

# Optical trapping of single quantum dots for cavity quantum electrodynamics

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Received 10 November 2017; revised 28 December 2017; accepted 15 January 2018; posted 17 January 2018 (Doc. ID 313145); published 27 February 2018

We report here a nanostructure that traps single quantum dots for studying strong cavity-emitter coupling. The nanostructure is designed with two elliptical holes in a thin silver patch and a slot that connects the holes. This structure has two functionalities: (1) tweezers for optical trapping; (2) a plasmonic resonant cavity for quantum electrodynamics. The electromagnetic response of the cavity is calculated by finite-difference time-domain (FDTD) simulations, and the optical force is characterized based on the Maxwell's stress tensor method. To be tweezers, this structure tends to trap quantum dots at the edges of its tips where light is significantly confined. To be a plasmonic cavity, its plasmonic resonant mode interacts strongly with the trapped quantum dots due to the enhanced electric field. Rabi splitting and anti-crossing phenomena are observed in the calculated scattering spectra, demonstrating that a strong-coupling regime has been achieved. The method present here provides a robust way to position a single quantum dot in a nanocavity for investigating cavity quantum electrodynamics. ©2018

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**OCIS codes:** (350.4855) Optical tweezers or optical manipulation; (240.6680) Surface plasmons; (270.5580) Quantum electrodynamics.

<https://doi.org/10.1364/PRJ.6.000182>

## 1. INTRODUCTION

Strong coupling between a quantum emitter and a cavity creates a hybridized state that not only attracts interest in the fundamental study of quantum electrodynamics (QED) phenomena but also finds a number of applications, such as quantum information processing [1–3] and ultrafast single-photon switches [4–6]. The strength of the coupling depends on the oscillator strength of the emitter as well as the ratio of the quality factor of the cavity to its mode volume [7]; therefore, a microcavity with high- $Q$  such as a photonic crystal cavity is favorable for approaching this regime [8–11]. In recent years, plasmonic cavities have gained increasing attention, with one reason being that the localized mode associated with the surface plasmon polaritons (SPPs) confines the light to a volume scaled down to cubic nanometers [12–14]. The confined electric field significantly enhances the strength of the emitter-photon interaction. Another reason for the use of a plasmonic cavity is its ease of integration with other photonic devices for optical information processing at nanoscales.

Although the small mode volume associated with a microcavity or a plasmonic cavity favors the realization of strong light-matter coupling, it also presents great challenges for positioning emitters at the field maximum of the cavity mode

for the strongest interaction. It is especially true when exploring the QED at single-emitter level [9,10,15–17]. Recently, strong coupling of single organic dye molecule with plasmonic nanocavities was observed by limiting the number of participant molecules either through host-guest chemistry [15] or by using the monolayer chain property of J-aggregate [17]. Other techniques of nano-positioning of a single molecule or a cavity for optimized coupling include scanning probe [18–20], self-assembly [21], and nanofabrication [8–10]. Besides, the significant confinement of light in a microcavity or a plasmonic cavity produces strong optical gradient force on particles that are present in the cavity. The optical force then traps the particles at the antinode or “hot-spot” of the cavity mode for the strongest light-matter interaction [22–24]. It is especially interesting that the presence of the particle itself imposes great influences on the local electric field and produces a new trapping mechanism, self-induced back-action (SIBA) optical trapping [25]. The active role of the particle enhances the trapping in SIBA by a magnitude of 1 order and has been used for trapping dielectric nanoparticles and quantum dots with reduced laser power [26,27]. Despite this advantage, the previous nanohole designs for SIBA trapping are not suitable for the study of strong emitter-cavity coupling.

Here we report a nanotweezer design for trapping single quantum dots and investigating their strong interaction with the plasmonic cavity. The optical force field of the nanotweezers is calculated by using finite-difference time-domain (FDTD) Maxwell equations solver. The parameters of the nanostructure are tuned to have a plasmonic resonant mode matching the emissions of quantum dots. When two quantum dots are trapped at the hot spots, their scattering spectrum exhibits Rabi splitting and anti-crossing, a signature of strong coupling regime. The proposed nanotweezers provide a robust way to reproducibly position single quantum dots in a plasmonic cavity for the study of the strong interaction between the emitter and the cavity.

