Low-threshold surface plasmon amplification from a gain-assisted core–shell nanoparticle with broken symmetry

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Abstract

We report that the gain threshold of a core–shell nanoparticle-based spaser can be reduced significantly by offsetting the gain-doped dielectric core within the metallic shell. By investigating the optical cross sections of the reduced symmetry core–shell nanoparticle with different levels of gain, we determined the gain threshold of the asymmetric nanoparticle-based spaser fueled by different plasmon modes. The calculation results indicate that when multipolar plasmon oscillations excited in asymmetric core–shell nanostructures are used as lasing modes, the gain threshold of an asymmetric spaser particle can drop by 30% as compared to the case of a perfect or symmetric particle. The underlying physics of low-threshold surface plasmon amplification is explained by investigating the Q factor and the optical field confinement and enhancement associated with the lasing mode.

Keywords: spaser, gain threshold, core–shell nanoparticle, symmetry breaking

(Some figures may appear in colour only in the online journal)

1. Introduction

Coherent light sources free from diffraction limitations have recently attracted significant attention due to their potential applications in biosensing, data storage, photolithography and optical communications [1–3]. Based on the strong interaction between nanosized emitters and localized surface plasmons (SPs), Bergman and Stockman first proposed that a coherent light field can be generated directly at the nano-scale through surface plasmon amplification by stimulated emission of radiation, named a spaser or plasmon laser [4]. With the idea of the spaser, many theoretical schemes and experimental studies have been carried out on the amplification of localized SPs or propagating SPs. Plasmon lasers using propagating SPs are usually constructed through waveguide configurations, by replacing the dielectric layers in waveguides with gain materials and forming a Fabry–Perot cavity to generate feedback [5, 6]. Plasmon lasers based on localized SPs are constructed by combining metal nanoparticles or nanostructure arrays with dielectric media incorporating gain [7–9]. The particle itself serves as a resonator (or a resonant cavity) and the adjacent gain medium delivers energy to the plasmon mode. Since the particle is considerably smaller than the wavelength, further reduction in both the physical laser size and the mode size is possible. As shown by Noginov and co-workers, a spaser particle with a...
gold core and a dye-doped silica shell could be reduced to the scale of just tens of nanometers [7]. So far, nanoparticle-based spasers comprising gain-assisted metal nanospheres [7, 10, 11], cubic nanoboxes [12], nanorods [13], nanoshells [14–18] or nanorings [19] have been demonstrated theoretically and experimentally.

While great progress has been made in spasers or plasmon lasers, the realization of a deep subwavelength size, low-threshold, efficient source of radiation that operates at room temperature has remained a challenge [1–3]. In order to trigger the amplification of SPs in a plasmonic structure, the gain strength of the active medium must be strong enough to prevail over the metallic loss, which is particularly strong at optical frequencies, and the radiation damping [20, 21]. A spaser with a large and/or unattainable lasing threshold due to high dissipative loss is undesirable for practical applications. One of the methods to reduce the metallic loss and thus improve the gain capability is to cool plasmonic lasing structures down to cryogenic temperatures [6, 22]. To effectively reduce the gain threshold, more attempts are required to further modify the design of these plasmonic nanostructures or to find SP modes with higher quality factors ($Q$) [13, 15, 23].

Since nanoparticles with different configurations, sizes or morphologies are capable of supporting different plasmon modes and thus different conditions of surface plasmon amplification, nanoparticle-based spasers are expected to display excellent flexibility when faced with the design of a functional nanolaser. Recently, Zhang et al demonstrated that a silica–gold–silica nanoshell-based spaser has a higher magnitude of surface plasmon amplification and lower gain threshold than a nanosphere-based or nanoegg-based spaser [14]. In their research, the dipole-like mode is used to realize the spaser. In contrast to a concentric core–shell nanoparticle [14], a particle with reduced structural symmetry will give rise to modified plasmonic features due to the change of interaction between primitive plasmon modes [24, 25]. Halas’ group have demonstrated that the near- and far-field properties of plasmonic nanoparticles can be modified by changing the core–shell offset due to the excitation of multipolar modes accompanied by stronger electric field enhancement [24, 25]. Therefore, the performance of a spaser based on a reduced symmetry core–shell nanoparticle that is fueled by a multipolar mode is intriguing. To the best of our knowledge, the optical amplification and laser action based on multipolar modes in asymmetric core–shell nanostructures have not been explored.

