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An Easy-to-Integrate Embedded Cascade Microspheres for Efficient Local Field Enhancement

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Abstract. Surface-enhanced Raman spectroscopy (SERS) is a non-destructive and ultra-sensitive tool for optical trace detection. However, the complex manufacturing process hinders the application of SERS. For this reason, an embedded cascaded microsphere structure is proposed in this paper, which is easy to integrate and process. Such a structure is composed of large microspheres and small microspheres with micropores cascaded. More importantly, the micropore diameter at the bottom of the large microspheres is the same as that of the small microspheres. Through the finite element analysis, it is concluded that compared with a single microsphere, the photonic nanojet generated by this structure is stronger, which can achieve local electric field enhancement more effectively. Meanwhile, different materials of the cascade microspheres used in the embedded cascade have various reinforcement effects. In addition, compared with the manual physical alignment of two microspheres in the traditional cascade structure, this structure is easier to process and integrate, convenient for future integrated applications.

Keywords: Surface-Enhanced Raman Spectroscopy, Microspheres, Photonic Nanojet, Cascade Microspheres.

1. Introduction

Raman spectroscopy is a vital spectroscopic technology to analyze the molecular structure and recognize the chemical fingerprint of molecules through the inelastic scattering of monochromatic light, which has significant applications in life science, material science, analytical science, pharmacy and other fields [1]. However, Raman scattering is extremely weak and difficult to detect due to the small interaction cross section, so it is difficult to be applied in practice [2]. Hence, the researchers developed surface-enhanced Raman (SERS) [3] to increase the Raman scattering signal of substances, making it easier to detect and analyze. Surface-enhanced Raman scattering is mainly based on two mechanisms, including electromagnetic enhancement and chemical enhancement. Due to the low enhancement factor of chemical enhancement, electromagnetic enhancement has always been the main enhancement mechanism of SERS. In the past, the electromagnetic enhancement mainly originated from the localized surface plasmon resonance (LSPR) excited by the external electric field acting on metal nanoparticles [4]. In recent years, to obtain low-cost Raman-enhanced substrates, researchers have paid attention to non-LSSR Raman-enhanced substrates. Besides, dielectric microspheres have been a popular topic in Raman-enhanced research for their unique light field regulation properties [5].

In 2007, the team of Lu Yongfeng first applied dielectric microspheres to Raman detection, which obtained strong Raman enhancement signals and concluded that Raman enhancement originated from PNJ of dielectric microspheres [6]. Subsequently, Bisht et al. conducted an in-depth study of Raman enhancement rate. They analyzed its relationship with the numerical aperture of the objective lens, pump wavelength, single microsphere size, and refractive index. According to their results, the Raman enhancement rate mainly depends on the matching between the laser spot size and the microsphere diameter. In particular, they found that the enhancement rate of Raman scattering can be significantly improved using short-wavelength pump light [7]. After developing a SERS substrate based on a 2D silica particle array for the detection of crystal violet (CV), Lombardi et al. found that when CV molecules are attached to the surface of the particle array, the enhancement effect of SERS can reach a significant 104 magnitude [8]. In 2022, Mengyuan Wang et al. realized a facile flexible coupling SERS substrate consisting of an array of dielectric microsphere cavities and random gold nanoparticles covered on polydimethylsiloxane films, which enables huge Raman enhancement [9]. The large difference in Raman enhancement factors between experimental and theoretical microsphere-enhanced LSPR is attributed to the collimation of near-field hotspots by the excitation light, where microsphere cavity focusing is essential for effective energy localization of nanoparticles. Thus, it is crucial to understand the localization of excitation energy and develop an autocollimated hybrid Raman enhancement, so as to realize ultra-sensitive Raman trace detection on microsphere-coupled SERS substrates.

