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Surface Plasmon Coupled Emission Enhancement with Nanoparticles in the Metal Layer

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***Abstract*—We show that it is possible to enhance surface plasmon coupled emission (SPCE) intensity by $\sim$55% using two dimensional periodic nanoparticle arrays in the glass-metal layer interface of an SPCE structure. The nanoparticles act as coupled nanoantennas. With optimized dimensions and periodicity, the nanoparticle arrays resonate and re-emit intensified SPCE. In this work, two types of nanoparticles such as cuboids and hemispheres are used in the arrays. The optimized dimensions and periodicity of the nanoparticle arrays that lead to enhanced intensity are calculated using the particle swarm algorithm. The increased SPCE intensity will be helpful for using SPCE as a biomolecule detection scheme.**

I. Introduction

Fluorophores placed near a thin metal layer introduce surface plasmon resonance (SPR) in the metal, which comes out in the other side of the metal layer as a cone-shaped light at a sharply defined angle. This directional emission is called surface plasmon coupled emission (SPCE) [1]. As SPCE is a potential microscopy and bio-sensing technique, it has received significant theoretical and experimental attention. Attempts have been made to increase the intensity of SPCE, as it would lead to more sensitive detection schemes. SPCE intensity is an involved process depending on many factors. The environment in which the fluorophores are located affects their emission significantly. To increase SPCE, the usual approach is to enhance the fluorescence by modifying the host environment. Metal nanoparticles suspended in the host medium are found to increase the fluorophore emission by about 35 folds [2]. Grating based couplers have been used instead of traditional excitation configurations and found to enhance SPCE sensitivity [3]. By reflecting the SPCE ring onto a single point using a conical mirror around the hemispherical prism, collection efficiency of SPCE can be increased and nearly 500 folds enhancement over the free space signal has been shown [4]. Recently, carbon nanodots are found to increase the luminescence by about 1000 folds, setting the benchmark for all fluorescence enhancement techniques [5]. The increase in the power coupling ratio has been found by using metal bi-layers of silver and gold [6].

In this work, we use two dimensional periodic nanoparticle arrays in the radiative side of the metal layer and show that this scheme increases the intensity of SPCE by $\sim$55% compared to that of a typical SPCE structure. We study the dynamics of a structure with metal nanoparticle arrays by full-field finite difference time domain (FDTD) simulations. The nanoparticles act as antennas and reradiate with the SPCE emission. The advantage of this strategy is that it can be used in addition to other enhancement schemes so that the SPCE intensity can be further enhanced.

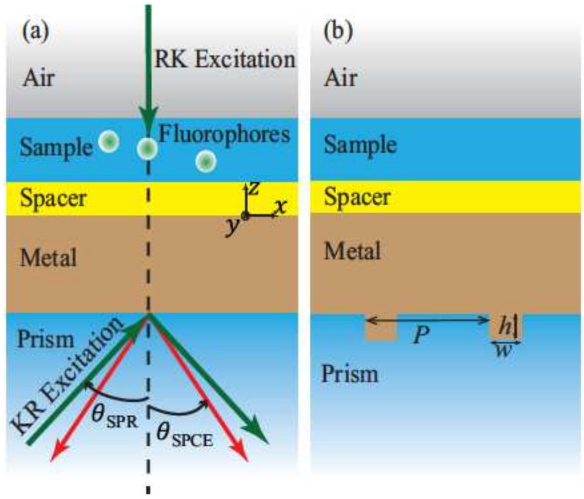

Fig. 1. (a) Typical structure used for SPCE generation and (b) Metal nanoantenna attached structure.

II. A typical SPCE structure

Figure 1(a) shows a typical structure used for SPCE generation. A 50 nm silver (Ag) layer is deposited on a hemispherical glass (SiO_2) prism. A 10 nm SiO_2 layer is deposited on the top of metal layer to act as a spacer between the metal and the molecules. The sample layer containing the fluorophores are placed above the spacer layer. In the typical setup, we assume that a poly-vinyl alcohol (PVA) layer of 30 nm thickness is used as the sample layer and Rhodamine B is used as the fluorophore. Rhodamine B absorbs maximum excitation light at wavelength 545 nm and emits maximum energy at wavelength 565 nm.

