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A polarization-insensitive plasmonic SECARS substrate with multiple hot spots

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ABSTRACT

A plasmonic substrate providing high, reproducible and stable Raman signals should be highly desirable for the development of surface enhanced coherent anti-Stokes Raman spectroscopy (SECARS). In this work, we theoretically present a design of SECARS substrate consisting of five different-sized gold nanodisks and investigate its enhancement properties under different excitation polarizations by using finite element method. The numerical results reveal that the pentamer supports a polarization-independent Fano-resonant scattering spectrum due to its symmetric geometrical arrangement. Multiple electromagnetic hot spots produced by the Fano resonance are overlapped spatially at three characteristic frequencies involved in SECARS process. Consequently, the theoretically estimated overall enhancement factor (EF) of SECARS nearly keeps the same order of magnitude up to $\sim 10^{14}$ for any horizontally excitation polarizations, and the relative root mean square error of the logarithm of the overall EF ($\mathrm{Log}_{10}\mathrm{EF}$) is less than 2%. Giant and polarization-insensitive SECARS enhancements enable the pentamer structure to be promising for plasmonic substrates in SECARS as well as other enhanced nonlinear optical process.

1. Introduction

Coherent anti-Stokes Raman scattering (CARS) is a well-known method in molecular identification and vibrational bio-imaging by means of their vibrational fingerprints [1–4]. In CARS process, the incident two beams including the pump (ω_p) and Stokes (ω_s) beams interact coherently through the third-order susceptibility of identified objects, thereby generating a spectrally blue-shifted anti-Stokes signal ($\omega_{as} = 2\omega_p - \omega_s$) with the intensity dependence of a third-order nonlinear optical process as follows [5,6],

$$I_{CARS} \propto \left|\chi^{(3)}\right|^2 I_p^2 I_s \tag{1}$$

where $\chi^{(3)}$ is the third-order susceptibility of identified objects, I_p and I_s are the intensities of the incident pump and Stokes beams, respectively. Usually, the CARS is orders of magnitude stronger than spontaneous Raman scattering due to its higher order dependence on incident intensities of two beams. However, the CARS sensitivity is still not enough for identifying or imaging molecules present in extremely low concentrations especially for bio-chemical applications as optical labels and drug-delivery vehicles.

The sensitivity of CARS can be increased by introducing an appropriately plasmonic substrate. Similar to surface enhanced Raman scattering (SERS) [7–11], the surface-enhanced CARS (SECARS) is achieved mainly via the localized electromagnetic (EM) field of the excited surface plasmon resonances generated on plasmonic nanostructured substrates [12–16]. For plasmonic SECARS substrates, a few studies with colloidal gold or silver nanoparticles [13,14], nanostructured metallic surfaces [5,15] and metallic nanoparticles arrays with single- or multi-layered constructions [16] have been made and a $10 \sim 10^7$ times signal enhancement relative to conventional CARS is usually observed in previous work.

Recently, Halas et al. proposed a Fano-resonant quadrumer as SECARS substrates and experimentally achieved an enhancement factor (EF) of SECARS signal up to $\sim 10^{11}$ due to a highly confined EM hot spot produced by the Fano resonance [17,18]. Then, He et al. theoretically achieved an EF of $\sim 10^{13}$ in a specially-designed Fano-resonant SECARS substrate by means of the spatially overlapped hot spot at three characteristic frequencies (ω_p, ω_s

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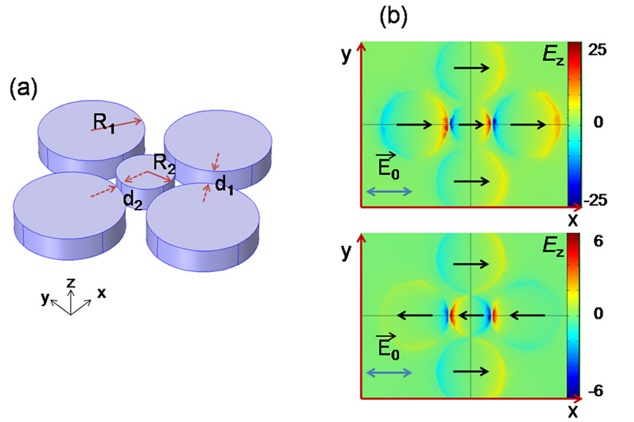

Fig. 1. (a) Sketch of the disk pentamer with the defined parameters and coordinate axis. (b) The E_z distributions on the pentamer's surface. The black arrow denotes a dipolar resonance in each disk.

and ω_{as}) involved in SECARS process [19]. Based on the idea above, Wang et al. recently designed a multi-band resonant SECARS substrate and theoretically obtained a higher SECARS EF up to $\sim 10^{16}$ [20]. In these works, the SECARS enhancement properties should be sensitive on the polarization of incident light due to the asymmetric geometrical arrangements of plasmonic substrates, consequently presenting a challenge for a flexible control of the excitation polarizations in practical SECARS implementations. Generally, any change in the direction of excitation polarization leads to a change in both intensity and spatial distribution of EM hot spots. Therefore, producing a polarization-insensitive SECARS enhancement from the polarization-dependent near-fields remains a challenging issue.

