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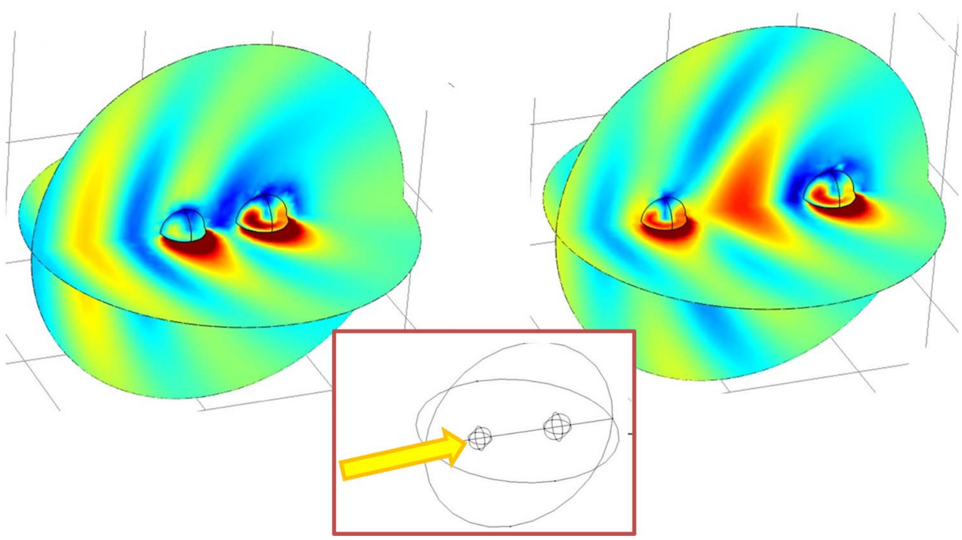

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Control of the Light Interaction in a Semiconductor Nanoparticle Dimer Through Scattering Directionality

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Abstract: Dimers of nanoparticles are very interesting for several devices due to the possibility of obtaining intense light concentrations in the gap between them. A dynamic control of this interaction to obtain either the maximum or minimum light through interferential effects could be also relevant for a multitude of devices such as chemical sensors or all-optical devices for interchip/intrachip communications. Semiconductor nanoparticles satisfying Kerker conditions present an anisotropic scattering distribution with a minimum in either the forward or the backward direction and prominent scattering in the contrary direction. The reduction or enhancement of the electromagnetic field in a certain direction can minimize or maximize the interaction with neighboring nanoparticles. In this paper, we consider a dimer of nanoparticles such that each component satisfies each one of the Kerker conditions. Depending on the arrangement of the nanoparticles with respect to the impinging light direction, we can produce a minimum or a maximum of the electric field between them, reducing or maximizing the interferential effects. The strong dependence of the directional conditions with external conditions, such as the incident wavelength, can be used to dynamically control the light concentration in the gap.

Index Terms: Nanophotonics and photonic crystals, semiconductor materials, backscattering, forward scattering.

1. Introduction

The interaction between resonant structures, both metallic and dielectric, is as important as resonances themselves for a large amount of applications. In fact, a lot of applications and devices based on nanoparticles exploit their mutual interaction with other elements, such as molecules or substrates. In particular, in the field of nanophotonics, we can find a myriad of examples. Simple dipole nanoantennas [1], complex Yagi-Uda antennas [2], and optical waveguides [3] are some of these examples for propagation effects. The interaction between nanoparticles has been also used to produce hotspots [4]. This strong concentration of the field is currently used to design ultra-sensitive biosensors [5] or optical nanotweezers [6] among other applications. The interaction between resonant nanoparticles is also present in the appearance of interesting effects like Fano resonances or to produce metamaterials [7]. Additionally, the interaction and coupling of light resonances of nanoparticles with other components (e.g., molecules or quantum dots) is also explored. Besides SERS (surface-enhanced Raman spectroscopy) techniques based on adsorbate-nanoparticle coupling [8], Förster resonance energy transfer (FRET) is other interesting process based on these effects. In this case, the emission of fluorophores is enhanced and quenched by the presence of plasmonic nanoparticles [9], [10]. In these works, the interaction between the components is mainly controlled through the distance between them, according to the intensity and range of the near field of the structures [11], [12].

