating a broken optical fiber tip prepared by heating while stretching in an electric fusion splicer. As the tip shape can be partially adjusted by adjusting the stretching force, the arc discharging current for breaking the fiber and the arc discharging current to form the sphere tip, the shape and focal length of the mirror are partly adjustable. Fig.2 (a), (b) and (c) show scanning electron microscope images of two tip-sphered optical fibers and one fabricated aluminum micro-concave mirror. Fig.2 (d) shows one fabricated aluminum micro-concave mirror focused the illumination light and formed a bright spot in an optical microscope image.
Figure 4. (a) The measured fluorescence spectra of a ∼8-µm diamond glued on a sphered optical fiber end in three different configurations: I. not using any mirror, II. facing an aluminum flat mirror and III. facing an aluminum micro-concave mirror. Single shot ODMR spectra of the diamond: (b) in facing a micro-concave mirror configuration, and (c) in a not using any mirror configuration. The laser power (∼0.49 mW) and magnetic field (∼2.74 mT) are the same. No lock-in amplifier is used in the ODMR measuring.

We used the Fig.3 setup to assess the NV center fluorescence excitation and collection efficiency enhancing. After passing through a 532-nm clean filter, the excitation laser (solid arrows in Fig.3) is coupled into the 10 output port of a 90:10 1×2 fiber optical coupler (Beijing XingYuan AoTe technology Co., Ltd, China) by an objective (10×, NA=0.1); ∼10% of its power is guided to the ∼8-um diamond glued with UV curing glue on the center of the sphered end of the graded index multi-mode fiber (GIF625, Thorlab). Collected by the same optical fiber end, the fluorescence (dashed arrows in Fig.3) is guided back into the input port of the fiber optical coupler; ∼90% of its power will pass the long-pass (LP) filter (cut wavelength 615nm) and finally reach the 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 HPHT (high pressure, high-temperature) type-Ib single crystal which was electron-irradiated and subsequently annealed. The aluminum micro-concave mirror is mounted on a position adjustable XYZ three dimension stage facing the diamond on the sphered fiber end (inset of Fig.3). For switching between accessions without using a lock-in amplifier and using a lock-in amplifier, connections shown by the dashed line and double-solid lines in Fig.3 were switched.

We recorded the collected spectra of the diamond on the sphered fiber end in three different configurations: I. not using any mirror (the diamond is far away from the mirror), II. facing an aluminum flat mirror (the diamond is facing the flat section of the mirror), and III. facing an aluminum micro-concave mirror (the diamond is approximately located at the focal point of the micro-concave mirror). The results are shown in I, II and III of Fig.4 (a). The laser power of the sphered fiber end was ∼0.49 mW. Compare with not using any mirror, over 25 times more of fluorescence have been collected in the facing an aluminum micro-concave mirror configuration (I and III in Fig.4 (a)). We also placed a magnet nearby and a microwave coil (RF coil) circling near the diamond, then scanned the optically detected magnetic resonance (ODMR) in both the not using any mirror and the facing micro-concave mirror configurations. Fig.4 (b) and (c) show the results obtained without using a lock-in amplifier. And Fig.5 shows the results obtained when using a lock-in amplifier to modulate the microwave amplitude (at a frequency of 25 KHz and a depth of 100%) and to read the signal directly from the photo-diode. When the lock-in amplifier is not applied, in the facing the micro-concave mirror configuration, we can easily obtain the ODMR spectrum in a single shot (Fig.4 (b)); while in the not using any mirror configuration, we can almost only get noise in a single shot at the same excitation laser power (Fig.4 (c)). When the lock-in technique was applied, we obtained the ODMR and even the Nitrogen hyperfine of the NV center in the ODMR spectra (inset in Fig.5) in both the configurations. Compare with not using any mirror, we can get much lower noise signals in the facing the micro-concave mirror configuration. (Fig.5). In Fig.5 (a), when the micro-concave mirror was
used, the collected fluorescence intensity is high and the
ODMR contrast (DC signal have been filtered) is much
higher than the auto offset level of the lock-in amplifier
(~1.5 × 10^{-5} V), the signal processing of the lock-in ampli-
er will lead the resonance peaks point upward and the
whole signal level is above the auto offset level. While in
Fig.5 (b), as the micro-concave mirror was not used, the
collected fluorescence intensity is low and the contrast of
the ODMR is lower than the auto offset level of the lock-
in amplifier, thus its ODMR peaks point downward and
the whole signal level is below the auto offset level.

