Nondecaying surface plasmon polaritons in linear chains of silver nanospheroids

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We consider propagation of surface plasmon polaritons in linear chains of equidistant metallic nanospheroids. We show that, for suitably chosen parameters, the propagation is free of spatial decay in spite of the full account of absorptive losses in the metal. © 2013 Optical Society of America

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Propagation of surface plasmon polaritons (SPPs) in chains of metallic particles has attracted significant attention due to the number of potential applications in waveguiding and nanoplasmonics [1–4], as well as in spectroscopy and chemical sensing [5–7]. Chains of nanoparticles have also attracted attention in quantum information processing and it was recently demonstrated that such chains can serve as unidirectional single-photon or single-plasmon emitters [8]. In this Letter, we investigate the spatial decay of SPPs in chains of spheroidal particles and evaluate ways to mitigate this decay by manipulating the aspect ratio of the particles.

SPPs (guided modes) are collective plasmonic excitations that can propagate as well-formed wave packets [9] in chains of metallic nanoparticles without radiative losses (at least, in perfectly periodic linear arrays). In [8], relatively short chains (nine nanoparticles total) were considered and SPP decay due to Ohmic losses did not play a significant role. In longer chains, which are the subject of this Letter, spatial decay of SPPs is a critically important factor. It is known that Ohmic losses in metal can result in some attenuation. According to conventional wisdom, Ohmic losses make the decay of SPPs inevitable. However, this decay is not a consequence of energy conservation or of any other fundamental law. Below, we show that propagation of SPPs without spatial decay is possible in chains of particles with sufficiently small aspect ratios. As an example of such particles, we used spheroids. Of course, it is not possible to manufacture nanoparticles in perfect spheroidal shapes. However, the effect reported by us is not critically dependent on the manufacturing precision or on the particle shape. What *is* important in our simulations is the *depolarization factor* of a particle. This quantity is well-defined as long as the particle possesses a dominating dipole resonance. A simple alternative example of such a particle is a truncated cylinder of height H and base radius R. This object has a single dominating dipole resonance in the two limits $H/R \ll 1$ (nanodisk) and $H/R \gg 1$ (nanoneedle). As will be discussed below, these two cases are of special interest to us.

In the propagation regime discussed in this Letter, all spheroids in the chain dissipate optical energy into heat (at about the same rate), yet there is no spatial decay of the SPPs. This result has not been noticed before, most likely, because the majority of theoretical treatments of the subject consider chains of spherical particles. We note that recently described [10] undamped SPPs in chains of nanospheres are probably similar to what we have termed "extraordinary" SPPs, which indeed propagate with only slow algebraic decay, but only after an initial precipitous drop of the amplitude (by a few orders of magnitude) [11].

We will provide a few numerical examples to support the above claim. We work in the frequency domain and the time-dependence factor $\exp(-i\omega t)$ is suppressed throughout. Consider a linear periodic chain of identical silver spheroids characterized by the eccentricity factor $e = \sqrt{1-(b/a)^2}$ $(b \le a)$, where b and a are the shorter and longer semi-axes. The spheroids can be either prolate or oblate, and we assume that the longer semi-axis is perpendicular to the chain in both cases. We will consider only the SPPs that are polarized transversely to the chain. For oblate spheroids, the two mutually orthogonal transverse polarizations are equivalent. However, this is not so for prolate spheroids. In the latter case, we will consider only the polarization aligned with the longer axis. In all cases, the shorter semi-axis is taken to be $b = 8$ nm and the center-to-center interparticle distance is taken to be $h = 3b = 24$ nm. Thus, the surface-to-surface distance between two nearest neighbors in the chain is equal to b. For transversely polarized SPPs, these parameters are within the range of validity of the dipole approximation, which will be used in all simulations.

