

# Photoluminescence Enhancement Dependent on the Orientations of Single NV Centers in Nanodiamonds on a Gold Film

Gengxu Chen, Yan Liu, Min Song, Botao Wu, E Wu, and Heping Zeng

**Abstract**—We studied the orientations of single Nitrogen-Vacancy (NV) color centers in nanodiamonds on a gold film by defocused imaging with a scanning confocal microscopy system and an electron-multiplying charge-coupled device. It was demonstrated that the perpendicular oriented dipole of single NV color center showed shorter fluorescence lifetime and higher fluorescence intensity than any other oriented ones on account of higher coupling efficiency of local electronic field induced by the surface plasmon resonance on the gold film for a vertical transition dipole.

**Index Terms**—Diamond, gold, image orientation analysis, photoluminescence, plasmons.

## I. INTRODUCTION

NOWADAYS, the research on stable, efficient, and practical single-photon sources is becoming one of the most important topics in quantum physics. Several single-photon sources have been developed, such as trapped single atoms [1], single molecules [2], quantum dots [3]–[6], etc., but these single-photon emitters either need extremely complicated setup or cannot stably work at room temperature. Single-color centers in diamond are regarded as a promising practical single-photon source in application because of its photostability at room temperature [7]–[12]. However, with the rapid progress of quantum information technique, more efficient and long-term stable single-photon sources are eagerly required.

In order to obtain more practical single-photon source, it has been demonstrated that the photophysical prosperities of NV color centers are obviously improved, such as enhanced fluorescence intensity and shortened fluorescence lifetime, by coupling to metallic nanoparticles or metallic film on nanoscale [13]–[17]. The interaction between metal nanostructures and incident beam generates surface plasmons, which can enhance the electronic field close to the surface and vary the electromagnetic environment. The coupling efficiency between surface plasmons

and photon emitters depends on several factors such as the size of the metallic nanostructures [18], the incident angle of the light [19], etc. Additionally, the spatial orientation of the emitter's absorption–emission dipole would also affect the coupling efficiency, as the photophysical parameters of a single-photon emitter, such as fluorescence lifetime and observable emission intensity, often depend on the emitter's absorption–emission dipole [20], [21]. Therefore, in order to have full control on the coupling between single emitters and surface plasmons, it is necessary to study the enhancement dependence on the orientation of the single-photon emitter's transition dipole.

In this paper, we investigated the photoluminescence properties of the NV color centers in nanodiamonds on a gold film dependent on the dipole orientations by defocused imaging on an EMCCD with a scanning confocal microscopy system. We observed that the photoluminescence properties of the metal-coupled single NV center got larger enhancement due to stronger surface plasmon resonance when the excited dipole was in perpendicular orientation.

## II. EXPERIMENTAL SETUP

NV color centers were generated by high-power electron irradiation and subsequently annealing on nanodiamond crystals [22]. The nanodiamond containing NV color center on a 50-nm-thick gold film was prepared in the same way as in [17].

As shown in Fig. 1, we studied the photoluminescence of the single NV color centers on a gold film by a scanning confocal microscope. Linear polarized continuous-wave excitation laser at 532 nm was focused on the nanodiamond sample by a microscope objective ( $\times 100$ , N.A. = 1.40, oil immersion, Olympus UPlanSApo) after passing through a half-wave plate that was used for changing the polarization of the excitation laser. The photoluminescence from the NV color center was collected by the same microscope objective. In order to obtain the best signal-to-background ratio, a long-pass filter cutting off at 600 nm was set in the detection channel for removing the excitation laser and the fluorescence from the gold film. After spatial and spectral filtering, the photoluminescence was sent to the detection part, which consisted of a Hanbury-Brown and Twiss setup [23] and a time-correlated single-photon counting device (TCSPC, PicoHarp 300). With this setup, we checked the unity of the fluorescent spot in the image by the second-order autocorrelation function measurement. If a deep dip was observed at zero delay in the second-order autocorrelation function, it would verify that the fluorescent spot in the image was a single NV center.