New applications in the field of optical communications and future optical computing can exploit these interactions to obtain dynamic devices. One of the first examples of these devices is the optical switch [13]. Other recent works show that nanoscale optical switches based in semiconductor nanoparticles are achieved by using ultrafast photoinjection of electron-hole plasma in a single nanoparticle [14] or by two-photon absorption to switch the magnetic Mie resonance [15]. In this sense, we also proposed an all-optical switch based on the directional scattering of semiconductor nanoparticles [16]. This effect, experimentally demonstrated in [17] and [18], shows that the directional control over the scattered field can be achieved at certain wavelengths in dielectric nanoparticles, thanks to the coherent interaction between electric and magnetic resonances. The appearance of these effects can be governed by means of the geometrical conditions of the nanoparticles, such as the shape [19] and the size [20]. In our proposal, an optimum arrangement of two nanoparticles satisfying Kerker's conditions, one being the minimum forward (MF) and the other one the zero backward scattering (ZB), was analyzed to produce a maximum contrast of light in the gap region when the directional conditions are satisfied or not.

An anisotropic scattering of nanoparticles, involving that the scattered light if preferably directed in a particular direction, also induces a different way of interaction with neighboring structures. In a dimer of nanoparticles, and taking into account the background field (i.e., the part of the incident field that is not scattered neither absorbed), two possible interferential effects could appear: the one between the scattered field of each nanoparticle and the background incoming field and the one between the scattered fields of each component of the dimer. Then, a series of constructive and destructive interferences appears when both nanoparticles get away from each other. If these nanoparticles may satisfy one of the Kerker conditions, we can manipulate these interferences, drastically changing the spatial distribution of light. In this work, we explore the evolution of the interferential effects between the different electromagnetic fields present in a dimer of semiconductor nanoparticles with directional scattering. The main score of this contribution is the control over the different interferences such that a maximum variation of light can be achieved in the gap region when the directional conditions are satisfied or not. Obtaining a measurable variation, the geometry can be used as the base for the design of devices like the proposed all-optical switch. In addition, a deep knowledge of the spatial distribution may be used to produce maximum hot-spots performing integrated, complementary metal-oxide semiconductor-compatible ultrasensitive chemical sensors.

2. Theoretical Background

As stated above, the interaction between small scatterers is a key feature for several applications. The control of the scattered field of these systems potentially may improve this interaction and hence their characteristics. This control could be performed through the two most important effects on the interaction between neighboring scatterers: the distance between them and the spatial distribution of their scattered fields. Current technologies allow a precise control over the distances. On the other hand, at the beginning of the 1980s, Kerker and co-workers showed up that the control of spatial distribution of the scattered fields can be obtained. They considered spherical particles in the Rayleigh limit, presenting both electric and magnetic response, and under certain conditions [21]. This study was based on the analysis of the relation between the scattering coefficients of the Mie theory. As it is well known, Mie theory uses a multipolar decomposition to describe both the scattering and absorption cross sections. In fact, the extinction (scattering+absorption, C_{ext}), scattering (C_{sca}) and absorption (C_{abs}) cross sections of a homogenous and isotropic spherical particle with a radius R illuminated by a linearly polarized plane wave with a wavelength λ are given by [22]

$$\begin{aligned} C_{\text{ext}} &= \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)\text{Re}\{a_n+b_n\} \\ C_{\text{sca}} &= \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)\left(|a_n|^2+|b_n|^2\right) \\ C_{\text{abs}} &= C_{\text{ext}} - C_{\text{sca}} \end{aligned} \tag{1}$$

where k is the wavenumber of the incident beam $(k = 2\cdot\pi/\lambda)$, and a_n and b_n are the n-polar Mie coefficients.

The multipolar character of this theory is related with the Mie coefficients, in such a way that a_n coefficients are usually associated to the electric behavior, while b_n coefficients corresponds to the magnetic one. Additionally, first order coefficients (a_1, b_1) are related to the dipolar character, while second order coefficients refer to the quadrupolar phenomena, and so on.

