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Back focal plane imaging of Tamm plasmons and their coupled emission

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Abstract

The unique optical properties of TPs – such as flexible wavevector matching conditions including in-plane wavevector within the light line, existing both S- and P-polarized TPs and ability of populating with KR and RK illuminations – facilitate them for direct optical excitation. The Tamm plasmon Coupled emission (TPCE) from a combined photonic-plasmonic structure sustaining both surface plasmons (SPs) and Tamm plasmons (TPs) is described. The sensitivity of TPCE to the emission wavelength and polarization is examined with back focal plane imaging and verified with the numerical calculations. The results reveal that the excited probe can couple with both TPs and SPs, resulting in SPCE and TPCE, respectively. The TPCE angle is strongly dependent on the wavelength allowing for spectral resolution using different observation angles. These Tamm structures provide a new tool to control the optical emission from dye molecules and have many potential applications in fluorescence based-sensing and imaging.

Keywords

Tamm Plasmons; Surface Plasmons; Tamm Plasmon-Coupled Emission Fluorescence; Back Focal Plane Imaging

1. Introduction

Surface plasmon resonance (SPR) and surface plasmon-coupled emission (SPCE) are widely used in the biosciences and material sciences [1-5]. Both SPR and SPCE depend on unique optical conditions to allow access to the surface plasmon modes. Surface plasmons (SPs) have higher wavevectors (shorter wavelengths) than that of freely propagating light with the same frequency. As a result, SPs exist outside the light line. Illumination through a high refractive index prism is needed to increase the incident wavevectors to match the SPs. In the case of SPCE the fluorophore must be within a sub-wavelength (near-field) distance of

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Supporting Information

Supporting Information is available online.

the metal so the high local wavevectors of the fluorophores can interact with the SPs. In the present report we examine the unique optical properties of Tamm plasmons (TPs). TPs, sometimes called Tamm plasmon polaritons (TPPs), are a trapped electromagnetic state that exists between a metal and a dielectric Bragg reflector where the electric-magnetic field is highly confined [6]. This location is different from the widely investigated SPs, which are coherent electron oscillations that exist at the metal-dielectric interface (such as a metal sheet in air) [7, 8]. TPs have wavevectors within the light cone, and thus it can be optically excited without the aid of prisms, gratings or small defects. In contrast to SPs, TPs can have either S- or P-polarization. For TPs, the electric-field confinement in the metal is achieved as a result of its negative dielectric constant. The confinement in the dielectric multilayer structure is due to the photonic stop band of the Bragg reflector [6]. Due to their strong localization normal to the interface and slow in-plane motion governed by a parabolic dispersion law, TPs can be seen as the slow and compact light [9], which makes them a promising candidate for numerous applications, such as absorbers [10], filters [11], and bistable switches [12]. TPs based sensors have achieved a sensitivity $\sim 900 \text{ nm} / \text{RIU}$ with high detection accuracy ($\sim 30 \mu\text{m}^{-1}$) [13]. One-way Tamm plasmons polaritons at the interface between magneto-photonic crystals and conducting metal oxides has also been theoretically reported [14]. Single quantum dots coupled to the TPs were shown to experience acceleration or inhibition of their spontaneous emission depending on their emission spectral shift from the resonant wavelength of the TPs [15]. Based on this finding, new kinds of metal/semiconductor lasers and the single photon source using confined Tamm Plasmon (TP) modes have been experimentally realized [16,17]. In this paper, combined photonic-plasmonic structures were fabricated which can support both SP and TP modes. We used back focal plane (BFP) imaging technique, which has the merits of high spatial resolution and ability of real-time measurement [18-22], to investigate the optical properties of the TP modes, especially their sensitivity to wavelength and polarization. Further, in the near-field the TP modes can couple with dye molecules which modulate the emitting direction, spectra and polarizations of the fluorescence (which are seldom reported before). These BFP imaging experiments reveal the different optical properties of SPs and TPs even when they are present in the same structure. Our experiments show that fluorophores on top of the metal film can couple with the TPs under the film and result in Tamm Plasmon Coupled Emission (TPCE). In contrast to SPCE, TPCE occurs within the light line and can even be directed normal to the surface. As a result we anticipate the use of TPCE in the biosciences, medical diagnostics, sequencing and imaging [23]. The present paper represents part of our continuing efforts to use near-field effects to obtain new opportunities and formats for fluorescence detection. We show that coupling between fluorophores and TPs can convert the usual omni-directional emission into directional emission and modify the polarization of the coupled emission without the use of any lenses or polarizers. The near-field coupling also results in the directional angle sorting of fluorescence emission at different wavelengths, originating at the site of emission, rather than with external optical components.

