CHIN. PHYS. LETT. Vol. 38, No. 2 (2021) 023301

Lower Exciton Number Strong Light Matter Interaction in Plasmonic Tweezers

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(Received 23 September 2020; accepted 7 December 2020; published online 27 January 2021)

The plasmonic nanocavity is an excellent platform for the study of light matter interaction within a sub-diffraction volume under ambient conditions. We design a structure of plasmonic tweezers, which can trap molecular J-aggregates and also serve as a plasmonic cavity with which to investigate strong light matter interaction. The optical response of the cavity is calculated via finite-difference time-domain methods, and the optical force is evaluated based on the Maxwell stress tensor method. With the help of the coupled oscillator model and virtual exciton theory, we investigate the strong coupling progress at the lower level of excitons, finding that a Rabi splitting of 230 meV can be obtained in a single exciton system. We further analyze the relationship between optical force and model volume in the coupling system. The proposed method offers a way to locate molecular J-aggregates in plasmonic tweezers for investigating optical force performance and strong light matter interaction.

DOI: 10.1088/0256-307X/38/2/023301

Ashkin and colleagues first reported the experimental demonstration of optical tweezers, created by using radiation pressure from a single laser beam.[1] Since then, optical tweezers have been used as indispensable instruments in many applications in various fields, on the basis that they can precisely control micrometer-sized objects.[2] For example, optical tweezers have been applied in the noninvasive manipulation of living cells in biosciences[3] and have enabled the assembly of nanoparticles in physics.[4] In conventional free space optical tweezers, objects mainly experience gradient force exerted by the focused laser beams. Their fundamental drawback is that the focal spot size is limited to the diffraction limit, which makes it difficult to trap objects at the nanoscale. Recently, the concept of plasmonic nanotweezers, based on surface plasmon polaritons (SPPs), has become a powerful scenario in which to circumvent the limitations of conventional optical tweezers.[5–11] However, the perturbation Rayleigh method used to calculate the optical force ignores the influence of the object itself on the local field. In 2009, Juan *et al.* demonstrated the self-induced back-action (SIBA) trapping mechanism for the first time.[12] Compared with the conventional trapping mechanism, the appearance of the object itself has a great impact on the local field, and plays an active role in the SIBA trapping mechanism. The SIBA mechanism provides a unique advantage in terms of the successful trapping of dielectric nanoparticles with lower light intensities than those used in previous approaches.[13]

Strong coupling is a special regime of light matter interaction, with many applications in nanoscience. It provides a potential platform for the study of fundamental quantum science phenomena.[14–18] In recent years, the plasmonic cavity has come to be seen as an appropriate candidate for achieving strong coupling between emitters, excitons in J-aggregates, and SPPs, since the localized mode of SPPs confines the light to a nanoscale volume, and the confined electromagnetic field significantly enhances the interaction between excitons and SPPs.[19–21] Nevertheless, achieving strong coupling in plasmonic cavity also represents a significant challenge. The challenge is locating J-aggregates at the field maximum of the electromagnetic mode for the strongest interaction, particularly for those studies investigating strong coupling at the lower exciton number level.

In this Letter, a structure of plasmonic tweezers based on SPPs is designed in order to trap molecular J-aggregates, and the strong light matter interaction between them is also investigated. The finite-difference time-domain (FDTD) method and the Maxwell stress tensor method are used to obtain the optical response and the trapping performance of the plasmonic tweezers. When the J-aggregates are trapped in the structure, the scattering spectrum exhibits Rabi splitting with the signature strong coupling effect. We use the coupled oscillator model and the virtual exciton theory to analyze the strong coupling behavior in lower exciton numbers. The proposed plasmonic tweezers offer a robust method to reproducibly locate molecular J-aggregates in a plasmonic cavity for the study of light matter interaction.