In this paper, we propose a reduced symmetry core–shell nanoparticle-based spaser, which comprises a gain-material-doped spherical silica core, a gold shell and an outer silica shell doped with the same gain material. The symmetry breaking is caused by offsetting the core within the shell. By investigating the gain-assisted scattering efficiency and near-field enhancement, we demonstrate that the asymmetric nanosystem enables the achievement of a lower threshold of SP amplification than its symmetric counterpart.

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2. Model and simulation method

Figure 1 shows the configuration of the nanoparticle-based spaser with reduced symmetry core–shell. The offset between the core center and the shell center is defined as the parameter $D$. The radii of the silica core, the metal shell and the outer silica shell are set to be $r_1 = 35$ nm, $r_2 = 40$ nm and $r_3 = 60$ nm, respectively. The core–shell particle is assumed to be exposed to the air. On the basis of classic linear electrodynamics, the gain medium can be considered as a dielectric with a negative imaginary part. We set the refractive index of the core and outer shell to be constant (non-dispersive), i.e. $n = 1.5 - ik$, where the real part represents the refractive index of silica and $k$ defines the level of optical gain. For simplicity, the experimentally measured dielectric function is utilized for the gold shell [26], neglecting the electron surface scattering effect, which would weaken and broaden the plasmon peaks but leave the peak wavelengths essentially unchanged [27].

The near- and far-field optical properties of the spaser particles were solved numerically in Comsol Multiphysics 4.2 with the RF module by calculating the optical scattering off of a passive or an active core–shell nanoparticle. The 3D spherical simulation domain consisted of the core, the shells of the particle system, the embedding medium and a perfectly matched layer (PML). The thickness of the air domain around the core–shell nanoparticle was equal to half the wavelength in free space. The PML with a thickness of half a wavelength in free space was outside of the air domain and acted as an absorber of the scattered field. The plane wave, used for excitation, was inserted on the inside of the PML with the polarization perpendicular ($X$-polarization) or parallel ($Y$-polarization) to the axis of the nonconcentric core–shell nanoparticle. The scatterer and air domain were meshed with tetrahedral elements. A sweep mesh was used for the PML. The maximum mesh size was 0.1 wavelengths in free space, and a local mesh refinement was applied for the core–shell nanoparticle with a maximum element size of 2 nm in the gold shell. The scattering cross-section ($\sigma_{\text{scat}}$)
is defined as the total integrated power contained in the scattered field normalized by the irradiance of the incident field, and the absorption cross-section ($\sigma_{\text{abs}}$) is defined by the net flux through a surface surrounding the concentric shells normalized by the incident field irradiance [17]. Both can be obtained by calculating the scattered and absorbed powers based on Poynting’s theorem [17]. An additional spherical surface was set in the air domain and adjacent to the PML for integral operation. The extinction spectra ($\sigma_{\text{ext}}$) were obtained by summing the scattering ($\sigma_{\text{scat}}$) and absorption cross sections ($\sigma_{\text{abs}}$). The quality factor was estimated by $Q = \lambda_{\text{res}}/\Delta\lambda$, where $\lambda_{\text{res}}$ is the resonant wavelength and $\Delta\lambda$ is the spectral linewidth obtained by fitting the multipeak data through a Lorentz function.

3. Results and discussion

3.1. Performance of the asymmetric core–shell nanoparticle-based spaser

Figure 2 shows the optical extinction spectra of core–shell nanoparticles with different core offset parameters ($D$) in the absence of gain material ($k = 0$). The index $l$ is used here to refer to the multipolar mode. For the symmetric nanoparticle with $D = 0$, one main peak is observed, corresponding to the dipolar mode ($l = 1$). For a nanoparticle with symmetry breaking ($D > 0$), several multipolar peaks ($l = 2, 3, 4$) appear and shift to red with increasing offset. This can be understood very well in terms of the hybridization theory that describes the interaction or ‘hybridization’ of elementary plasmons in complex nanostructures of arbitrary shape [25, 28, 29]. Specifically, for a spherically symmetric nanoshell, plasmon hybridization only occurs between the primitive plasmon modes of the same angular momentum, i.e. the cavity plasmons supported by the inner surface of the shell and the sphere plasmons of the outer surface. For a nanoshell with an offset core, the reduction in symmetry relaxes these selection rules, allowing for an admixture of dipolar components in all plasmon modes of the particle. The hybridization strengths between these plasmons become stronger with increase of offset, leading to a core offset-dependent multipeaked spectrum [25, 28].