The principal values of the polarizability tensor of each spheroid are given by $\alpha = (\alpha_{\mathrm{LL}}^{-1} - 2ik^3/3)^{-1}$, where α_{LL} is quasi-static Lorentz–Lorenz polarizability of a spheroid and the term $-2ik^3/3$ is the first nonvanishing radiative correction [12]. The former is defined by

$$\alpha_{\mathrm{LL}} = (a^p b^{3-p}/3)[\nu + 1/(\epsilon - 1)]^{-1}. \quad (1)$$

Here, ϵ is the dielectric permittivity of spheroids at the working frequency ω, $k = \omega/c$ is the free space wave number, $p = 1$ for prolate spheroids, $p = 2$ for oblate spheroids, and ν is the appropriate depolarization factor. We now introduce the following notations: for the spatial direction aligned with the spheroid axis of symmetry (the longer or shorter axis for prolate or oblate spheroids, respectively), we write $\nu = \nu_\parallel$ and for either of the two linearly independent transverse polarizations, $\nu = \nu_\perp$. Then, in the case of prolate spheroids, we can use the formula $\nu_\perp = (1 - \nu_\parallel)/2$, where $\nu_\parallel = g^2(e)[(1/2e)\ln((1+e)/(1-e)) - 1]$. In the case of oblate spheroids, we can use $\nu_\parallel = 1–2\nu_\perp$, where $\nu_\perp = (g(e)/2e^2)[\pi/2 - \arctan g(e)] - g^2(e)/2$. In these expressions, $g(e) = \sqrt{1/e^2 - 1}$.

We also need to define the dielectric permittivity ϵ, which appears in Eq. (1). To this end, we use the Drude formula $\epsilon = \epsilon_0 - \omega_p^2/\omega(\omega + i\gamma)$, where ω_p is the plasma frequency, γ is the relaxation constant, and $\epsilon_0 - 1$ is the contribution to the permittivity due to the interband transitions. In numerical simulations, we use the parameters for silver, $\omega_p/\gamma = 526.3$ and $\epsilon_0 = 5.0$.

We now turn to the transmission properties of chain waveguides of finite length N. Assume that the first particle in the chain ($n = 1$) is illuminated by a near-field external source, which creates a fixed and known amplitude A of the electric field. We are interested in the dipole moment amplitude of the last ($n = N$) particle of the chain, which can be "read out", for example, by a near-field scanning optical microscope (NSOM) tip. We will use the formalism of normalized Green's function [11], $\mathcal{F}_n = |\mathcal{D}_{n,1}/\mathcal{D}_{1,1}|$. Here, the Green's function $\mathcal{D}_{nm}$ gives the dipole moment d_n of the nth spheroid due to the localized external electric field incident on the mth spheroid. In other words, if the external electric field at the site r_n (center of the nth spheroid) is given by $E_n = A\delta_{nm}$, then $d_n = \mathcal{D}_{nm}A$. The Green's function $\mathcal{D}_{nm}$ can be obtained in finite chains numerically by solving the coupled-dipole equations [11]. Here we consider finite chains and compute $\mathcal{D}_{nm}$ numerically by direct inversion. We will refer to the quantity $\mathcal{F}_N$ as the transmission of the chain.

The spectral dependence of transmission for a chain containing $N = 1001$ spheroids of different shapes and eccentricities is illustrated in Fig. 1. It can be seen that SPPs tend to propagate with less decay for smaller values of b/a (at a given amplitude of the external field, A). This finding may be interesting, but the truly striking results can be observed when $0.05 < \omega/\omega_p < 0.15$ (for prolate spheroids) or $0.05 < \omega/\omega_p < 0.25$ (for oblate spheroids), when the transmission $\mathcal{F}_N$ is of the order of unity. This result is obtained for $b/a = 0.2$ in the case of prolate spheroids and for $b/a \le 0.3$ in the case of oblate spheroids.

