Nanoscale localization of the near-surface nitrogen vacancy center assisted by a silicon atomic force microscopy probe

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Abstract

The nitrogen vacancy (NV) center in diamond has wide applications in sensing, imaging and quantum information processing. One of the bases of these applications is to localize the NV center in diamond with high precision. In this work we demonstrate a method for nanoscale imaging and locating near-surface NV centers on diamond waveguides based on an atomic force microscopy (AFM) combined confocal system. The resultant lateral resolution for imaging the NV center is 31.6 nm and the precision of locating the NV center in diamond is 0.7 nm. Finally, the position of the NV center is indicated in AFM images of the diamond waveguide. These results provide a useful characterization tool for optimizing the diamond nanostructure for quantum information processing and quantum sensing.

1. Introduction

The nitrogen vacancy (NV) center in diamond is an atomic-sized defect with remarkable properties at ambient conditions, including long electronic spin coherence times, single-photon emission, optical initialization and readout of the spin state. These properties of NV centers are desirable for quantum information and sensing applications [1–11], including quantum computing networks [12, 13] and highly sensitive nanoscale quantum magnetometers [14–17].

One of the important requirements in these applications is nanoscale localization of the NV center in diamond. Many efforts have been devoted to developing nanoscale localization techniques. On the one hand, the fluorescence property of the NV center allows its optical imaging and localization through confocal microscopy. By using well-developed super-resolution optical imaging technology, the imaging resolution can be greatly improved from the optical diffraction limitation around 250–2.4 nm [18, 19]. On the other hand, the spin property of the NV center enables its imaging through magnetic resonance detection under a gradient magnetic field. The stronger the field gradient is, the more sensitive the NV center is to its spatial localization. Based on this principle, scanning-field-gradient MRI can achieve an imaging resolution of 8.9 nm and a positioning precision of 0.2 nm in a field gradient of 1 G nm\(^{-1}\) [20], while Fourier magnetic imaging technology can achieve an imaging resolution of about 30 nm [21]. Despite these great achievements, precisely determining the relative position of the NV center in diamond is still challenging. While optical imaging requires another nanoscale coordinate reference with optical fluorescence, magnetic imaging requires pre-knowledge of the nanoscale spatial distribution of the gradient magnetic field. Both requirements are non-trivial tasks.

The other possibility is to use a hybrid approach. Near-field optical imaging combined with atomic force microscopy is a promising method to address this issue. While a scanning atomic force microscope (AFM)
can image the diamond surface or the sample on the diamond surface at the nanoscale, optical imaging of the NV center is also at nanoscale resolution in close proximity to an AFM tip. Recently, this hybrid approach was experimentally reported using a metal AFM tip, which is based on plasmonic effects \[22, 23\]. In this work, we demonstrate nanoscale localization of near-surface NV centers in diamond waveguides using silicon AFM tips. Unlike when using a metal tip, the plasmonic effect in our experiment is not required. The resulting lateral imaging resolution and locating precision were 31.6 nm and 0.7, respectively. In this work, the fluorescence intensity is reduced by the silicon tip with a high refractive index and the topographic information for the diamond surface at the corresponding position is given at the same time.

2. Experimental details

The diamond used in this work is a (100)-oriented ultrapure electronic grade chemical vapor deposition diamond (Element Six). The NV center was created by implantation of 5 keV N\(^+\) ions and annealing for 2 h at 800 °C in a vacuum. Based on the stopping and range of ions in matter simulations \[24\], the depth of the NV center was estimated to be 5–11 nm. Nano-pillar waveguides were then fabricated on the diamond by the top-down fabrication method combining electron beam lithography and inductively coupled plasma etching. This structure is commonly used in nanoscale magnetic resonance spectroscopy \[25\], nanoscale magnetic imaging \[26\] and searching for axions or axion-like particles at the nanoscale \[27\] to increase the fluorescence collection efficiency.