In figure 2, both X-polarized and Y-polarized light excitations are considered due to the anisotropy of the optical extinction resulting from the symmetry breaking of the nanostructure. It is noticed that the $l = 2$ mode in an asymmetric nanoparticle with $D = 4$ nm is centered at about 779 nm for $X$-polarization and 785 nm for $Y$-polarization, approaching the wavelength of the $l = 1$ mode (797 nm) in the symmetric case ($D = 0$). Considering the dispersion of the metal permittivity in optical frequencies, the asymmetric nanoparticle-based spaser fueled by the $l = 2$ mode is first focused on by us for the sake of comparing with its symmetric counterpart.

We calculated the optical cross-section spectra of scattering ($\sigma_{\text{scat}}$) and absorption ($\sigma_{\text{abs}}$) for the reduced symmetry core–shell-based spaser ($D = 4$ nm) with different levels of optical gain ($k$) under $X$-polarized light excitation.

When $k = 0$, the peak value of $\sigma_{\text{abs}}$ is much larger than that of $\sigma_{\text{scat}}$ for both the dipolar and the multipolar mode, indicating a strong absorption characteristic of the core–shell nanoparticle (figure 3(a)). When the gain medium is introduced into the silica core and the outer shell ($k > 0$), the magnitude of $\sigma_{\text{scat}}$ for the $l = 2$ mode increases rapidly and the linewidth decreases remarkably, resulting in a greatly enhanced quality factor (not shown here). The absorption of light by metal ($\sigma_{\text{abs}}$) increases at first and then decreases due to the gain medium doped in the silica compensating the energy loss of the localized SPs by resonance energy transfer from the gain medium to the gold nanoshell [12]. When $k$ further increases, $\sigma_{\text{abs}}$ exhibits a negative absorption and the nanoparticle starts to operate as an optical amplifier. At $k = 0.056\,\text{177}$, a significant resonance takes place for the $l = 2$ mode ($\lambda_{\text{res}} = 778.8$ nm), as shown in figure 3(b). The scattering cross-section reaches $6.07 \times 10^5\,\text{m}^2$, $10^5$ orders of magnitude higher than that with $k = 0$. Meanwhile, the linewidth drastically drops to a small value of below 0.2 nm and the quality factor of the LSPR is greatly enhanced, indicating that the strength of SPs associated with the $l = 2$ mode is significantly amplified by the energy transferred from the gain medium. When the gain coefficient continues to grow and exceeds the critical value, the $l = 2$ mode becomes off resonance, with the peak values of $\sigma_{\text{scat}}$ and $\sigma_{\text{abs}}$ dropping and the linewidth increasing significantly, due to the light amplification surpassing the energy dissipation of the nanosystem [13, 14]. The optical cross sections of the $l = 2$ mode as a function of the gain $k$ are plotted in figure 3(c), which indicates clearly that $\sigma_{\text{scat}}$ and $\sigma_{\text{abs}}$ reach their highest magnitude, while the optical extinction cross-section $\sigma_{\text{ext}}$, i.e. the sum of $\sigma_{\text{scat}}$ and $\sigma_{\text{abs}}$, is close to zero at the point $k = 0.056\,\text{177}$, meaning a zero-level net optical gain. For stable operation of the spaser, the net amplification of the system should be zero [30]. Therefore, the value of $k = 0.056\,\text{177}$ can be viewed as the gain threshold ($k_{\text{thre}}$) of the $l = 2$ mode for the spaser, which gives the maximum lasing efficiency.

In the same way, the optical cross-section spectra of $\sigma_{\text{scat}}$ and $\sigma_{\text{abs}}$ under $Y$-polarized light excitation are calculated for the same asymmetric core–shell-based spaser ($D = 4$ nm),
Figure 3. Optical cross-section spectra of the asymmetric core–shell nanoparticle-based spaser \((D = 4 \text{ nm})\) with \(k = 0\) (a) and \(k = 0.056 177\) (b) under X-polarized light excitation. (c) The optical cross sections of the \(l = 2\) mode as a function of \(k\) under X-polarization.

