Enhanced Second-Harmonic Generation from Fanolike Resonance in an Asymmetric Homodimer of Gold Elliptical Nanodisks

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ABSTRACT: In this article, we have investigated the enhanced second-harmonic generation (SHG) from Fanolike resonance in an asymmetric homodimer of gold elliptical nanodisks using a three-dimensional finite element method. We have found that the broken symmetry will cause Fanolike resonances in the extinction spectrum, resulting in the enhancement of SHG efficiency. When one of the gold elliptical nanodisks rotates, the SHG efficiency increases first and then decreases. In addition, we have also shown that the SHG signal blue-shifts with the reduction of efficiency when the separation between two nanodisks increases. Furthermore, when the nanodisks become thicker, the SHG signal also blue-shifts with the increase of efficiency. The SHG signal from this simple plasmonic structure with high efficiency and tunability may pave a way toward practical applications in sensing and generating a new light source.

1. INTRODUCTION

Plasmonic nanostructures support surface plasmon resonances with highly localized energy in nanoscale geometries, leading to a variety of unique optical properties, such as negative refraction,1,2 hyperbolic dispersion,3,4 magnetic resonance,5−7 and perfect absorption.8 Besides these linear phenomena, the strong enhancement of electric field near the plasmonic nanostructure also leads to nonlinear optical processes.8−10 Among them, second-harmonic generation (SHG) is of great interest in two aspects.11−17 In one aspect, second-order nonlinearity, such as SHG, is explicitly forbidden in bulk crystals with a centrosymmetric lattice regardless of the intensity of the applied field. This seems in contradiction to both simulated and measured SHG effects from a large number of plasmonic nanostructures, showing pronounced SHG emission owing to surface plasmon resonances. In another aspect, second-order nonlinear effects are usually stronger than the higher-order ones, making it easier to be observed and more feasible for practical applications.

Over the past few years, SHG from plasmonic nanostructures has been widely investigated. It has been demonstrated that the SHG is originated from centrosymmetry breaking at the metal surface.14,18 In addition, the near-field enhancement and the low radiation at the fundamental frequency are the key factors to boost the SHG efficiency. However, the near-field enhancement and the scattering efficiency usually reach the maximum value simultaneously.19 Namely, it is difficult to obtain enhanced near field and suppressed radiation loss at the same time. Nevertheless, plasmonic nanostructures with Fano resonance may satisfy both these requirements, resulting from a destructive interference between a bright mode and a dark mode in plasmonic nanostructures.20−24 Fano resonance provides a framework for studying light−matter interaction at the nanoscale and many fascinating applications, such as sensors,25,26 filters,27 color routing,28 and amplifying optical force.29 At the Fano dip, the radiation loss will be efficiently suppressed and the near field will be significantly enhanced, satisfying the key factors to boost the SHG efficiency.30 Thus, enhanced SHG from plasmonic nanostructures with Fano resonance has been extensively studied.11,31−33 Walsh et al. studied the enhanced SHG by the coupling between long-range photonic resonances and localized surface plasmon in periodic arrays of Au nanoparticles.34 Li et al. proposed to boost the SHG efficiency by combining the advantages of Fano resonance and mode-matching in a metasurface.35 Magnetic Fano resonance, which has relatively low radiation,36 also has been introduced to enhance SHG in plasmonic metamolecules.37,38 In simple terms, these works achieved enhanced SHG emission with mode-matching,35,38 lattice resonance,33,34 and high-order resonances32,37,38 requiring complex structures. Besides the plasmonic structure, all-dielectric nanostructure-based nonlinearity has also been widely studied with the Mie resonances in symmetric or asymmetric clusters and oligomers.39 Nevertheless, it could be still applicable to investigate the enhancement of SHG in plasmonic structures from Fano resonance.

In this article, we propose and demonstrate that the Fano resonance in an asymmetric homodimer, consisting of two gold elliptical nanodisks, could enhance the SHG efficiency owing to the hybridization of plasmonic modes. Using a three-dimensional (3-D) simulation method, we investigate the effect of plasmon hybridization on the emitted SHG signal. When one of the gold elliptical nanodisks rotates, the SHG efficiency increases first and then decreases. In addition, we...
also show that the SHG signal blue-shifts with the reduction of efficiency when the separation between two nanodisks increases. Furthermore, when the nanodisks become thicker, the SHG signal also blue-shifts with the increase of efficiency.

2. ELECTROMAGNETIC THEORETICAL METHODS

Figure 1 schematically shows the geometry of the homodimer of gold elliptical nanodisks. Here, \( a = 5 \) nm and \( b = 25 \) nm denote the minor and major axis of the elliptical nanodisk, respectively. The separation between the two nanodisks is \( S \). The homodimer is normally illuminated by a plane wave propagating along the \( z \)-axis with polarization along the \( x \)-axis.