To determine the orientations of the NV color centers, we used defocused imaging method. Up to now, several methods

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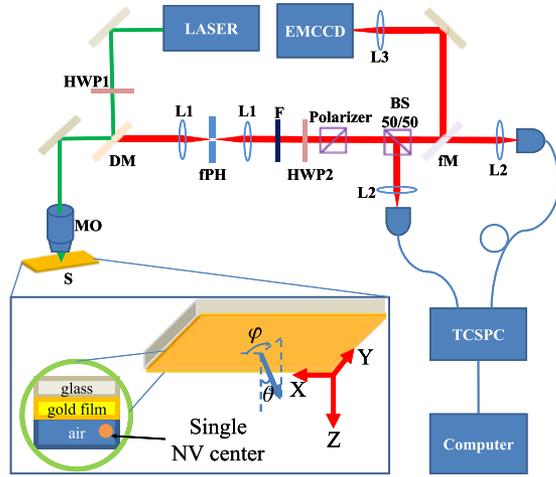


Fig. 1. Experimental geometry and coordinate system. LASER: laser emitting at 532 nm; HWP1: half-wave plate at 532 nm; DM: dichroic mirror; MO: microscope objective; S: sample; L1: achromatic lens,  $f = 50$  mm; fPH: pinhole with  $50 \mu\text{m}$  diameter on a flip-flop mount; F: long-pass filter cutting off at 600 nm; HWP2: achromatic half-wave plate at 450–800 nm; P: polarizer; BS: nonpolarized beam splitter; fM: flip-flop mirror; L2: achromatic lens,  $f = 20$  mm; L3: achromatic lens,  $f = 100$  mm; APD: silicon avalanche photodiode in photon counting regime; EMCCD: electron multiplying charge-coupled device; TCSPC: time-correlated single-photon counting device. The  $x$ -axis is parallel to the polarization direction of the excitation beam and the  $z$ -axis is perpendicular to the interfaces. The orientation of the simplified transition dipole of a single NV center is defined by the pair of angles  $(\theta, \varphi)$ .

have been proposed for determining single-molecule orientations by fluorescence imaging. The essence of these approaches is to obtain the information about the angular distribution of the fluorescence emission by causing the image to deteriorate, either by introduction of aberration [24]–[27] or by defined image defocusing [20], [28], [29]. In both cases, the intensity distribution of the blurred image contains information about the molecule's emission dipole orientation. Here, we employed the same approaches to investigate the dipole orientations of single NV color centers. Furthermore, the patterns on the defocused image plane were also simulated for deducing the orientations of the NV defects.

For NV defects, the photoluminescence is associated with two orthogonal dipoles, which are located in a plane perpendicular to the NV defect symmetry axis [30]. Locating the laser focused on the fluorescent spot, we optimized the signal by using the polarization selectivity to excite one dipole predominantly since the excitation laser was linearly polarized. The transmission of the polarizer in this case was defined as the *longitudinal polarization* ( $L$ ) direction. The *transverse polarization* ( $T$ ) direction was defined as the emission of orthogonal orientation. We removed the pinhole that was placed on a flip-flop mount and switched on the flip-flop mirror to guide the photons to the EMCCD (iXon3 897, Andor Tech.) so that we were able to obtain the defocused image of the single NV center with the EMCCD. Since the pixel size of the EMCCD was  $16 \times 16 \mu\text{m}^2$ , we placed the EMCCD 17 mm behind the focus of the lens L3. By tuning the angle of HWP2, we could get the defocused pattern of the single NV center at two orthogonal polarization directions.

The simulation of the patterns on the defocused image plane was based on surface plasmon-coupled emission method [31], [32]. Since only one of the two orthogonal dipoles was excited predominantly, we considered the excited single NV center as

