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Hybridization of Singular Plasmons via Transformation Optics

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Abstract

In this paper, we develop a new coupled mode theory for hybridization of singular plasmons. We show that the hybridization of these singular plasmons increases the density of the spectrum thus overcoming non-locality. We propose a metasurface design for broadband light absorption. The proposed model demonstrates an elegant interplay between the hybridization picture and transformation optics.

1 Manuscript

Surface plasmons are resonant oscillations of charge densities on metallic nanoparticles. The plasmons of interacting particles have been extensively studied and utilized for sub-wavelength control of light. The frequency spectrum of the plasmons is discrete and the hybridization model can describe its structure in a way analogous to molecular orbital theory. When particles are brought together and become close-to-touching, the spectrum changes significantly: the discrete spectrum becomes more dense and eventually becomes continuous in the limit, a phenomenon which can be exploited for broadband light-harvesting. Recently, the application of Transformation Optics (TO) has revealed that the continuous nature of the spectrum for close-to-touching particles is due to the singular character of the geometry, that is, the narrow gaps between the particles. However, a fundamental difficulty is encountered in the quest to achieve a dense spectrum. The non-local effect, which has a quantum origin, smooths out the singularity resulting in a less dense spectrum. The gap distance is effectively non-zero when the particles are touching. Many studies have been carried out with the goal of modeling the nonlocal effect accurately. Here we ask a different question: can we increase the density of the spectrum without requiring that the geometry becomes more singular?

In this work, we develop a new hybridization model for plasmons of strongly interacting particles. Our model provides a means of circumventing the issue of non-locality. Specifically, we show that the spectral density of a many-particle system can be enhanced through careful adjustment of its geometrical configuration, while keeping inter-particle gap distances unchanged. This new model of hybridization lends itself naturally to a promising design principle for plasmonic metamaterials. With this guiding principle in mind, we propose an original metasurface design that successfully overcomes the issue of non-locality.

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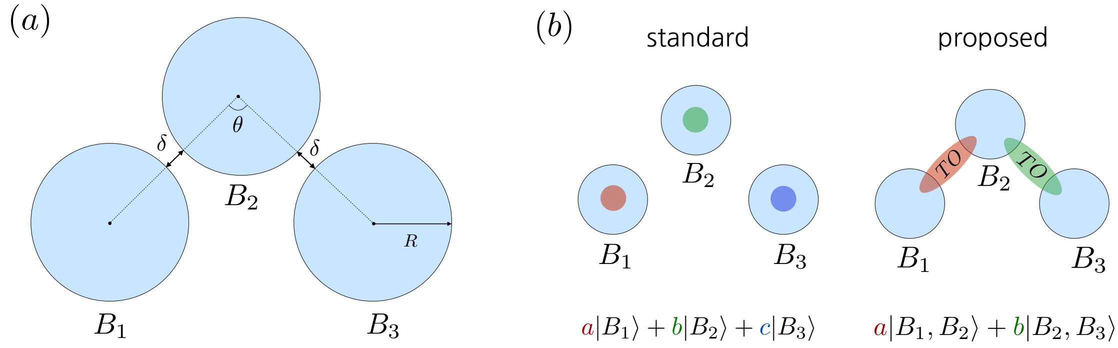

Figure 1: (a) Geometry of the trimer. (b) Comparison of our model with the standard hybridization model.

We briefly discuss some challenges in understanding plasmons in systems of interacting particles. The standard hybridization model provides a simple and insightful physical picture, however, in the case of close-to-touching particles, the picture becomes complicated. The TO approach, on the other hand, is effective for the close-to-touching case, yet TO alone cannot be applied to systems featuring three (or more) particles. The authors of this work have recently developed an efficient numerical scheme for strongly interacting many-particle systems.

We now outline the first key part of this work which is our proposed model for hybridization which we call the *Singular Hybridization Model*. In the standard hybridization model, a plasmon of the system is a combination of plasmons of individual particles. On the contrary, in our model, the basic building blocks are the gap-plasmons of a pair of particles, and for these we use the TO approach. We consider a trimer in Figure 1a, it being the simplest example for our model (we emphasize that our model can be applied to any configuration of particles). The trimer plasmon is now treated as a combination of two gap-plasmons. This simple conceptual change is the fundamental key to solving the aforementioned challenges. For each gap, the singular behavior of a gap-plasmon is captured by a TO solution. The gap-plasmons are strongly confined in their respective gaps and the two gaps are well-separated, which means the two gap-plasmons do not overlap significantly. Hence, the change of the spectrum from the TO spectrum is not significant. Therefore, we can expect that we still get a simple picture even in the close-to-touching case. On the contrary, in the standard hybridization model, a large number of plasmons (of individual particles) are needed to describe the hybrid plasmons of close-to-touching particles.