In this work, we theoretically present a design of the Fano-resonant plasmonic pentamer and investigate its SECARS enhancement properties under various excitation polarizations by using the finite element method (FEM). It is found that the pentamer supports a polarization-insensitive Fano-resonant scattering spectrum due to its rotational symmetry in structural configuration. Multiple EM hot spots by the Fano resonance are located at the same spatial positions for three different frequencies (ω_p, ω_s and ω_{as}). More importantly, the theoretically estimated overall SECARS EF nearly keeps the same order of magnitude up to $\sim 10^{14}$ for any horizontally excitation polarizations. The polarization-insensitive SECARS enhancement brings in more flexibility for polarization control in practical implementations, enabling the pentamer to be promising for applications in SECARS substrates as well as other enhanced nonlinear optical process.

2. Structure design and simulation method

Fig. 1(a) gives the schematic of the proposed pentamer with the defined geometrical parameters, which consists of five metallic disks. The four outer-disks have the same size with the radius of R_1. The center disk has the radius of R_2. The gap distance among these four outer-disks is defined as d_1. The gap distance between the center and outer disks is defined as d_2. These four parameters are not fully independent variables, one of which can be determined by other three parameters according to the formula below.

$$R_2 = \frac{2R_1 + d_1}{\sqrt{2}} - R_1 - d_2 \qquad (2)$$

In addition, these five nanodisks have the same thickness defined as h. Gold is chosen to be the disk's metallic material due to its relatively low intrinsic loss in the considered spectral range.

Numerical simulations were carried out based on the finite element method (FEM) by employing the commercial software of COMSOL Multiphysics 5.3a with wave-optics module. In the simulation model, an isolated pentamer surrounded by a spherical Perfect Matched Layer (PML) was adopted, where PML with a nearly half-wavelength thickness was used to avoid spurious boundary reflections. For simplifying calculations, the pentamer has no standing substrates and the surrounding medium was assumed to be air with the refractive index of $n = 1$. The optical constants of gold were taken from the experimental data provided by Johnson and Christy [21]. Notice that introducing a dielectric substrate would not modify the plasmonic properties of the pentamer, but only cause a red-shift of resonant wavelengths accompanied with a slight increase in resonant linewidth due to the dielectric screening [22,23].

For all simulations, the pentamer structure was illuminated normally by plane wave. The scattering cross section was calculated by integrating the far field in the framework of the scattered formulation. The scattering efficiency (Q_{scat}), defined by the scattering cross section divided by πr_{eff}^2 (r_{eff} is the effective radius of the pentamer by satisfying the relation of $V = 4\pi r_{eff}^3/3$, where V is the pentamer's volume), was plotted as a dimensionless quantity in the scattering spectrum. In addition, the size of the FEM meshgrid, which slightly influences the value of the calculated SECARS EF, was chosen to be always consistent in all calculations for providing a relatively fair comparison.

3. Results and discussions

3.1. *Fano resonance and spectral tunability*

Plasmonic oligomers supporting Fano resonance have attracted great interest in recent years due to their unique optical properties, the remarkable potential in LSPR sensing, the mature fabrication technology and being an important model system to understand the nature of electromagnetic coupling between adjacent metallic nanoparticles [24–27]. Fano resonance in plasmonic oligomers usually origins from the destructive interference between a superradiant (bright) and a subradiant (dark) plasmon resonant mode [22,26,27]. To identify the nature of excited Fano-resonant mode in the proposed pentamer, a normal component of electric-field vector (that is, E_z) is given in x–y plane, as shown in Fig. 1(b). In general, one has to study the mode in all three spatial dimensions. However, it has proven sufficient to only investigate the normal components of the near fields (including H_z) which nearly fully reveal the mode character [28,29]. Obviously, it is found in Fig. 1(b) that the E_z distribution of each metallic disk exhibits a clear dipolar pattern along the direction parallel to the excitation polarization (along the x-axis, denoted by the black arrows). These dipolar resonances in each disk hybridizes in-phase (see the top picture in Fig. 1(b)) and out-of-phase (see the bottom picture in Fig. 1(b)), forming the superradiant and subradiant mode of Fano resonance, respectively.

Fano resonance can be tuned to various wavelengths by varying the geometrical parameters of the pentamer, as shown in Fig. 2. It is found in Fig. 2(a) that with increasing R_1 from 95 to 125 nm, a significant broadening in the profile line-width of the shoulders accompanied with an obvious red-shift of the dip can be observed in the scattering spectrum of the Fano resonance. This is mainly attributed to the increased plasmonic coupling among the outer-disks and the enlarged radiation damping by the increased size in the outer-disks. When changing d_1 from 20 to 50 nm, it is shown in Fig. 2(b) that the Fano modulation depth becomes more and more remarkable and the Fano dip shifts to red slightly. A weakened coupling among the outer adjacent disks caused by the increased d_1 leads to a decrease of the net dipole moment of the

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