III. Factors Controlling SPCE Intensity

Fluorophores have distinct frequencies for excitation and emission. The emission wavelength is greater than the emission wavelength by an amount called stokes shift. Two frequently implemented excitation configurations for fluorophores are Kretschmann (KR) and reverse Kretschmann

(RK) schemes. In RK configuration, the fluorophores are excited directly from the sample layer side by shining excitation frequency light at normal incidence. The excitation light is absorbed as it passes through the fluorophore-infused sample layer. In KR configuration, excitation is done from prism side. It is an attenuated total reflection mode, where light of excitation frequency incident from the prism side at the SPR angle (θ_{SPR}) creates SPR in the sample layer side. The horizontal and vertical components of the evanescent field decay exponentially inside air or water. As light is absorbed by the surface plasmons, a reflectance minimum is found at θ_{SPR} for p-polarized light. For any of these configurations, this field excites the fluorophores, and the fluorophores emit at their emission frequency. This emission interacts with the metal layer and comes out as SPCE in an angle called the SPCE angle (θ_{SPCE}). As SPCE is the reverse process of SPR, reflectance measured from prism side at emission frequency is minimum at θ_{SPCE}. The angles θ_{SPR} and θ_{SPCE} are different, because θ_{SPR} is the angle for reflectance minimum for excitation and θ_{SPCE} is the angle for emission from the metal surface.

The SPCE intensity from a single fluorophore in the sample layer depends primarily on two factors: The local excitation electric field and distance dependent coupling ratio. The SPCE intensity from molecules fluorescing the same energy but situated at different elevations from the metal layer will not be the same due to the distance-dependent power coupling ratio. Fluorescence is also proportional to the excitation field intensity, as well as very sensitive to the host medium environment and the presence of any neighboring nanoantenna. The molecular dipole moment of the fluorophores is in the direction of its local excitation field. The coupling ratio is dependent on the distance from the metal layer and the direction of the moment [7]. If the distance from the metal layer is less than 10 nm, a very small amount of power is coupled as a result of fluoroscence quenching by the metal layer.

In this work, we are mainly interested in increasing the SPCE intensity by the improvement of power coupling efficiency. As horizontal dipoles contribute a very small amount of power to SPCE, we do not discuss the intensity improvement due to the horizontal dipoles [7]. As the power coupling changes significantly with the position of the dipole, for fair comparison with the typical SPCE structure, the dipole is kept at the same height from the metal layer in all cases.

IV. Metal Cuboids in the Prism Side

We add metal cuboids to the metal layer in the metal-prism interface and study the effects on the properties of SPCE. The motivation for this patterning is that the plasmon mode in prism-metal interface is radiative, and cuboids will act as nanoantennas and will enhance the emission intensity. The schematic diagram of the SPCE structure with cuboid nanoantennas is shown in Fig. 1(b). The performance parameters discussed below, e.g., peak SPCE intensity and coupled power in prism are normalized by that in typical structure without any nanoparticle array.

The variables of the metal cuboid are the width of the base (w), height (h), and periodicity (P) of the two dimensional grid. For $h = 10$ nm and $P = 750$ nm, we show the peak SPCE intensity and coupled power for a vertical dipole for different w in Figs. 2(a) and 2(b). We note that the smaller w leads to an increase in the peak intensity. It is possible to get 55% increase in peak intensity by using 50 nm wide nanocubes. The power coupled in the prism side with the nanoantenna also increases from that of a typical SPCE structure. The maxima in peak SPCE intensity and coupled power curves occur at the same w.

Figures 2(c) and 2(d) show peak SPCE intensity and coupled power for different periodicity of the two dimensional cuboid array with $w = 50$ nm and $h = 10$ nm. It is noted that for any periodicity, the peak SPCE intensity of the structure with nanoantennas is greater than that of the typical SPCE structure. Also periodicity at multiples of 750 nm gives maximum intensity and coupled power. The coupled power increases by a maximum of $\sim 10\%$. Figures 2(e) and 2(f) show peak SPCE intensity and coupled power with cuboid height for $w = 50$ nm and $P = 750$ nm. We note that both SPCE intensity and coupled power have a maximum value when $h = 10$ nm. The cuboid size and periodicity for maximum peak intensity have been optimized by particle swarm optimization technique. This leads to $w = 50$ nm, $h = 10$ nm, and $P = 750$ nm.

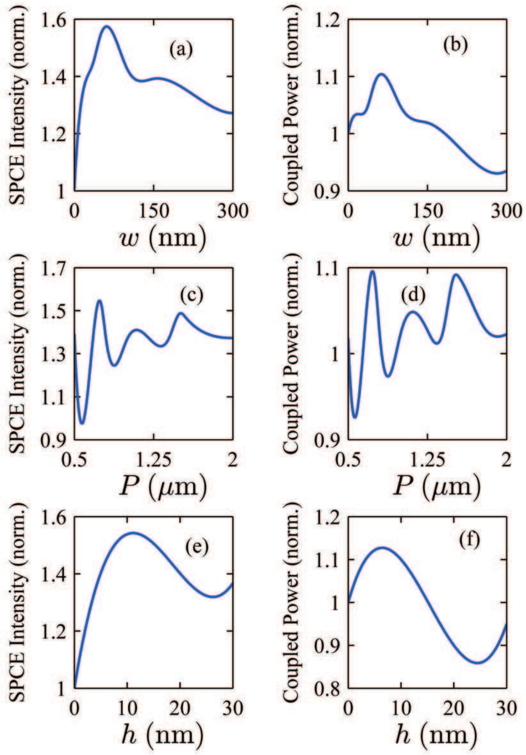

Fig. 2. Peak SPCE intensity and coupled power, respectively, with the variation of cuboid nanoantenna (a, b) width (w), (c, d) periodicity (P), and (e, f) height (h).