Figure 4. Optical cross-section spectra of the asymmetric core–shell nanoparticle-based spaser \((D = 4 \text{ nm})\) with \(k = 0\) (a) and \(k = 0.056 391\) (b) under Y-polarized light excitation. (c) The optical cross sections of the \(l = 2\) mode as a function of \(k\) under Y-polarization.

as shown in figure 4. Assisted by the optical gain, the \(l = 2\) mode reaches the maximum magnitude of \(\sigma_{\text{scat}}\) and \(\sigma_{\text{abs}}\) at \(k = 0.056 391\) (figures 4(b) and (c)). At this critical point, the scattering cross-section of the \(l = 2\) mode reaches \(7.52 \times 10^3 \text{ \mu m}^2\), similar to the case of X-polarized light excitation.

For comparison, the dependences of \(\sigma_{\text{scat}}\) and \(\sigma_{\text{abs}}\) on the value of \(k\) for the dipole mode \((l = 1)\) in a symmetric core–shell nanoparticle \((D = 0 \text{ nm})\) are also investigated, as shown in figure 5. The gain threshold is determined to be \(k = 0.080 128\), apparently higher than the asymmetric case \((k_{\text{thre}} = 0.056 177\) for X-polarization and \(k_{\text{thre}} = 0.056 391\) for Y-polarization). In particular, the gain threshold of the core–shell nanoparticle-based spaser is reduced by 30% by breaking the symmetry of the nanosystem and exploiting the excited multipolar plasmon resonance as the lasing mode. Figure 5 also indicates that the maximum magnitude of \(\sigma_{\text{scat}}\) at the gain threshold \((k_{\text{thre}})\) is one order of magnitude higher than those obtained through the \(l = 2\) mode (figures 3(c) and 4(c)), showing more efficient radiation of the spaser fueled by the dipolar mode.

### 3.2. The physical mechanism of a low-threshold spaser realized by an asymmetric core–shell nanoparticle

As mentioned above, in order to realize stimulated emission of SPs, the electromagnetic energies offered by the gain medium are required to compensate the overall losses of plasmonic structure experienced by the lasing mode. Lower
dissipation losses will lead to a smaller gain threshold. To understand the lower gain threshold of the \( l = 2 \) mode in an asymmetric core–shell nanostructure than that of the \( l = 1 \) mode in the symmetric case, we calculated the passive \( Q \) factors of each mode, because the \( Q \) value is dominated by the dissipation loss. We found that the \( Q \) values of the \( l = 1 \) mode in a symmetric core–shell nanoparticle and the \( l = 2 \) mode in a reduced symmetry case (\( D = 4 \) nm) are 16.4 and 22.3 (\( X \)-polarization) and 21.8 (\( Y \)-polarization), respectively, suggesting that the \( l = 2 \) mode suffers lower dissipation losses than the \( l = 1 \) mode, consistent with the fact of the lower gain threshold associated with the \( l = 2 \) mode. Since the main sources of losses in a spaser are dissipation in the metal and radiation of electromagnetic waves, the quality factor is limited by these two factors and can be decomposed as \( Q_{\text{tot}}^{-1} = Q_{\text{mat}}^{-1} + Q_{\text{rad}}^{-1} \). where \( Q_{\text{mat}} \) and \( Q_{\text{rad}} \) account for the metallic and radiation loss, respectively [21]. Due to approaching the resonance frequencies at around 790 nm, and thus the similar imaginary parts of the metal permittivity, both modes, i.e. the \( l = 1 \) mode in the symmetric and the \( l = 2 \) mode in the asymmetric nanostructure, are assumed to suffer similar metallic loss, and the difference in \( Q_{\text{tot}} \) is mainly dominated by the \( Q_{\text{rad}} \) factor. In view of the characteristic of multipolar mode that can be regarded as a ‘subradiant’ mode (or dark mode) owing to weak coupling (or no coupling) to the far-zone optical field and thus less radiation loss [16, 31], the higher radiation quality factor \( Q_{\text{rad}} \) and consequently higher value of \( Q_{\text{tot}} \) for the \( l = 2 \) mode than the \( l = 1 \) mode or dipolar mode (also known as the superradiant or bright mode) in the symmetric case is easy to understand.