## 2. DOUBLE-HOLE NANOTWEEZERS

The structure of our nanotweezers is similar to that of the previous designs, with the difference being that the aperture is fabricated in a thin metal patch rather than a film. This structure is designed to have two functionalities: (1) the tweezers for trapping particles; (2) the plasmonic cavity for quantum electrodynamics. As shown in Fig. 1(a), double elliptical holes are fabricated in a rectangle silver patch deposited on a silicon substrate. The silver patch has a thickness of 40 nm, and its length and width are 80 and 70 nm, respectively. The double holes have a semi-major and a semi-minor axis of 30 and 10 nm, respectively, and are separated by a distance of 30 nm. A 20 nm wide slot is carved in the middle of the patch to connect the two holes. Both of the parameters of the patch and those of the holes are adjusted to have a relatively narrow plasmonic resonance in the visible light range, and the optimization comes from FDTD simulation data. In the simulations, the permittivity of the silver is obtained from Ref. [28]. Because the nanotweezers are supposed to operate in a solution environment, the background index is set to 1.33. The light source for

optical trapping illuminates the holes downward from the top, and its polarization is chosen to be along the  $x$  direction such that the localized plasmonic mode rendering enhanced electric field on the two tips can be excited. Figure 1(b) shows the scattering spectrum of the aperture without quantum dots when the above settings are used. A resonant peak with a full width at half-maximum (FWHM) of 80 nm is observed at 650 nm, a wavelength consistent with the emission of common commercial quantum dots. When the structure is illuminated by a laser at around 650 nm, a significant localized electric field is confined at the edges of the two tips, as shown in Figs. 1(c) and 1(d).

When particles appear in the holes, the field intensity gradient can produce strong optical gradient forces on them. Moreover, the active role of a particle produces a possible self-induced back-action effect and thus enhances the trapping [25]. To demonstrate this point, we calculate the optical force of the confined light acting on a quantum dot. The electromagnetic field distribution is calculated by the FDTD simulation, and the optical force is evaluated by integrating the Maxwell's stress tensor  $\vec{T}$  over a surface  $\Omega$  that encloses the quantum dot [23]:

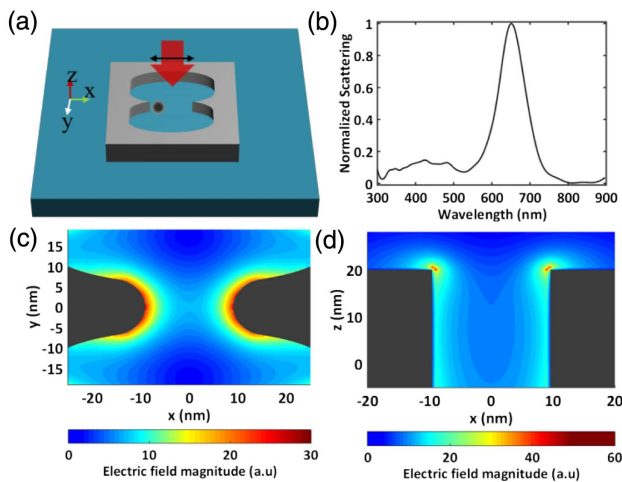
$$\vec{F} = \oint_{\Omega} \vec{T} \cdot dS, \quad (1)$$

where  $\vec{T}$  has the form

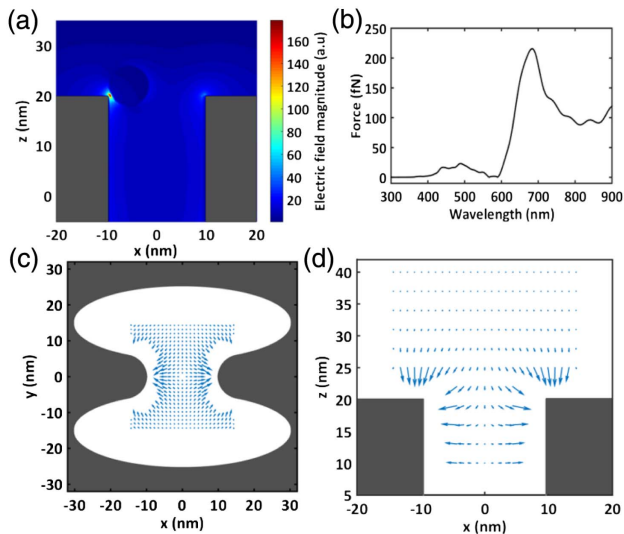
$$\vec{T} = \epsilon \vec{E} \vec{E} + \mu \vec{H} \vec{H} - \frac{1}{2}(\epsilon E^2 + \mu H^2) \vec{I}, \quad (2)$$

