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A Bright Single-Photon Source from Nitrogen-Vacancy Centers in Diamond Nanowires *

Shen Li(李燊)[1,2], Cui-Hong Li(李翠红)[1,3,4], Bo-Wen Zhao(赵博文)[1,2], Yang Dong(董杨)[1,2], Cong-Cong Li(李聪丛)[1,2], Xiang-Dong Chen(陈向东)[1,2], Ya-Song Ge(葛亚松)[3,4], Fang-Wen Sun(孙方稳)[1,2]**

[1]*Key Lab of Quantum Information, Chinese Academy of Sciences, University of Science and Technology of China, Hefei 230026*

[2]*Synergetic Innovation Center of Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei 230026*

[3]*Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029*

[4]*University of Chinese Academy of Sciences, Beijing 100049*

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Single-photon flux is one of the crucial properties of nitrogen vacancy (NV) centers in diamond for its application in quantum information techniques. Here we fabricate diamond conical nanowires to enhance the single-photon count rate. Through the interaction between tightly confined optical mode in nanowires and NV centers, the single-photon lifetime is much shortened and the collection efficiency is enhanced. As a result, the detected single-photon rate can be at 564 kcps, and the total detection coefficient can be 0.8%, which is much higher than that in bulk diamond. Such a nanowire single-photon device with high photon flux can be applied to improve the fidelity of quantum computation and the precision of quantum sensors.

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With stable photon emission and long decoherence time at room temperature, nitrogen vacancy (NV) centers in diamond have been applied as an important candidate for practical quantum information techniques. A single NV center can be a single photon source for optical quantum communication.[1−4] The electronic spin state in the NV center can provide one of the promising solid qubits for quantum computation.[5] Moreover, the quantum state of such an electronic spin is very sensitive to the outer environment, which can act as a high precision quantum sensor for electromagnetic field,[6,7] temperature[8,9] and pressure.[10,11] With super-resolution microscopy,[12−16] nanoscale spatial resolution can be achieved for such a quantum sensor,[17] which has been well applied in biology applications.[9,18] However, all of these advantages of spin state in NV centers are based on the optical detection method, called optically detected magnetic resonance, where the spin state is read out from the emitted photons.[19] Therefore, to improve the performance of quantum information processes based on the spin state of NV centers, it is necessary to enhance detected photon numbers. The higher the photon number is, the higher the fidelity in quantum computation and the higher the precision in quantum sensors.

Because of the large refractive index ($n \approx 2.4$) of diamond material, the emitted photons from NV centers deeply in bulk diamond may be totally reflected at the diamond surface. In this case, the photon collection rate is very low.[20] Optical techniques by using a solid immersion lens[21] or side-coupling method[22] were proposed to improve the photon collection rate. In addition to the passive optical coupling, it is promising to enhance the emission rate of NV centers by optical nanostructures.[2,23−25] It has been well known that the interaction between the emitter and confined optical mode in nanostructure would greatly shorten the emission lifetime because of the Purcell effect. Also, the emission direction can be modified to further enhance the collection rate. As a result, the detected photon numbers are enhanced.

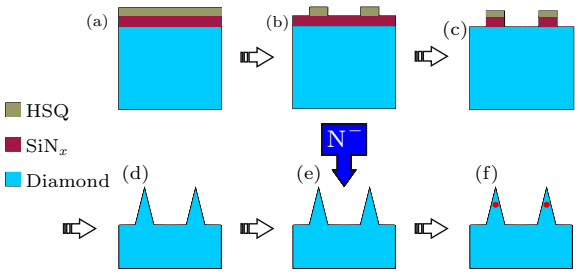

Fig. 1. Pivotal fabrication processes: (a) the bulk diamond with SiN_x and HSQ layers, (b) the HSQ layer patterned and developed, (c) the SiN_x layer etched by RIE, (d) ICP etched diamond for the nanowire, (e) N-ion implantation, and (f) the sample annealed at a vacuum chamber for NV.