Considering dipole-like particles, only the two first Mie coefficients $(a_1,\ b_1)$ are not negligible. Under this assumption, the particle interaction with light can be modeled as two superimposed dipoles, an electric dipole and a magnetic one. Kerker and co-workers [21] realized that these dipoles can interfere producing a zero scattering in either the forward or the backward direction. Those two interferential conditions, known as Kerker's conditions, are given, respectively, by

$$a_1 = -b_1 \tag{2}$$

$$a_1 = b_1. \tag{3}$$

This study remained in oblivion due to the impossibility of finding natural materials with electrical and magnetic response in the visible range. Nowadays, this fact was overcome with the discovery of electric and magnetic resonances in high-refractive-index semiconductor nanoparticles (e.g. Si, Ge, etc.) [23]. Departing from this analysis, the spatial distribution of the scattered field of semiconductor nanoparticles satisfying these conditions can be studied through several complex approaches, e.g., a coupled electric-magnetic dipole approach. However, in order to avoid loss of information, in this work, we consider FEM simulations, solving the Maxwell equations in the considered geometry.

3. Geometrical Conditions of the Considered System

The considered system is composed of two semiconductor nanoparticles, in particular silicon spheres, separated a distance d. For the sake of clarity, Fig. 1 shows this geometry and a view of the near field scattered by each single nanoparticle obtained by FEM calculations (COMSOL Multiphysics). Particle sizes are such that the first particle $(R_1 = 82\text{ nm})$ satisfies the zero-backward (ZB) scattering condition, while the second one $(R_2 = 97\text{ nm})$ fulfills the minimum forward scattering condition (MF) when the incident wavelength is 700 nm. As can be seen, the ZB condition is well defined and there is practically no scattered electrical field in the backward direction. Otherwise, MF condition involves a drastic reduction of the scattered field in the forward direction, but there is still an appreciable electric field. These distributions show that nanoparticles can direct the scattered radiation in a very selective way, and when using them in a dimer, the overlapping of the fields can be very important.

Depending on the impinging direction of light and considering the anisotropic scattering of both nanoparticles, scattered light can be mainly directed outwards the gap [see Fig. 1(a)] or towards

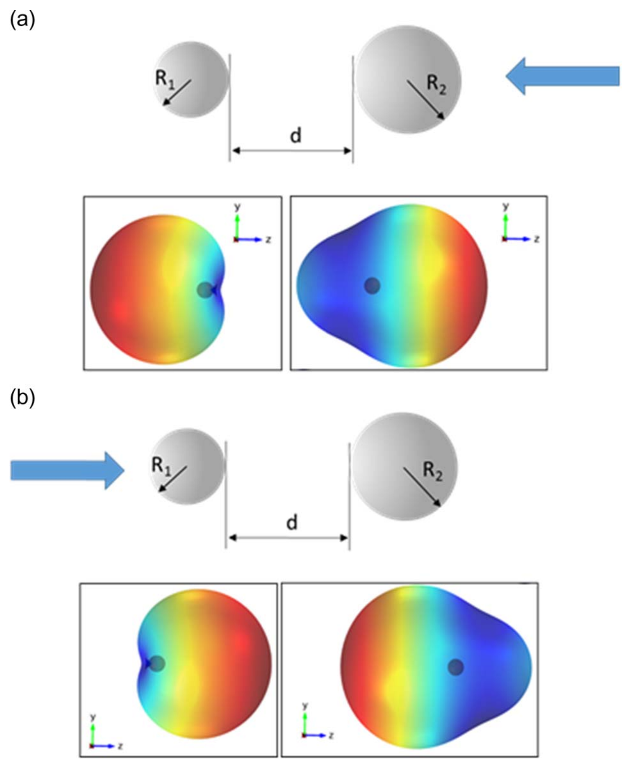

Fig. 1. Scheme of the considered geometry. The system is composed of two silicon nanoparticles located at a distance *d* between them. The particle sizes are such that, while one nanoparticle has zero backscattering, the other one satisfied the MF scattering at the same incident wavelength ($\lambda = 700$ nm). The bottom figures show the 3-D spatial distribution of the light scattering of each isolated nanoparticle, with blue being the lowest intensity and red being the highest intensity. Depending on the incident direction, from right to the left (a) or *vice versa* (b), the light concentration on the gap drastically changes.