Our experiment used an optically detected magnetic resonance setup combined with an atomic force microscope (MFP-3D Asylum Research). As illustrated schematically in figure 1, the diamond was set on a two-dimensional piezo-electric stage that was used for fluorescence and AFM imaging in the experiment. Green laser light (\(\lambda = 532\) nm) was delivered via a single-mode fiber, reflected by a dichroic mirror and focused by a long-work-distance objective (Olympus, LUCPLFN 60X, NA 0.7). The scanning mirror and a z-direction nano-positioner on the objective were used to align the focus of the laser beam to the silicon tip. The lens pair for 4f imaging was implemented in the setup to provide good mapping of sample and image planes. Fluorescence with a wavelength of 650–800 nm was collected through the same path and directed onto an avalanche photodiode (Perkin Elmer SPCM-AQRH-14) after passing through the spectral filters and confocal pinhole. The number of photons was recorded by a counter (PI). The tip apex was cone shaped and the tip size (the diameter of the tip at its top) of the silicon AFM probe (AC240TS Asylum Research) was about 20 nm; the half angle of the tip apex was about 10°.

In this experiment, the AFM was operated in tapping mode with progressive scanning. The counter only recorded the number of photons when the probe vibrated to the bottom, which was synchronized by a PulseBlaster (Spincore). Information on the surface topography and fluorescence intensity of the NV center was recorded at the same time as the two-dimensional piezo-electric stage was scanned.

3. Results

We first performed confocal imaging of the diamond waveguide through the confocal system by moving the diamond while keeping the AFM tips fixed. A typical image is shown in figure 2(a), in which the bright spots indicate diamond waveguides with NV centers inside. By performing a fine scan of these waveguides we see a detailed fluorescence spot, as shown in figure 2(b). In contrast to the standard fluorescence spot satisfying a Gaussian distribution, a tiny dark area is displayed in the bright fluorescence spot due to the presence of AFM tips. Figures 2(c) and (d) show the typical pattern along the x direction. The fluorescence intensity at each coordinate position in the image can be expressed as

\[
I_{\text{pl}} (r) = \text{PSF}_{\text{confocal}} (r - r_c) + \text{PSF}_{\text{tip}} (r - r_t)
\]

where \(r\) is the displacement of the two-dimensional piezo-electric stage and \(r_c\) and \(r_t\) are the position vectors of the NV center in the confocal image and the AFM tip-enhanced resolution image, respectively. \(\text{PSF}_{\text{confocal}} (r)\) and \(\text{PSF}_{\text{tip}} (r)\) are the point spread functions of confocal imaging and AFM tip-enhanced imaging, which can be described by the Gaussian distributions Gauss\(_c\) (\(P_x\)) and Gauss\(_t\) (\(P_x\)), respectively:

\[
I_{\text{pl}} (P_x) = \text{Gauss}_{c} (P_x) + \text{Gauss}_{t} (P_x)
\]

\[= \left[ I_{0c} + \frac{A_c}{\omega_c \sqrt{\pi/2}} e^{-\frac{\left(P_x - P_{x0}\right)^2}{2\omega_c^2}} \right] + \left[ I_{0t} + \frac{A_t}{\omega_t \sqrt{\pi/2}} e^{-\frac{\left(P_x - P_{x0}\right)^2}{2\omega_t^2}} \right].\]

From the fitting, the resolution of the AFM-enhanced confocal image of the NV center reaches about 31.6 nm, which is about 20 times greater than that of the confocal image. This value is comparable to the...
previous results with metal AFM tips [22, 23], thus showing that the plasmonic effect is not necessary for resolution enhancement.

To further obtain the precise position of the NV center in diamond we then obtained an AFM image of the diamond profile while recording its confocal image at the same time. In such a way, the confocal image and the AFM image are in the same coordinate system. Figure 3 shows the results of simultaneous imaging of the diamond waveguide and the NV center.

By fitting the center position of the PSF_{tip}, the localization precision is about 0.7 nm and 1.8 nm for the x and y directions, respectively. The difference in localization precision between the x and y directions can be explained by the possibility of a small drift between successive x-oriented scan lines.

To understand the weakening of AFM tip-induced fluorescence in our experiment, the finite-difference time-domain (FDTD) method (Lumerical FDTD Solutions) was used to numerically calculate the influence of the silicon tip on the excitation and radiation collection of the NV center.

As shown in figure 4, the changes in laser excitation and dipole radiation during the whole process of silicon tip sweeping through NV position were simulated. The waveguide model (in figures 4(a) and (d)) exactly follows the actual waveguide structure, which was acquired through a scanning electron microscope. According to the energy of ion implantation, we set the depth of the NV center at 8 nm. The distance between the center of the silicon tip and the position of the diamond surface directly above the NV center is x.

