Published in final edited form as:

Opt Lett. 2014 August 1; 39(15): 4341–4344.

Extracting surface wave-coupled emission with subsurface dielectric gratings

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Abstract

Conventional surface plasmons (SPs) or Bloch surface waves (BSWs) have a wave vector exceeding that of light in vacuum, and, therefore, the surface plasmon-coupled emission (SPCE) or Bloch surface wave-coupled emission (BSWCE) cannot escape from the corresponding structures. With the aid of a high-refractive-index prism or an oil-immersion objective, the SPCE or BSWCE can be coupled into free space. But the large volumes of the prism and objective are certainly unfavorable for miniaturization of the optical systems or inconvenient for applications such as the optical displays. Here we experimentally demonstrate a new method to extract the SPCE or BSWCE with a subsurface dielectric grating. The experimental results verify that the chip-like substrate with two decorated sides can bring out the directional fluorescence emission in free space. The emitting direction and emitting patterns can be tuned by the period size and dimensionality of the gratings. Our work provides a new strategy to realize free-space directional fluorescence emission at a very low cost and compact configuration, which has potential applications in fluorescence-based sensing, imaging, light-emitting diodes, optical displays, and other near-field optical devices.

Fluorescence detection is widely used in biosciences, medical diagnostics, sequencing, and imaging. However, fluorescence detection is reaching some natural limits, and only incremental improvements in sensitivity, using classical far-field free-space optics, can be expected [1]. To circumvent these limits, near-field effects have been widely adopted to control emission from dye molecules, which is due to rapid development of plasmonics [2,3]. The strong resonant coupling between the excitation light and noble metal nanostructures leads to a number of remarkable optical effects, including a high near-field enhancement of fluorescence around the metal surface. The optical absorption and scattering properties of metallic nanostructures can be used to control the radiative decay rate of a fluorophore emission [4]. For example, a significant increase in the fluorescence signal from single fluorophores controllably linked to the ends of Au nanorods [5] or inside nanometric

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apertures milled in gold film [6] was reported. The gold nanoapertures surrounded by periodic corrugations transform standard fluorescent molecules into bright unidirectional fluorescence emission sources [7]. A plasmonic spiral antenna can beam the fluorescence from quantum dots (QDs) with controllable polarization states [8]. The plasmonic interaction between Ag nanocubes and the film can induce a significant fluorescence enhancement [9]. For these fluorescence emission-controlling methods, the related positions of the fluorescent molecules to the nanostructures are very crucial. On the other hand, a thin, continuous metallic film can induce directional emission from fluorescent molecules, known as surface plasmon-coupled emission (SPCE) [2]. Additionally, we have shown recently that Bloch surface waves (BSWs) loaded on one-dimensional photonic crystals (1DPCs) with appropriate alternate low- and high-dielectric layers can interact with the nearby fluorophores and convert the usual omnidirectional emission into directional emission, known as Bloch surface wave-coupled emission (BSWCE) [10–12]. These directional emissions are both related to the confined surface wave (SW) on the planar films, so they can be named as SW-coupled emission (SWCE). In contrast to metallic nanostructures, SWCE does not require precise nanoscale features, and it is not necessary to precisely control the lateral location of the fluorescent molecules. When they are within near-field distances from the metallic film or 1DPC, the interaction of the fluorophores with the surface plasmons (SPs) or BSWs will induce directional emission. A simple planar structure is more favorable for high-throughput imaging or sensing. However, the wave vector of the SWs is larger than that of light in vacuum, so the SWCE cannot propagate into free space without the aid of a high-refractive-index medium such as a prism or an oil-immersion objective [13]. It is evident that a large size of the prism or objective is unfavorable for integration or miniaturization of the optical system. To resolve this problem, here we propose a new coupling configuration to extract the SWCE to free space with substrate engineering, which will not destroy the planar surface of the metallic film or 1DPC, and will preserve the directional emission. The new substrate is more compact and can be easily integrated into larger devices.