this region [see Fig. 1(b)]. Thus, a maximum or a minimum of the scattered field could be observed in the midst point of the system. While the first case [see Fig. 1(a)] is called "direct/0°" configuration, the second configuration [see Fig. 1(b)] is referred as "inverse/180°" configuration. The incident beam of 1 V/m is considered.

As it was previously commented, the gap distance in this geometry is a key point. For very low values ($d \ll R$), the individual scattering profiles of each nanoparticle are avoided. On the contrary ($d \gg R$), nanoparticles are considered as isolated and there is no interaction between them, thus the scattering profile should be similar to those shown in Fig. 1. Distances in the middle range allow the appearance of interferential phenomena, which control is the objective of the present work.

4. Results

In a previous study, we showed that the directionality of light scattering of semiconductor nanoparticles can be used to produce either "hot-spots" or "dark-spots" in a dimer geometry [16]. This effect, together with the strong sensitivity of Kerker's conditions with the wavelength, allows the design of a new generation of nanodevices for futuristic applications, such as an all-optical nanoswitch [16]. In addition, the concentration or lack of the scattered light in the gap region affects to the interaction between the nanoparticles, in particular due to interferential phenomena. Coming up next, we show that the analysis of this interaction is fundamental to obtain an optimized maximum/minimum intensity of the total field in the gap region of the dimer.

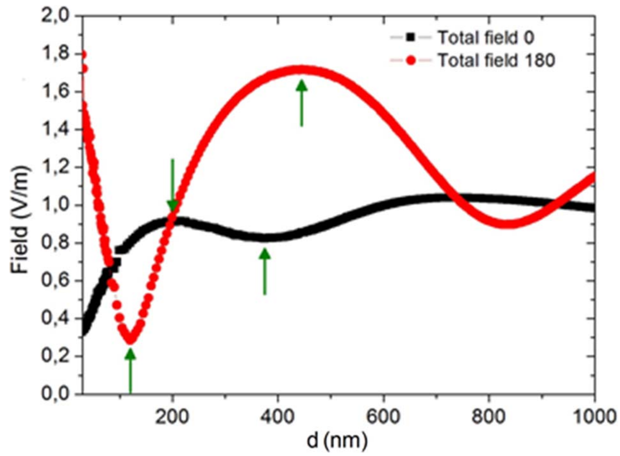

Fig. 2. Total electric field (scattered + background) at the middle point of the silicon dimer nanoswitch as a function of the distance between nanoparticles. Incident light impinging at (black squares) the direct/0° and (red circles) the inverse/180° configurations. Green arrows highlight the gap distances that will be considered hereinafter.

4.1. Gap Distance Effect

For this study, we consider a dimer of nanoparticles such that the Kerker's conditions are satisfied at an incident wavelength of $\lambda = 700$ nm, as it was shown in Fig. 1. Fig. 2 shows the numerical calculation of the modulus of the total electric field (scattered+background) in the middle point between the two nanoparticles composing the dimer as a function of the gap distance (d) between them and for both considered configurations, direct/0° (black squares) and inverse/180° (red circles). An incident beam of 1 V/m has been considered, so every point under 1 is considered as attenuating the incident field, and over 1 reinforcing it. As it was previously explained, for the direct/0° case [see Fig. 1(a)] a minimum of the electric field is expected in the midst point of the gap because the scattered field is directed outwards, while the inverse/180° configuration [see Fig. 1(b)] produces a maximum in the gap region due to the directionality of light into the gap. The change between the interferential profiles of both configurations is clearly observed, with a significant change in the visibility (V) of the interferences.