The fabrication of micro-concave mirror on aluminum
film is simple and direct forward, but its relatively large
size (bigger than the optical fiber diameter) will reduce
the compactness of the optical fiber-based diamond NV
center system. To solve this issue, we fabricated a micro-
concave mirror on an optical fiber directly facing the dia-
mond on a fiber flat end facing an on-fiber micro-
concave mirror in a silica tube under the fiber guided green
laser illumination. (c).Principle schematic diagram of combin-
ing the solid immersion lens with the micro-concave (parabolic
shape) mirror to further enhance the NV center fluorescence
excitation and collection efficiency.

Figure 6. (a) scanning electron microscope image of an
on-fiber etching-coating fabricated micro-concave mirror. (b)
Optical microscope image of the side view of a ∼10µm di-
amond on a optical fiber flat end facing an on-fiber micro-
concave mirror in a silica tube under the fiber guided green
laser illumination. (c).Principle schematic diagram of combin-
ing a parabolic shape micro-concave mirror and a
fluorescence extracting solid immersion lens to further
enhance the diamond NV center’s fluorescence excitation
and collection.

In conclusion, we have demonstrated a simple method
based on using a micro-concave mirror to enhance the
laser excitation and also fluorescence collection in an
optical fiber-based NV quantum sensor system. Using
this technique, we experimentally collected over 25 times
more fluorescence from a NV enriched micrometer-sized
diamond attached to an optical fiber end than that from not using any mirror. This efficiency-enhancing technique can be used solely
or combined with other exist fluorescence collection
enhancing techniques, such as solid immersion lens (SIL)
and circular Bragg grating, to further improve laser
excitation and fluorescence collection of NV center in
diamond or other solid state quantum emitters such
as silicon-vacancy center in diamond, quantum dots,
and rare-earth ion-based color centers. Additionally, we
demonstrated that using a 1 × 2 fiber optical coupler to
replace the bulky optical elements based optical path
splitter in the fiber-based NV quantum sensor system,
can reduce the sensor systems’ complexity and enhance
its portability and robustness.

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1 L. Childress, M. V. Gurudev Dutt, J. M. Taylor, A. S.
Zibrov, F. Jelezko, J. Wrachtrup, P. R. Hemmer, M. D.
Zibrov, F. Jelezko, I. Popa, M. Domhan, A. Grud-

Lukin, ” Coherent Dynamics of Coupled Electron and Nu-
clear Spin Qubits in Diamond,” Science.314(5797), 281–
285 (2006).

2 F. Jelezko, T. Gaebel, I. Popa, M. Domhan, A. Gru-

"dduan@gwdg.de
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ber, J. Wrachtrup, "Observation of Coherent Oscillation of a Single Nuclear Spin and Realization of a Two-Qubit Conditional Quantum Gate," Phys. Rev. Lett. 93(13), 130501(2004).

3 Gopalakrishnan Balasubramanian, I. Y. Chan, Roman Kolesov, Mohamad Al-Hmoud, Julia Tisler, Chang Shin, Changdong Kim, Aleksander Wojcik, Philip R. Hemmer, Anke Krueger, Tobias Hanke, Alfred Leitenstorfer, Rudolf Bratschitsch, Fedor Jelezko and Jrg Wrachtrup. "Nanoscale imaging magnetometry with diamond spins under ambient conditions," Nature.455, 648(2008).

4 J. R. Maze, P. L. Stanwix, J. S. Hodges, S. Hong, J. M. Taylor, P. Cappellaro, L. Jiang, M. V. G. Dutt, E. Togan, A. S. Zibrov, A. Yacoby, R. L. Walsworth, M. D. Lukin, "Nanoscale magnetic sensing with an individual electronic spin in diamond," Nature.455, 644(2008).

5 Mayeu Ch pay, Alexandre Tailleire, Jocelyn Achard, Sbastien Pezzagna, Jan Mejier, Vincent Jacques, Jean-Francois Roch, Thierry Debuisschert, "Magnetic imaging with an ensemble of nitrogen-vacancy centers in diamond," The European Physical Journal D. 69(7),166(2015).

6 Gopalakrishanan Balasubramian, Philipp Neumann, Daniel Twitchen, Matthew Markham, Roman Kolesov, Norikazu Mizuochi, Junichi Isoya, Jocelyn Achard, Johannes Beck, Julia Tisler, Vincent Jacques, Philip R. Hemmer, Fedor Jelezko and Jrg Wrachtrup, "Ultralong spin coherence time in isotopically engineered diamond," Nature Materials, 8, 383(2009).

7 F. Dolde, H. Fedder, M. W. Doherty, T. Nbauer, F. Rempp, G. Balasubramanian, T. Wolf, F. Reinhard, L. C. L. Hollenberg, F. Jellezko and J. Wrachtrup, "Electric-field sensing using single diamond spins," Nature Physics, 7, 459(2011).