To gain a deeper physical understanding, we develop a coupled mode theory for the hybridization of singular plasmons. For simplicity, we only consider 2D structures, however, our theory can be extended to the 3D case. We assume the Drude model for the metal permittivity: $\varepsilon = 1 - \frac{\omega_p^2}{\omega^2}$, where ω_p is the bulk plasma frequency and the background permittivity is $\varepsilon_0 = 1$. We also adopt the quasi-static approximation by assuming the particles to be small compared to wavelength of the incident light.

We briefly review the TO description of gap-plasmons which are the basic building blocks of our proposed model. Consider a dimer of cylinders of radii R separated by a distance δ.

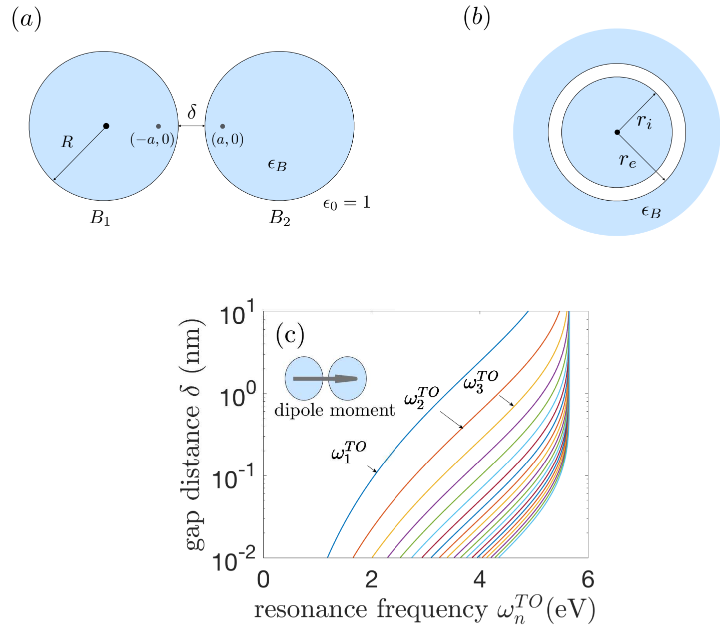

Figure 2: (a) Geometry of the dimer. (b) The concentric annulus in the transformed frame. (c) The resonance frequencies of the dimer plasmons computed by the TO approach. We set $\omega_p = 8$ eV.

The conformal transformation Φ given by

$$x' + iy' = \Phi(x + iy) = \frac{x + iy + a}{x + iy - a}, \qquad a = (\delta(R + \delta/4))^{1/2},$$

maps the dimer to a concentric annulus whose inner radius is $r_i = e^{-s}$ and outer radius is $r_e = e^{s}$, where $\sinh s = a/R$. The transformation Φ reveals a hidden symmetry of the dimer plasmons and provides a simple analytical description of the spectrum. We only consider the resonance modes whose dipole moment is aligned parallel to the dimer axis since these are the modes which contribute to the optical response significantly. The resonance frequencies ω_n^{TO} of the modes are given by

$$\omega_n^{TO} = \omega_p \sqrt{e^{-ns}\sinh(ns)}, \quad n = 1, 2, 3, \cdots .$$

When the gap distance δ gets smaller, as shown in Figure 3c, the frequencies ω_n^{TO} are red-shifted in a singular fashion and the spectral density increases, which allows for broadband light-harvesting. In the touching limit, the spectrum becomes continuous if we neglect the nonlocal effect. Thus, the TO description captures the singular behavior of dimer plasmons $|\omega_n^{TO}\rangle$.

We now turn to our model. Consider the trimer shown in Figure 1a. The plasmons of the trimer are specified as a superposition of the TO dimer plasmon of the pair (B_1, B_2) and that of the pair (B_2, B_3). We let (a_n, b_n) represent the following linear combination of the dimer plasmons: $a_n|\omega_n^{TO}(B_1, B_2)\rangle + b_n|\omega_n^{TO}(B_2, B_3)\rangle$. This hybridization of these two dimer plasmons