The above finding is unexpected and radically different from the results previously obtained for chains of spherical particles. In all previous studies, the Green's function $\mathcal{D}_{nm}$ drops sharply with $|n - m|$ if realistic losses are taken into account. We illustrate this point in Fig. 2 by plotting $\mathcal{F}_n$ as a function of n for chains constructed of spheroids with $b/a = 0.2$ and of spheres. The working frequency is chosen to be $\omega = 0.15\omega_p$ for the case of

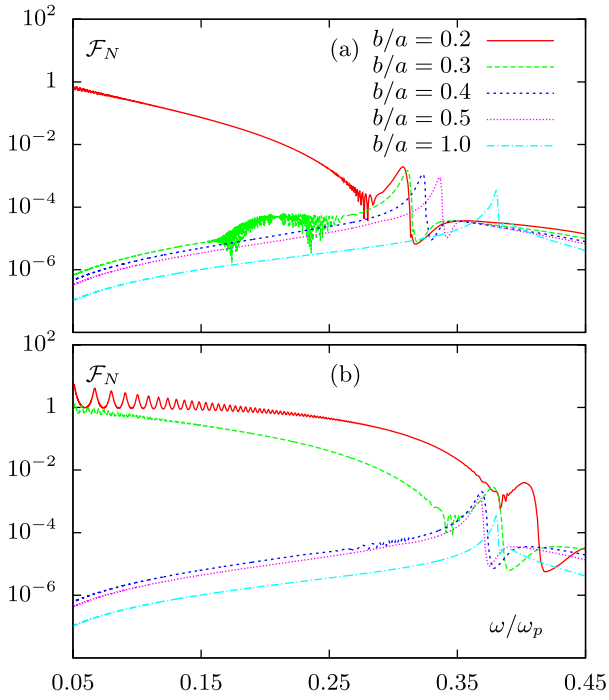

Fig. 1. Transmission spectra for transversely polarized SPPs in chains built from prolate spheroids (a) and oblate spheroids (b) for different aspect ratios b/a.

spheroids and $\omega = 0.38\omega_p$ for the case of spheres. This choice is explained as follows. In the case of spheroids, the dispersion curve is approximately linear when $0.05 < \omega/\omega_p < 0.25$ and the working frequency has been chosen at the center of this interval (data not shown but see qualitatively similar dispersion curves in [9]). In chains of spheres, well-formed wave packets can not be created at any frequency, and so we have chosen the working frequency for which the decay of $\mathcal{F}_n$ is the slowest. Still, it can be seen that, in the case of spheres, $\mathcal{F}_n$ drops precipitously with n. The curves for spheroids exhibit much slower decay, or no decay at all.

It may even seem that the data of Figs. 1,2 contradict conservation of energy. However, this is not so. In our simulations, the expected dissipation of optical energy into heat occurs in each spheroid. Yet, this process does not result in spatial decay of $\mathcal{D}_{nm}$. How is this possible? The answer to the above question becomes obvious if we

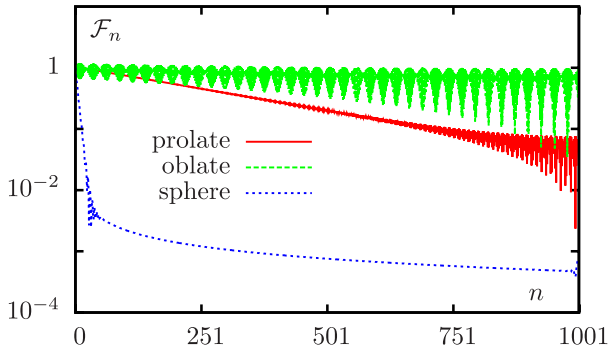

Fig. 2. Normalized Green's function $\mathcal{F}_n$ for linear chains of spheroids with aspect ratio $b/a = 0.2$ at the frequency $\omega/\omega_p = 0.15$ and for a chain of spheres at $\omega/\omega_p = 0.38$.

recall that, in the definition of the Green's function $\mathcal{D}_{nm}$, we assume that the external field amplitude A is fixed and independent of the actual distribution of dipoles in the chain. When the decay of the Green's function is slow or absent, all spheroids dissipate energy approximately at the same rate and, correspondingly, this energy must be supplied by an external source. Therefore, to maintain a given amplitude of the external field, one must supply the system with the optical energy, which is proportional to the length of the chain, N. The physical situation is somewhat similar to the case when N resistors are sequentially connected to an ideal source of current. The power dissipated by each resistor is in this case the same and independent of the number of resistors. In this analogy, the external optical source of fixed amplitude A plays the role of an ideal source of current.