In this work, we fabricated a diamond conical nanowire structure to significantly enhance the detected photon flux. Through the coupling between the tightly confined optical mode in the nanowire and NV centers, the lifetime of emitted photons is shortened from 25.6 ns in nano-diamond crystal to 14.3 ns in the nanowire. Also, the collection coefficient was

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**Corresponding author. Email: fwsun@ustc.edu.cn

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improved, compared with the NV centers in bulk diamond. Based on the saturation measurement, the photon rate from a single NV center in the nanowire can be at 564 kcps and the total detection coefficient can be 0.8%. However, in bulk diamond, the counts are 94.7 kcps and the collection coefficient is about 0.1%.

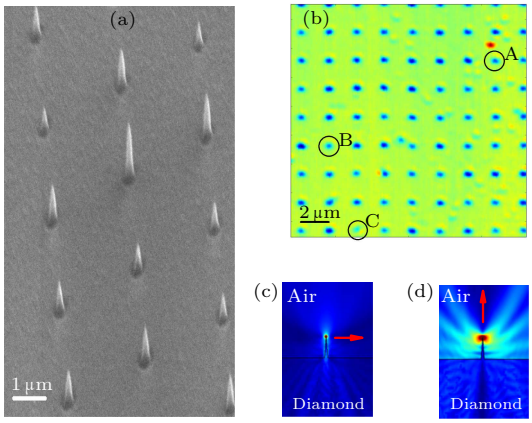

Fig. 2. (a) Scanning electron microscopy (SEM) analysis for the diamond nanowire sample. (b) Confocal microscope scanning image of $\sim 16 \times 16\,\mu m^2$ area of the sample. (c) Representative field profile of a radial component of the E-field in the case of a 1.2-μm-long 200-nm-bottom-diameter conical diamond nanowire with an on-axis s-polarized dipole emitting at $\lambda = 680$ nm. (d) The same case as (c) but having a p-polarized dipole (vertical red arrow) positioned on the nanowire axis. Both (c) and (d) assumed an objective lens collecting upward emitted photons in air, and were Comsol Multiphysics simulations of the diamond nanowire with built-in NV centers (50 nm below the top tip because of 30 KeV N-ion implantation).

The conical nanowire structure was fabricated in a high-quality electronic grade chemical-vapor-deposition (CVD) diamond (Element 6 Corporation, (100) crystal plane, 2 mm×2 mm×0.5 mm size), with nitrogen concentration < 5 ppb. To realize CMOS-process-compatible diamond nanowires, a 300-nm-thick SiN_x layer was deposited on diamond by plasma enhanced chemical vapor deposition (PECVD),[26,27] and a fox-15 negative electron-beam resist (HSQ) layer was then spun on SiN_x to form the resist layer, as shown in Fig. 1(a). Circle areas with diameter 200 nm were patterned using an electron-beam-lithography (EBL) system at a base dosage of 3500 μC/cm^2. Then HSQ was developed in 25% tetra-methyl ammonium hydroxide (TMAH). After that, the sample was placed in a reactive-ion etching (RIE) system and the SiN_x layer was etched for pattern transferring. Then the sample was etched by inductively coupled plasma (ICP) for 15 min with 30 sccm of oxygen gas, 100 W bias power, 700 W ICP power at a chamber pressure of 10 mTorr to fabricate conical nanowire structures with lengths of ∼1.2 μm.[28] Finally, HF wet etch was used to remove the residue of HSQ or SiN_x.

To fabricate NV centers,[29,30] diamond nanowires were subjected to an N-ion implantation vertically with a dose of 5×10^{10} cm^{-2} and an energy of 30 KeV. Post implantation samples were annealed for 6 h at 800℃ and 6 h at 1200℃, both at a chamber pressure of 1×10^{-6} Pa. Lastly, an additional acid bath treatment was performed before device testing.