The lack of scattered field in the gap region in the direct/0° arrangement (dark squares) involves a weak interaction between both nanoparticles and then a weak interference, with a low visibility of it. When gap distances are quite small, the inability of the incident field to penetrate into the gap, together with the absence of scattered fields due to the Kerker's conditions, produce a pronounced minimum of the electric field in the midpoint, as expected. The drawback of this gap distance range is the fabrication and physical complexity as the nanoparticles become closer [24]. After a complete sweep of the gap distance, we considered that the gap distance limit of our approach without losing noticeable phenomena is located around 40 nm. As the gap distance increases over this limit, the intensity of the light in the gap also increases as the incoming light enters into it. In addition, as the minimum forward condition cannot provide a zero scattering [23], MF nanoparticle is driving some scattered field into the gap (bottom Fig. 1(a), particle on the right side), and then, a slight variation of light intensity can be modulated by its interference with the incident field. However, its visibility is quite small, as can be seen in Fig. 2. As a consequence, at distances at which an interference can be observed between the scattered and the incident fields, a minimum, corresponding with a destructive interference, is obtained at $d = 375$ nm and a maximum, a constructive interference, is seen at $d = 200$ nm (green arrows point out these conditions in Fig. 2). However, the difference between the maximum and minimum value is less than a 9%.

Both situations can be clearly observed in the 3-D distribution of the electromagnetic field around the system in the destructive [see Fig. 3(a)] and constructive interference [see Fig. 3(b)].

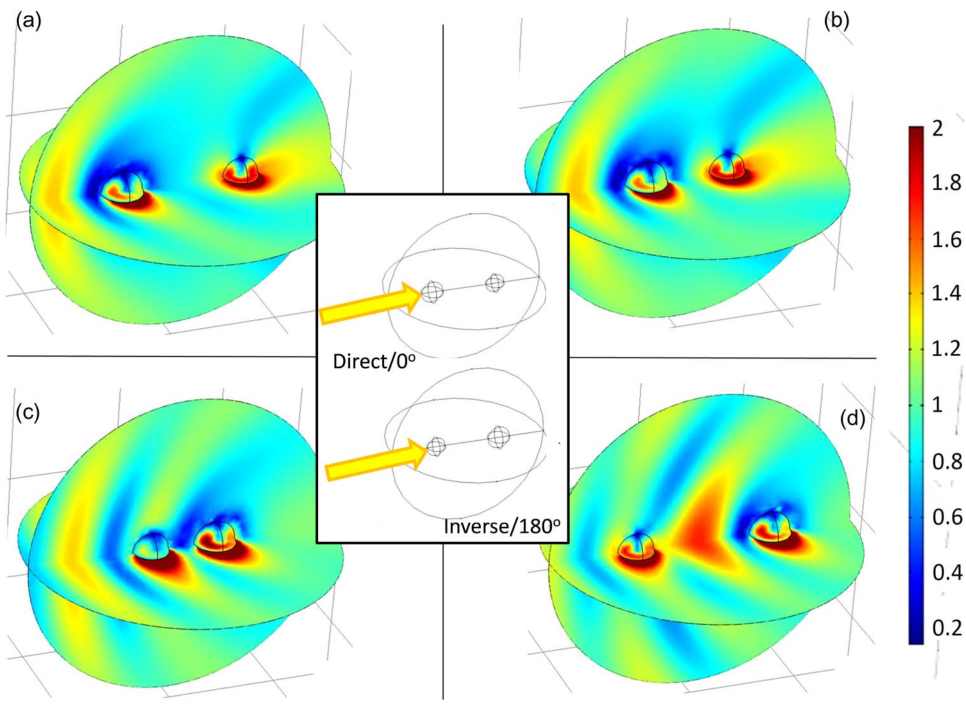

Fig. 3. Distribution of the electric field in the incident and orthogonal (including the middle point of the gap) planes in the region between the nanoparticles considering an incident beam of $\lambda = 700$ nm. The geometry and view are shown in the center. (a) and (b) Direct/0° configurations with gap distances of $d = 375$ nm and $d = 200$ nm, respectively. (c) and (d) Inverse/180° configurations with gap distances of $d = 120$ nm and $d = 445$ nm, respectively.