8 Gopalakrishnan Balasubramian, Andrii Lazariev, Sri Ranjini Arumugam and De-wen Duan, "Nitrogen-Vacancy color center in diamondemerging nanoscale applications in bioimaging and biosensing," Current Opinion in Chemical Biology, 20, 67−77(2014).

9 Florian Dolde, Marcus W. Doherty, Julia Mitch, Ingmar Jakobi, Boris Naydenov, Sebastien Pezzagna, Jan Mejier, Philipp Neumann, Fedor Jelezko, Neil B. Manson and Jrg Wrachtrup, "Nanoscale Detection of a Single Fundamental Charge in Ambient Conditions Using the NV− Center in Diamond," Phys. Rev. Lett.112, 097603(2014).

10 V. M. Acosta, E. Bouchard, L.-S. Pezzagna, E. D. Budker, C. F. Wang, Y-S. Choi, J. C. Lee, E. L. Hu, J. Yang, "Observation of Whispering Gallery Modes in Nanodiamonds using Solid Immersion Lenses," Applied Physics Letters, 104(7), 07080(2010).

11 G. Kucsko, P. C. Maurer, N. Y. Yao, M. Kubo, H. J. Noh, P. K. Lo, H. Park, M. D. Lukin, "Nanometre-scale thermometry in a living cell," Nature. 500, 54(2013).

12 P. Neumann, I. Jakobi, F. Dolde, C. Burk, R. Reuter, G. Waldherr, J. Honert, T. Wolf, A. Brunner, J. H. Shim, D. Suter, H. Sumiya, J. Isoya, J. Wrachtrup, "High-Precision Nanoscale Temperature Sensing Using Single Defects in Diamond," Nano Letters,13(6),2738−2742(2013).

13 I. V. Fedotov, S. Blakley, E. E. Serebyannikov, N. A. Safarova, V. L. Velichansky, M. O. Scully, A. M. Zeltikov, "Fiber-based thermometry using optically detected magnetic resonance." Applied Physics Letters 105(26), 261109 (2014).

14 Tim Schröder, Sara L. Mouradian, Jiabao Zheng, Matthew E. Trusheim, Michael Walsh, Edward H. Chen, Luozhou Li, Igal Bayn and Dirk Englund, "Quantum nanophotonics in diamond [Invited]," J. Opt. Soc. Am. B. 33(4), B65–B83(2016).

15 S. Ates, L. Sapienza, M. Davanco, A. Badolato and K. Srinivasan, "Bright Single-Photon Emission From a Quantum Dot in a Circular Bragg Grating Microcavity," IEEE Journal of Selected Topics in Quantum Electronics 18(6), 1711-1721(2012).

16 Luozhou Li, Edward H. Chen, Jiabao Zheng, Sara L. Mouradian, Florian Dolde, Tim Schrder, Sinan Karaveli, Matthew L. Markham, Daniel J. Twitchen, Dirk Englund, "Efficient Photon Collection from a Nitrogen Vacancy Center in a Circular Bullseye Grating," Nano Letters 15(3), 1493-1497(2015).

17 J. P. Hadden, J. P. Harrison, A. C. Stanley-Clarke, L. Marseglia, Y.-L. D. Ho, B. R. Patton, J. L. O’Brien, J. G. Rarity, "Strongly enhanced photon collection from diamond defect centers under microfabricated integrated solid immersion lenses," Applied Physics Letters 97(24), 241901(2010).

18 Mohammad Jamali, Ilja Gerhardt, Mohammad Rezaei, Karsten Frenner, Helmut Fedder, Jrg Wrachtrup, "Microscopic diamond solid-immersion-lenses fabricated around single defect centers by focused ion beam milling," Review of Scientific Instruments85(12),123703(2014).

19 A. Young, C. Y. Hu, L. Marseglia, J. P. Harrison, J. L. O’Brien, J. G. Rarity, "Cavity enhanced spin measurement of the ground state spin of an NV center in diamond," New Journal of Physics, 11, 013007(2009).

20 S. Furuyama, K. Tahara, T. Iwasaki, M. Shimizu, J. Yaita, M. Kondo, T. Kodera, M. Hatano, "Improvement of fluorescence intensity of nitrogen vacancy centers in self-formed diamond microstructures," Applied Physics Letters, 107(16), 163102(2015).

21 C. F. Wang, Y-S. Choi, J. C. Lee, E. L. Hu, J. Yang, J. E. Butler, "Observation of whispering gallery modes in nanocrystalline diamond microdisks," Applied Physics Letters, 90(8), 081110(2007).