2. Experiments and results

The combined photonic-plasmonic structure (for simplicity, we name it as Tamm structure) was fabricated using plasma-enhanced chemical vapor deposition (PECVD) of SiO₂ and Si₃N₄ on standard glass cover slips (0.17 mm thickness). Prior to PECVD of the Tamm structure, the glass cover slips were cleaned with piranha solution and then rinsed with nano-pure deionized (DI) water and dried with an air stream. The Ag film was evaporated onto the dielectric Bragg reflector with a thermal vapor deposition. The Rhodamine 6G (Rh6G) doped PMMA was spin-coated onto the Ag film with a final thickness of about 15 nm. The thickness of each layer is illustrated in Figure 1.

2.1. Experimental apparatus

The schematic diagram of the experimental set-up is shown in Figure 2. The white light source (Thorlab, OSL-I, spectrum ranging from 450 - 700 nm) combined with a set of band pass filters are used to obtain light at different wavelengths. The center wavelengths of the filters are 570, 590 and 610 nm with 10 nm spectral bandwidth. A lens and beam-expander are used to collimate the white light which then illuminates the full rear aperture of an oil-immersed objective (60X, N.A = 1.42). By this way, the incident angle of the white light on the Tamm sample will range from -69° to 69° [24]. The reflected white light from the Tamm sample is collected by the same objective and then imaged onto a sCMOS camera by a lens system. The position of the sCMOS is adjusted to image the back focal plane (BFP) of the objective. A polarizer is placed after the white light source to obtain different polarizations of the incident light. The white light source also can be used to measure the transmission spectrum of the Tamm structure. In this case, a fiber with a collector linked with the spectrometer is placed on the Tamm sample. We remove the filters and the objective so that the white light can incident on the Tamm sample at zero angle. Unless otherwise noted all incident light and observations are through the Bragg reflector side of the sample.

2.2. Transmission and fluorescence spectra

The measured transmission spectrum of the Tamm structure is presented in Figure 3. For comparison, the transmission spectrum of the dielectric Bragg reflector (without the Ag and PMMA film) is also shown in Figure 3 (black curve). The Bragg reflector can reflect the light at wavelengths ranging from 540 nm to 600 nm and there is no transmission peak at 590 nm. Addition of the Ag layer to the Bragg reflector results in a new transmission band near 590 nm (red curve). The appearance of the transmission peak at 590 nm is consistent with the numerical simulation shown in Figure S1. When the light at 590 nm wavelength strikes the Tamm sample at zero angle, a decrease in the reflectivity curve is noticed. At this incident angle, the 590 nm wavelength light can excite the TPs between the Ag film and Bragg reflector. The TP field can penetrate the thin Ag film (42 nm thickness) and then radiate into the upper space of the Tamm sample. As a result, a transmission peak at 590 nm wavelength appears.

To investigate the interactions between the dye molecules and the Tamm structure, a 532 nm wavelength laser is expanded and focused onto the same position of the Tamm structure as the white light source was focused. The dye Rh6G doped in the PMMA film can be excited

by the 532 nm laser. The spectrum of the fluorescence emitting into the upper space was measured with the fiber collector and the spectrometer. The fluorescence spectrum collected by objective can also be measured with the same spectrometer. The emission angle of the fluorescence at different wavelengths can be derived from the fluorescence BFP image of the objective. In this case, the band-pass filters and the polarizer used in white light experiments is moved to the position before the relay-lens system as shown in the dash box of Figure 2.

