

Strong coupling in silver-molecular J-aggregates-silver structure sandwiched between two dielectric media

Kunwei Pang(庞昆维)[1,2], Haihong Li(李海红)[2], Gang Song(宋钢)[1,2], Li Yu(于丽)[1,2]

Citation: Chin. Phys. B . 2019, 28(12): 127301 . **doi:** 10.1088/1674-1056/ab4fda
Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

What follows is a list of articles you may be interested in

Resonant surface plasmons of a metal nanosphere treated as propagating surface plasmons

Yu-Rui Fang(方蔚瑞), Xiao-Rui Tian(田小锐)

Chin. Phys. B . 2018, 27(6): 067302 . **doi:** 10.1088/1674-1056/27/6/067302

Highly stable two-dimensional graphene oxide: Electronic properties of its periodic structure and optical properties of its nanostructures

Qin Zhang(张琴), Hong Zhang(张红), Xin-Lu Cheng(程新路)

Chin. Phys. B . 2018, 27(2): 027301 . **doi:** 10.1088/1674-1056/27/2/027301

Diffraction properties of binary graphene sheet arrays

Yang Fan(樊洋), Cong Chen(陈聪), Ding-Guo Li(李定国)

Chin. Phys. B . 2017, 26(1): 017302 . **doi:** 10.1088/1674-1056/26/1/017302

Different optical properties in different periodic slot cavity geometrical morphologies

Jing Zhou(周静), Meng Shen(沈萌), Lan Du(杜澜), Caisong Deng(邓彩松), Haibin Ni(倪海彬), Ming Wang(王鸣)

Chin. Phys. B . 2016, 25(9): 097301 . **doi:** 10.1088/1674-1056/25/9/097301

Compact surface plasmon amplifier in nonlinear hybrid waveguide

Shu-shu Wang(王曙曙), Dan-qing Wang(王丹青), Xiao-peng Hu(胡小鹏), Tao Li(李涛), Shi-ning Zhu(祝世宁)

Chin. Phys. B . 2016, 25(7): 077301 . **doi:** 10.1088/1674-1056/25/7/077301

Strong coupling in silver-molecular J-aggregates-silver structure sandwiched between two dielectric media*

Kunwei Pang(庞昆维)[1,2], Haihong Li(李海红)[2,†], Gang Song(宋钢)[1,2], and Li Yu(于丽)[1,2,‡]

[1] State Key Laboratory of Information Photonics and Optical Communications, Beijing University of Posts and Telecommunications, Beijing 100876, China

[2] School of Science, Beijing University of Posts and Telecommunications, Beijing 100876, China

(Received 17 September 2019; revised manuscript received 19 October 2019; published online 22 November 2019)

We theoretically investigate the strong coupling in silver-molecular J-aggregates-silver structure sandwiched between two dielectric media by using classical methods. Fresnel equations are employed to solve our proposed structure. The results show that both the reflection and transmission spectra show a Rabi splitting-like line shape, revealing the strong coupling phenomenon. Furthermore, the radiative angle *versus* incident wavelength exhibits a Fano line shape. The strong coupling phenomenon can be well tuned by controlling the surface plasmon excitation, such as the incident angle and the thickness of the silver films. Our structure has potential applications in quantum networks, optical switches, and so on.

Keywords: surface plasmons, Krestchmann configurations, strong coupling

PACS: 73.20.Mf **DOI:** 10.1088/1674-1056/ab4fda

1. Introduction

Strong interactions between cavities and quantum emitters open a new branch to manipulate light, which has much applications such as quantum networks,[1] single-atom lasers,[2] ultrafast single-photon switches,[3] and quantum information processing.[4–6] Recently, plasmonics cavities are widely used to realize strong coupling phenomenon with emitters in both theory and experiment at room temperature, which would lead the cavity and emitter mode hybridization and Rabi splitting.[7] There are lots of plasmonic cavities, such as single metallic nanoparticle/metallic nanoparticle array,[8–14] metallic gratings,[15,16] subwavelength hole arrays,[17] and the most popular structures of Krestchmann configurations.[18–22]