22 Thomas M. Babiniec, Jennifer T. Choy, Kirsten J. M. Smith, Mughees Khan, Marko Lonar, "Design and focused ion beam fabrication of single crystal diamond nanobeam cavities," Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena, 29(1), 010601(2011).

23 B. J. M. Hausmann, B. J. Shields, Q. Quan, Y. Chu, N. P. de Leon, R. Evans, M. J. Burek, A. S. Zibrov, M. Markham, D. J. Twitchen, H. Park, M. D. Lukin, M. Loncer, "Coulpling of NV Centers to Photonic Crystal Nanobeams in Diamond," Nano Letters, 13(12), 5791−5796(2013).

24 Lars Liebermeister, Fabian Petersen, Asmus v. Mochow, Daniel Burchardt, Juliane Hermelbrach, Toshiyuki Tashima, Andreas W. Schell, Oliver Benson, Thomas Meinhardt, Anke Krueger, Ariane Stiebeiner, Arno Rauschenbeutel, Harald Weinfurter, Markus Weber, "Tapered fiber coupling of single photons emitted by a deterministically positioned single nitrogen vacancy center," Applied Physics Letters, 104(3), 031101(2014).

25 Masazumi Fujiwara, Hong-Quan Zhao, Tetsuya Noda, Kazuhiro Ikeda, Hitoshi Sumiya, Shigeki Takeuchi, "Efficient Photon Collection from a Nitrogen Vacancy Center in a Circular Bragg Grating," Nano Letters 15(3), 1493-1497(2015).

26 Erik Janitz, Maximilian Ruf, Mark Dimock, Alexandre Bourassa, Jack Sankey, Lilian Childress, "Fabry-Perot mi-
crocavity for diamond-based photonics,” Phys. Rev. A, 92(4), 043844(2015).

27 Roland Albrecht, Alexander Bommer, Christoph Pauly, Frank Mcklich, Andreas W. Schell, Philip Engel, Tim Schrder, Oliver Benson, Jakob Reichel and Christoph Becher, “Narrow-band single photon emission at room temperature based on a single nitrogen-vacancy center coupled to an all-fiber-cavity,” Applied Physics Letters, 105(7), 073113(2014).

28 Janik Wolters, Gnter Kewes, Andreas W. Schell, Nils Nsse, Max Schoengen, Bernd Lchel, Tobias Hanke, Rudolf Bratschitsch, Alfred Leitenstorfer, Thomas Aichele, Oliver Benson, “Coupling of single nitrogen-vacancy defect centers in diamond nanocrystals to optical antennas and photonic crystal cavities,” physica status solidi (b), 249(5), 918–924(2012).

29 Stefan Schietinger, Michael Barth, Thomas Aichele, Oliver Benson, “Plasmon-Enhanced Single Photon Emission from a Nanoassembled Metal-Diamond Hybrid Structure at Room Temperature,” Nano Letters, 9(4), 1694–1698(2009).

30 Michael Barth, Stefan Schietinger, Tim Schrder, Thomas Aichele, Oliver Benson, “Controlled coupling of NV defect centers to plasmonic and photonic nanostructures,” Journal of Luminescence, 130(9), 1628–1634(2010).

31 I. V. Fedotov, S. M. Blakley, E. E. Serebryannikov, P. Hemmer, M. O. Scully, A. M. Zheltikov, “High-resolution magnetic field imaging with a nitrogen-vacancy diamond sensor integrated with a photonic-crystal fiber,” Opt. Lett.41(3), 472–475(2016).

32 Feng Chen, Hewei Liu, Qing Yang, Xianhua Wang, Cong Hou, Hao Bian, Weiwei Liang, Jinhai Si and Xun Hou, “Maskless fabrication of concave microlens arrays on silica glasses by a femtosecond-laser-enhanced local wet etching method,” Opt. Express, 18(19), 20334–20343(2010).

33 Andreas Muller, Edward B. Flagg, John R. Lawall and Glenn S. Solomon, “Ultra-high-finesse, low-mode-volume Fabry-Perot microcavity,” Opt. Lett. 35(13), 2293-2295(2010).

34 D. Hunger, C. Deutsch, R. J. Barbour, R. J. Warburton and J. Reichel, “Laser micro-fabrication of concave, low-roughness features in silica,” AIP Advances, 2(1), 012119(2012).

35 Daniel Najer, Martina Renggli, Daniel Riedel, Sebastian Starosielec and Richard J. Warburton, “Fabrication of mirror templates in silica with micron-sized radii of curvature,” Applied Physics Letters, 110 (1), 011101(2017).