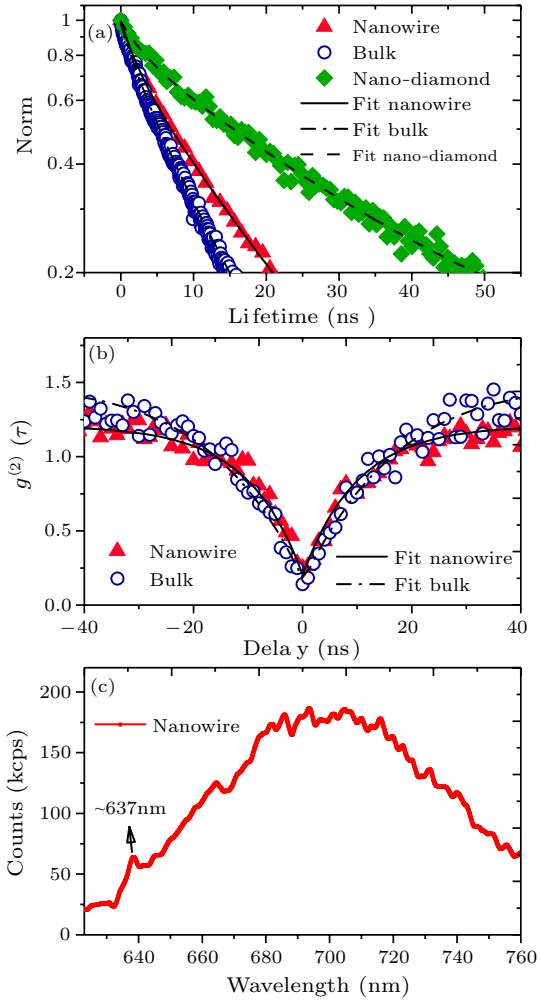

Fig. 3. (a) The experimental result of lifetimes of single NV centers in diamond nanowires, bulk diamond and nano-diamond. Each lifetime was fitted by exponential function, demonstrating the lifetimes of 11.0 ns, 14.3 ns and 25.6 ns, respectively. (b) For single NV centers (Fig. 2(b) marked point C) in the diamond nanowire $g^{(2)}(0) = 0.16$, and typical fitted value in the bulk diamond is $g^{(2)}(0) = 0.21$. (c) Spectrum of the marked point C showing the negatively charged (637 nm) NV centers.

The emission of NV centers at the top part (50 nm below the tip because of 30 KeV N-ion implantation) of the 1.2-μm-long 200-nm-bottom-diameter conical diamond nanowire was simulated with Comsol Multiphysics (as shown in Figs. 2(c) and 2(d)), where the emission modes were quite modified. In the experiment, the NV center was imaged with a home-built confocal microscopy. An N.A. = 0.9 objective was used to collect the photons from NV centers. Figure 2(b) shows the scanning result, where each marked point (B, C) is a single NV center in the nanowire (Table 1). The quantum statistics of such single NV centers was verified by the measurement of intensity autocorrelation function $g^{(2)}(\tau)$ based on the Hanbury–Brown–

Twiss interferometer. Strong photon anti-bunching ($g^{(2)}(0) < 0.5$) demonstrated the single photon emission from the single NV centers. Also, such a result indicated that the coupling between the single NV centers and the nanowire structure dominated over all other backgrounds, including scattering light, dark counts of single-photon detector and substrate fluorescence.

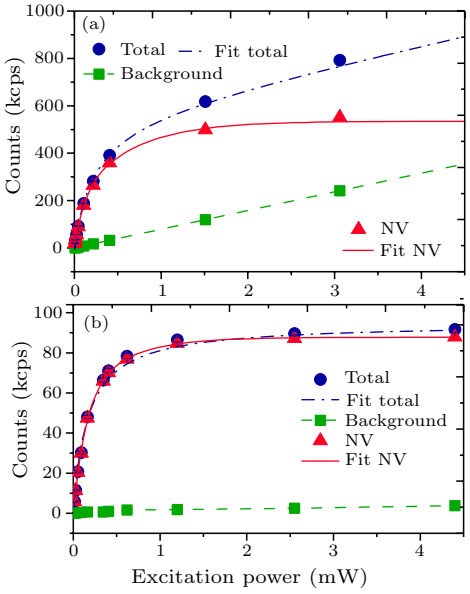

Fig. 4. Fluorescence saturation of NV centers versus excitation power: (a) for NV centers in the diamond nanowire (point C), and (b) for NV centers in bulk diamond.

