Surface Plasmons in Ordered and Disordered Chains of Metal Nanospheres

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Abstract: We describe two types of surface plasmons in ordered and disordered chains. The second kind is mediated by far-field interaction and is affected by Ohmic and radiative losses much less than the first kind.

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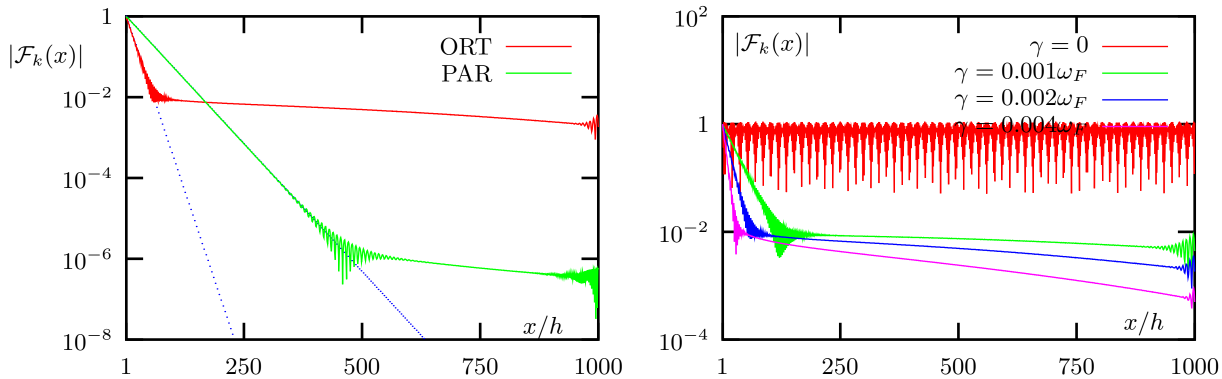
Surface plasmons (SPs) are states of polarization that can propagate along metal-dielectric interfaces without radiative losses. Polarization in an SP excitation can be spatially confined on scales that are much smaller than the free-space wavelength. This property proved to be extremely valuable for manipulation of light energy on subwavelength scales and miniaturization of optical elements. SP excitations in ordered one-dimensional arrays of nanoparticles have attracted significant attention in recent years due to numerous potential application in nanoplasmonics. For example, a periodic chain of high conductivity metal nanospheres can be used as an SP wave guide - an analog of an optical waveguide [1]. High-quality SP modes in ordered and disordered chains may be utilized in random lasers [2]. Electromagnetic forces acting on linear chains of nanoparticles can produce the effect of optical trapping [3]. Various spectroscopic and sensing applications have also been discussed [4,5].

Although, under ideal conditions, SP excitations can propagate without loss of energy, in practice this is not so. There are two physical effects that can result in decay of SP excitations. The first effect is Ohmic losses due to the finite conductivity of the metal. The second effect is radiative losses due to disorder in the chain (scattering from imperfections). The latter effect is more subtle and is closely related to the phenomenon of localization. We first focus on decay due to Ohmic losses and show that it can be suppressed at sufficiently large propagation distances. The main idea is based on exploiting an exotic non-Lorentzian resonance in the chain which originates due to radiation-zone interaction of nanoparticles [6]. From the spectroscopic point of view, the non-Lorentzian resonances are manifested by very narrow lines in extinction spectra [4]. The author has argued previously that the small integral weight of the spectral lines associated with these resonances precludes them from being excited by a near-field probe [6]. However, numerical simulations reveal that the corresponding SP has relatively small yet nonzero amplitude and is also characterized by very slow spatial decay. Therefore, in sufficiently long chains, this SP becomes dominant and can propagate, without significant further losses, to remarkable distance. We stress that the non-Lorentzian SP is an excitation specific to discrete systems; it does not exist, for example, in metal nanowires. But the above consideration applies only to ordered chains. Therefore, we consider next the effects of disorder. To isolate radiative losses due to scattering on imperfections from Ohmic losses, we consider nanoparticles with infinite conductivity (equivalently, zero Drude relaxation constant). An equivalent system can be constructed experimentally by embedding metallic particles into a dielectric medium with positive gain.