To better illustrate the above point, we now consider the ratio of radiative energy loss to the total energy loss in the system. Recall that, in an infinite chain, SPPs propagate without radiative losses. In a finite chain, radiative losses occur at both ends of the chain. The characteristic parameter to consider is the quality factor $\eta = Q_s/Q_e$, where Q_s and Q_e are the efficiencies of scattering and extinction. In terms of the dipole moments, these quantities are defined as follows:

$$Q_e = \frac{4k}{NR^2}\,\mathrm{Im}\sum_n \frac{d_n E_n^*}{|A|^2} = \frac{4k}{NR^2}\,\mathrm{Im}\,\frac{d_1 A^*}{|A|^2}, \tag{2a}$$

$$Q_s = \frac{8k^4}{3NR^2}\sum_n \frac{|d_n|^2}{|A|^2}. \tag{2b}$$

Here, E_n represents the amplitudes of the external electric field at the center of the nth sphere and we have used the expression $E_n = A\delta_{n1}$ to obtain the second equality in Eq. (2a). R is an arbitrary constant of dimensionality of length; its value does not affect η. Note that Q_s is proportional to the total optical energy scattered by the chain into free space per unit time.

In Fig. 3, we plot η as a function of frequency for chains of spheroids and spheres. For both prolate and oblate spheroids, we have $\eta \approx 1/N \approx 10^{-3}$ at $\omega = 0.15\omega_p$. For spheres, the quality factor is approximately 50 time larger at the same frequency. This indicates that, in the case of spheroids, almost all energy supplied by the source is absorbed and the absorption is equally divided between the spheroids comprising the chain; hence, we see no significant spatial decay of $\mathcal{F}_n$.

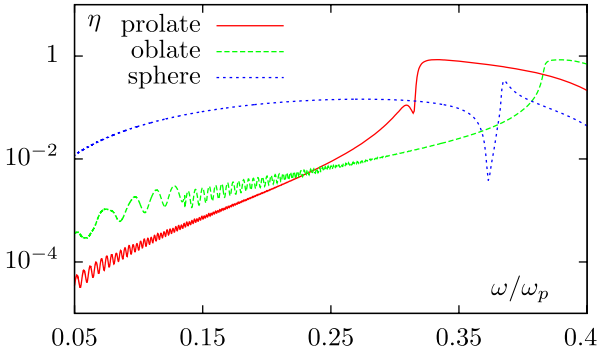

Fig. 3. Quality factor $\eta = Q_s/Q_e$ for chains of spheres and spheroids with the aspect ratio $b/a = 0.2$.

The slow decay of SPPs reported above can be understood in the framework of previously known theoretical results. In [11], we have shown that the Green's function in an infinite chain can be found analytically by Fourier transform and has the form

$$D_{n0} = \int_{-\pi/h}^{\pi/h} \frac{\exp(iqhn)}{1/\alpha - S(\omega,q)}\frac{h\,dq}{2\pi}. \tag{3}$$

In this expression, the illuminated particle is assumed to be labeled as $m = 0$ and $S(\omega,q)$ is the dipole sum (self energy). The detailed definition and plots of $S(\omega,q)$ are given in [11]. Equation (4) can be used to estimate the SPP propagation length, ℓ. This can be done with the use of the quasi-particle pole approximation. Namely, we assume that the dominating input to the integral is obtained in the small vicinity of the Bloch wave number q_1, which is the root of the equation $\mathrm{Re}[1/\alpha - S(\omega,q)] = 0$ (selected with the additional condition that q_1 is not close to the logarithmic singularity of $S(\omega,q)$). The root q_1 thus determined is the Bloch wave number of the *ordinary* SPP. The dipole sum is an analytical function of q in the vicinity of q_1 and it can be expanded in a Taylor series. Note that the imaginary part of the denominator is small and constant in the vicinity of q_1. We then extend the integration in Eq. (3) to the whole real axis using contour integration to obtain the following expression:

