NANO LETTERS

2008
Vol. 8, No. 8
2369-2372

Frequency-Controlled Localization of Optical Signals in Graded Plasmonic Chains

A. V. Malyshev,[†,‡,§] V. A. Malyshev,[†] and J. Knoester*[,†]

Centre for Theoretical Physics and Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands, and GISC, Departamento de Física de Materiales, Universidad Complutense, E-28040 Madrid, Spain

Received April 28, 2008; Revised Manuscript Received June 9, 2008

ABSTRACT

We study theoretically the optical response of graded linear arrays of noble metal nanospheres in which the center-to-center distances and/or the radii of the spheres change linearly along the chain. A strong asymmetry of the system response with respect to the direction of incidence of the incoming light is revealed. We show that for light propagating from smaller to larger spheres the optical signal can be localized in a controlled way at an arbitrary subset of a few neighboring spheres by adjusting the wavelength of the incoming field. This opens new opportunities to control the flow of electromagnetic energy at the nanometer scale.

Recent advances in nanotechnology allow one to structure and characterize metals at the nanometer scale. Arrays of metallic nanoparticles (referred to as plasmonic arrays), whose optical excitations are surface plasmon polaritons, are widely recognized as potential building blocks for nanoscale optical circuits. Particular interest in plasmonic arrays has been sparked by Quinten et al.[1] and Brongersma et al.[2] who suggested that such systems may be used for guiding optical signals at a subwavelength scale. During the past decade, this branch of surface plasmon physics experienced an enormous development[3–17] (see ref 18 for an overview). Nanofocusing of optical energy in short graded plasmonic chains,[4,5] plasmon antennae,[8–10] tunable nanoscale switching of the transmittance of plasmon particle arrays,[11,13] and guiding the signal over a large distance[16] have been investigated.

Clearly transmission and guiding of electromagnetic energy on a subwavelength scale is important for photonic applications. However, it would open an even more exciting perspective for optical circuits if we could localize the guided signal at any given point of a plasmonic array. This would, for instance, offer the opportunity to branch electromagnetic energy into other waveguides in a controlled way. In this communication, we show that controlled localization of the optical signal is possible in graded noble metal nanoparticle chains with a linear variation of nanoparticle sizes and center-to-center distances. This variation gives rise to a gradient of the nanoparticle polarizabilities and resonance frequencies, which drastically affects the response of the plasmonic chain. In particular, if the incident field propagates in the direction from the smallest to the largest sphere, the optical signal undergoes strong localization on only a few particles. The location of the wave packet can be controlled by the wavelength of the incoming field; for typical system parameters, the control region covers almost the entire visible spectrum. The system operates therefore as a broadband antenna with selective localization of the response signal.

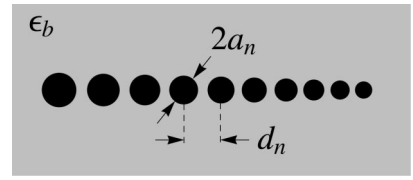

Figure 1. Schematics of a graded plasmonic array composed of spherical metal nanoparticles with variable radii a_n and center-to-center distances d_n. The array is embedded in a dielectric medium with background dielectric constant ε_b. We consider two types of graded arrays: (i) with linearly varying radii and constant center-to-center distances (simple graded array) and (ii) with linearly varying radii a_n and distances d_n, keeping the ratio a_n/d_n constant (invariantly graded array).

Figure 1 presents the schematics of a linear graded plasmonic array: a linear chain of N spherical metal nanoparticles of radii a_n located at positions x_n ($n = 1,..., N$). We focus on two types of plasmonic chains. In type (i), the simple graded array, the radii a_n vary linearly with n while the center-to-center distance $d_n = x_{n+1} - x_n$ is the same for

* Corresponding author.
† University of Groningen.
‡ Departamento de Física de Materiales Universidad Complutense.
§ On leave from Ioffe Physiko-Technical Institute St. Petersburg, Russia.