with  $\vec{I}$  being the unit dyad, and  $\epsilon$ ,  $\mu$  being the permittivity and permeability of the medium, respectively. The quantum dots used in this simulation has a diameter of 8 nm, and its emission is resonant with the plasmonic cavity. The dielectric function of quantum dots is described approximately by a Lorentz model with a bound electron permittivity of 6.1, Lorentz resonance of 1.91 eV, and a linewidth of 0.02 eV. Due to the small size of the quantum dots, a grid of 0.2 nm is used in the numerical simulations. The presence of a quantum dot in the holes will change the local electromagnetic field distribution. A comparison between Figs. 1(d) and 2(a) indicates that, when a quantum dot is brought closely to one of the tips, the electrical field in the gap between the tip and the quantum dot is further enhanced. Therefore, the optical force of the tweezers on a quantum dot depends not only on the optical response of the bare nanocavity but also on the optical properties of the quantum dot. This active role of a particle in the light-matter interaction exhibits strong dependence of the optical force on the excitation wavelength [25]. To determine the optimal trapping of a quantum dot, we numerically calculate the optical force as a function of the excitation wavelength. The quantum dot is located at  $x = -5.5$  nm,  $y = 0$  nm, and  $z = 22$  nm. As shown in Fig. 2(b), the magnitude of the optical force on the quantum dot reaches its maximum at 675 nm, which is different from the resonant peak of the bare cavity without a quantum dot, evidence of the active role of the quantum dot on the trapping.

Once the optimal trapping wavelength is determined, we then investigate the optical force acting on the quantum dot located at different positions in the nanoholes. As indicated in Figs. 2(c) and 2(d), the optical force tends to push the quantum dot toward the two edges of the tips. As long as this optical force is strong enough, the quantum dot will be trapped at the



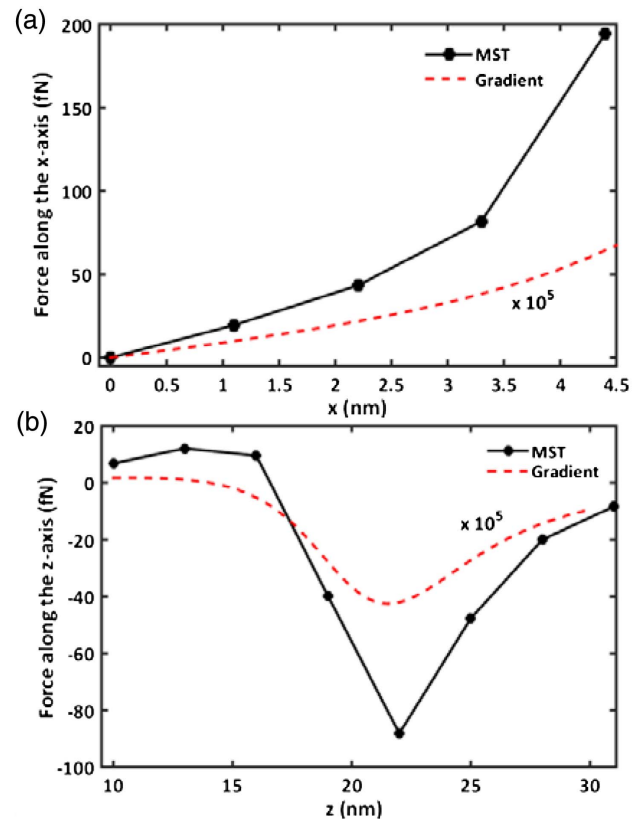
**Fig. 1.** Optical trapping of quantum dots for the study of the strong light-matter interaction. (a) Nanotweezers with double holes in a silver patch. (b) Scattering spectrum of the nano-structure without quantum dots. The spectrum is normalized to its maximum. (c) Electric field distribution in the  $x$ - $y$  plane located at the top surface of the silver patch. (d) Electric field distribution in the  $x$ - $z$  plane bisecting the nanostructure. Gray areas indicate the structures.



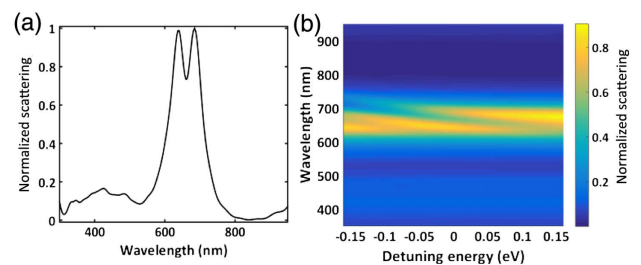
**Fig. 2.** Optical force generated by the localized surface electromagnetic field. (a) Electrical field distribution of the nanocavity in the  $x$ - $z$  plane bisecting the structure when a quantum dot is present at one of the tips. (b) Optical force on the quantum dot as a function of trapping laser wavelength. (c) Optical force vector field in the  $x$ - $y$  plane located at the top surface of the silver patch. (d) Optical force vector field in the  $x$ - $z$  plane bisecting the structure.