Besides the \( Q \) factor, the threshold gain required to compensate the dissipation losses is also related to the confinement factor of the mode in the gain region. The high local field associated with the lasing mode is the underlying physical reason for the very strong feedback in the spaser [30]. Figures 6(a)–(c) show the electric field distributions of the \( l = 2 \) mode in an asymmetric nanosystem (\( D = 4 \) nm) and the \( l = 1 \) mode in its symmetric counterpart (\( D = 0 \) nm) at \( k = 0 \). The field enhancement factor is defined as \( |E|/|E_0| \), where \( |E| \) and \( |E_0| \) represent the amplitude of the local electric field with and without the core–shell nanoparticle. It can be seen clearly that the \( l = 2 \) mode in the nanostructure with broken symmetry enables more efficient localization and enhancement of the
E-field than the \( l = 1 \) mode in the symmetric nanoparticle, which is confirmed by the smaller value of the mode volume for \( l = 2 \). The mode volume of the core–shell nanoparticle can be calculated using the formula \[ V_{\text{eff}} = \frac{\int \varepsilon(r)|E(r)|^2 d^3r}{\max[\varepsilon(r)|E(r)|^2]} \]

where \( \varepsilon(r) \) is the dielectric constant at the position \( r \), \( |E(r)|^2 \) is the corresponding field intensity and the integration is over the gain medium. Using the FEM software package, we can obtain the field distribution and then estimate the mode volume. The \( V_{\text{eff}} \) of the \( l = 2 \) mode is calculated to be 0.000052(\( \lambda/n \))^3 for X-polarization and 0.000028(\( \lambda/n \))^3 for Y-polarization, two orders of magnitude smaller than that of the \( l = 1 \) mode in the symmetric case \( V_{\text{eff}} = 0.0012(\lambda/n)^3 \), where \( n \) is the refractive index of the core and the outer shell (%(n = 1.5)). Since the high \( Q \) factor along with strong mode confinement can lead to enhancement of the spontaneous emission rate (i.e. the Purcell factor \( F = F \propto Q/V_{\text{eff}} \)) \cite{22, 32}, the low-threshold surface plasmon (SP) amplification by exploitation of the multipolar mode can be understood.

We also noticed that the value of \( V_{\text{eff}} \) associated with the \( l = 2 \) mode for X-polarization is roughly two times that for Y-polarization, but the corresponding gain thresholds are almost the same \( (k_{\text{thre}} = 0.056\ 177 \) for X-polarization and \( k_{\text{thre}} = 0.056\ 391 \) for Y-polarization). As pointed out by Hill and others \cite{32}, caution needs to be used when applying \( V_{\text{eff}} \) to predict the Purcell factor because it is only valid for emitters placed at the modal field maximum. For the case presented here where emitters are distributed evenly over the gain medium volume, \( V_{\text{ave}} \) can also be used to estimate the Purcell factor, which is defined similarly to \( V_{\text{eff}} \) but with the denominator in \( V_{\text{eff}} \) (equation (1)) replaced by the average value of \( \varepsilon(r)|E(r)|^2 \) in the gain medium \cite{32}. As a result, we obtained \( V_{\text{ave}} = 0.005\ 85(\lambda/n)^3 \) for X-polarization and \( V_{\text{ave}} = 0.005\ 70(\lambda/n)^3 \) for Y-polarization, and thereby similar \( F \) values and gain thresholds of the \( l = 2 \) mode for both polarizations. Anyway, it is undoubted that the \( l = 2 \) mode in an asymmetric nanostructure enables a much smaller mode volume than the \( l = 1 \) mode in the symmetric case. Efficient localization and enhancement of the electromagnetic field are desired for low gain threshold.

In addition, there is a huge enhanced electromagnetic field associated with the lasing mode \( (l = 2) \) as the gain increases to the value of the threshold, reaching more than \( 10^5 \) E-field enhancement \( (|E|^2/|E_0|^2) \) under both polarizations, as shown in figures 6(d) and (e). Despite the significant difference of the magnitude of E-field enhancement between the active and the passive nanosystem, both of them exhibit the same field profiles and hot-spot distributions. The giant enhancement of the local electric field serves as additional evidence for a spaser.