10.1021/nl8011962 CCC: $40.75
Published on Web 07/22/2008

all n. In type (ii), the invariantly graded array, both a_n and d_n vary linearly with n, in such a way that the ratio a_n/d_n is kept constant.

We will assume that the array is embedded in a homogeneous dielectric background with a dielectric constant ε_b (which is assumed to be a smooth function of frequency in the vicinity of the plasmon resonances of the particles). We consider a linearly polarized plane wave excitation $E = E_0 \exp(ikx)$ with a constant amplitude E_0 and a wave vector $k = 2\pi\sqrt{\varepsilon_b}/\lambda$ (λ is the wavelength in vacuo) propagating along the chain, so that only transverse plasmon modes polarized along the incoming field are excited. The generalization to arbitrary angles of incidence is straightforward. The ratio a_n/d_n is chosen to obey $a_n/d_n \leq 1/3$, allowing us to use the point dipole approximation. The validity of this approximation has been investigated theoretically in ref 19. The authors calculated the surface plasmon dispersion relations for a chain of metallic nanoparticles as a function of a/d and showed that, for $a/d \leq 1/3$, the point dipole approximation gives results indistinguishable in graphs from those calculated including up to 80 multipoles in the expansion of the secondary field. Moreover, in ref 13, the point dipole approximation was demonstrated to fit very well the experimental data on frequency driven switching in a linear plasmonic array of 10 silver nanoparticles with $a =$ 55 nm and $d = 150$ nm.

The particles are characterized by their polarizabilities $\alpha_n(\omega)$, where ω is the frequency of the incident field. Under steady state conditions, the set of coupled equations for the induced dipole moments p_n of spheres reads:

$$\sum_m \left(\frac{1}{\varepsilon_b \alpha_n}\delta_{nm} + G_{nm}\right) p_m = E_n \tag{1}$$

Here, $E_n = E_0 \exp(ikx_n)$ is the incident field at the position x_n of the nth dipole and the matrix G_{nm} is the retarded Green's function of the electric field in the dielectric background.[20] Thus, the term $\Sigma\, G_{nm}p_m$ represents the field of all other dipoles at the position n. The Green's function G_{nm} for the geometry under study is given by (see, e.g., ref 11)

$$G_{nm} = \frac{1}{\varepsilon_b}\left(\frac{k^2}{|x_n - x_m|} + \frac{ik}{|x_n - x_m|^2} - \frac{1}{|x_n - x_m|^3}\right)\exp(ik|x_n - x_m|) \tag{2}$$

while $G_{nn} = 0$. We take the polarizability of the nth sphere in the form

$$\frac{1}{\alpha_n} = \frac{1}{\alpha_n^{(0)}} - \frac{k^2}{a_n} - i\frac{2}{3}k^3 \tag{3}$$

where $\alpha_n^{(0)}$ is the bare polarizability of the nth sphere, the term k^2/a_n describes the depolarization shift,[21] while $(2/3)ik^3$ accounts for the radiative damping, which is important for energy conservation (see, e.g., the discussion in ref 16). The bare polarizability $\alpha_n^{(0)}$ is expressed in terms of the dielectric constant of the particle material, ε_n, as follows:[22,23]

$$\alpha_n^{(0)} = a_n^3 \frac{\varepsilon_n - \varepsilon_b}{\varepsilon_n + 2\varepsilon_b} \tag{4}$$

where the dielectric constant is chosen in a generalized Drude form [12,15]

$$\varepsilon_n = \varepsilon - \eta\frac{\omega_p^2}{\omega^2 - i\omega\gamma_n} \tag{5}$$

Here, ω_p and γ_n are the plasma frequency and Ohmic damping (collision frequency) in the bulk metal, while ε and η are adjustable parameters.