Figure 4 (a) presents the fluorescence spectrum emitting into the upper space of the Tamm sample, which is similar to the emission spectrum of Rh6G on a glass substrate with peak wavelength at about 570 nm, Figure 4 (a). For comparison, the emission spectrum of the fluorescence emitting through the Tamm sample is also shown in Figure 4 (b). The difference in these spectra reveals that the emission spectrum is modulated due to the coupling with the Tamm samples. The wider emission spectrum observed through the Tamm structure using the objective may be due to the wider acceptance angle and wider range of collectible wavelengths. Changes in the shape of emission spectra with a Tamm structure were reported in reference [25] and for photonic structures in references [26, 27]. It is difficult to separate the effects of angular wavelength separation from changes in the total emission, but references [25-27] represent growing evidence that the total emission spectrum can be redistributed due to near-field interactions with these structures.

2.3. Tamm plasmons and surface plasmons excited by the white light

The BFP images of the reflected white light from the Tamm sample are shown in Figure 5. The incident wavelengths are 570, 590, and 610 nm (selected from white light by using band pass filters) the same as those used in the numerical simulations (Supporting Information). The polarization direction of the incident light is shown by the white double-headed arrow lines. There are pair of partial dark arcs towards the outer edge on all three BFP images, which are the typical signatures of P-polarized surface plasmons. From the diameter of dark arcs and N.A of the objective, the polar (or radial) angle of the dark arcs at the three wavelengths can be derived as 46.12° (570 nm), 45.75° (590 nm), 45.37° (610 nm), which are consistent with the angles of the SP dips on the reflectivity curves shown in Figure S1 (b). These SP resonant angles decrease with increasing wavelengths, but are only weakly dependent on wavelengths.

In contrast to the dark SP arcs, there is a dark, slightly elliptical ring appeared on the BFP image as shown in Figure 5 (a), which rotates with the polarization direction of the incident light (not shown here). In our experiment, the expanded white light is linearly polarized and focused by the axially symmetric objective. Then, some of the spots on the expanded beam will be P-polarized and some are S-polarized with respect to the x-z or y-z planes, respectively. For example, we select two points (red points, labeled with P and S) on the elliptical ring as shown in Figure 5 (a). The polarization direction of the point S is along the tangential of the dark ring and that of point P is along the radial of the ring. The corresponding incident angles related with the points P and S can be derived as 17.32° and 18.32° , respectively, which are consistent with the corresponding resonant angles of the dips on reflectivity curves (Figure S1). So the appearance of the dark elliptical ring on Figure 5

(a) is due to the excitation of the TP mode using 570 nm wavelength light. In contrast to the partial SP arcs, this ring has nearly the same depth at all angles around the z-axis. This continuous ring occurs because the TPs exist for both S- and P-polarization. When the incident light is 590 nm, the dark elliptical ring shrinks into the center of the BFP image and becomes round disc as shown in Figure 5 (b). This phenomenon means that the TPs resonant angle decreases to near-zero for both S- and P-polarized 590 nm incident light, which is consistent with the numerical simulations shown in Figure S1. When the wavelength is further increased to 610 nm, the dark rings or disks disappears. Once again, this is consistent with the dips related with the TP mode on the reflectivity curves shown in Figure S1 that are much less visible at 610 nm. The Tamm structure in Figure 1 becomes unable to support TPs above 610 nm. The consistence between the numerical and experimental results clearly verifies the presence of both SP and TP modes on the proposed Tamm structure. Different from the SP modes, which can be excited over a broad wavelength range, the TP modes exist for a narrow range of wavelengths. The resonant angle of the TP mode is more sensitive to the incident wavelength than the SP mode.

2.4. Tamm plasmon-coupled emission and surface plasmon-coupled emission

We now consider the interactions of fluorophores with the Tamm structure. It is known that excited state fluorophores can display near-field coupling with surface plasmons on smooth metal films. This interaction results in directional fluorescence emission, which is commonly known as the surface plasmon-coupled emission (SPCE) [28, 29]. It is a reverse process of the SPs excitation. We reasoned that excited state fluorophores could couple with TPs to yield Tamm plasmon-coupled emission (TPCE). The fluorescence spectrum observed through the substrate side of the Tamm sample is given in the Figure 4 which yields a broad band emission. Three band-pass filters are used to select the fluorescence wavelengths. The corresponding fluorescence intensity distributions at the BFP of the objective are shown in Figure 6. On the three images, the outer bright rings have typical signatures of the SPCE and hence are due to emission coupled to SPs mode [30, 31]. With a polarizer before the sCMOS, the rings are split into two arcs due to the unique polarization of the SPCE. The bright rings on the BFP images represent the directional emission from Rh6G. The emitting angle can be derived as 44.70° (610 nm), 45.16° (590 nm), 45.78° (570 nm) which are consistent with the numerical simulation (Figure S1, Supporting Information) and the white light experimental results shown in Figure 5.