Methods. Figure 1 depicts the structure of the nanotweezers, which comprise a gold ring and two rounded triangles. The gold ring has a thickness (t) of 30 nm, the inner radius ($r_{\rm in}$) of 50 nm and outer radius

Supported by the National Key Research and Development Program of China (Grant No. 2016YFA0301300), the Fundamental Research Funds for the Central Universities (Grant No. 2019XD-A09), and the National Natural Science Foundation of China (Grant No. 11574035).
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($r_{\rm out}$) of 80 nm. The two gold rounded triangles are attached to the ring, whose thickness and side lengths are 30 nm and 50 nm, respectively. Two triangular tips extend in the middle of the structure, forming a gap (L) 40 nm wide. The whole structure is supported on a glass (SiO_2) substrate, and is intended to be operate while immersed in a solution, so we set the background index to 1.33, with a grid measuring 1 nm in each direction. The permittivity of gold is obtained from Johnson and Christy.[22] The molecular J-aggregates are treated as a dielectric particles, whose radius is 8 nm, and whose dielectric function can be characterized by a Lorentz model,[23] with a bound electron permittivity of 6.1, Lorentz resonance of 1.71 eV, and a linewidth of 0.02 eV. Utilizing a light source illuminates the bare structure from the top, we can observe only one resonant peak at 690 nm in the normalized scattering spectrum, as shown in Fig. 2(a). When the structure is illuminated by a light source at 690 nm, the two triangular tips can confine a giant localized electromagnetic field, as illustrated in Figs. 2(b) and 2(c). The appearance of the particle in the structure affects the surrounding refractive index, thereby altering the local electromagnetic field distribution, as shown in Fig. 2(d). As we can see from Figs. 2(c) and 2(d), when the particle is close to one of the tips, the electromagnetic field in the gap between the particle and the tip is further enhanced. The vast majority of the electromagnetic energy is confined in the gap.

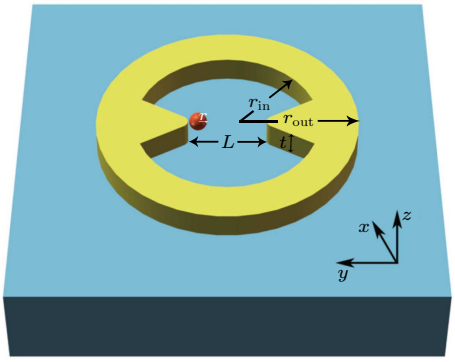

Fig. 1. Schematic diagram of the nanotweezers structure.

Owing to the strong plasmonic resonance, when a particle appears in the structure, the highly localized electromagnetic field generates a strong optical gradient force on it. Conversely, the particle itself produces an SIBA effect that improves trapping.[12,13] The total induced optical force on the particle is evaluated via the Maxwell stress tensor method, without approximation. We obtain the total force $\boldsymbol{F}$ on a particle by integrating the time-averaged Maxwell stress tensor T on the surface S enclosing particle as[24] $\boldsymbol{F} = \oint_s \overleftrightarrow{T}\, ds$ and $\overleftrightarrow{T}$, $\overleftrightarrow{T} = \varepsilon \boldsymbol{E}\boldsymbol{E} + \mu \boldsymbol{H}\boldsymbol{H} - \frac{1}{2}(\varepsilon E^2 + \mu H^2)\ \overleftrightarrow{I}$, where ε and μ denote the permittivity and the permeability of the medium, respectively; $\overleftrightarrow{I}$ is the unit dyad, and the electromagnetic field distribution parameters $\boldsymbol{E}$ and $\boldsymbol{H}$ are taken from the FDTD simulation. However, simultaneously with optical force, the particle is also subjected to thermal energy. As an additional trapping condition, we therefore add the requirement that the trapping potential created by the optical force must overcome the thermal energy. The trapping potential energy is numerically estimated in areas near the equilibrium point along the trapping length direction, which is expressed as $U = -\int \boldsymbol{F} \cdot d\boldsymbol{r}$. In the following sections, we adopt the absolute value of U to show the depth of the trapping potential well.