Throughout this communication, silver nanospheres are considered. Using the parameters obtained in refs 12 and 15, we thus have $\varepsilon = 5.45$, $\eta = 0.73$, $\omega_p = 1.72 \times 10^{16}$ rad/s, and $\gamma_n = \gamma = 8.35 \times 10^{13}$ 1/s (the correction of the damping rate because of the finite size of the sphere is neglected). The above parameters provide a good fit to tabulated optical data for silver[24] throughout the visible and infrared. We address chains of nanospheres with radii a_n decreasing linearly with n from 50 to 25 nm, sizes that are quite typically used in experiment. In the case of a simple graded array, we use a center-to-center distance $d_n = d =$ 150 nm, while for the invariantly graded array $d_1 = 150$ nm and the ratio radius/center-to-center-distance is kept to be $a_n/d_n = 1/3$. The dielectric constant of the host medium is chosen to be $\varepsilon_b = 2.25$.

Figure 2 shows the wavelength dependence of the response (distribution of the square moduli of the induced dipoles, $|p_n|^2$) calculated for the incoming field propagating in two opposite directions: from the largest to the smallest sphere (upper panel) and vice versa (lower panel). The propagation direction is indicated by arrows in the figure. The figure demonstrates a strong dependence of the array response on the direction of incidence. This property opens the opportunity to design various "asymmetric" devices, such as an optical diode or sensors with anisotropic response characteristic. Apart from the asymmetry, the behavior of the response resembles the one that was predicted in ref 11 and demonstrated experimentally later[13] for a homogeneous plasmonic chain of 10 identical silver nanospheres. In those papers, the $|p_n|^2$ distribution was found to localize either at

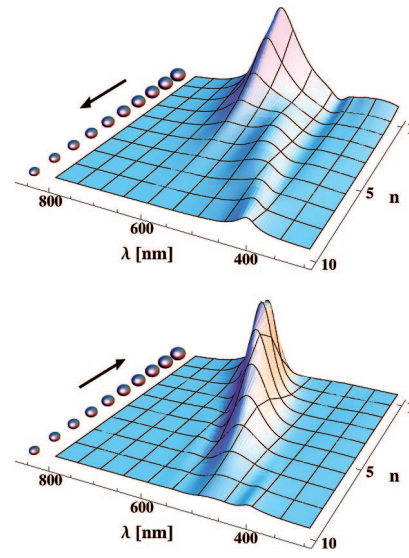

Figure 2. Wavelength dependence of the response ($|p_n|^2$) for a simple graded plasmonic chain of $N = 10$ silver nanospheres calculated for two directions of incidence of the incoming field indicated by the arrows. The radii a_n decrease linearly with n from 50 to 25 nm. The center-to-center distance $d_n = 150$ nm.

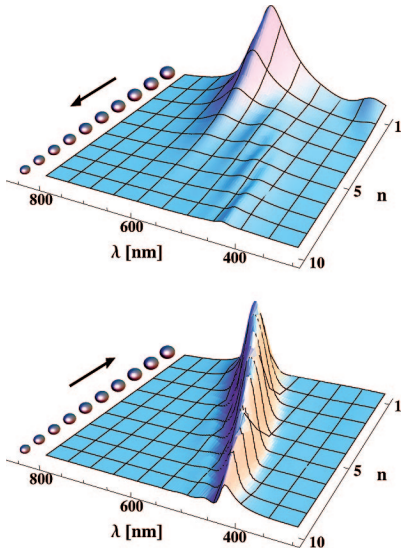

Figure 3. Same as in Figure 2 calculated for an invariantly graded plasmonic chain with radii varying linearly from 50 to 25 nm, $d_1 = 150$ nm, and $a_n/d_n = 1/3$.

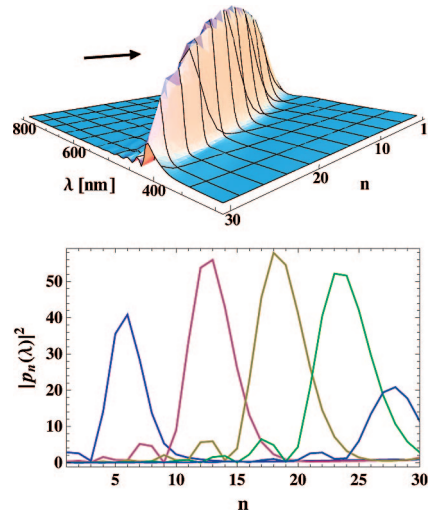

Figure 4. Upper panel: same as in the lower panel of Figure 3 calculated for an invariantly graded plasmonic chain of $N = 30$ silver nanospheres with radii varying linearly from 50 to 25 nm, $d_1 = 200$ nm, and $a_n/d_n = 1/4$. Lower panel: cross sections of the response $|p_n|^2$ from the upper panel. The curves (from left to right) correspond to wavelengths λ ranging from 650 to 450 nm in steps of 50 nm.