Kretschmann configuration is a fundamental structure to generate surface plasmon polaritons (SPPs).[23–26] Lots of Kretschmann configurations are widely used in nanotweezers,[27] four-wave mixing,[28] and optical bistability.[29] The nonlinear effect of the lateral shift or Goos–Hänchen (GH) shift[30] is reported in Kretschmann configuration involving Kerr nonlinear medium.[29,31] If the molecular J-aggregates is sandwiched between two Kretschmann configurations instead of the dielectric medium, the relation between input and output turns to be very interesting and is a great point of exploration.

In this paper, we investigate the strong coupling in the structure based on two Kretschmann configurations involving molecular J-aggregates. The proposed structure is made up of a molecular J-aggregate layer sandwiched between two Kretschmann configurations. Incident light is injected from one Kretschmann configuration with an incident angle, and the output is detected in the other configuration. Fresnel equations are employed to solve our proposed structure. The parameters of molecular J-aggregates and Ag film are changed in order to show the impact on the output. Our structures have potential applications in quantum networks, optical switches, and so on.

2. Calculation models and results

The sketch of our calculation model is shown in Fig. 1. An Ag film–molecular J-aggregate layer–Ag film structure is sandwiched between dielectric media, which is denoted from L1 to L5 from the down to the top. Incident light is injected from the dielectric medium with an incident angle $\theta_{\rm i}$ and the output angle is θ_0. The thicknesses of Ag film and molecular J-aggregate layer are d_1 and d_2, respectively. The dielectric constant function (ε_2) of Ag is obtained in Ref. [32]. The index of the dielectric medium ($\sqrt{\varepsilon_1}$) is chosen to be $\sqrt{3.6}$. The molecular J-aggregates were modeled by a single Lorentzian oscillator[8,11]

$$\varepsilon_3 = \varepsilon_\infty + \frac{f\omega_x^2}{\omega_x^2 - \omega^2 - {\rm i}\gamma_x\omega}, \tag{1}$$

where, ε_∞=1.44^2 comes from the bound electron permittivity, $\omega_x = 2\pi c/\lambda_x$, c is the speed of light, λ_x is the oscillator wavelength of 580 nm, and $\gamma_x = 2.45\times10^{13}$ [rad/s] is the damping constant.[8] f is the oscillator strength and related with the density of molecular J-aggregates.

*Project supported by the Science Fund from the Ministry of Science and Technology of China (Grant No. 2016YFA0301300) and the Fundamental Research Funds for the Central Universities, China.

†Corresponding author. E-mail: haihongli@bupt.edu.cn

‡Corresponding author. E-mail: yuliyuli@bupt.edu.cn

© 2019 Chinese Physical Society and IOP Publishing Ltd

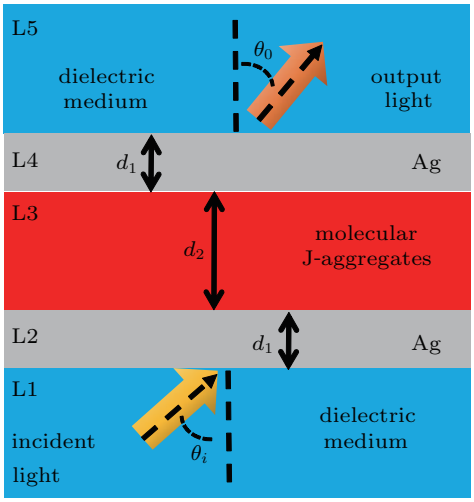


Fig. 1. The sketch of our calculation model.