As can be seen, although there is a variation between these two cases, it is very slight, and the electric field distribution is almost unaltered as the distance changes in this configuration.

On the other hand, considering an inverse/180° arrangement the situation is very different. According to Fig. 1(b), the contrary disposition of each particle, regarding the incoming light, directs the light scattered from one particle towards the other one. This involves a high concentration of the electric field in the gap region and then a strong interaction between both scattered fields. The result of this strong interaction is observed in Fig. 2 (red circles) through remarkable minimum and maximum, resulting of an intense either destructive or constructive interference. For very small distances, the strong interaction of the nanoparticles produces an intense hot-spot in the middle point of the gap. As the gap distance increases and the incident field enters into the gap region, the interferential behavior is noticeable, with the first destructive interference appearing at $d = 120$ nm while a first maximum, or constructive interference, is located at $d = 445$ nm (again two green arrows point out them). In this case, the contrast between both cases is very remarkable, reaching a 140% of the incident field. This can be also clearly observed in the 3D near-field distribution of the electric field, see Fig. 3(c) and (d). In addition, the strong interaction enables the appearance of interferential effects at larger distances than in the other arrangement. However, these ones are less important than those at shorter distances.

4.2. Optimizing the Contrast: Wavelength Effects

As it was previously explained, the sensitivity of the Kerker's conditions [see (2) and (3)] with the incident wavelength makes this parameter a convenient one to control the spatial distribution of the scattered field. Although the wavelength control is not trivial experimentally, in this section, we explore the changes of the light concentration in the middle point of the dimer as a function of a slight variation of the incident wavelength, because Kerker's conditions are no

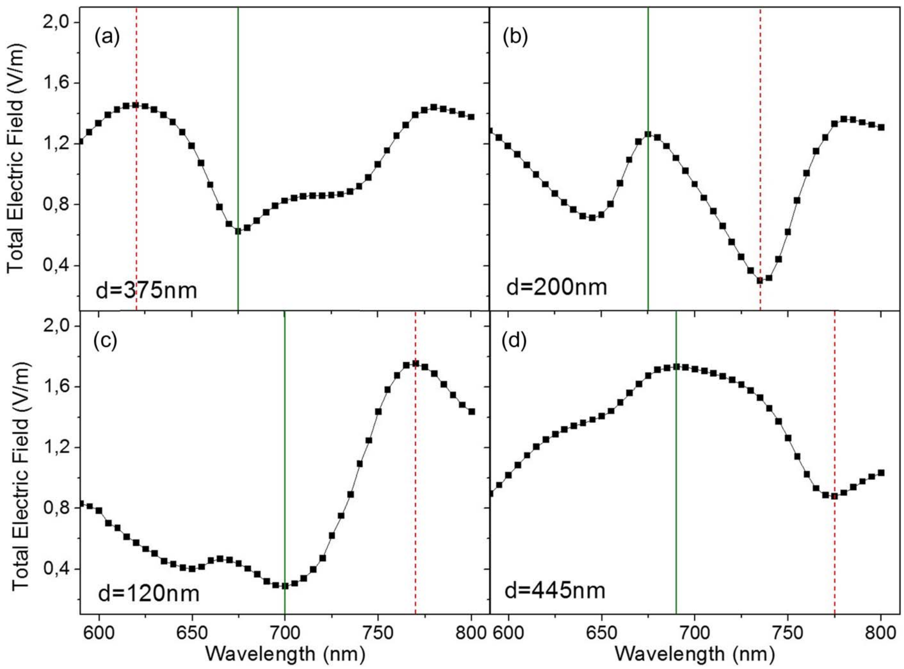

Fig. 4. Total electric field (scattered + background) at the middle point of the considered dimer as a function of the incident wavelength around the target one $(\lambda = 700\ \text{nm})$ for the remarkable gap distances observed in Fig. 2. (a) and (b) Distances of the minimum (375 nm) and maximum (200 nm) of the total electric field at the direct/0° configuration, respectively. (c) and (d) Distances providing a minimum (120 nm) and a maximum (445 nm) of the total electric field at the inverse/180° configuration, respectively. The vertical lines highlight the optimum states concerning the contrast.

longer satisfied. In particular, our aim is the optimization of the dimer structure to obtain a maximum contrast between the two above remarked states, taking advantage of either a small or a strong interaction between the nanoparticles. In this optimization, we explore several parameters such as the optimum arrangement or the gap distance and search for a pair of incident wavelengths maximizing this contrast.