An embedded cascaded microsphere is proposed in this study, which consists of large microspheres and small microspheres with holes. According to the results of numerical simulation analysis, compared with a single microsphere, the local electromagnetic enhancement is higher; compared with the ordinary cascade of large and small microspheres, the embedded cascade can ensure the electromagnetic enhancement and is easier to integrate and align. This study provides a convenient method for the design and fabrication of cost-effective hybrid Raman enhancement for spectral detection, which is conducive to the development of new high-efficiency hybrid nanophotonic devices in the future.

1.1 Scheme

In this study, the photonic nanojet (PNJ) of dielectric microspheres is numerically simulated and analyzed by the finite element method (FEM) of multi-physical field simulation using the COMSOL. The monochromatic light with a wavelength of 500nm propagates vertically from top to bottom through the medium sphere surrounded by air. At the same time, a perfect matching layer is used on the boundary to eliminate the influence of stray light at the boundary. Finally, the electric field distribution diagram of PNJ is obtained.

1.2 Length of PNJ

For three-dimensional microspheres, the length of PNJ will increase with the increasing microsphere diameter, while the maximum strength will hardly change [10]. In order to investigate the length of PNJ and the diameter of dielectric microspheres, we made numerical simulations for the length of PNJ of a single SiO_2 microsphere with different diameters of the same material, which obtained the simulation results in Figure 1. Figure 1 (a)-(e) shows the simulation results when the microsphere diameters are 1μm, 3μm, 5μm, 7μm, and 9μm, respectively. Figure (f) shows the fitting line between the jet length and the sphere diameter, indicating that the jet length has a good linear relationship with the diameter of the medium sphere in a certain range. This may be due to the sharp oscillation in the intensity and length of the jet at the radius of the resonant particle for the formation of a resonant jet when one or more whispering gallery modes (high-mass Michaelis resonance) are excited inside the particle. This linear relationship has a certain guiding significance for the subsequent design of related experiments to select the appropriate diameter of the medium sphere.

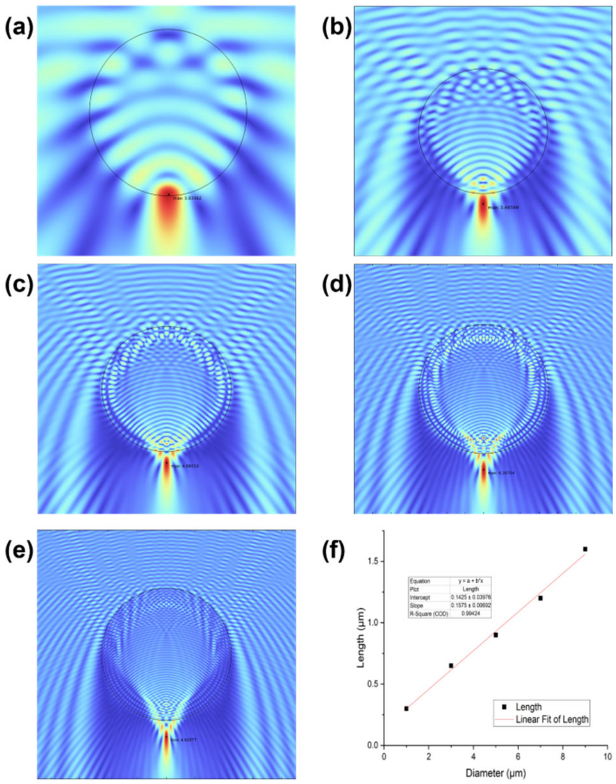

Figure 1 Simulation Results of Length of PNJ and Different Dielectric Microsphere Diameters, with Dielectric Microsphere Diameters as (a) 1μm, (b) 3μm, (c) 5μm, (d) 7μm, (e) 9μm Respectively. (f) is a Linear Fitting Plot