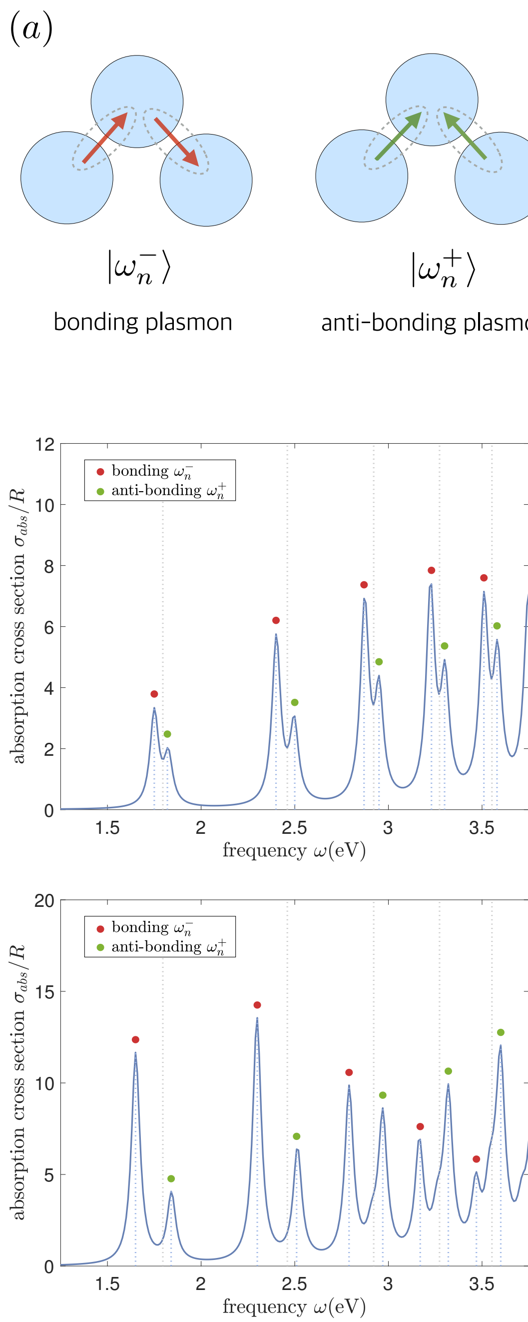

Figure 3: Trimer plasmons.

is characterized by the following coupled mode equations:

$$\begin{bmatrix} (\omega_n^{TO})^2 & \Delta_n \\ \Delta_n & (\omega_n^{TO})^2 \end{bmatrix} \begin{bmatrix} a_n \\ b_n \end{bmatrix} = \omega^2 \begin{bmatrix} a_n \\ b_n \end{bmatrix}.$$

Here, Δ_n represents the coupling between the two TO modes, its expression is given in the Supplementary Material. This coupled mode system is derived using the spectral theory of the Neumann-Poincaré operator and TO (see Supplementary Material). We emphasize that the above equation is a simplified version of our theory. Although we require additional TO modes for improved accuracy, we shall see that this simplified characterization can already capture some essential physics. Solving the equation, we obtain the hybrid modes for the trimer:

$$|\omega_n^{\pm}\rangle \approx \frac{1}{\sqrt{2}}\Big(|\omega_n^{TO}(B_1, B_2)\rangle \mp |\omega_n^{TO}(B_2, B_3)\rangle\Big),$$

and their resonance frequencies

$$\omega_n^{\pm} \approx \omega_n^{TO} \pm \Delta_n.$$

The TO resonance frequency ω_n^{TO} splits into two frequencies.

Our theory predicts that the density of the spectrum increases as the particles B_1 and B_3 become closer (but not too close). As the distance between the two gap-plasmons decreases, the coupling strength Δ_n increases. We call $|\omega_n^-\rangle$ and $|\omega_n^+\rangle$ the *bonding plasmon* and *anti-bonding plasmon*, respectively. These modes are very different from the bonding and anti-bonding modes of a dimer in the standard hybridization model. They are trimer plasmons and are capable of capturing the close-to-touching interaction via TO.

We validate our model with numerical examples. We set the radius of the particles to be $R = 50$ nm and the inter-particle gap distance to be $\delta = 0.25$ nm. Notice that the ratio δ/R is very small as the particles are nearly touching. We plot the absorption cross section for the trimer when the bonding angle is $\theta = 140°$ (weak coupling) in Figure 3b and $\theta = 50°$ (strong coupling) in Figure 3c, respectively. In the latter case, the coupling strength is stronger since the gaps are closer to each other. We also plot the values of ω_n^- and ω_n^+ (red and green circles) computed by a complete version of our theory. They are in excellent agreement with the location of local peaks. The gray dots represent the dimer frequency ω_n^{TO} computed using the TO approach. In Figure 3b and 3c, the splitting of the TO resonance frequencies ω_n^{TO} is clearly shown. In the strong coupling case (Figure 3c), the splitting is more pronounced resulting in a denser spectrum than that of the dimer case. Hence, the numerical results are consistent with the prediction of our proposed *Singular Hybridization Model*.

Next, we consider as an application the design of broadband metasurfaces, which is the second key part of this work. Recently, Pendry et al. proposed a broadband absorption metasurface based on geometrical singularities. They interpreted its broadband response as a realization of compacted dimensions. Our metasurface geometry, shown in Figure 4a, is a 1D periodic array consisting of two particles with different radii. This array is a combination of two sub-arrays, $B_1^{\#}$ which consists of larger particles, and $B_2^{\#}$ which consists of smaller particles.