edges to interact with the enhanced electrical field. Figure 3 shows the dependence of the optical force on the location of the quantum dot. As indicated, the optical force increases significantly when the quantum dot is brought closely to the edges. When the quantum dot is at  $x = 4.4$  nm,  $y = 0$  nm, and  $z = 19$  nm (a location at which the dot almost contacts with the tip considering the finite size of the quantum dot), the optical force can be as high as 200 fN if the tweezers operate at a laser intensity of  $1 \text{ mW}/\mu\text{m}^2$ . As in the case of other nanotweezers featured with nanoholes, the main mechanism behind this strong trapping is the self-induced back-action instead of the electric field intensity gradient [25]. To clarify this point, we also show in Fig. 3 the optical force calculated from the electric field intensity gradient of the bare cavity [29]. Although the gradient force follows the same tendency when the quantum dot is moved toward the tips, the magnitude of the gradient force is smaller by five orders as compared with that calculated from the Maxwell's stress tensor.

The capability of trapping quantum dots in the localized electromagnetic field provides a convenient way of positioning a single quantum dot in the nanocavity for the study of strong emitter-cavity coupling, which is greatly challenged in the single molecule level. To demonstrate this possibility, we calculate the scattering spectrum of the nanocavity with one and two quantum dots trapped at the edges of its two tips, respectively. As shown in Fig. 4(a), the resonant peak of the bare cavity splits into two peaks when the two dots are trapped at the “hot spots”, and a smaller splitting is also observed with a single dot (the curve is similar and is not shown). The Rabi splitting in the scattering spectrum is usually observed when the strong-coupling regime is reached. To further confirm this point, we investigate the scattering spectrum of the cavity-dots system



**Fig. 3.** Optical force on a quantum dot located at different positions in the cavity. (a)  $X$  component of the optical force as a function of the  $x$  coordinates of the quantum dot. The  $y$  and  $z$  coordinates of the quantum dot are fixed at 0 and 19 nm, respectively. (b)  $Z$  component of the optical force as a function of the  $z$  coordinates of the quantum dot. The  $x$  and  $y$  coordinates of the quantum dot are fixed at 4.4 and 0 nm, respectively. The solid curves are calculated from the Maxwell's stress tensor, and the dashed lines are calculated from the electric field intensity gradient of the bare cavity. The intensity of the trapping laser used in the simulation is  $1 \text{ mW}/\mu\text{m}^2$ . The dashed curves are magnified by a magnitude of five orders for display. MST, Maxwell's stress tensor method; gradient, field intensity gradient method.



**Fig. 4.** Scattering spectra of the nanocavity and the trapped quantum dots. (a) Scattering spectrum of the nanotweezers with two quantum dots trapped at the edges of the cavity's tips. Quantum dots are resonant with the nanocavity. (b) Scattering spectra of the nanotweezers with two trapped quantum dots having various emissions. Spectra are ordered by the detuning energy of the quantum dots from the plasmonic cavity.

when the quantum dots are fixed at the tips but with their emissions detuned from the cavity resonance. The results present in Fig. 4(b) indicate that, when the emission of the quantum dots is detuned across the resonance of the plasmonic cavity, the two peaks in the scattering spectra are never combined, and this anti-crossing phenomenon is a signature of strong-coupling regime.

### 3. CONCLUSION

In conclusion, we reported a nanostructure for trapping single quantum dots and studying their strong interaction with the plasmonic mode of the structure. The nanostructure was featured with two elliptical holes in a thin Ag patch and a slot connecting the holes. This structure is used both as tweezers for optical trapping of nanoparticles and as a plasmonic cavity for studying cavity quantum electrodynamics. The characteristics of the nanostructure were evaluated by FDTD simulations. When this structure was illuminated by light at 650 nm, a plasmonic resonance can be excited with strong electromagnetic field confined at the edges of its tips. The structure reported here also exhibited strong optical force on the quantum dots in the holes, and an optical force of up to 200 fN can be obtained by a trapping laser at an intensity of  $1 \text{ mW}/\mu\text{m}^2$ . When two quantum dots were trapped at the edges of the two tips, the scattering spectrum was calculated and the Rabi splitting was observed. Detuning the emission of the quantum dots from the plasmonic resonance displayed an anti-crossing phenomenon in their scattering spectra, confirming that a strong-coupling regime had been reached. The method presented here provides a robust way to position a single quantum dot in a nanocavity for investigating cavity quantum electrodynamics and has the potential to be used for single-photon generation and quantum information processing.

**Funding.** National Key R&D Program of China (2016YFA0301300).

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