1.3 Enhancement Effect of Cascade Microspheres

The PNJ produced by a medium sphere can amplify the light intensity within a certain range. This amplification is similar to the focusing effect of a lens. Considering that the lens can be cascaded, the PNJ produced by a microsphere may also have cascaded amplification. To this end, we used a 5μm sphere as the first stage of the cascade, and took microspheres with different diameters for the second-stage cascade microspheres, so as to analyze the relationship between the maximum field intensity enhancement (B_{max}) of PNJ and the diameter of the cascade microspheres. In particular, we will align the centers of the two microspheres and stick them together for the sake of study. Figures (a)-(c) are the simulation results of the cascade spheres with diameters of 0.4μm, 0.2μm, and 0.8μm, respectively. Figure (d) is a histogram of the diameter and the maximum field intensity of the PNJ. When the diameter is 0.4μm, the electric field intensity is significantly higher than that of other microspheres. The width of the PNJ produced by the first-stage microsphere is 0.4μm. The general situation of PNJ is that the parallel light incident on the upper surface of the sphere is similar only when the diameter of the second-stage cascade sphere is smaller than the width of the PNJ. Otherwise, once it is larger than this width, the distribution of the PNJ in the sphere will be greatly reduced, no longer similar to the general situation. However, when the diameter is smaller than the width of the PNJ, the two-stage cascade sphere cannot receive all the energy in the first-stage PNJ region, so the maximum field intensity enhancement decreases. In summary, when the diameter of the secondary cascade microsphere is equal to the width of the PNJ of the primary microsphere, the maximum field intensity enhancement can be obtained, and the intensity enhancement rate depends on the matching of the spot size and the diameter of the microsphere.

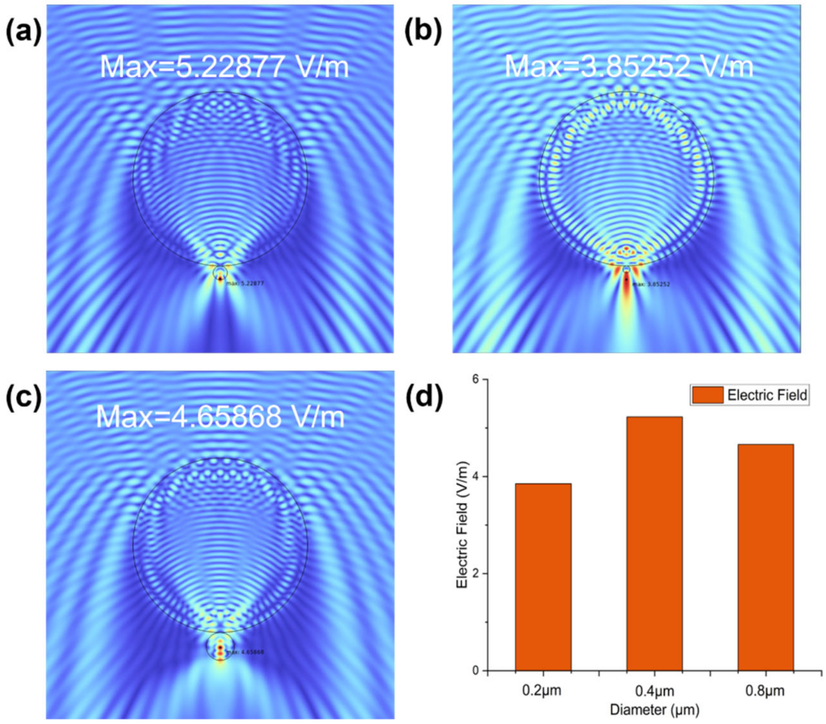

Figure 2 Simulation Results of Different Diameters and Electric Field Strengths of the Cascade Microspheres. The Diameters of the Cascade Microspheres are: (a) 0.4μm, (b) 0.2μm, and (c) 0.8μm Respectively. (d) Diagram of Relation Between Electric Field Intensity and Diameter of Cascade Microspheres