Figure 1(a) presents the schematic of the structure with two decorated sides. The Ag film of 40 nm thickness is evaporated on the upper surface of a glass substrate with electron beam evaporation [2,3]. Rhodamine B (RhB) molecules are doped in a polymethyl methacrylate (PMMA) film coated on the Ag film. The thickness of the RhB-doped PMMA film is about 60 nm. At the subsurface of the glass, large-area dielectric gratings (SX AR-N 7520/4, ALLRESIST GmbH, Germany) are fabricated by two-beam interference lithography with a laser beam (442 nm wavelength) from a He–Cd laser (Kimmon, Japan). The green laser emitting at 532 nm wavelength is used to excite the RhB molecules. Several bandpass filters (with a bandwidth of 10 nm) are used to filter the fluorescence wavelengths and reject the laser beam. Figure 1(b) shows the atomic force microscopy (AFM) image of the grating and the cross-section profile along a white line. The grating period (P) is about 505 nm and depth 70 nm.

The excited dye molecules will couple with the SPs inside the Ag–PMMA–air multilayer film, which results in directional emission into the glass substrate at the surface plasmon resonant (SPR) angle [θ shown in Fig. 1(a)]. As we know, the SPR angle is larger than the

total internal reflection angle at the glass–air interface. When the directional fluorescence beam reaches the lower surface of the glass, it will be totally reflected back to the glass and cannot escape into free space. As illustrated in Fig. 2(a), the red circular ring R_0 represents the directional SPCE into the glass substrate at the same radial angle θ but a different azimuthal angle ϕ . The diameter of the ring is equal to the effective index ($n_{sp} = K_{sp}/K_0$) of the excited SPs. The red ring is corresponding to the SPCE ring on the back focal plane (BFP) image using an oil-immersion objective from the same sample shown in Fig. 1(a), but without the bottom grating [14]. This situation will change if a dielectric grating is fabricated on the lower surface of the glass. As the fluorescence beam reaches the lower surface, it will generate a propagating evanescent wave at the glass–air interface, which interacts with the grating and forms the fluorescence Bloch waves. The wave vector of the Bloch waves is $\mathbf{K} = \mathbf{K}_{sp} \pm n * \mathbf{G}$, where $\mathbf{K}_{sp}$ is the wave vector of the SPCE, the grating vector $\mathbf{G}$ is along x axis, with value $(\lambda_0/P)*K_0$, and n is an integer. Here we only consider the case $n = 1$; λ_0 is the free-space (air) wavelength of the fluorescence [15]. Then the wave vectors of the Bloch waves are illustrated as the red rings R_{-1} and R_1 , which clearly show that a part of the fluorescence Bloch waves has wavenumbers smaller than that of light in vacuum, so it can enter free space at an angle β [see Fig. 1(a)].

BFP imaging (BFPI) is used to measure the emitting angles of the coupled fluorescence [10, 16]. The blue ring (blue dashed circle) in Fig. 2(a) illustrates the collection range of the objective used in the BFPI (60 $\times$, N.A. = 0.70). The N.A. of the objective is considered to be smaller than 1, so it can only collect fluorescence within the light cone. The period of the grating is 505 nm. Figures 2(b)–2(e) present the BFP images of the fluorescence at wavelengths of 560, 600, 630, and 640 nm, respectively, where two bright arcs appear in each image and are consistent with the pattern in Fig. 2(a). The left arc is related to the ring R_1 and right one to R_{-1} . As we know, each spot on the BFP images represents an emitting angle (β and ϕ) of the fluorescence; so the bright arcs indicate that the coupled fluorescence only radiates into free space at certain angles, which is a typical feature of directional fluorescence emission.

The two bright arcs are symmetrical and intersect in the BFP images where the cross area is brighter than the other parts of the arcs. From Figs. 2(b)–2(e), with increase in wavelength, the left arc moves toward $+x$ direction and the right one moves toward $-x$ direction. The cross areas of the two arcs move toward the center of the BFP images, which means that the fluorescence is emitted into free space at small angles. This phenomenon can be simply explained with Fig. 2(a). With increase of wavelength (λ_0), the value of the grating vector (G/K_0) increases and the ring R_1 moves to $+x$ direction and R_{-1} to $-x$ direction. Meanwhile, the diameter of the ring decreases (a longer wavelength is corresponding to a smaller SPR angle in the visible light band). These results demonstrate that the substrates proposed are more sensitive to wavelength than the SPCE from the thin Ag film deposited on a prism.