3.3. The performance of asymmetric core–shell nanoparticle-based spasers fueled by different modes

Next, we investigated the spaser performance of a gain-assisted asymmetric core–shell nanoparticle fueled by the \( l = 1 \) or \( l = 3 \) mode. For ease of comparison, the resonant wavelength \( \lambda_{\text{res}} \), passive quality factor \( Q \) and gain threshold \( k_{\text{thre}} \) of each mode in symmetric \( (D = 0) \) and asymmetric \( (D = 4 \) nm) core–shell nanoparticles are listed in table 1. It can be seen that the asymmetric nanoparticle-based spaser fueled by a dipolar or multipolar mode shows a lower gain threshold than its symmetric counterpart. In our considered modes, the \( l = 3 \) mode excited in the asymmetric nanostructure exhibits the highest \( Q \) value and the lowest gain threshold \( k_{\text{thre}} \) of 0.054\ 006 for X-polarization and \( k_{\text{thre}} = 0.054\ 526 \) for Y-polarization). Compared with the core–shell concentric spherical nanoparticle, the asymmetric nanostructure is capable of generating a more efficiently localized and enhanced E-field by using a dipolar or multipolar mode (the electric field distributions induced by the \( l = 1 \) and \( l = 3 \) modes are not shown here). The optical gain coefficient at the laser threshold is inversely proportional to the optical confinement factor and the quality factor \( Q \) of the lasing mode \cite{21}, therefore the low-threshold SP amplification can be achieved through the reduced symmetry nanostructure.

A low gain threshold brings more choices of appropriate gain materials, such as dye molecules, rare earth ions and semiconductor quantum dots. In these systems, the gain coefficient \( k \) is generally related to both the emission cross-section (\( \sigma_e \)) and the concentration (\( N \)) of the gain medium. The formula can be written as \( k = \lambda N \sigma_e/4\pi \) \cite{12}. Therefore, the gain coefficient can be controlled in experiments by changing the gain concentration as the gain cross-section is known \cite{12}. In addition, the modeled core–shell nanoparticle with reduced symmetry can be fabricated by electron-beam-induced ablation of metallic nanoshells, which allows us to

<table>
<thead>
<tr>
<th>Core offset</th>
<th>Mode index</th>
<th>( \lambda_{\text{res}} ) (nm)</th>
<th>( Q )</th>
<th>( k_{\text{thre}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>D = 0 nm</td>
<td>( l = 1 )</td>
<td>797.5</td>
<td>16.4</td>
<td>0.080 128</td>
</tr>
<tr>
<td>X-polarization</td>
<td>( l = 1 )</td>
<td>914.4</td>
<td>19.6</td>
<td>0.058 531</td>
</tr>
<tr>
<td>( E \parallel x )</td>
<td>( l = 2 )</td>
<td>778.8</td>
<td>22.3</td>
<td>0.056 177</td>
</tr>
<tr>
<td></td>
<td>( l = 3 )</td>
<td>697.2</td>
<td>24.5</td>
<td>0.054 006</td>
</tr>
<tr>
<td>D = 4 nm</td>
<td>( l = 1 )</td>
<td>952.5</td>
<td>15.5</td>
<td>0.060 441</td>
</tr>
<tr>
<td>Y-polarization</td>
<td>( l = 2 )</td>
<td>785.2</td>
<td>21.8</td>
<td>0.056 391</td>
</tr>
<tr>
<td>( E \parallel y )</td>
<td>( l = 3 )</td>
<td>699.4</td>
<td>24.5</td>
<td>0.054 526</td>
</tr>
</tbody>
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carefully and systematically thin the top of an individual nanoshell in a highly controlled manner [33].

4. Conclusion

In summary, we have proposed and numerically studied an asymmetric core–shell nanoparticle-based spaser. Our calculations demonstrated that by reducing the structural symmetry by offsetting the dielectric core within the metallic shell, the gain threshold of a core–shell nanoparticle-based spaser can be reduced significantly. In particular, by increasing the core offset from \( D = 0 \) to \( D = 4 \) nm, the gain threshold of a core–shell nanoparticle particle can be decreased by 0.080 128 to 0.056 177 (X-polarization) or 0.056 391 (Y-polarization) at around the wavelength of 790 nm, dropping by almost 30%. This is because the reduced symmetry nanostructure supports multipolar plasmon oscillations, which possess higher quality factors (lower dissipation losses), and more significant \( E \)-field confinement and enhancement than the dipolar mode excited in a symmetric nanostructure. Therefore, a lower threshold for surface plasmon amplification can be achieved as the multipolar modes are used for lasing. In addition, more than \( 10^3 \) times scattering enhancement and \( E \)-field enhancement can be achieved for the asymmetric core–shell nanoparticle when the optical gain \( k \) reaches the threshold. Although we proposed an approach of structural symmetry breaking to realize low-threshold surface plasmon amplification through a single core–shell nanoparticle, this work also gives insights into the achievement of a source of coherent optical radiation based on an array of nanostructures with reduced symmetry.

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