In figure 4(b), the enhancement coefficient of the electric field $|E|^2/|E_0|^2$ is used to show the change in field intensity at the NV position when the silicon tip sweeps over the diamond surface. Here $|E|^2$ is the electric field intensity with a silicon tip and $|E_0|^2$ is the electric field intensity without a silicon tip. In the dipole radiation simulation we set the monitor below the diamond waveguide as shown in figure 4(d), and obtain the far-field projection from the monitor. Considering the reflection and refraction of fluorescence on the interface, we recalculated the far-field projection based on Snell’s law and the Fresnel equations. The radiation collection intensity of fluorescence in figure 4(e) is a total effect of both the reduction of the spontaneous rate ($R$) and the collection efficiency ($\eta$).

The simulation results show that the presence of the silicon tip leads to a decrease in both the excitation and radiation collection, resulting in the dark fluorescence observed in figure 2(b). The decrease in the spontaneous emission rate ($R$) indicates that the silicon tip reduces the local density of states at the NV. The decrease in collection efficiency ($\eta$) indicates that the silicon tip also changes the direction of
Figure 2. Experimental results for AFM tip-enhanced resolution imaging. (a) Scanned confocal image of the diamond waveguide array. (b) The fine scan image, indicated by the red circle in (a). (c), (d) The different ranges of the profile data at the red dashed line in (b). The profile data are represented by a solid black line and the solid red line represents the fitting of the experimental data; \( P_x \) is the displacement vector of the piezo-electric stage in the \( x \) direction. The FWHMs of PSF_{confocal} and PSF_{tip} are 573.9 (26.3) nm and 31.6 (1.8) nm, respectively.

Figure 3. Experimental results of locating the NV center. The red asterisk indicates the position of the NV center in the AFM image of the diamond waveguide. The precision is about 0.7 nm and 1.8 nm for the \( x \) and \( y \) directions, respectively.

fluorescence radiation. We believe that the slight difference in resolution between the experiment and the simulation is due to slight wear of the silicon tip. As further shown in figures 4(c) and (f), the resolution of imaging is dependent on the dimension of the silicon tip. For a silicon tip size of about 10 nm, a higher resolution of about 20 nm is expected.
Figure 4. (a) FDTD simulation for the excitation analysis. The Gaussian beam source ($\lambda = 532$ nm) focused on the top of the diamond waveguide is set to simulate the laser exciting the NV center. The polarization of light is perpendicular out-of-plane and the point monitor is set at the position of the NV center to detect the near-field electric field intensity. (b) The enhancement coefficient of the electric field $|E|^2/|E_0|^2$ as a function of $x$. The FWHM of the valley is 28.7 nm (the tip size is 20 nm). (c) The FWHM of the valley in (b) as a function of tip size. (d) FDTD simulation for the radiation collection. The dipole ($\lambda = 637$ nm) is located at the position of the NV center, oriented perpendicular out-of-plane and the monitor is set below the diamond waveguide to obtain the radiation collection intensity of fluorescence. (e) The radiation collection intensity of fluorescence as a function of $x$. The FWHM of the valley is 28 nm (the tip size is 20 nm). The inset shows the spontaneous emission rate ($R$) and collection efficiency ($\eta$) as a function of $x$. (f) The FWHM of the valley in (e) as a function of tip size.

We also extended the method to the case of the NV deviating from the center of the waveguide and the case of using a bulk diamond in the FDTD simulation. In both cases our method is still effective and can achieve almost the same spatial resolution. This indicates that the waveguide mode has little effect on the method, and our method can be extended to general localization of NV in bulk diamond.

4. Conclusion

In conclusion, we have demonstrated a new method for imaging and locating near-surface NV centers in diamond waveguides using silicon AFM probes. The image and location of the NV center are realized by the reduction in silicon tip-induced fluorescence intensity. In the experiment, we observed the dark spot caused by the silicon tip in confocal imaging. The final imaging resolution and location precision were 31.6 nm and 0.7 nm, respectively. The results agree well with the results of the FDTD simulation. The simulation results show that the silicon tip can cause a decrease in excitation and radiation collection. The imaging resolution can be further improved by using a smaller silicon tip probe, which can reach about 20 nm. These results could help us understand the role of the NV center position in the diamond photonics system and provide a characterization tool to improve its performance.

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