By solving the wave equation for the magnetic field at the interface between L2 and L3 ($H_{3,\text{down}}$), we can obtain the relation between $H_{3,\text{down}}$ and the magnetic field at the interface between L3 and L4 ($H_{3,\text{up}}$) as

$$H_{3,\text{up}} = H_{3,\text{down}}\zeta = H_{3,\text{down}}\,\mathrm{e}^{-k_0\mathrm{Re}(g)d_2}, \tag{2}$$

where $g^2 = k_0^2\varepsilon_3 - \varepsilon_1\sin^2\theta_i$, and k_0 is the wave vector in vacuum. According to the Fresnel equations of this kind of Kretschmann configurations and decoupling from SPPs to light,[23,29] we can obtain the transmission of our proposed structure

$$T = \eta_i \times \eta_0 \times A_i \times A_0\,\mathrm{e}^{-2\mathrm{i}\mathrm{Re}(g)d_2}, \tag{3}$$

where, $\eta_i \propto 1 - |r_{123}(\theta_i)|^2$, $\eta_0 \propto 1 - |r_{543}(\theta_0)|^2$, r_{123} and r_{543} are the reflection coefficients[23]

$$r_{abc} = \frac{r_{ab} + r_{bc}\exp(2\mathrm{i}k_b d_1)}{1 + r_{ab}r_{bc}\exp(2\mathrm{i}k_b d_1)}, \quad a = 1,\ 5,\ b = 2,\ 4,\ c = 3, \tag{4}$$

where $r_{ij} = (\varepsilon_j k_i - \varepsilon_i k_j)/(\varepsilon_j k_i + \varepsilon_i k_j)$, $i, j = 1,\ 2,\ 3,\ 4,\ 5$. $k_b = \sqrt{k_0^2\varepsilon_b - k_\perp^2}$, $k_\perp = k_0\sqrt{\varepsilon_a}\sin\theta$, and k_0 is the vacuum wave vector. A_i and A_0 are related with the dissipation of Ag films (A_i, $A_0 \sim \mathrm{e}^{-d_1/\delta}$), and δ is the intensity skin depth. Considering the decoupling from SPPs to light, θ_0 is close to $\theta_{\text{SPPs}} = \sqrt{\varepsilon_1^{-1}\frac{\varepsilon_2\varepsilon_3}{\varepsilon_2+\varepsilon_3}}$.[33] The reflection R is related with $|r_{123}|^2$. Hence, we can calculate R and T based on the above equations.

First, we calculate the structures without and with molecular J-aggregates. The thicknesses of Ag films and molecular J-aggregates are 20 nm and 50 nm, respectively. The incident angle is adopted as 40°. f is chosen as 0.01. We calculate the reflection and the transmission in the corresponding surfaces. The reflection (R), the transmission (T), and the output angle (θ_0) *versus* the incident wavelength (λ) of the structures without and with molecular J-aggregates are shown in Fig. 2, respectively.

As shown in Figs. 2(a)–2(d), the strong coupling phenomenon happens in forms of Rabi splitting. The curve of θ_0 *versus* λ shows a Fano line shape. The strong coupling in Kretschmann configuration has been reported in the previous works, which shows two splitting peaks in the reflection spectrum in the structure involving molecular J-aggregates. The additional Kretschmann configuration (made up of L3, L4, and L5) converts SPPs excited in Kretschmann configuration (L1, L2, and L3) into light in L5, and the Rabi splitting appears in the transmission spectrum as shown in Fig. 2(d).

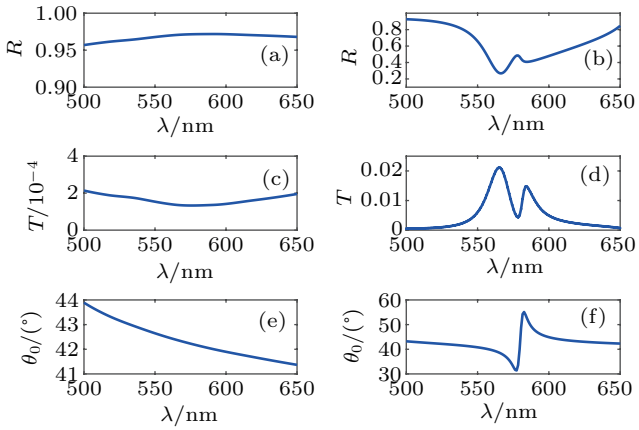


Fig. 2. The reflection R [(a) and (b)], the transmission T [(c) and (d)] and the output angle θ_0 [(e) and (f)] *versus* the incident wavelength λ without and with molecular J-aggregates.