$$\ell = \frac{1}{\delta}\left|\mathrm{Re}\,\frac{\partial S(\omega,q)}{\partial q}\right|_{q=q_1}, \qquad \delta = -\mathrm{Im}\,\frac{1}{\alpha_{\mathrm{LL}}}. \tag{4}$$

The data presented in this Letter are fully consistent with the formula in Eq. (4). Indeed, for the polarizability function defined by Eq. (1), we have

$$h^3\delta = \frac{3(h/b)^3(b/a)^p\gamma\omega\omega_p^2}{\Omega^4 + (\epsilon_0 - 1)^2(\gamma\omega)^2}, \tag{5}$$

where $\Omega^2 = (\epsilon_0 - 1)\omega^2 - \omega_p^2$. For fixed h/b, ℓ can be increased when (i) the frequency ω is decreased or (ii) the aspect ratio b/a is decreased while the ratio h/b is fixed (in this case, the dependence is stronger for oblate spheroids). For spheroidal particles, resonance excitation can occur only in close vicinity of the Frohlich frequency ω_F, where $\omega_F^2 = [\nu/(1 + (\epsilon_0 - 1)\nu)]\omega_p^2$. For spherical particles, $\nu = 1/3$ and the frequency of resonant excitation can not be arbitrarily reduced while the spherical shape is fixed. However, we can significantly reduce the resonant frequency by utilizing spheroids with $\nu < 1/3$, and, simultaneously, achieve an additional gain in ℓ due to the increased factor $(a/b)^p$. For the parameters used in Fig. 2, we can estimate $\ell \approx 44$ μm (oblate spheroids), $\ell \approx 9.1$ μm (prolate spheroids), and $\ell \approx 0.6$ μm (spheres). These estimates are in a good agreement with the data of Fig. 2. Note that the entire length of the chain is $L = h(N - 1) \simeq 24$ μm.

We can also explain the phenomenon just discussed from a physical point of view. We first note that replacing spherical particles by spheroids increases the strength of electromagnetic interaction of neighboring particles because the coupling coefficients (polarizabilities) can be increased while keeping the center-to-center distance constant. This results in a significant increase of the group velocity of SPPs, as was already predicted in [9]. This phenomenon is analogous to the increase of the velocity of sound in solids when the elastic modulus is increased. Correspondingly, a wave packet (if we consider the time-domain dynamics) spends less time travelling from one end of the chain to another. If we further assume that the amplitude of the wave packet decays with time at a constant rate, then we can expect a larger amplitude of the SPP at the far end of the chain. Upon Fourier-transforming the SPP field into the frequency domain, we find that the Green's function decays slower in chains with spheroidal particles. Another helpful factor is that the introduction of nonsphericity allows one to shift the Frohlich frequency of the spheroids toward the red, where metal nanoparticles are know to be better (higher-quality) resonators.

We conclude that the use of nonspherical particles can be advantageous for the design of nanoparticle waveguides. Gaussian SPP wave packets can propagate along a chain of spheroids with negligible spatial decay. These two features are desirable in many of the applications considered in the literature. Unfortunately, this regime of propagation requires large energy input (proportional to the number of particles in the chain, N) and the fraction of incident energy converted to the useful signal scales as $1/N$. It is difficult to estimate the actual power needed for reliable detection of wavepackets at the far end of a given chain, since this estimate requires the knowledge of the threshold incident power at which the nanoparticles are destroyed and of the level of electromagnetic noise at the detector. Overall, there are many factors that influence waveguiding properties of plasmonic chains. One such factor, which is practically important but has been overlooked in the past, is the particle nonsphericity.

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