2-15-2007

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Vadim A. Markel
University of Pennsylvania, vmarkel@mail.med.upenn.edu

Andrey K. Sarychev
Ethertonics Incorporated

Recommended Citation

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Abstract

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Disciplines

Biomedical Engineering and Bioengineering | Engineering

Comments

Propagation of surface plasmons in ordered and disordered chains of metal nanospheres

Vadim A. Markel*

Departments of Radiology and Bioengineering, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA

Andrey K. Sarychev†

Ethertronics Incorporated, San Diego, California 92121, USA

We report a numerical investigation of surface plasmon (SP) propagation in ordered and disordered linear chains of metal nanospheres. In our simulations, SPs are excited at one end of a chain by a near-field tip. We then find numerically the SP amplitude as a function of propagation distance. Two types of SPs are discovered. The first SP, which we call the ordinary or quasistatic, is mediated by short-range, near-field electromagnetic interaction in the chain. This excitation is strongly affected by Ohmic losses in the metal and by disorder in the chain. These two effects result in spatial decay of the quasistatic SP by means of absorptive and radiative losses, respectively. The second SP is mediated by longer range, far-field interaction of nanospheres. We refer to this SP as the extraordinary or nonquasistatic. The nonquasistatic SP cannot be effectively excited by a near-field probe due to the small integral weight of the associated spectral line. Because of that, at small propagation distances, this SP is dominated by the quasistatic SP. However, the nonquasistatic SP is affected by Ohmic and radiative losses to a much smaller extent than the quasistatic one. Because of that, the nonquasistatic SP becomes dominant sufficiently far from the exciting tip and can propagate with little further losses of energy to remarkable distances. The unique physical properties of the nonquasistatic SP can be utilized in all-optical integrated photonic systems.

I. INTRODUCTION

Surface plasmons (SPs) are states of polarization that can propagate along metal-dielectric interfaces or along other structures 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,[1,2] miniaturization of optical elements,[3] and achieving coherent temporal control at remarkably short times.[4,5] SP excitations in ordered one-dimensional arrays of nanoparticles have attracted significant attention in recent years due to numerous potential application in nanoplasmonics.[6–10] A periodic chain of high conductivity metal nanospheres can be used as an SP wave guide—an analog of an optical waveguide.[11] High-quality SP modes in ordered and disordered chains may be utilized in random lasers.[8,12] Electromagnetic forces acting on linear chains of nanoparticles can produce the effect of optical trapping.[13] Various spectroscopic and sensing applications have also been discussed.[14–16]

In this paper we study theoretically and numerically propagation of SP excitations in long ordered and disordered chains of nanospheres. Although, under ideal conditions, SP excitations can propagate without loss of energy, in practical situations this is not so. There are two physical effects that can result in decay of SP excitations as they propagate along the chain. 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). This effect is more subtle and is closely related to the phenomenon of localization. In this paper we discuss both effects and illustrate them with numerical examples.

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[14,17] and cannot be understood within the quasistatics, even when both the nanoparticles in the chain and the interparticle spacing are much smaller than the wavelength. From the spectroscopic point of view, the non-Lorentzian resonances are manifested by very narrow lines in extinction spectra.[14,15] One of the authors (V.A.M.) 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.[17] This property would make the non-Lorentzian resonance a curiosity which is rather useless for nanoplasmonics. However, numerical simulations shown below 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.

However, 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). Although such metals do not exist in nature, an equivalent system can be constructed experimentally by embedding metallic particles into a dielectric medium with positive gain.[12] We show that while the ordinary (defined more precisely in the text below) SP excitations are very sensitive to off-diagonal (position) disorder, the SP due to the non-Lorentzian resonance is not. Diagonal disorder (disorder in nanoparticle properties) is also considered.

The paper is organized as follows. In Sec. II, we describe the theoretical model and introduce basic equations. Conditions for resonance excitation of SPs in a chain are considered ins Sec. III. Numerical results for propagation in ordered and disordered chains are reported in Secs. IV and V, respectively. Finally, Sec. VI contains a summary of obtained results.