The Fano line shape is induced by the interaction between SPPs and molecular J-aggregates. Light is injected from L1 onto L2 and is changed to SPPs which provides a dark mode, while molecular J-aggregates support a bright mode.[34–36] The interaction of the two modes makes the output angle *versus* the incident wavelength showing a Fano line shape with SPPs decoupling in the structure made up of L3, L4, and L5.

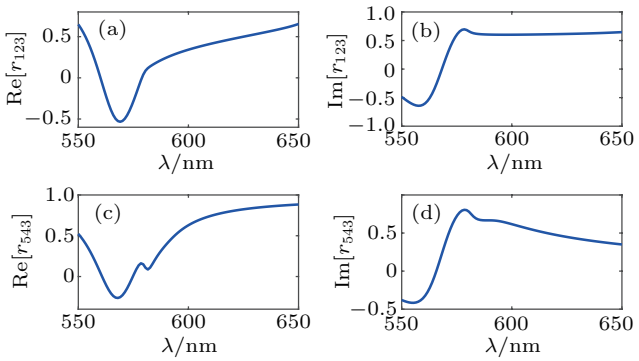


Fig. 3. The real and the imaginary parts of r_{123} and r_{543} with $d_1 = 20$ nm, $d_2 = 50$ nm, and $\theta_i = 40°$, respectively.

We also plot the real and the imaginary parts of r_{123} and r_{543} in Fig. 3 according to Eq. (4). As figure 3 shows, the values of the real and the imaginary parts of r_{123} and r_{543} are close to zero at the wavelength around λ_x. This is because of the strong resonance of molecular J-aggregates making the light confined in the structure. We also noticed that the peak

value of the Rabi splitting is dissymmetry and varies with conditions. This is because the absorption of metals varies with both the thickness of the metal and the incident wavelength.

Furthermore, we use finite-difference time-domain (FDTD) method to calculate our proposed structure. We choose three wavelengths of 550 nm, 580 nm, and 583 nm, which covers the wavelengths around λ_x. The incident angle θ_i and the thickness of Ag film d_1 are 40° and 20 nm. The thickness of molecular J-aggregates d_2 is 50 nm. The indies of Ag and molecular J-aggregates are adopted as above. The gird is 0.5 nm. The output angle θ_0, the transmission T, and the reflection R are listed in Table 1. Comparing these two methods, there are small differences. FDTD method needs to mesh the structure in the simulation processes and it mainly introduces a small errors in the simulation. Hence, there are small differences between these two methods.

Table 1. Comparisons between FDTD and calculation.

Wavelength/nm	θ_0 (FDTD)	T (FDTD)	R (FDTD)	θ_0	T	R
550	43.3	0.005	0.62	41.7	0.006	0.65
580	38.6	0.003	0.51	35.9	0.004	0.48
583	45.5	0.011	0.44	48.5	0.015	0.41

Then, we change the incident angle θ_i to show the changes of the reflection spectrum R and the transmission T. The incident angles 38°, 40°, and 42° are adopted, R and T *versus* λ are shown in Fig. 4. As figure 4 shows, the Rabi splitting denoted as $\Omega = |\omega_+ - \omega_-|$ increases with the increase of the incident angle and the intensity of the transmission decreases, where ω_+ and ω_- are the frequencies of the two splitting peaks.