In addition to the split SPCE rings, a slightly elliptical ring of emission appears on Figure 6 (a), similar to the elliptical dark ring on the Figure 5 (a). The corresponding emission angles of the point P and S on the elliptical ring can be derived as 17.67° and 18.71° . The polarization direction of the fluorescence on point S is along the tangential of the ring and that of point P is along the radial of the ring. So the fluorescence point S is of S-polarization and point P of P-polarization with respect to the surface of the Tamm structure. We note that the polarization at spots S and P on Figure 6a is due to the emission polarizer because both polarizations appear at nearby the same angle. The polarization and the emitting angles of the spots on the elliptical ring are consistent with those (point S and P) on the dark elliptical ring on Figure 5 (a). The consistence also appears between Figures 5(b), (c) and Figures 6(b), (c), respectively. So the dark elliptical ring on Figure 5 (a) and bright elliptical ring on

Figure 6 (a) can be attributed as the TPCE. In our experiment, the Rh6G doped PMMA is on the Ag film whereas the optical field of the TP modes is confined below the Ag film (Figures S2 and S3). To couple with the TP modes, the fluorescence must couple through the Ag film and is probably less efficient due to the longer distance. On the other hand, the optical field of the SP mode is localized on the Ag film (Figure S3 (d)), and within the PMMA film, so it can more easily couple with the dye molecules above the metal film. For this reason the intensity of the SPCE is stronger than that of the TPCE. Due to the small in-plane wave-vector of the TP mode, the TPCE could be noticed normal to the surface or at small angles away from the normal axis of the planar Tamm sample. When comparing with the plasmonic lenses made of metallic nano-structures which also can beam the fluorescence emission [32], the Tamm structure is easier to be fabricated and of lower cost. And also the large planar surface of the Tamm sample is more favorable for the high throughput optical sensing and imaging.

From the BFP images of the white light and fluorescence, we found that the TP mode is very sensitive to the wavelength. A small shift of the wavelength will induce large change of the TP resonant angle or the TPCE angle. For example, at 570 nm, the TPCE angle is about 18°, whereas, at 590 nm, the TPCE angle is near zero. So the Tamm sample is also very useful in directional sorting of fluorescence emission which was previously realized by the plasmonic antenna composing of corrugated metallic nano-apertures with grooves [33]. For each emission wavelength, the fluorescence beam can be directed along a specific direction. Comparing with this kind of antenna, the Tamm structure is more sensitive to wavelength, so the fluorescence at closer wavelengths can be easily sorted by the observation angle.

2.5. Wavelength-tunable Tamm plasmon-coupled emission with surface normal directionality

In many applications, such as DNA or protein arrays, it will be useful to obtain zero-angle fluorescence emission (normal to the surface of the sample) at different wavelengths. To accomplish this with plasmonic antennas, we have to change the structural parameters at nanoscale which will be time consuming and of high cost [34-36]. Herein, we demonstrate that this can be easily realized with a single planar Tamm structure by fine-tuning the dimensions of layers in Tamm structure. For this we considered using the Tamm structure with varied silver layer thickness. This can be realized by the thermal evaporation of the Ag film on to slightly tilted glass substrate with Bragg reflector. By this way, the thickness of the evaporated Ag film on the Bragg reflector will be non-uniform. The Ag film becomes gradually thicker from one side to the other side of the sample. The inset graph of Figure 7 presents schematic of the new Tamm structure, where thickness of the Ag film is gradually varied. Subsequently, we spin-coated the Rh6G doped PMMA film that may be a little different from 15 nm due to the non-uniform thickness of the Ag film, but the following experiments will verify that the thickness of the dielectric PMMA layer above the Ag film has no influence on the TP modes and TPCE. We then measured the transmission spectra at four selected spots on the sample as shown in Figure 7, which present peaks at 560, 570, 580 and 600 nm, respectively. The four spots are labeled as A, B, C and D. The transmission peak shifts to longer wavelengths from point A to D, which is consistent with the spatial distribution of the Ag film's thickness. From A to D the thickness of the Ag film is