The previous section showed that, although we expected a minimum concentration in the direct/0° configuration [see Fig. 1(a)] and a maximum in the inverse/180° arrangement [see Fig. 1(b)] due to the directionality conditions, the interferential phenomena highlight two other gap distances producing the inverse results, a maximum and a minimum electric field intensity, respectively. Thus, hereinafter, we will analyze these four geometries (two gap distances per each arrangement) in order to determine the optimum one. Accordingly, a numerical analysis of the total electric field in the gap as a function of the incident wavelength around the target one $(\lambda = 700\ \text{nm})$ has been made at the four interesting distances listed before (see green arrows in Fig. 2).

Fig. 4 shows the modulus of electric field in the middle point of the gap as a function of incident wavelength for the four considered cases. While Fig. 4(a) and (b) correspond to the direct/0° configuration, Fig. 4(c) and (d) are those corresponding with the interesting distances at the inverse/180° configuration (in accordance with Fig. 3(a)–(d) configurations, respectively). According with the defined geometry of each figure and following the results of Fig. 2, either a maximum or a minimum is expected at 700 nm. However, a slight wavelength blue-shift is observed for these maxima/minima due to the joint effect of both the directional scattering conditions and the interferential phenomena (green solid lines). This shift is always smaller than 25 nm and it is more pronounced in the case of the direct/0° configuration.

The considered wavelengths producing the expected maximum/minimum in each case are highlighted with a solid vertical line. As explained, we want to obtain a strong contrast of the

TABLE 1

CP values of the considered configurations. *d* is the gap distance

Direct Configuration	Inverse Configuration
14.50 (d = 200 nm)	15.13 (d = 120 nm)
9.40 (d = 375 nm)	5.63 (d = 445 nm)

light intensity in the gap region by changing the incident wavelength. Consequently, directional conditions could not be fulfilled and/or an opposite interference may appear. Thus, we explore the minima/maxima at wavelengths close to the previous ones. The evaluation of this contrast, in order to find convenient wavelength shifts, takes into account two important magnitudes: the variation of the intensity of the electric field, ΔE, and the wavelength range between extreme values, $\Delta\lambda$. In this sense, we define the following parameter, which is called the contrast parameter (CP)

$$\mathrm{CP} = \frac{\frac{\Delta E}{\overline{E}}}{\frac{\Delta\lambda}{\overline{\lambda}}} \tag{4}$$

where $\Delta\lambda = \lambda - \lambda_0$ and $\Delta E = E - E_0$. λ_0 and E_0 are the incident wavelength and the electric field at the midst point of the dimer at the considered maximum/minimum described above (solid lines in Fig. 4). On the other hand, λ and E correspond to the incident wavelength and the electric field in the midst point, respectively, when the distribution of the electric field has changed enough to produce the opposite case (minimum/maximum; see vertical dashed lines in Fig. 4). $\overline{\lambda}$ and $\overline{E}$ are the average wavelength and electric field, respectively, between the two previous extreme values, which means that $\overline{\lambda} = (\lambda + \lambda_0)/2$ and that $\overline{E} = (E + E_0)/2$.

At first sight from Fig. 4, the contrast between the values of the electric field in the gap region seems better for short distances [see Fig. 4(b) and (c)], where scattered fields dominate and the effect of the incident field is still small. Furthermore, comparing the two possible arrangements, the direct/0° [see Fig. 4(a) and (b)] and the inverse/180° configuration [Fig. 4(c) and (d)], no remarkable difference appears. However, from a practical point of view, we are also interested on the wavelength shift to produce this variation: the smaller the shift, the higher the operation speed of a possible device based on this dimer. For this reason, we included this effect in the definition of the parameter CP. Following these criteria, we found the corresponding wavelengths related to maximum values of CP at each considered configuration, which are the vertical dashed red lines highlighted in Fig. 4. The values of the CP parameters are summarized in Table 1.