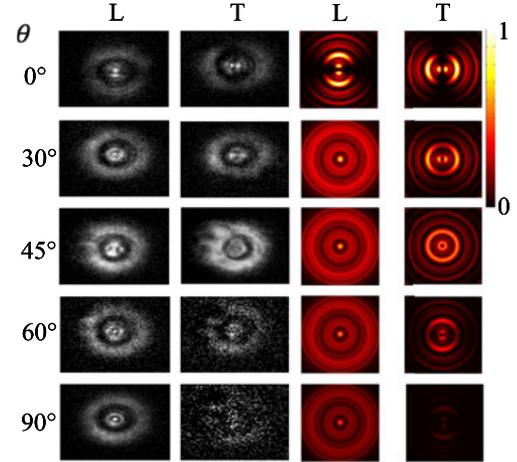


Fig. 2. (Left) Experimental defocused fluorescence patterns and (right) simulated images of single NV centers for different dipole orientations.  $L$  stands for longitudinal polarization, while  $T$  stands for transverse polarization.

a single dipole for simplifying our model. The dipole was set close to the gold film, and the detection of the emission was performed on the opposite side of the metal film. Similar to the simulation in [31], different defocused patterns of different dipole orientations could be acquired with different angles  $(\theta, \varphi)$  (as shown in Fig. 1) by calculating their point spread functions. The entire computational domain was gridded. On each grid, the electric field vector and the fluorescence intensity at orthogonal polarization on the image plane were solved with a given angle pair  $(\theta, \varphi)$ . We set  $\varphi = 0$  as  $L$  polarization and presented the dipole images dependent on  $\theta$  values at  $L$  and  $T$  polarizations. The simulation was conducted on a square grid with  $10 \mu\text{m}$  discretization step in  $1 \text{mm}^2$  range. In the simulation, the thickness of a gold film was 50 nm, and the distance of single dipole away the gold film was 20 nm. After considering that the reflective index of air, gold, and substrate/oil were 1,  $0.32+2.83i$ , and 1.515, respectively, the defocused intensity patterns of a single NV color center for different orientations were simulated in the form of  $L$  and  $T$  polarization directions separately.

### III. RESULTS AND DISCUSSIONS

The defocused fluorescence imaging was measured for 30 individual single NV centers. Here, we present five NV centers with high image contrast patterns in Fig. 2(left) together with the simulated images with different angles of  $\theta$  from  $0^\circ$  to  $90^\circ$  (right). The experimental results exhibited an excellent agreement with the simulations of the defocused fluorescence patterns for the single NV color centers on the gold film. By comparing the  $L$  and  $T$  situation, it could be observed that the intensity of the  $L$ -polarized dipole was stronger than that of the  $T$ -polarized dipole (except  $\theta = 0$ ), which corresponded to the orientation of maximum fluorescence count for each single NV center as its  $L$  polarization situation. Particularly for a perpendicularly oriented excited dipole ( $\theta = 0$ ), there existed symmetric distribution of the  $L$  and  $T$  polarization directions and the same peak amplitudes were obtained for both situations. Such a symmetry could be expected for a perpendicularly oriented dipole because the axial symmetry would lead to radial

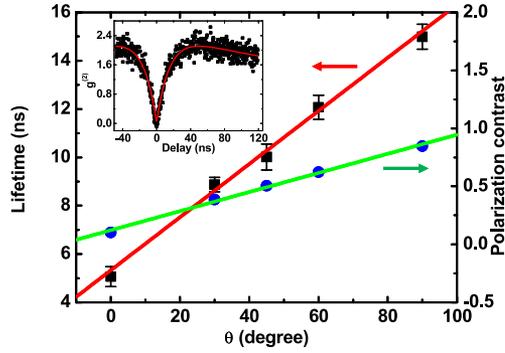


Fig. 3. Fluorescence lifetime (black square points with red fitted line, the error bar presents the lifetime fitting error for each NV center) and polarization contrast (blue circle points with green fitted line) of single NV centers dependent on transition dipole orientation. (Inset) Corrected antibunching trace of a single NV center with a fit.

patterns when projected on to the image plane. The intensity of the patterns increased with the decrease in angle  $\theta$ , i.e., the perpendicularly oriented dipole showed higher photoluminescence intensity than any other oriented ones, indicating that the photoluminescence enhancement efficiency of single NV color center induced by surface plasmon on the gold film was much higher when the dipole was perpendicular to the gold film than when it was parallel to the surface. Furthermore, the intensity difference between the  $L$  and  $T$  polarization directions for each dipole got larger as the increase of the angle  $\theta$ . We figured out the polarization contrast of the fluorescence by:

$$P = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \quad (1)$$

where  $I_{\max}$  was recorded by the APDs with the fluorescence in  $L$  polarization and  $I_{\min}$  in  $T$  polarization. The polarization contrast increased linearly with the angle as shown in Fig. 3. It could be explained that the dipole component in  $L$  polarization projected on the image plane increased with the increase in angle  $\theta$ , while the dipole component in  $T$  polarization was maintained, resulting in the change of the polarization contrast.