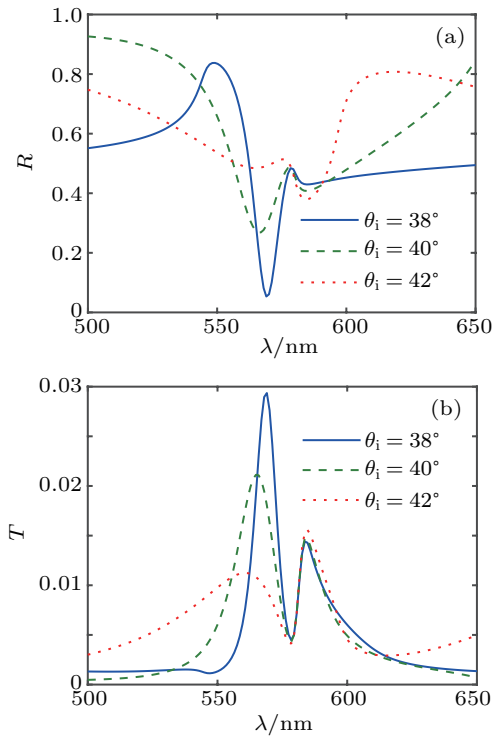


Fig. 4. The reflection spectra (a) and the transmission spectra (b) with $\theta_i = 38°$, 40°, and 42°, respectively.

Rabi splitting in Kretschmann configuration is related with the interaction between SPPs and molecular J-aggregates. The intensities of SPPs for each wavelength are determined by the difference between the incident angle and θ_{SPPs} ($\Delta\theta = \theta_i - \theta_{SPPs}$). If $\Delta\theta$ is less than 0 and θ_i is less than θ_{SPPs} in all wavelength regimes, there are few SPPs excited in Kretschmann configuration. θ_{SPPs} is changed from 30° to 55° in the wavelength regime from 500 nm to 650 nm. With the incident angle increasing from 38° to 42°, the wavelength regime for the best SPP excitation increases and there more and more SPPs interact with molecular J-aggregates. Hence, the Rabi splitting increases with the increase of the incident angle. With the increase of θ_i, much more energy of incident light is used to generate SPPs propagating along the interface between molecular J-aggregates and Ag film. Hence, the transmission decreases.

Then, we show the impact of f on the reflection spectra. The f is considered as 0.005, 0.01, and 0.015, and the results are shown in Fig. 5. As figure 5 shows, Rabi splitting Ω linearly increases with the increase of f. The f is determined by the molecular J-aggregates involving in the interaction with SPPs. If f becomes larger, the number of molecular J-aggregates in the interaction increases and Rabi splitting becomes larger. Comparing with the analysis above, the change of f can be treated as the change of incident angle. The increase of f in the interaction seems that SPPs increase by the increase of the incident angle with molecular J-aggregates fixed. Rabi splitting can be amplified by the increase of SPPs or molecular J-aggregates in the interaction around the resonant wavelength of molecular J-aggregates.

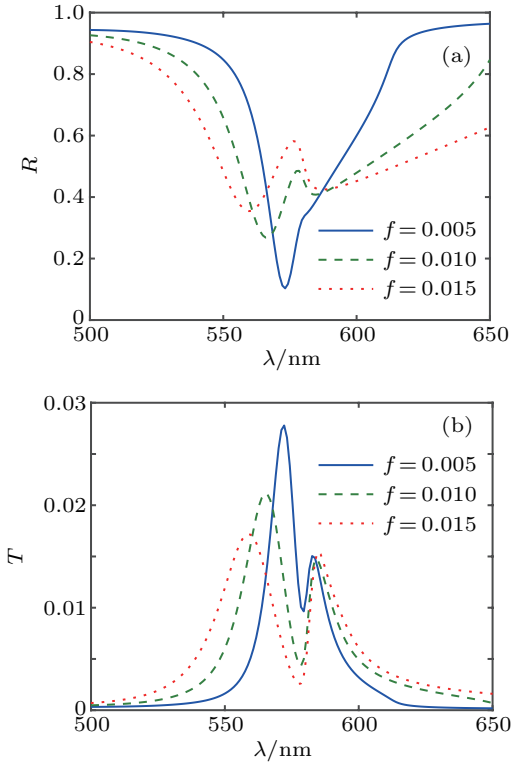


Fig. 5. The reflection spectra (a) and the transmission spectra (b) with $f = 0.005$, 0.01, and 0.015, respectively.