1.4 Sphere Position in Cascade Microsphere Structure

It is shown that photonic nanojets generated by the high refractive index microspheres are non-decaying and non-resonant beams, which are focused on the sub-diffraction transverse size (~λ/3) and can travel a distance over the light wavelength, so the specific position of the small spheres in the cascade will affect the electromagnetic strength of the cascade microspheres. In this study, spheres with diameters of 5μm and 0.2μm are selected as the first and second stages of the cascade respectively, and the positions of the second-stage microspheres are numerically simulated, with the simulation results shown in Figure 3. Figure 3 (a)-(h) shows the simulation results of the distance between the two spheres of 0.1μm, 0.2μm, 0.3μm, 0.4μm, 0.5μm, 0.6μm, 0.7μm, and 0.8μm, respectively. Figure (i) shows the relationship between the distance of the second-stage microsphere and the first-stage microsphere and the electromagnetic field intensity. In this figure, when the distance between two spheres is 0.4μm, the cascade microspheres have the largest electric field intensity. At this time, the center of the second-stage microsphere is just at the PNJ focus of the first-stage microsphere, and the electric field at this position is the most intense. A dimensional second-stage microsphere can generate the largest electric field, with the electric field intensity reaching 5.58 V/m. In addition, we believe that the PNJ beams generated by microspheres are similar to Gaussian beams, with the smallest divergence angle and the strongest intensity at the beam waist, which are closest to parallel light. Therefore, when the microspheres are placed at this time, the cascade microspheres can obtain the largest electric field intensity.