the first or at the last sphere, depending on the excitation wavelength. In the case of the simple graded array, however, the signal can also be localized at the inner part of the chain, which was not possible for the homogeneous array. Still, as is observed, the localization is not very pronounced.

The situation changes drastically for the invariantly graded array (see Figure 3). When the incoming light propagates in the direction from the smallest to the largest sphere (lower panel of Figure 3) a strong localization of the response signal is observed. As a matter of fact, the signal can be localized at an arbitrary subset of a few neighboring spheres by tuning the excitation wavelength λ.

It should be stressed that the observed localization can not be explained by sheer difference in the resonance plasmon frequencies of the spheres. Rather, the controlled localization as well as the asymmetry of the response arises from an interplay of the resonance and interference effects, as we argue in the following. Consider the homogeneous system of equations corresponding to eq 1: $\Sigma[(\varepsilon_b\alpha_n)^{-1}\delta_{nm} + G_{nm}]p_m = 0$. Although the matrix $(\varepsilon_b\alpha_n)^{-1}\delta_{nm} + G_{nm}$ is not Hermitian, it can be diagonalized. All eigenvectors of the matrix are localized in space due to the strong size dependence of the polarizabilities (see eq 3 and eq 4) that appear on the diagonal of the matrix. The solution of the system eq 1 is therefore an expansion of the plane wave in its right-hand side in terms of these localized eigenvectors. It is clear then that expansions of two plane waves propagating in opposite directions are not equivalent, as they acquire different spatial phases $\pm kx_n$, which results in the observed asymmetry of the response. Moreover, because k is the wavevector of the incoming light, this set of phases can be changed by the excitation frequency, providing therefore a mechanism to control the response signal.

The response of graded arrays can be modified by several parameters that can easily be adjusted in experiment. In particular, the radii of the spheres determine their resonance frequencies, and therefore, the total width of the response band can be tuned by the amount of radius grading. On the other hand, the interparticle distance determines the interaction between particles and also affects the shape of the response characteristic.

To demonstrate the latter, we considered an invariantly graded chain of $N = 30$ nanospheres with radii varying linearly from 50 to 25 nm, taking $d_1 = 200$ nm and $a_n/d_n = 1/4$. Figure 4 shows that the controlled localization effect persists for longer and sparser chains. The lower panel of Figure 4 presents several cross sections of the response signal from the upper panel to demonstrate the widths of the $|p_n|^2$ distributions at different wavelengths. The full width at half-maximum remains almost constant ($\approx$5) within a wide interval of the wavelength λ, making it possible to strongly localize the optical signal at an arbitrary subset of a few neighboring spheres. We also note that the response characteristic (the dependence of the maximum of $|p_n|^2$ on the wavelength) is relatively flat as compared with the above considered cases, which is advantageous for applications.

In conclusion, we have theoretically studied the optical response of graded chains of noble metal nanospheres. We found a strong dependence of the response signal on the direction of incidence of the incoming field. We demonstrated that, in the case when the incoming light propagates along the array axis in the direction from the smallest to the largest sphere, the response signal undergoes strong localization: the full width at half-maximum of the signal measures typically a few center-to-center distances. The position of the maximum can be changed over the full length of the array (dozens of nanoparticles) by adjusting the wavelength of the light. The arrays we focused on respond over a broad range of wavelengths ($\lambda \approx 450-650$ nm) covering almost the entire visible spectrum. The predicted frequency-controlled selective excitation of a few nanospheres opens the possibility to design various optical devices such as

broadband antennae, optical diodes, and directional sensors. Finally, we showed that different parameters, such as chain length, grading, and interparticle distances, affect different aspects of the response function of the array. In addition, the background dielectric function alters the overall frequency range of the system. Because these parameters can easily be controlled in experiment, the response characteristics can be designed and optimized for a particular application. The latter suggests that graded plasmonic arrays are promising candidates for building blocks of subwavelength optical circuitry.