Consider a linear chain of N nanospheres with radiuses a_n centered at points x_n and work in the dipole approximation which is valid if $x_{n+1} - x_n \gtrsim (a_{n+1} + a_n)/2$. The n-th nanosphere is characterized by a dipole moment with amplitude d_n oscillating at the electromagnetic frequency ω. The dipole moments are coupled to each other and to external field by the coupled-dipole equation [7] $d_n = \alpha_n \left[E_n + \sum_{n' \neq n} G_k(x_n, x_{n'}) d_{n'} \right]$, where α_n is the polarizability of the n-th nanosphere, E_n is the external electric field at the point x_n, $k = \omega/c$ is the free space wave number and $G_k(x, x')$ is the appropriate element of the free space, frequency-domain Green's tensor for the electric field. The polarizability of the n-th sphere is taken in the form $\alpha_n^{-1} = a_n^{-3}(\epsilon_n + 2)/(\epsilon_n - 1) - 2ik^3/3$, where the first term is the Lorenz-Lorentz quasistatic polarizability of a sphere of radius a_n and $2ik^3/3$ is the first non-vanishing radiative correction. We further adopt the Drude model for ϵ_n, namely, $\epsilon_n = 1 - \omega_{\mathrm{p}n}^2/\omega(\omega + i\gamma_n)$, where ω is the electromagnetic frequency, $\omega_{\mathrm{p}n}$ is the plasma frequency, and γ_n is the Drude relaxation constant in the n-th nanosphere.



Left: propagation of a SP in an ordered chain of $N = 1000$ nanospheres for orthogonal (ORT) and parallel (PAR) polarization of oscillations with respect to the chain. Parameters: $\omega = \omega_F$, $\gamma/\omega_F = 0.002$, $\lambda = 10h$, $h = 4a$. Right: same as in the left panel but for different ratios γ/ω_F and for SP polarized orthogonally to the chain. The slowly-decaying segments of the curves are due to the extraordinary SP discussed in the text.

Suppose that SP is excited at a given site (say, $n = m$) by a near-field probe. Then the external field can be set to $E_n = E_0\delta_{nm}$. The solution with $E_n = \delta_{nm}$ is, essentially, the Green's function for polarization. We denote this Green's function by $\mathcal{D}_k(x_n, x_m)$. We also define the normalized Green's function $\mathcal{F}_k(x_n - x_m; x_m) = \mathcal{D}_k(x_n, x_m)/\mathcal{D}_k(x_m, x_m)$. In an infinite periodic chain, this function is independent of the second argument; in finite or disordered chains, such dependence exists, but will be suppressed in the list of formal arguments.

In the case of infinite ordered chains, the coupled-dipole equation can be solved analytically by Fourier transform. For finite and disordered chains, it must be solved numerically. This has been done for ordered and disordered chains of length N up to 10^4. The simulations reveal the existence of two types of plasmons: ordinary (quasistatic) and extraordinary (non-quasistatic) SPs. The ordinary SP is characterized by short-range interaction of nanospheres in a chain. The retardation effects are inessential for its existence and properties. The ordinary SP behaves as a quasistatic excitation. It can not radiate into the far zone in perfectly periodic chains because its wave number is larger than the wavenumber $k = \omega/c$ of free electromagnetic waves. However, it can experience decay due to absorptive dissipation in the material. The second, extraordinary, SP propagates due to long-range (radiation zone) interaction in a chain and may experience some radiative loss but is much less affected by absorptive dissipation and disorder. As a result, it can propagate to much larger distances along the chain. A cross over from ordinary to extraordinary SP is illustrated in the Figure. Finally, simulations suggest that even small disorder in the position or properties of nanoparticles results in localization of the ordinary SP. However, the extraordinary SP appears to remain delocalized for all types and levels of disorder considered.

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