In the previous work, it was shown that there would be a variation in the fluorescence lifetime when single NV centers coupled to plasmonic structures due to different coupling efficiency [14], [15]. Although a dielectric mirror could be used to optimize the collection efficiency by reflecting the emission spectrum of an NV color center, it would not affect any NV color center emission properties including the emission lifetime [33], [34]. Therefore, we believe the modification of the emission lifetime was due to the coupling to surface plasmon. In order to confirm the coupling efficiency of single dipole and surface plasmon, we investigated the NV color center fluorescence lifetime with the second-order autocorrelation function  $g^{(2)}(t)$ , which could be fitted by:

$$g^{(2)}(t) = 1 - (1 + a)e^{-t/\tau_1} + ae^{-t/\tau_2} \quad (2)$$

where  $t$  was the delay time,  $a$  a parameter dependent on the transition rates among the energy levels,  $\tau_1$  the fluorescence lifetime, and  $\tau_2$  corresponds to the decay time of the metastable state that caused the bunching effect in long time scale. In this way, we obtained the fluorescence lifetime of the different oriented dipoles shown in Fig. 2. Since the excited surface plasmon

traveled only along the surface, there existed a dependence of the coupling efficiency on the orientation angle. As shown in Fig. 3, the fluorescence lifetime increased with the angle  $\theta$ . The linear fit showed that with the increase of the angle, the emission lifetime prolonged. The fluorescence lifetime  $\tau_1$  was determined by both radiative rate  $R_{\text{rad}}$  and nonradiative rate  $R_{\text{nonrad}}$ , i.e.:

$$\tau_1 = \frac{1}{R_{\text{rad}}} + \frac{1}{R_{\text{nonrad}}}. \quad (3)$$

For a dipole oriented perpendicularly to the gold film surface, the radiative rate increased due to the constructive interference from the local fields of the dipole and the dipole induced by surface plasmon resonance on the gold film. On the contrary, the radiation of the dipole and the surface-plasmon-induced dipole was out of phase when the dipole was parallel to the gold film surface and the radiative rate did not change. Although the surface plasmon would lead to an increase of the nonradiative rate for the parallel-oriented dipole, the nonradiative rate would increase for vertical polarization as well due to energy transfer [29]. Therefore, the fluorescence lifetime trended to be shorter under the circumstance of perpendicular orientation.

However, due to the same reasons mentioned earlier, the increase of radiative rate would result in luminescence enhancement, while the increase of nonradiative rate would lead to luminescence quenching. Therefore, a luminescence quenching happened if the energy transfer was the dominating effect, while the luminescence was enhanced if the increase of  $R_{\text{rad}}$  reigned [35]. Nevertheless, as mentioned in [36], high fluorescence enhancement only occurred in the strong coupling of the NV centers to surface plasmon local field where radiative and nonradiative rates were larger as well. For this reason, there existed much higher probability of high photoluminescence enhancement when the transition dipoles of the metal-coupled NV centers were oriented perpendicularly to the metal film.

#### IV. CONCLUSION

In conclusion, we investigated the orientation of single NV color centers in nanodiamonds on a gold film by the defocused images, which were in perfect agreement with the simulation model. Due to the higher enhancement efficiency of local field induced by surface plasmon resonance effect on the gold film, the single NV centers with perpendicularly oriented transition dipoles trended to have shorter decay lifetime and higher fluorescence intensity than other oriented ones. This study on the orientation dependence of surface plasmon-coupled dipole emission would benefit the preparation of more practical single-photon sources.

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