decreased. The peak wavelengths of the transmission spectra mean that the fluorescence emission at these wavelengths will be beamed normal to the surface of the Tamm substrate. The corresponding fluorescence and white light BFP images shown in Figure S4 verify this directional emission and transmission for each of these wavelengths at near zero degrees. We recently noticed wavelength insensitivity towards the metal layer thickness when the probe is positioned below the metal layer. However, the present study indicates that the TPs mode in the fabricated structure is indeed Ag layer thickness dependent. This discrepancy in the observed wavelength sensitivity might be due to multiple factors influencing the TPCE. One such factor might be the thickness change in the top dielectric layer^[11]. Also the other study involves with the probe below metal layer and for the present study we are using the probe above the metal layer. We are not sure of the effect of probe location on the observed phenomenon. Subsequently, understanding the differences in the observation is more complex than we think and we may need to address this issue more carefully that can be the aspect for the future publication.

Finally, a water solution mixed with Rh6G molecules is dropped on the surface of the Tamm structure shown in the inset graph of Figure 7, which can represent a biological sample. As we know, biological entities, such as cells, labeled with fluorescence probes are always cultured in the solution with approximately the same refractive index as the water. To prevent the flow and evaporation of the water solution, a cover glass is placed on the Tamm sample. Figure 8 presents the fluorescence BFP images from four selected spots on the Tamm sample. At each spot, the fluorescence – at 560 (a), 570 (b), 580 (c), and 600 nm (d) wavelengths respectively – coupling with the TPs mode is designed to beam normal to the surface (Figure S4). For example, the bright disk on the center of BFP image presented in Figure 8(a) shows that the TPCE at 560 nm propagates normal to surface of the structure. The appearance of the center bright disks at the same wavelengths as those in Figure S4 demonstrates that the water layer on the Tamm structure does not influence the TP resonant angles, which is because the TP modes exist below the Ag film. Different from the fluorescence BFP images shown in Figure S4 or Figure 6, the SPCE rings do not appear with water layer on the top of the metal layer. The larger refractive index ($n = 1.33$) of water than that of air ($n = 1$) made the wavevector of the SP at Ag-water interface larger than that for Ag-Air interface. As a result the SPCE cannot be collected by the objective due to the limited numerical aperture of the objective and refractive index of the glass substrate. Compared to Figure S4 (or Figure 6), the bright TPCE disks at the center of the BFP images in Figure 8 becomes more visible. It is difficult to compare the brightness of the disks in Figures 8 and Figure S4 because we are yet to know the distances over which fluorophores can couple with the Tamm structures. These experimental results (Figure 8 and Figure S4) clearly verify the advantage of TPCE over the SPCE due to the smaller or near-zero in-plane wavevector of the TPs modes. It should be noted that some of the emission appearing at larger angles may be due to coupling to the internal cavity modes such as S2 in Figure S1^[24]. If the band-pass filter and the black-white sCMOS are replaced with a 532 nm long pass filter and a color CCD, the fluorescence at different wavelengths that is reaching CCD can be observed simultaneously. Figure 8 (a2) shows the corresponding color BFP image taken from the spot where the transmission peak locates at 560 nm wavelength. The center of the BFP appears green which means that the short wavelength (such as 560 nm

wavelength) is beamed normal to the glass substrate. A red ring appears around this green center disk, which means the fluorescence at longer wavelength (such as 600 nm) emits into a narrow angular ring in the direction with a non-zero angle to the normal axis of the glass plane. For comparisons, the color BFP image from the spot D where the transmission peak locates at 600 nm wavelength is presented in Figure 8 (d2). Here the center disk is red and a green ring appears around the red disk which means that the long wavelength light (600 nm) are beamed normal to the glass substrate. These two color BFP images verify the wavelength sorting of directional fluorescence emission is still preserved when the Tamm structure is covered with the water solution.