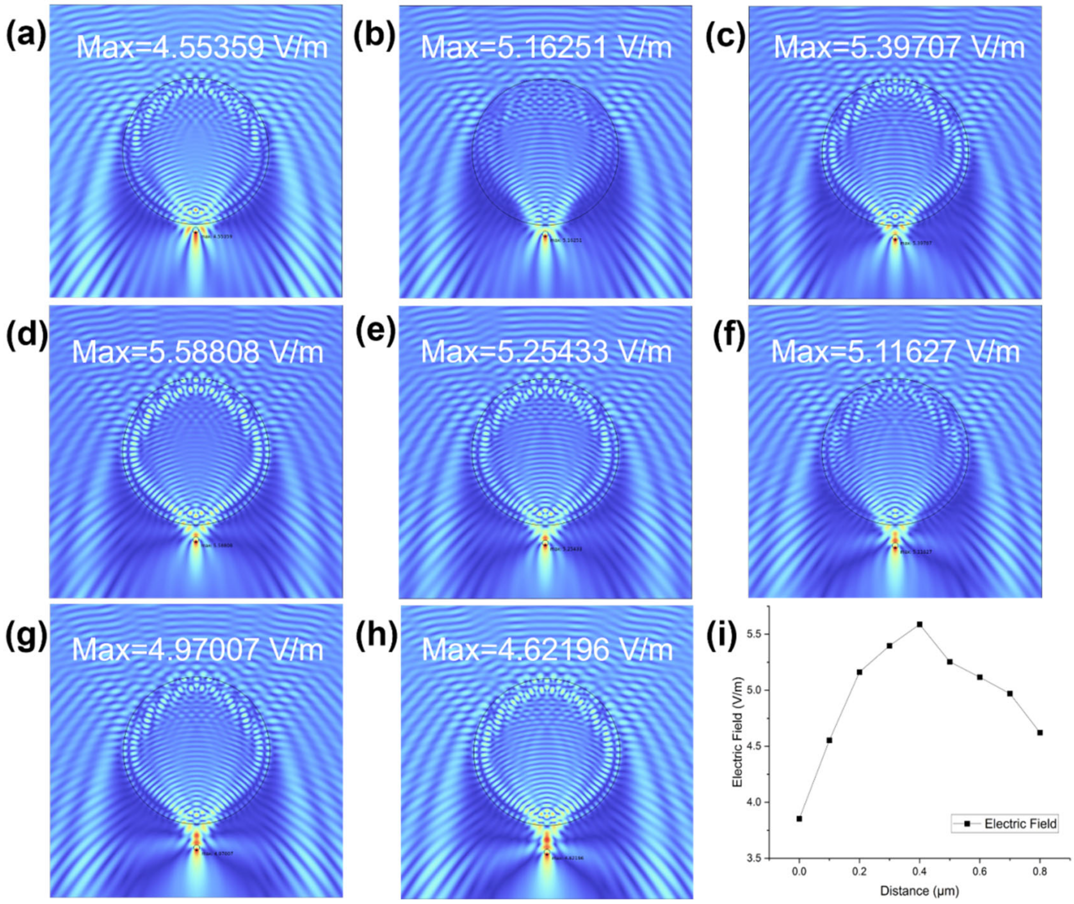

Figure 3 Simulation Results of Position of the Second-Stage Microsphere and the Maximum Field Intensity. The Positions of the Two Spheres are: (a) 0.1μm, (b) 0.2μm, (c) 0.3μm, (d) 0.4μm, (e) 0.5 μm, (f) 0.6μm, (g) 0.7μm, (h) 0.8μm, (i) The Maximum Field Intensity Corresponding to the Position of the Second-Stage Microsphere

1.5 Design of Embedded Cascade Structure

In LSPR technology [11], manufacturing of suitable nanostructures mainly aims to synthesize metal nanostructures with controllable shapes and sizes or to prepare nanostructures on the surface of materials through chemical methods. The former is difficult to precisely control the spacing of nanoparticles, and the latter is expensive to manufacture. For dielectric microspheres, considering their shape and microsphere diameter, it is difficult to stack two microspheres to achieve alignment in technology, which is not conducive to its application. To solve this problem, based on the previous research, we designed an embedded cascade microsphere structure, which facilitates the positioning and assembly of the two spheres. Based on the previous simulation analysis structure, to prove that the cascade structure has a higher local electric field enhancement than a single microsphere, we conducted a numerical simulation on the embedded cascade microsphere structure. In this simulation, we take the microsphere with a diameter of 5μm as the first stage and the microsphere with a diameter of 0.8μm as the second stage, with the simulation results obtained in Figure 4. (a) the electric field intensity of a single microsphere; (b) the B_{max} of the electric field intensity of the cascade microspheres. It can be seen that the electric field intensity generated by the cascade microsphere structure is 6.056 V/m, which is much greater than that of a single microsphere. In the future, in terms of process design, we will consider using femtosecond laser ablation to ablate a small hole under the first-stage microsphere, and embed the second-stage microsphere into it. Finally, the cascade structure of the embedded microsphere processing is completed.

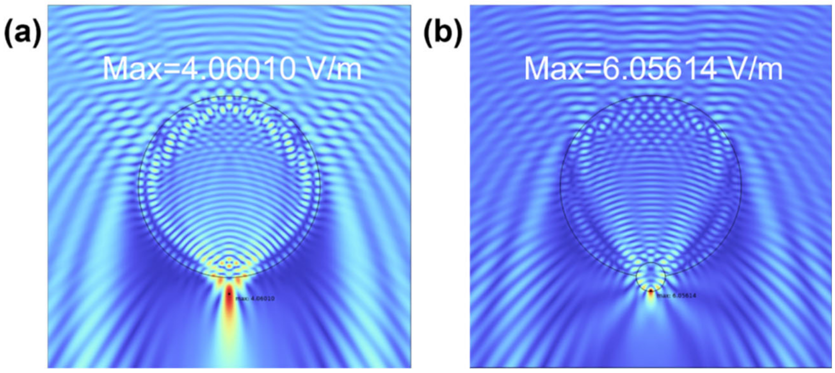

Figure 4 Comparison of the Enhancement of the Maximum Field Intensity Between the First-Stage PNJ and the Embedded Cascade Microsphere PNJ

1.6 Cascade of Small Spheres of Different Materials

The materials of the dielectric microspheres mentioned above are all SiO_2, and the materials commonly used in PNJ include fused silica (FS), polystyrene (PS), polymethylmethacrylate (PMMA), etc. [12]. Hence, it is urgent to verify the effects of cascade spheres made of these materials. In this study, the reinforcement effect of the second-stage microspheres of different materials used in the embedded cascade microsphere structure is numerically simulated and analyzed for different materials, with the simulation results in Figure 5. Figures (a)-(d) are the simulation results of spheres made of SiO_2, FS, PS, and PMMA respectively. Figure (e) is the electric field intensity generated by the second-stage microspheres cascaded with different materials. In these figures, the electric field intensity of the sphere made of FS is the smallest, and that of the microsphere made of PS is the largest. However, no matter which material is made of the sphere, a better cascading effect can be obtained by this method.