To explain the motivation behind our metasurface, we begin by considering the array of larger particles $B_1^{\#}$. As the particles comprising $B_1^{\#}$ become closer, the spectrum becomes more dense, however, the effective gap distance cannot be smaller than 0.25 nm due to non-locality.

Our idea is to introduce the array of smaller particles $B_2^{\#}$ and position them close to $B_1^{\#}$.

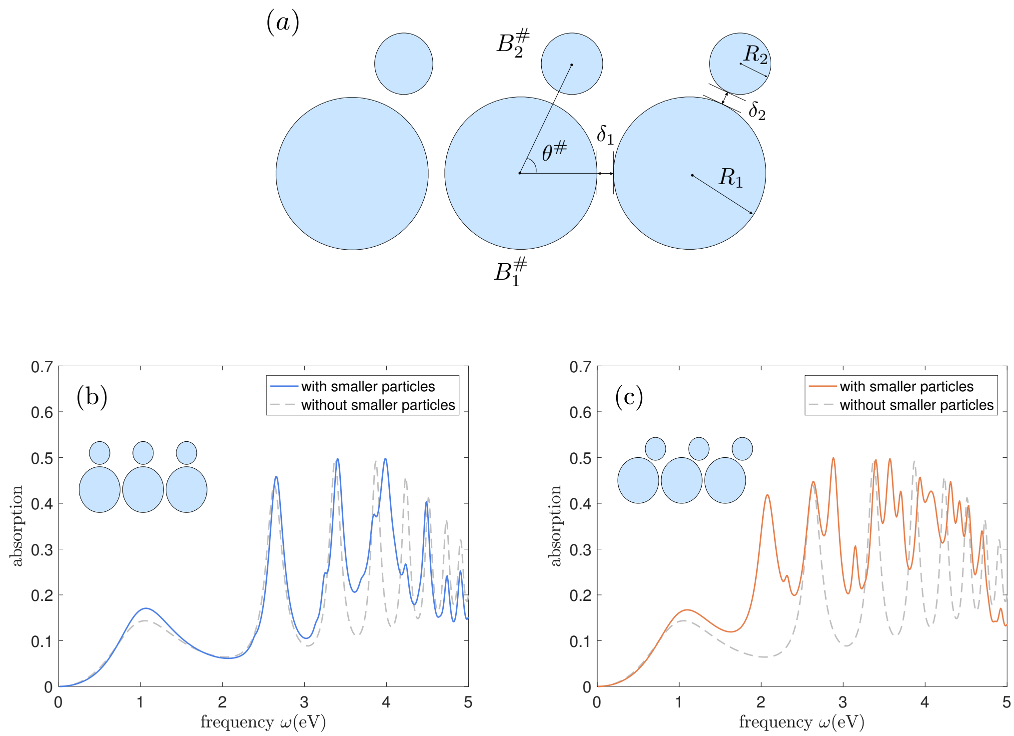

Figure 4: Metasurface.

This leads to the formation of singular plasmons in each gap. As in the case of a single trimer, it is natural to expect that the spectrum becomes more dense as the bonding angle $\theta^{\#}$ decreases. We verify this prediction with numerics. We set the radii to be $R_1 = 30$ nm, $R_2 = 23$ nm, and the inter-particle gap distances to be $\delta_1 = \delta_2 = 0.25$ nm. We consider two cases with different bonding angles: $\theta^{\#} = 90°$ (weak coupling) and $\theta^{\#} = 60°$ (strong coupling). We plot the absorption of the weak and strong coupling cases in Figures 4b and 4c, respectively, assuming normal incidence (see the Supplementary Material for the details). We also compare these results with the absorption of a metasurface consisting of only the larger particles $B_1^{\#}$ (gray dotted lines). Although this metasurface already features a relatively dense spectrum, the spectral density is enhanced due to the hybridization of singular plasmons by the introduction of the particles $B_2^{\#}$. In the weak coupling case (Figure 4b), the introduction of the array of particles $B_2^{\#}$ results in a somewhat minor alteration of the spectrum. In the strong coupling case (Figure 4c), however, due to the strong hybridization of singular plasmons, the spectrum clearly becomes significantly more dense.

Conclusion. In summary, we have proposed the *Singular Hybridization Model* for understanding singular plasmons of strongly interacting particles, and presented a new design principle for broadband metamaterials. Our model combines the power of both the hybridization picture and the TO approach. By controlling the hybridization of singular plasmons, we can circumvent the difficulty arising due to the non-local effect. Our proposed new metasurface design demonstrates that spectral density can be increased without requiring that the inter-particle gap distance becomes smaller.

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