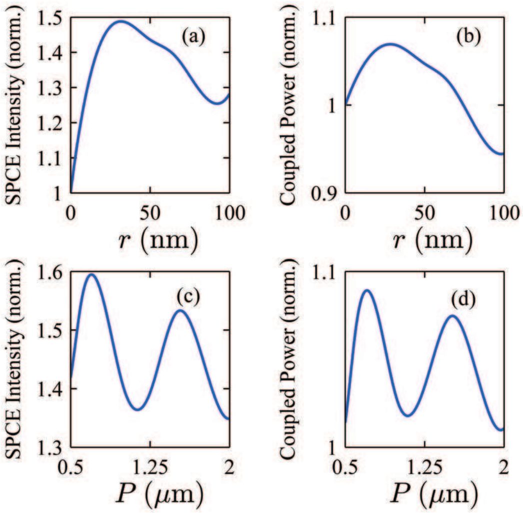

Fig. 3. Peak SPCE intensity and coupled power, respectively, with the variation of hemisphere nanoantenna (a, b) radius (r) and (c, d) periodicity (P).

V. Metal Hemispheres in the Prism Side

We now use two dimensional periodic metal hemisphere arrays in the metal-prism interface. The flat side of the hemispheres are attached to the metal layer. We vary the radius (r) of the hemispheres and the periodicity (P) of the array to increase SPCE intensity and coupled power.

Peak SPCE intensity and coupled power with hemispheres of different radius (r) are shown in Figs. 3(a) and 3(b). The periodicity of the array is kept fixed at $P = 750$ nm. We note that the SPCE intensity increases by $\sim$50% in case of 30 nm radius hemispheres. Figures 3(c) and 3(d) show peak SPCE intensity and coupled power for different periodicity (P) of the hemisphere array. An increase of $\sim$60% is found for a periodicity of 600 nm. The SPCE intensity also increases when the periodicity is multiples of 600 nm. Using particle swarm optimization, we find that the maximum SPCE can be found for hemispherical nanoparticles of radius $r = 30$ nm and periodicity $P = 600$ nm.

Figures 4(b) and 4(c) show the electric field profiles in the structure with cubes and hemispheres. Compared to the fields in the typical structure in Fig. 4(a), it is evident that both the cubes and hemispheres act as nanoantennas. For both cuboid and hemisphere arrays, the SPCE intensity becomes flat for higher periodicity. Because closely spaced nanoparticles interact by creating coupled modes, and as they move further and further, they individually start to act as a single nanoantenna. Therefore, when the periodicity increases, SPCE intensity becomes independent of periodicity. Also, hemisphere-shaped nanoparticles are capable of far reaching coupled modes than cuboids.

VI. Conclusion

We used metal nanoparticles in the metal layer to modify the behavior of SPCE and found enhanced performance in radiation intensity than that in the traditional plane metal layer SPCE. The results remain qualitatively the same when metal nanoparticles of different shapes such as cuboids and hemispheres are used.

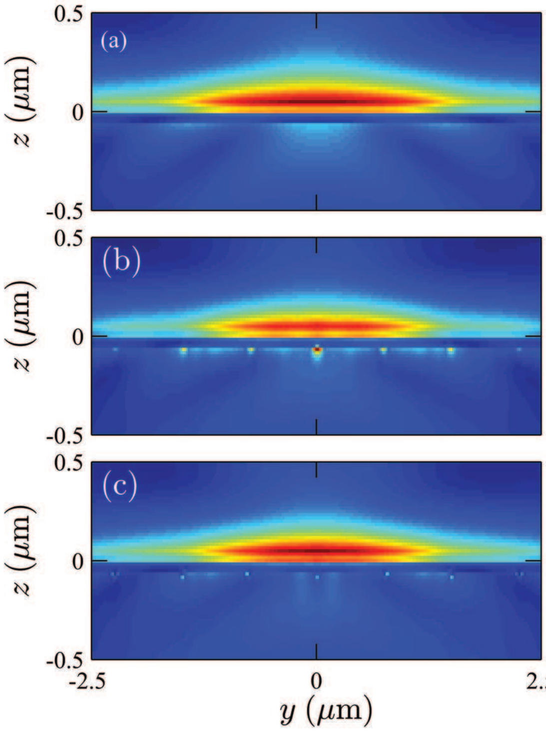

Fig. 4. Electric field profile in (a) typical structure, (b) with cuboid array, and (c) with hemisphere array.

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