The refractive index of gold is taken from experimental data. Without loss of generality, the surrounding dielectric material is set to be air. Indeed, SHG is theoretically forbidden in centrosymmetric media, such as gold, in the dipolar approximation. Nevertheless, the centrosymmetry could be locally broken at the surface of gold nanostructures, allowing SHG emission. Considering the nonlocal effect, the surface contribution to the SHG may be approximated by an effective nonlinear current at the surface of gold nanostructure, which is described as

\[
J_{NL} = \frac{i \omega}{n_0 c} \left[ \hat{t} (P_{1}^\parallel P_{1}^\parallel) + \hat{n} \frac{1}{2} \frac{3 \omega}{2 \omega + i \gamma} (P_{1}^\perp)^2 \right]
\]

where \( \hat{t} \) and \( \hat{n} \) are the unit vectors parallel and normal to the nanostructure surface, respectively. \( P_{1}^\parallel = \hat{t} \cdot P_{1} \) and \( P_{1}^\perp = \hat{n} \cdot P_{1} \), where \( P_{1} \) is the linear polarization. This approach could well describe the SHG process that may be easily implemented for full-field 3-D simulations. Note that there are some other theoretically allowed but negligible contributions to the SHG signal.

All numerical simulations were carried out using a three-dimensional (3-D) finite element method in two steps. First, the scattering field of the homodimer was simulated assuming a plane wave incidence at the fundamental frequency. The perfect matched layer condition was used around the homodimer to avoid nonphysical reflection of the outgoing electromagnetic wave. Second, an effective nonlinear current, as described by eq 1, at the surface of the nanostructure was applied as the source of the second-harmonic scattering.
process. Then, both the near-field and far-field distributions can be computed with a scattering model at SH frequency.

The SHG efficiency in this article is defined as $\eta_{\text{SHG}} = \frac{S_{\text{caSH}}}{S_{\text{caSH}} + \text{Ext}_{\text{FH}}}$, where $S_{\text{caSH}}$ and $\text{Ext}_{\text{FH}}$ are the scattering and extinction cross sections around the second and the fundamental harmonic wavelengths, respectively.\(^2\) Note that it is different from the traditional definition in experiments, where the SHG efficiency is calculated by the intensity of SHG emission divided by that of the pump source.

3. RESULTS AND DISCUSSION

We start from investigating the linear and nonlinear optical properties of a rotated gold elliptical nanodisk with a thickness ($t$) of 10 nm. Figure 2a,b shows the extinction cross section (ECS) spectra and the near-field intensity of a single gold elliptical nanodisk at the fundamental resonance wavelength (marked by the dashed line) as a function of the rotation angle $\theta$, respectively. At $\theta = 0$, the incident field is polarized along the minor axis of the elliptical nanodisk. Indeed, the plasmon resonance of the minor axis is located at a higher frequency. No significant resonance peak appears in the wavelength range 600–800 nm. At $\theta = \pi/2$, the incident field is polarized along the major axis of the elliptical nanodisk and a strong resonance peak appears at a wavelength of 706 nm. When $\theta \neq 0$ or $\pi/2$, we could still observe only the resonance peak of the major axis at a wavelength of 706 nm. Note that the resonances of minor and major axes are both bright modes, whose eigenfrequencies and intensities are dependent on the particle size. Figure 2c,d shows the SHG efficiency and the near-field intensity of a single gold elliptical nanodisk at the corresponding SHG wavelength (marked by the dashed line) as a function of the rotation angle $\theta$, respectively. When $\theta$ increases from 0 to $\pi/2$, the SHG efficiency gradually increases owing to the major axis plasmon resonance of the gold elliptical nanodisk. The near-field intensity at the SHG wavelength may further demonstrate that the major axis resonance is stronger than the minor axis resonance.

Besides these two bright modes, the gold elliptical nanodisk may also support a dark quadrupole mode, which can be excited by a near-field dipole source rather than a far-field incident wave. In addition, the plasmon resonance of the minor axis is far away from that of the major axis, indicating the difficulty of coupling between them. In the case of the asymmetric homodimer, the plasmon resonance of the major axis of the rotated nanodisk may act as a near-field dipole source and excite the dark mode in the other nanodisk. This means that a super-radiant bright mode strongly interacts with a subradiant dark mode via near-field coupling, producing a low-energy (bonding) and a high-energy (antibonding) hybridized mode.\(^41,42\) As a result, a Fano-like resonance appears in the linear ECS spectrum and enhances the near-field intensity at both bonding and antibonding modes, thereby boosting the SHG efficiency.

To demonstrate this, Figure 3a shows the ECS spectrum of the homodimer as a function of the rotation angle of one gold elliptical nanodisk. The separation ($S$) of the homodimer and the thickness ($t$) of the nanodisk are fixed as 32 and 10 nm, respectively. At $\theta = 0$, the homodimer is a symmetric structure with respect to the incident electromagnetic field. It is well known that we may not observe the Fano-like resonance in this case, as demonstrated by the black line in Figure 3a. When $\theta$ increases, the symmetry of the homodimer is broken, resulting in the hybridized subradiant and super-radiant modes. Hence, we can observe a Fano-like resonance in the ECS spectrum. When the value of $\theta$ approaches $\pi/2$, the homodimer recovers to be a symmetric structure with respect to the incident field and, consequently, the Fano dip becomes shallow and narrow.