To simplify and qualitatively represent the relation between the emitting angle, wavelength, and period of the grating, we selected point P_{-1} in Fig. 2(b) as an example. This point is in the middle of the left fluorescence arc (related to ring R_{-1}). The azimuthal angle (ϕ) of this point is the same, 0 degree. The radial angle (β) of the fluorescence emitted can be derived from the known N.A. of the objective used in the BFPI [14,17]. From Figs. 2(b)–2(e), we

find that the point P_{-1} moves with change of the wavelength of the fluorescence, which means different β at different wavelengths. The emitting angle also can be calculated through the equation $\mathbf{K} = \mathbf{K}_{sp} \pm \mathbf{G} \cdot \mathbf{K}_{sp}$ can be derived simply through leakage radiation microscopy or the thickness of the PMMA film [16,17], and $\mathbf{G}$ can be calculated through the period of the gratings. Three gratings with periods 414, 505, and 614 nm are inscribed on the lower surface of the glass substrate. Ten bandpass filters with center wavelengths ranging from 550–640 nm (10 nm steps) are used to select the wavelengths of the fluorescence emission. As shown in Fig. 3, both the experimental (414 nm-E, 505 nm-E, and 614 nm-E) and calculated data (414 nm-C, 505 nm-C, and 614 nm-C) of the radial angle β are nearly consistent. The small discrepancy may be due to the error in deriving the emitting angle from the length between P_{-1} and the zero point (O) because the bright arc is a little thick.

The emitting angle β of a selected point P_{-1} changes with both the period and wavelength. At a fixed wavelength, the radial angle (β) increases with increase of period size. At a fixed period size, β decreases with increasing wavelength. This phenomenon provides a method to tune the emission angle of the fluorescence through grating periods. Because the grating is on the lower surface of the substrate and does not contact the samples, it will not disturb the optical properties of the fluorescent molecules.

Next, we have fabricated a structure in which the silver layer is replaced with a 1DPC. The 1DPC consists of alternate layers of low- (SiO_2 , 126 nm thickness) and high-dielectric layers (Si_3N_4 , 78 nm thickness). The top layer of the 1DPC is SiO_2 with a thickness of 152 nm. There are 14 layers in the structures as shown in Fig. 4(a). For this kind of a 1DPC structure, BSWs at around 600 nm can be generated on the surface of the top SiO_2 layer. The RhB-doped PMMA film is coated on the 1DPC, which is of the same thickness as that in Fig. 1(a). As described in a previous work, the coupling between the dye molecules and the BSWs will induce the BSWCE whose wave vector is also larger than that of light in vacuum [10]. To extract the BSWCE from the structure, a dielectric grating with a period of 614 nm close to the BSW wavelength was inscribed on the sub-surface of the glass. Figures 4(b)–4(e) show the corresponding BFP images of the fluorescence emission at four imaging wavelengths (610, 620, 630, and 640 nm). Similar to those shown in Fig. 2, a pair of bright arcs appear, which can also be ascribed to the diffraction of the BSWCE by the dielectric grating. The wave vector of the outcoupled fluorescence can be written as $\mathbf{K} = \mathbf{K}_{BSW} \pm n \cdot \mathbf{G}$, where $\mathbf{K}_{BSW}$ is the wave vector of the BSWCE inside the glass. With increase of wavelength, the left arc moves toward $+x$ direction and the right one toward $-x$ direction. The formation and movement of the bright arcs also can be explained as the case of SPCE. The emitting angle also can be tuned with wavelength and the period of the grating, similar to the results shown in Fig. 3 (data not shown).

There are two differences between Figs. 2 and 4. The first one is that the bright arcs related to the BSWCE are narrower than that related to the SPCE. This can be attributed to the lower loss of BSWs over that of SPs. Second, we can see another two pairs of fluorescence arcs in Fig. 4. As we know, for the 1DPC, along with the BSWCE, there also exists internal mode-coupled emission (IMCE), whose wavenumber is larger than that of the BSWCE [10]. They can interact with the grating, resulting in bright arcs with large diameters. The

intensity of the IMCE is weaker than that of the BSWCE [10], so the corresponding arcs in Fig. 4 are of different brightness.