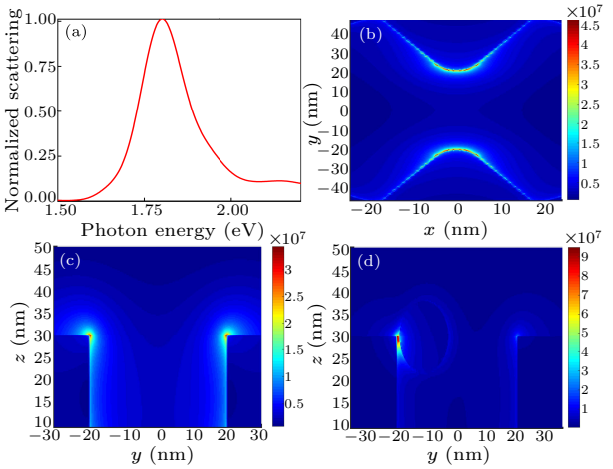

Fig. 2. (a) Normalized scattering spectrum of the nanotweezers' structure. The spectrum is normalized to its maximum. (b)–(d) Electromagnetic field distribution for $\lambda = 690$ nm: (b) $z = 30$ nm cross section, (c) $x = 0$ nm cross section, (d) $x = 0$ nm cross section, when a particle is located at one of the tips.

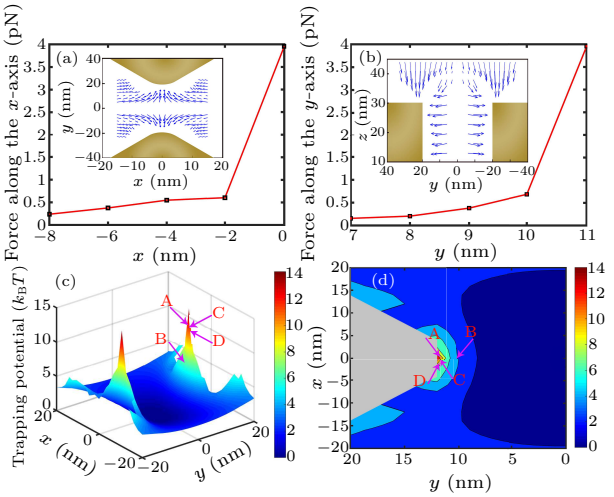

Fig. 3. (a) and (b) Optical force in the x–y plane along $y = 11$ nm located at $z = 30$ nm cross section, and in the y–z plane along $z = 30$ nm at $x = 0$ nm cross section, respectively. The pictures set in (a) and (b) indicate the optical force vector field distribution in the x–y plane at $z = 30$ nm cross section, and in the y–z plane at $x = 0$ nm cross section, respectively. (c) Trapping potential energy $|U|$ at the surface of the nanostructure in the x–y plane. (d) Magnified (c), with the gray part indicating the tip.

Results and Discussions. For the purpose of investigating the trapping behavior of our nanotweezers, we calculate the optical force on the particle at different positions in the nanotweezers' structure, and analyze their trapping potential energy distribution. The pictures set in Figs. 3(a) and 3(b) show the dis-

tribution of the optical force vector in the x–y plane at $z = 30\,\text{nm}$ cross section, and in the y–z plane at $x = 0\,\text{nm}$ cross section, respectively. Based on the optical force vector distribution, we know that the optical force is pushing the particle toward the tip in both the x–y plane and the y–z plane at $z = 30\,\text{nm}$ cross section. Moreover, as shown in Figs. 3(a) and 3(b), the value of optical force rises considerably when the particle approaches the tip. As we move the particle in the structure, the particle should be trapped when the trapping potential well is deep enough. The trapping potential resulting from the optical force determines the stability of the optical trap, and defines an important figure of merit for an optical trap. We put the particle at point A (0, 11, 30), where the distance between the particle and the tip is only 1 nm. The optical force exerted on the particle, measuring 3.8 pN, by the trapping laser intensity is $I = 1\,\text{mW}/\mu\text{m}^2$, and the trapping potential energy in the x–y plane at $z = 30\,\text{nm}$ is shown in Fig. 3(c). In principle, $1k_\text{B}T$ trapping potential energy is sufficiently large to overcome the thermal energy of the particle and confine it in the trap: $|U| \gg 1k_\text{B}T$,[25,26] where U is the trapping potential energy, k_B is the Boltzmann constant, and T is the temperature. However, owing to random Brownian motion, in order to achieve stability in trapping the particle, a larger trapping potential energy is required. It has been proposed that stable optical trapping requires a potential energy of approximately $10k_\text{B}T$.[1] We therefore consider $|U| \gg 10k_\text{B}T$ as the threshold for stable optical trapping. In Figs. 3(c) and 3(d), we present the trapping potential energy, $|U|$, experienced by the particle at $z = 30\,\text{nm}$ cross section. As shown in Figs. 3(c) and 3(d), the trapping potential energy is larger than $10k_\text{B}T$ near the tips. When we move the particle along $y = 11\,\text{nm}$ from $x = -2\,\text{nm}$ to point A (0, 11, 30), the trapping potential energies are $9.68k_\text{B}T$ at point D (−2, 11, 30), $10.53k_\text{B}T$ at point C (−1, 11, 30), and $11.14k_\text{B}T$ at point A (0, 11, 30), respectively. If we then move the particle along $x = 0\,\text{nm}$ from $y = 10\,\text{nm}$ to point A (0, 11, 30), the trapping potential energies are $6.86k_\text{B}T$ at point B (0, 10, 30), and $11.14k_\text{B}T$ at point A (0, 11, 30), respectively. When the particle contacts with one of the tips, the trapping potential energy is approximately $14k_\text{B}T$, which leads to the conclusion that the nanotweezers can achieve stable particle trapping in the area near the tips, facilitating the further study of light matter interaction.