The most remarkable contrast is observed in the inverse/180° configuration [see Fig. 4(c)]. In this case, the destructive interference at the target wavelength $(\lambda_0 = 700\ \mathrm{nm})$ produces a minimum value of the electric field in the middle point of the gap $(E_0 = 0.28\ \mathrm{V/m})$. A shift from 700 nm to 770 nm produces a 6-fold change of the electric field ($E = 1.75$ V/m for a CP of 15.13). It is a paradox that the best value of CP corresponds to a configuration that was not considered in previous works since under this arrangement a maximum of the electric field in the gap was expected. This highlights the importance of the interferential effects and the coupling between nanoparticles in the optimization of this system. The intense presence of the scattered fields in the gap region due to their directionality generates strong destructive interferences producing sharp minima and a convenient contrast.

There is another case presenting a similar value of CP, this corresponds with the gap distance producing a constructive interference in the direct/0° configuration [see Fig. 4(b)]. Again, we obtain a high contrast (CP 14.50) in a geometry in which the interference is as important as the directionality of the scattered fields. In addition, in this case there is another set of possible working conditions producing an optimum contrast. This appears at large wavelengths:

switching from 735 nm to 780 nm, and it has a CP of 21.50. Although it has the highest value of CP, the operation wavelengths are displaced enough from the target wavelength to avoid the satisfaction of the directionality conditions. For this reason, a deeper study of this situation is necessary.

The other two considered cases in Fig. 4 have low values of the contrast parameter: 9.40 and 5.63. The most important handicap of these cases is the wide wavelength shift that is necessary to have a large contrast and also the large gap distance between the nanoparticles, allowing the stronger effect of the incident field and the reduction of the contrast.

5. Conclusion

The importance of the interferential effects between the electromagnetic fields of neighboring nanostructures is a key feature in several applications due to the appearance of "*hot-spots*". In this work, we analyzed the interferential effects between two semiconductor nanoparticles satisfying the directional scattering conditions. In particular, we consider a dimer of silicon nanoparticles with radius of $R = 82$ nm and $R = 97$ nm, such that the zero-backward and the minimum forward conditions are satisfied at $\lambda = 700$ nm, and separated a certain distance.

By analyzing the joint effect of both the scattering directionality and the interference between either the scattered fields or the incident and the scattered fields, we observed that the directional conditions may enhance or reduce the interferential coupling between the nanoparticles. In this sense, we showed that a certain arrangement, direct/0° configuration, has no interesting interferential effects between the components of the dimer, while the opposite one, inverse/180°, presents remarkable interferential bands.

In addition, we showed the influence of a slight shift of the wavelength on the concentration of light in the gap region, such that the directional conditions are not satisfied anymore. In particular, we looked for a geometrical configuration such that a maximum variation of the intensity of the light in the gap is observed under a shift of the incident wavelength. The optimum conditions in terms of contrast correspond to a geometrical arrangement (inverse/180° and gap distance of 120 nm) producing intense electromagnetic fields in the gap region, but the selected wavelength range (from 700 nm to 770 nm) is taking advantage of the destructive interference of these fields. This highlights the need to take into account both phenomena in these studies: directionality and interference. In addition, a pure interaction of the scattered fields produces better contrast situations, for this reason short distances are preferred than large distances, at which the penetration of the background field in the gap region is not negligible.

Another interesting case was also found. A significant contrast of CP is obtained for a direct/0° arrangement with a gap distance of 200 nm, and a wavelength shift from 735 nm to 780 nm. However, a deeper study of this case is necessary to understand the relative importance of the directionality on it.

In summary, it is demonstrated that the dimer geometry can be optimized to obtain a high contrast in the gap region considering a slight wavelength shift. The joint effect between interferential and directional phenomena to produce intense or negligible intensities of the electromagnetic field is of great interest for several photonic devices such as ultra-high sensitive sensors, optical tweezers, or optical switches.

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