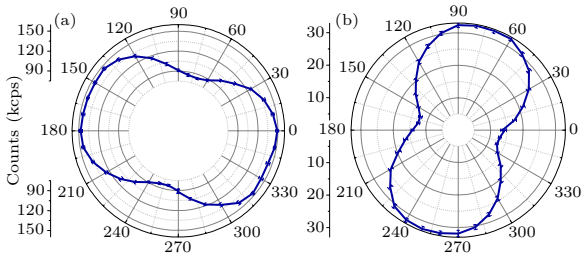

Fig. 5. Luminescence intensity versus excitation polarization for the single NV center (point C) in the diamond nanowire (a) and bulk diamond (b).

The lifetime of photons from NV centers were measured to further demonstrate the coupling between the NV center and the nanowire structure. Also the lifetime shows an upper bound on the number of single photons that can be collected. For such an optical coupling structure, the measured photon lifetime is 14.3 ns, which is much shorter than that in nanodiamond crystal (25.6 ns, Fig. 3(a)[31,32]), corresponding to the dipole in air ($n = 1$). As a comparison, the lifetime of photons from the NV center in bulk diamond is 11.0 ns because $n \approx 2.4$.

To obtain the advantages of this NV-nanowire coupling structure, it is necessary to compare the single photon flux from an NV center in the nanowire and bulk diamond crystal. Two key parameters are $P_{\rm sat}$, corresponding to how much optical power must be used to saturate the NV center response, and $I_{\rm sat}$, corresponding to the number of single photon counts per second (cps) that can be detected. These two parameters can be extracted from the measurement of the photon count rate with different pump powers. As shown in Fig. 4, after a sharp increase in the low pump power ($P < P_{\rm sat}$) region, the number of detected photons reaches saturation ($I_{\rm sat}$) in the high power region because of the finite NV center emission rate. This behavior can be described as[1]

$$I(P) = \frac{I_{\rm sat}}{1 + P_{\rm sat}/P} + \alpha P, \tag{1}$$

where αP describes the linear dependence of the background from the pump power. This single photon counting behavior is shown in Fig. 4. It demonstrates that $I_{\rm sat} = 94.7$ kcps and $P_{\rm sat} = 0.168$ mW for the NV center in bulk diamond. The result for a typical NV-nanowire structure is shown in Fig. 4(a), where $I_{\rm sat} = 564$ kcps and $P_{\rm sat} = 0.229$ mW. It indicates 0.1% detected photons per NV center lifetime in the bulk diamond and 0.8% in the diamond nanowire. Here the detection efficiency includes the imperfect coupling from the NV center, loss in optical path and the non-unity detection efficiency of a single-photon detector. It is indicated that the coupling efficiency is much higher in the nanowire than that in bulk diamond, since the lifetime is comparable.

Moreover, we measured the luminescence intensities versus the excitation polarization as shown in Fig. 5, which can be described by[33]

$$I(\varphi) \propto \left[\sin^2(\varphi - \theta) + \frac{2}{3}\cos^2(\varphi - \theta)\right], \tag{2}$$

where φ is the polarization of excitation laser, and θ is the projection of NV axis onto the sample surface.

Table 1. Photon statistical properties of the NV center in the nanowire and bulk diamond. The marked NV centers in the nanowire are shown in Fig. 2(b). From $g^{(2)}(0)$, point A probably contains two NV centers.

Diamond	Nanowire			Bulk
mark	A	B	C	
$g^{(2)}(0)$	0.55	0.32	0.16	0.21
Lifetime (ns)		15.6	14.3	11.0
$I_{\rm sat}$ (kcps)	353	490	564	94.7
$P_{\rm sat}$ (mW)	0.132	0.290	0.229	0.168

In summary, we have fabricated diamond conical nanowires to greatly enhance the single-photon emission rate and the detection rate for single NV centers. Such a nanowire single-photon device with high photon flux can be applied to improve the performance of quantum information processes. More importantly, through the spin-photon interaction, the remote spin state in diamond can be entangled. High single-photon flux from the NV center will offer an efficient solution to scalable quantum information processing.

Sample fabrication and SEM analysis were carried out at the USTC Center for Micro and Nanoscale Research and Fabrication.

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