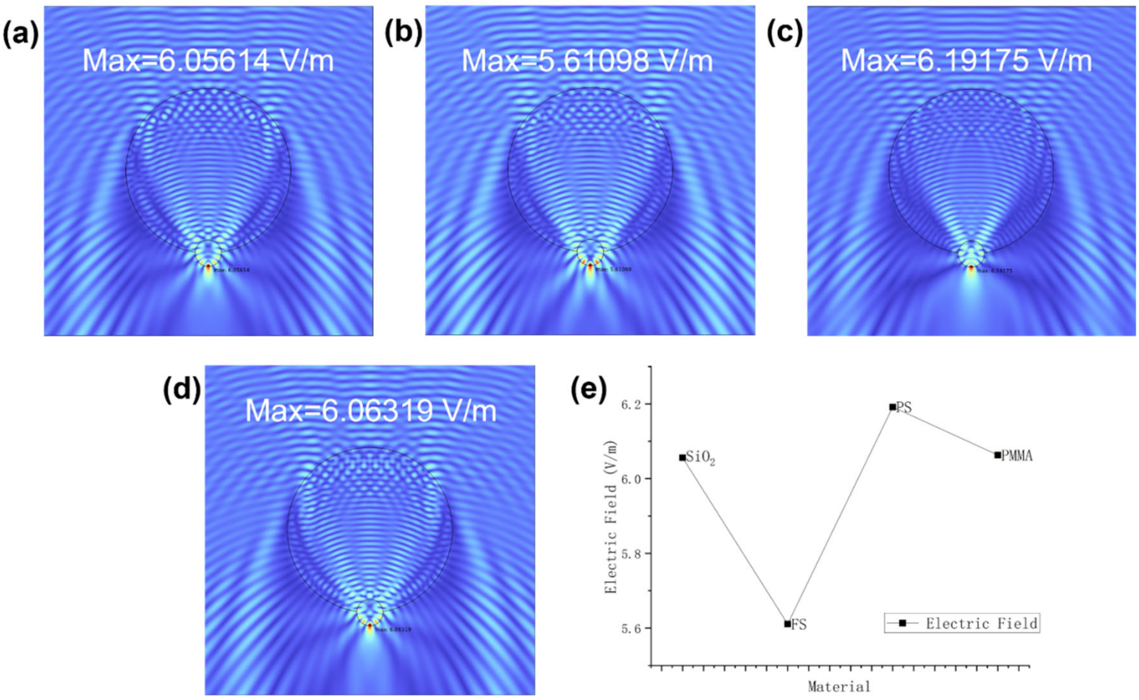

Figure 5 Simulation Results of PNJ Maximum Field Intensity Enhancement for Different Dielectric Microsphere Materials, Which are: (a) SiO2, (b) FS, (c) PS, (d) PMMA. (e) The Relationship Between Field Intensity and Different Media Microsphere Materials

2. Conclusions

In this paper, the linear relationship between the diameter of a single dielectric microsphere and its length of PNJ is obtained. Besides, a way of cascade microspheres is proposed to further enhance the local electromagnetic field. We found that when the cascade spheres are located in the focus of the first-stage PNJ, the maximum electric field enhancement can be obtained. However, considering the need for application and simplification of the process, we propose an embedded cascade method, which can enhance the electric field mode to 6.05614 V/m with a high value of potential application in Raman enhancement. In addition, for different materials of dielectric microspheres, the embedded cascade we designed has the effect of electric field enhancement. Thus, the structure design of embedded cascade microspheres provides a new cascade scheme for Raman enhancement, which is easier to integrate and process with potential applications in microfluidic chips, biomedicine and other fields.

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