Finally, we change the thickness of Ag film to show how Rabi splitting changes. We adopt $d_1 = 15$ nm, 20 nm, and 25 nm, and the reflection and the transmission spectra are shown in Fig. 6. As shown in Fig. 6, Rabi splitting increases with the increase of d_1 in both reflection and transmission spectra.

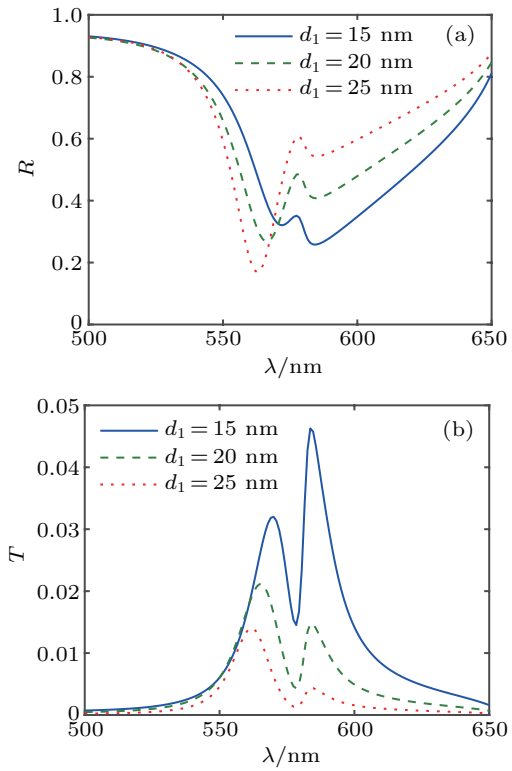


Fig. 6. The reflection spectra (a) and the transmission spectra (b) with $d_1 = 15$ nm, 20 nm, and 25 nm, respectively.

As figure 6 shows, the transmission increases with the decrease of d_1. A part of reflection spectra shows the same behavior with the transmission. With the decrease of d_1, the dissipation of Ag film (A_i and A_0) decreases, especially in the regime from 580 nm to 650 nm. Hence, the transmission and the reflection in this regime show a linear relationship with d_1.

3. Summary

In summary, we theoretically investigate the strong coupling in silver–molecular J-aggregates–silver structure sandwiched between two dielectric media by using Fresnel equations. Results show that both the reflection and the transmission spectra show the strong coupling phenomenon. The radiative angle *versus* the incident wavelength exhibits a Fano line shape around the resonant wavelength of molecular J-aggregates. The incident angle not only impacts on the intensity of the transmission, but also influences on the Rabi splitting. The oscillator strength and the thickness of Ag films are also used to tune the strong coupling. Our structure has potential applications in quantum networks, optical switches, and so on.