Finally, the 1D grating shown in Fig. 1(a) is replaced by a 2D grating (or a square lattice) as shown in Fig. 5(a). The square lattice presents periodicity along x ; y ; u , and v directions. The periods are 500 nm (x , y) and 720 nm (u , v). Figure 5(b) shows four pairs of bright arcs at 620 nm wavelength appearing in the BFP image, which can be attributed to the periodic properties of the square lattice. As can be seen in Fig. 2(a), the two rings R_1 and R_{-1} separate from each other with increase in the grating period. This is the reason why the two pairs of bright arcs related to the periods along u and v directions are located at the boundary of the images. When compared with Fig. 2, the pattern has changed.

In conclusion, a new substrate with two decorated sides is proposed to direct fluorescence emission into free space, which has a small volume and ease of integration. Fabrication of this kind of a substrate can be low-cost and can be extended into many areas. The fabrication can be simplified further by separate manufacture of the 1DPC (or the Ag film) and the subsurface gratings as the distance between them does not need to be controlled rigorously. The planar and larger surface area of the substrate is favorable for high-throughput sensing or imaging. Excitation can be accomplished from the sample side of the structure, and the continuous metal film rejects the emission from the bulk sample not within a near-field distance from the metal. Excitation can also be accomplished from below the structure (in progress). Our experimental results have demonstrated that the emitting angles or pattern of the SWCE can be modulated easily with the patterns inscribed on the subsurface of the glass substrate. The light extracted with the dielectric patterns can also be used to enhance the outcoupling efficiency of organic light-emitting diodes [18] and in fluorescence-based super-resolution imaging [16,19,20].

Acknowledgments

This work was supported by the National Key Basic Research Program of China under grant nos. 2013CBA01703, 2012CB921900, and 2012CB922003; the National Natural Science Foundation of China under grant nos. 11374286, 61377053, and 61036005; and NIH Grants RO1HG002655 and RO1EB006521.

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Fig. 1.

(a) Schematic configuration of the substrate with two decorated sides. RhB molecules are doped in the PMMA film. (b) AFM image of the inscribed one-dimensional (1D) grating and the cross-section profile along the white line. The grating direction is along x axis. The period of the grating is 505 nm.

Fig. 2.

(a) Schematic graph of the wave vectors modulated by the 1D grating. K_0 is the wavenumber of the fluorescence in vacuum. K_x and K_y are the x - and y -components of the emitting wave vector ($\mathbf{K}$). The ring R_0 represents the wave vector of the SPCE, while R_1 ($\mathbf{K} = \mathbf{K}_{sp} + \mathbf{G}$) and R_{-1} ($\mathbf{K} = \mathbf{K}_{sp} - \mathbf{G}$) are the fluorescence Bloch waves generated by the grating. The small ring in the center (blue-dashed circle) represents the N.A. of the objective (0.7). (b)–(e) BFP images of the fluorescence at different wavelengths (560, 600, 630, and 64 nm). P_{-1} represents the selected points on the fluorescence arcs. The red elliptical rings in (c)–(e) show the cross areas of the bright arcs.

Fig. 3.

Change of radial angle (β) (for point P_{-1} in Fig. 2) with respect to change of the gratings' periods (414, 505, and 614 nm) and fluorescence wavelength (550–640 nm). Both experimental data estimated from the BFP images and calculation results from the equation are plotted.

Fig. 4.

BFP images of the coupled fluorescence from the 1DPC substrate with the grating on the subsurface of the glass. (a) Schematic of the 1DPC, which is used instead of Ag layer in Fig. 1(a). The fluorescence wavelengths are (b) 610 nm, (c) 620 nm, (d) 630 nm, and (e) 640 nm. The period of the 1D grating is 614 nm. The ring labeled with the white-dashed circle (b) represents the N.A. of the collection objective (0.70).

Fig. 5. (a) AFM image of the inscribed two-dimensional (2D) grating. There is periodicity along x , y , u , and v directions. The white scale bar is $1\ \mu\text{m}$. (b) Corresponding BFP image of the fluorescence. The ring labeled with the white dashed circle represents the N.A. of the collection objective (0.70).