We calculate the scattering spectrum of the nanotweezers' structure when a single particle is trapped at point A (0, 11, 30), where the trapping potential is over 10 $k_\text{B}T$, and the particle is resonant with the plasmonic cavity. By comparing Figs. 2(a) and 4(a), we can observe a clear splitting, which indicates that coupling between the excitons of the particle and the plasmonic cavity produces new hybrid modes. In order to provide a physical insight into the coupling phenomenon, we analyze the scattering spectrum by fitting the calculation results with a phenomenological coupled oscillator model.[21,27] In this model, the plasmonic cavity resonance and the particle's excitons are each described as a damped harmonic oscillator. Essentially, these two oscillators represent the polarization of the plasmonic cavity and the particle, respectively. The coupled oscillator model can be qualitatively described in terms of energy form:[27,28]

$$\begin{vmatrix} E_\text{Pl} - i\gamma_\text{Pl}/2 & g \\ g & E_J - i\gamma_J/2 \end{vmatrix} \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = E \begin{pmatrix} \alpha \\ \beta \end{pmatrix}, \quad (1)$$

where E_Pl and E_J are the energies of the plasmon and excitons, respectively, g is the coupling strength, and γ_J and γ_Pl are the corresponding damping rates; E is the eigenvalue corresponding to the energies of the new hybrid states, and α and β are the eigenvector components (Hopfield coefficients) satisfying $|\alpha|^2 + |\beta|^2 = 1$. The eigenvalues E can be obtained from the equation:

$$E_\pm = \frac{1}{2}\Big[E_\text{Pl} + E_J - i\Big(\frac{\gamma_\text{Pl}}{2} + \frac{\gamma_J}{2}\Big) \pm \sqrt{4g^2 + \Big(E_\text{Pl} - E_J - i\Big(\frac{\gamma_\text{Pl}}{2} - \frac{\gamma_J}{2}\Big)\Big)^2}\Big]. \quad (2)$$

When the particle is resonant with the nanotweezers, i.e., $E_\text{Pl} = E_J$, the Rabi splitting energy is defined as $\Omega = h\Omega_\text{R} = \sqrt{4g^2 + (\frac{\gamma_\text{Pl}}{2} - \frac{\gamma_J}{2})^2}$. Moreover, if $\Omega = 2g > |\gamma_\text{Pl}/2 + \gamma_J/2|$, the coupling system achieves a strong coupling regime.[29] At point A (0, 11, 30), γ_Pl and γ_J are 170 meV and 20 meV for the uncoupled plasmon resonance and the excitons, respectively. In our system, using Eq. (2) and data collected from Fig. 4(a), we obtain the Rabi splitting energy $\Omega = 2g = 230\,\text{meV}$ and $|\gamma_\text{Pl}/2 + \gamma_J/2| = 95\,\text{meV}$, indicating that the system achieves a strong coupling regime.