Acknowledgment. This work is supported by NanoNed, a national nanotechnology program coordinated by the Dutch Ministry of Economic Affairs.

Supporting Information Available: Tabulated data for all 3D figures. This material is available free of charge via the Internet at http://pubs.acs.org.

References

(1) Quinten, M.; Leitner, A.; Krenn, R. M.; Aussenegg, F. R. *Opt. Lett.* **1998**, *23*, 1331.
(2) Brongersma, M. L.; Hartman, J. W.; Atwater, H. A. *Phys. Rev. B* **2000**, *62*, R16356.
(3) Maier, S. A.; Kik, P. G.; Atwater, H. A. *Phys. Rev B* **2003**, *67*, 205402.
(4) Li, K.; Stockman, M. I.; Bergman, D. J. *Phys. Rev. Lett.* **2003**, *91*, 227402.
(5) Stockman, M. I. *Phys. Rev. Lett.* **2003**, *93*, 137404.
(6) Weber, W. H.; Ford, G. W. *Phys. Rev. B* **2004**, *70*, 125429.
(7) (a) Citrin, D. S. *Nano Lett.* **2004**, *4*, 1561. (b) Citrin, D. S. **2005**, *5*, 985. (c) Citrin, D. S. *J. Opt. Soc. Am B* **2005**, *22*, 1763. (d) Citrin, D. S. *Opt. Lett.* **2006**, *31*, 98.
(8) Schuck, P. J.; Fromm, D. P.; Sundaramurthy, A.; Kino, G. S.; Moerner, W. E. *Phys. Rev. Lett.* **2005**, *94*, 017402.
(9) Mühlschlegel, P.; Eisler, H.-J.; Martin, O. J. F.; Hecht, B.; Pohl, D. W. *Science* **2005**, *308*, 1607.
(10) Cubukcu, E.; Kort, E. A.; Crozier, K. B.; Capasso, F. *Appl. Phys. Lett.* **2006**, *89*, 093120.
(11) Hernández, J. V.; Noordan, L. D.; Robicheaux, F. J. *J. Phys. Chem. B* **2005**, *109*, 15808.
(12) Sweatlock, L. A.; Maier, S. A.; Atwater, H. A.; Pennikhof, J. J.; Polman, A. *Phys. Rev. B* **2005**, *71*, 235408.
(13) de Waele, R.; Koenderink, A. F.; Polman, A. *Nano Lett.* **2007**, *7*, 2004.
(14) van Hulst, N. *Nature* **2007**, *448*, 141.
(15) Koenderink, A. F.; de Waele, R.; Prangsma, J. C.; Polman, A. *Phys. Rev. B* **2007**, *76*, 201403.
(16) Markel, V. A.; Sarychev, A. K. *Phys. Rev. B* **2007**, *75*, 085426.
(17) Fung, K. H.; Chan, C. T. *Opt. Commun.* **2008**, *281*, 855.
(18) Maier, S. A.; Atwater, H. A. *J. Appl. Phys.* **2005**, *98*, 011101.
(19) Yong, S.; Stroud, D. *Phys. Rev. B* **2004**, *69*, 125418.
(20) The importance of retardation effects for the adequate description of the linear plasmonic waveguides was discussed in a number of papers (see, e.g., refs 7, 11, 13, and 16).
(21) Meier, M.; Wokaun, A. *Opt. Lett.* **1983**, *8*, 581.
(22) Bohren C. F. and Huffman, D. R. *Absorption and Scattering of Light by Small Particles*; Wiley: New York, 1983.
(23) Mie, G. *Ann. Phys.* **1908**, *25*, 377.
(24) *Handbook of Optical Constants of Solids*; Palik, E. D., Ed.; Academic: San Diego, 1985.

NL8011962