References

[1] Tiecke T G, Thompson J D, de Leon N P, Liu L R, Vuletić V and Lukin M D 2014 *Nature* **508** 241
[2] McKeever J, Boca A, Boozer A D, Buck J R and Kimble H J 2003 *Nature* **425** 268
[3] Volz T, Reinhard A, Winger M, Badolato A, Hennessy K J, Hu E L and Imamoglu A 2012 *Nat. Photon.* **6** 605
[4] Imamoglu A, Awschalom D D, Burkard G, DiVincenzo D P, Loss D, Sherwin M and Small A 1999 *Phys. Rev. Lett.* **83** 4204
[5] Khitrova G, Gibbs M, Kira M, Koch S W and Scherer A 2006 *Nat. Phys.* **2** 81
[6] Chang D E, Vuletic V and Lukin M D 2014 *Nat. Photon.* **8** 685
[7] Hennessy K, Badolato A, Winger M, Gerace D, Atatüre M, Gulde S, Fält S, Hu E L and Imamoglu A 2007 *Nature* **445** 896
[8] Eizner E, Avayu O, Ditcovski R and Ellenbogen T 2015 *Nano Lett.* **159** 6215
[9] Trügler A and Hohenester U 2008 *Phys. Rev. B* **77** 115403
[10] Zengin G, Wersäll M, Nilsson S, Antosiewicz T J, Käll M and Shegai T 2015 *Phys. Rev. Lett.* **114** 157401
[11] Lin Q Y, Li Z, Brown K A, N ÓBrien M, Ross M B, Zhou Y, Butun S, Chen P C, Schatz G C, Dravid V P, Aydin K and Mirkin C A 2015 *Nano Lett.* **15** 4699
[12] Schlather A E, Large N, Urban A S, Nordlander P and Halas N J 2013 *Nano Letters* **13** 3281
[13] Zengin G, Johansson G, Johansson P, Antosiewicz T J, Käll M and Shegai T 2013 *Sci. Rep.* **3** 3074
[14] Chikkaraddy R, de Nijs B, Benz F, Barrow S J, Scherman O A, Rosta E, Demetriadou A, Fox P, Hess O and Baumberg J J 2016 *Nature* **535** 127
[15] Vasa P, Pomraenke R, Cirmi G, De Re E, Wang W, Schwieger S, Leipold D, Runge E, Cerullo G and Lienau C 2010 *ACS Nano* **412** 7559
[16] Symonds C, Bonnand C, Plenet J C, Bréhier A, Parashkov R, Lauret J S, Deleporte E and Bellessa J 2008 *New J. Phys.* **10** 065017
[17] Dintinger J, Klein S, Bustos F, Barnes W L and Ebbesen T W 2005 *Phys. Rev. B* **71** 035424
[18] Pockrand I, Brillante A and Möbius D 1982 *J. Chem. Phys.* **77** 6289
[19] Bellessa J, Bonnand C, Plenet J C and Mugnier J 2004 *Phys. Rev. Lett.* **93** 036404
[20] Ouyang Q, Zeng S, Jiang L, Hong L, Xu G, Dinh X Q, Qian J, He S, Qu J, Coquet P and Yong K T 2016 *Sci. Rep.* **6** 28190
[21] Sarkar M, Besbes M, Moreau J, Bryche J F, Olivéro A, Barbillon G, Coutrot A L, Bartenlian B and Canva M 2015 *ACS Photon.* **2** 237
[22] Memmi H, Benson O, Sadofev S and Kalusniak S 2017 *Phys. Rev. Lett.* **118** 126802
[23] Kretschmann E 1971 *Z. Physik* **241** 313
[24] Yang Y, Liu J and Li Z Y 2015 *Chin. Phys. B* **24** 074201
[25] Lu J H and Wang G H 2016 *Chin. Phys. B* **25** 117804
[26] Liu X and Yang D X 2016 *Chin. Phys. B* **25** 047301
[27] Juan M L, Righini M and Quidant R 2011 *Nat. Photon.* **5** 349
[28] Palomba S and Novotny L 2008 *Phys. Rev. Lett.* **101** 056802
[29] Zhou H, Chen X, Hou P and Li C F 2008 *Opt. Lett.* **33** 1249
[30] Goos F and Hänchen H 1947 *Ann. Phys.* **436** 333
[31] Artmann K V 1948 *Ann. Phys.* **2** 87
[32] Palik E 1985 *Handbook of Optical Constant of Solids* (San Diego: Academic Press)
[33] Maier S A 2007 *Plasmonics: Fundamentals and Applications* (Springer)
[34] Mingbo P, Yinghui G, Xiong L, Xiaoliang M and Xiangang L 2018 *ACS Photonics* **5** 3198
[35] Huang Y J, Luo J, Pu M B, Guo Y H Y, Zhao Z Y, Ma X L, Li X and Luo X G 2019 *Adv. Sci.* **6** 1801691
[36] Guo Y H, Ma X L, Pu M B, Li X, Zhao Z Y and Luo X G 2018 *Adv. Opt. Mater.* **6** 1800592