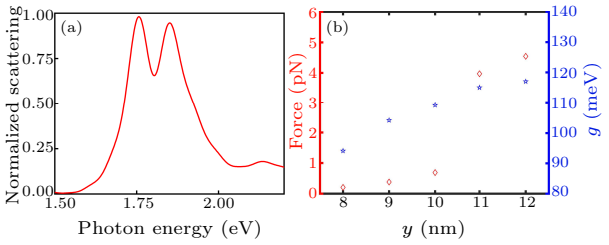

Fig. 4. (a) Normalized scattering spectrum of the nanotweezers' structure when a particle is trapped at point A. (b) Calculated values of optical force and coupling strength with a change in y.

Here, we analyze the coupling behavior when the distance between the particle and the tweezers' tip is changed. The coupling strength can be directly calculated by[30]

$$g = \sqrt{N}\mu_\text{e}|E_\text{vac}|, \quad (3)$$

where N is the number of excitons contributing to the coupling process, μ_e is the molecular dipole moment, $|E_\text{vac}| = \sqrt{\hbar\omega/2\epsilon\epsilon_0 V_\text{m}}$ is the vacuum field, $V_\text{m} = \frac{\int \epsilon(r)|E(r)|^2 dV}{\max[\epsilon(r)|E(r)|^2]}$ is the mode volume, and the dipole moment of the particle is assumed to be $\mu_\text{e} = 15D$.[31] As the particle moves along $x = 0$ from point

E (0, 8, 30) to one of the tips (0, 12, 30), we can calculate the mode volume, coupling strength, optical force, and the average number of excitons, as shown in Fig. 4(b) and Table 1.

Table 1. Calculation results.

Distance (nm)	4	3	2	1	0
$V_{\rm m}$ (10^{-25} m^3)	4.82	3.37	2.28	0.59	0.17
g (meV)	95	104	109	115	117
F (pN)	0.2	0.38	0.68	3.95	4.53
N (average)	4.84	4.15	3	0.89	0.27

When the particle approaches the tip, there is an upward tendency for both coupling strength and optical force, but a decreasing current for the model volume and average numbers. In the coupling progress, we find that the optical force depends greatly on the model volume and the coupling strength, and the relationship between them can be expressed as $f = A \cdot g \cdot V_{\rm m}^{-3/2}$, where A is a scaling parameter. According to Eq. (3), the coupling strength $g \propto \sqrt{\frac{N}{V_{\rm m}}}$. Interestingly, when the distance is less than 2 nm, the average number of excitons is less 1, which is at variance with our previous understanding. At lower coupling strength, the states of two hybrid modes in the system can act as 'dressed states', i.e., superpositions of two states containing both plasmons and excitons of molecular J-aggregates. These two states contain the same exciton numbers. However, with the increase in coupling strength, all excited states are dressed by multiple states containing different exciton numbers, leading to the plasmons and excitons of molecular J-aggregates in ground state becoming virtual.[32] As shown in Table 1, the force is six times stronger at a distance of 1 nm than the force at a distance of 2 nm. The model volume, however, is one-quarter that found at a distance of 2 nm. The sudden change in the force here is primarily contributed by the SIBA effect.[33,34] The key physics of the SIBA effect in this case is based on the fact that the position of the trapped particle alters the electromagnetic field, which can in turn affect the optical force on the particle, together with the emissions of that particle.

In summary, we have demonstrated how a plasmonic nanostructure may be used to construct optical tweezers for trapping molecular J-aggregates, and investigated the strong light matter interaction between SPPs and the excitons. An optical force of 3.95 pN, and a trapping potential energy of $11.14k_{\rm B}T$ at the trapping point can be obtained with an intensity of $I = 1\,{\rm mW}/\mu{\rm m}^2$. When the particle is trapped in the structure, with the help of the coupled oscillator models, a strong coupling Rabi splitting as large as 230 meV can be obtained from the spectrum, the relationship between optical force, model volume, and coupling strength is also investigated. Finally, the virtual exciton theory is used to explain the lower exciton number coupling progress. The proposed nanotweezers offer a robust method to reproducibly locate molecular J-aggregates in a plasmonic cavity for the study of light matter interaction, and have potential applications in quantum information processing and single-photon generation.

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