3. Conclusions and outlook

This work reports the facile and easy fabrication of large area hybrid photonic-plasmonic structure, Tamm structure, which shows both TP and SP modes. Back focal plane imaging technique is used to investigate the near-field interactions of fluorophores with these modes and the observed results are in agreement with the simulation results. TPCE exhibits both S- and P-polarization with light illumination under normal incidence and angular incidence within the light line. Whereas the SPs mode only can be populated with P-polarized light at angle larger than the critical angle and it provides typical P-polarized SPCE at SPR angle. When the resonant angle of the TPs mode is zero, the S-polarized and P-polarized TPs demonstrate the same electric field distribution despite the polarizations of the incident light. The TPCE angle is more sensitive to the emission wavelength. Whereas the corresponding shift SPCE angle is small. We further investigated the effect silver layer thickness on the emission wavelength, and the refractive index on effect on the TPCE and SPCE. The TP field intensity is confined within the structure and insensitive the refractive index of the surface layer and can sustain TP modes in aqueous environment as shown in the present study. Whereas the SPCE is sensitive to the top layer refractive index and SPCE could be not observed for the Tamm structure with the aqueous environment.

The hybrid plasmonic-photonic structures as described herein provide opportunities for spatial and spectral control of fluorophores which originates at the site of emission, rather than external optical devices that operate on the free-space emission. In contrast to the plasmonic antennas developed for beaming fluorescence emission [32-36], our structure used only thin one-dimensional films which were deposited by vapor deposition and spin-coating. Also, it contains no nanoscale features and thus large area structures can be fabricated at low cost. The Tamm modes are assembled within the light line, from either direction, and does not require a prism to couple the light into or out of the structures. These unique features of Tamm structures will have potential applications biological and clinical sensing using high-throughput formats, micro-well plate and for fluorescence microscope-based studies. The low angles are also favorable for wide-field imaging or observation with low NA objectives, which is useful in development of TPs-mode related lasers or single-photon sources. High angle-dependent emission dispersion ability of the TPCE is favorable for the wavelength sorting and multiplexing formats. The dual modes (SPs and TPs) of Tamm structures are expected to provide new opportunities for the plasmon-enhanced applications [37, 38].

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Figure 1. Schematic of the Tamm structure and layer dimensions. $Z = 0$ is at the metal-Bragg reflector interface.

Figure 2.

Sketch of the experimental set-up. In the case of the white light BFP measurement, the polarizer and the filters (in the dash line boxes) are placed before the white light source. For fluorescence BFP measurement, they are placed before the relay-lens.

Figure 3.
Measured transmission spectra of the Tamm structure and the dielectric Bragg reflector.

Figure 4.

Fluorescence spectrum from the Rh6G molecules measured from the upper space (a, red line) and from the substrate side of the Tamm sample (b). The emission spectrum of Rh6G molecules on a glass substrate is also shown in ((a), black dotted line).

Figure 5.

BFP images of the reflected 570 (a), 590 (b) and 610 nm (c) light from the Tamm structure. A white light source with a 570, 590 and 610 nm band-pass filter is used to illuminate the Tamm structure through the Bragg reflector. The incident light is linearly polarized as represented by the white arrow lines. Two points P and S are selected on the slightly elliptical ring on (a).

Figure 6.

BFP images of the fluorescence from the Rh6G molecules on the Tamm structures. The center wavelengths of band pass filters are 570 (a), 590 (b) and 610 nm (c). The white arrows on (a), (b), (c) represent the direction of the polarizer placed before the CCD (Figure 2).

Figure 7. Transmission spectra of the Tamm structure at four different spots (A, B, C and D). The Rh6G molecules are doped in the PMMA film.

Figure 8.

Fluorescence BFP images of four selected spots on the Tamm samples covered with water solution. The Rh6G molecules are dissolved in the water solution. The center wavelengths of band pass filters are 560 (a), 570 (b), 580 (c), and 600 nm (d). (a2) and (d2) are the color BFP images taken from the same spot as (a) and (d) respectively. No polarizer is used before the CCD.