Figure 3b shows the corresponding SHG efficiency as a function of the rotation angle of one gold elliptical nanodisk. At $\theta = 0$, the SHG efficiency is not significantly improved because of the absence of the Fano-like resonance. When the value of $\theta$ increases, the SHG efficiency is boosted because of the enhanced Fano resonance and near-field intensity at the surface of the nanostructure. At around $\theta = \pi/4$, the SHG efficiency is the highest, resulting from the maximum symmetry breaking and the deepest Fano resonance. When the value of $\theta$ approaches $\pi/2$, the capacitive coupling between the hybridized subradiant and super-radiant modes reduces because the homodimer becomes symmetrical in this case, thereby leading to a decreased SHG efficiency.

For further demonstration, Figure 4 plots the near-field and far-field distributions of the electric field intensity for the cases $\theta = \pi/4$ and $\theta = 0$ at different SHG wavelengths (as marked by points I, II, and III in Figure 2b). From Figure 4a–d, we can easily see that the SHG signal is significantly enhanced in the asymmetric homodimer in comparison to that in the symmetric case because of the excitation of Fano-like resonance and the near-field enhancement at the fundamental wavelength. In addition, the difference between the far-field patterns in symmetric and asymmetric cases indicates that the radiated direction of the SHG signal could be manipulated by engineering the plasmon hybridization. This property is critical to the design of practical applications in the nonlinear regime.

According to the plasmon hybridization model, the capacitive coupling strength between super-radiant and subradiant modes is dependent on the separation $S$. Therefore, we investigate the influence of the Fano-like resonance on the SHG efficiency by tuning the separation between the gold elliptical nanodisks in the asymmetric homodimer. The thicknesses of nanodisks and the rotation angles are fixed as $t = 10$ nm and $\theta = \pi/4$, respectively. As shown in Figure 5a, a decrease in the separation between the two nanodisks makes the Fano dip in the ECS spectra wider and deeper. This is because the capacitive coupling between the super-radiant and subradiant modes becomes stronger, thereby concentrating more energy in the near-field of the homodimer. In addition,
the enhanced coupling leads to red and blue shifts of the low- and high-energy peaks, respectively. These linear optical properties are consistent with the plasmon hybridization theory. Figure 5b plots the SHG efficiency at various separation distances, S, of 28, 30, 32, 34, and 36 nm. When the nanodisks move close to each other, the emitted SHG signals from the bonding and antibonding modes are red- and blue-shifted, respectively. Furthermore, the SHG efficiency is significantly enhanced because of the strong capacitive coupling at a small separation distance. Figure 5c,d presents the near- and far-field distributions, respectively, of the electric field at the high-energy SHG peak with separation distances, S, of 36, 34, 32, 30, and 28 nm, further validating the enhanced SHG efficiency from the Fanolike resonance. Note that the SHG efficiency could increase further when the separation S is reduced. The smallest value of S should make sure that the narrowest gap between two plasmonic nanoparticles is larger than 2 nm, under which the quantum tunneling effect between two plasmonic nanoparticles should be taken into consideration.43

The capacitive coupling strength between the super-radiant and subradiant modes also depends on the thicknesses of the nanodisks. Figure 6a plots the linear ECS spectrum with different nanodisk thicknesses, t = 8, 10, 12, 14, and 16 nm, while keeping the rotation angle and separation fixed as $\theta = \pi/2$ and $S = 32$ nm, respectively. It is obvious that both the subradiant and super-radiant modes red-shift with the increasing nanodisk thickness, which agrees well with the LC circuit theory.44,45 Besides, the Fano dip becomes wider and deeper because of the enhanced coupling strength. These
phenomena demonstrate that more electromagnetic energy is localized at the surface of nanostructures, which is beneficial for SHG efficiency. Figure 6b shows the corresponding SHG emission with different nanodisk thicknesses. We could observe that the SHG emission moves to higher frequency as the nanodisks become thicker. Owing to the increased coupling strength between subradiant and super-radiant modes, the SHG efficiency is significantly enhanced by a factor of about 5, whereas the thickness of nanodisks is increased from $t = 8$ to 16 nm.

4. CONCLUSIONS
In summary, we have proposed and demonstrated the enhanced SHG from a homodimer, consisting of two gold elliptical nanodisks, using a 3-D simulation method. The results show that the Fano-like resonance in an asymmetric homodimer may result in enhanced SHG efficiency owing to the plasmon hybridization. In addition, the enhanced SHG signal, including the radiation direction and frequency, could be manipulated by engineering the coupling between hybridized super-radiant and subradiant modes. Our results may provide an effective approach to enhance SHG efficiency in plasmonic structures and also find practical applications in nonlinear optical devices.

Author Contributions
The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes
The authors declare no competing financial interest.

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