II. THEORETICAL MODEL

Consider a linear chain of N nanospheres with radii a_n centered at points x_n. We work in the dipole approximation which is valid if $x_{n+1} - x_n \gtrsim (a_{n+1} + a_n)/2$ and has been widely used in the literature.[7,8,10,18–20] The nth nanosphere is then 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[14]

$$d_n = \alpha_n \left[E_n + \sum_{n' \neq n} G_k(x_n, x_{n'}) d_{n'} \right], \quad (1)$$

where α_n is the polarizability of the nth 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 latter is translationally invariant with respect to spatial variables, namely, $G_k(x, x') = G_k(x - x', 0) = G_k(x' - x, 0)$. For an SP polarized perpendicular and parallel to the chain, the respective functions $G_k^\perp$ and $G_k^\parallel$ are given by

$$G_k^\perp(x, 0) = \left(\frac{k^2}{|x|} + \frac{ik}{|x|^2} - \frac{1}{|x|^3} \right) \exp(ik|x|), \quad (2)$$

$$G_k^\parallel(x, 0) = \left(-\frac{2ik}{|x|^2} + \frac{2}{|x|^3} \right) \exp(ik|x|). \quad (3)$$

Polarizability of the nth sphere is taken in the form

$$\alpha_n = \frac{1}{1/\alpha_n^{(LL)} - 2ik^3/3}, \quad (4)$$

where $\alpha_n^{(LL)}$ is the Lorenz-Lorentz quasistatic polarizability of a sphere of radius a_n and $2ik^3/3$ is the first nonvanishing radiative correction to the inverse polarizability; account of this correction is important to ensure that the system conserves energy.[21] The Lorenz-Lorentz polarizability is given, in terms of the complex permeability of the nth nanosphere ϵ_n, by

$$\alpha_n^{(LL)} = a_n^3 \frac{\epsilon_n - 1}{\epsilon_n + 2}. \quad (5)$$

We further adopt, for simplicity, the Drude model for ϵ_n:

$$\epsilon_n = 1 - \frac{\omega_{pn}^2}{\omega(\omega + i\gamma_n)}, \quad (6)$$

where ω is the electromagnetic frequency, ω_{pn} is the plasma frequency, and γ_n is the Drude relaxation constant in the nth nanosphere. The inverse polarizability of the nth nanosphere is then given by

$$\text{Re}\left(\frac{1}{\alpha_n} \right) = \frac{1}{a_n^3} \left[1 - \left(\frac{\omega}{\omega_{Fn}} \right)^2 \right],$$

$$\text{Im}\left(\frac{1}{\alpha_n} \right) = -i \left(\frac{2k^3}{3} + \frac{1}{a_n^3} \frac{\omega \gamma_n}{\omega_{Fn}^2} \right), \quad (7)$$

where $\omega_{Fn} = \omega_{pn}/\sqrt{3}$ is the Frohlich frequency. The SP resonance of an isolated nth nanosphere takes place when $\omega = \omega_{Fn}$. Polarizability α_n at the Frohlich resonance is purely imaginary; if, in addition, there are no Ohmic losses in the material ($\gamma_n = 0$), the resonance value of the polarizability becomes $\alpha_n = \alpha_{res} = -i3/2k^3$, irrespective of the particle radius.

Suppose that SP is excited at a given site (say, $n = n_0$) by a near-field probe. Then the external field can be set to $E_n = E_0 \delta_{n,n_0}$. Of course, this is an idealization: the field produced even by a very small near-field tip is, strictly speaking, nonzero at all sites. However, this approximation is physically reasonable because of the fast (cubic) spatial decay of the dipole field in the near-field zone. The solution with $E_n = \delta_{n,n_0}$ is, essentially, the Green's function for polarization. We denote this Green's function by $\mathcal{D}_k(x_n, x_{n_0})$. It satisfies

$$\mathcal{D}_k(x_n, x_{n_0}) = \alpha_n \left[\delta_{n,n_0} + \sum_{n' \neq n} G_k(x_n, x_{n'}) \mathcal{D}_k(x_{n'}, x_{n_0}) \right], \quad (8)$$

where either Eq. (2) or (3) should be used for G_k, depending on polarization of the SP. In the case of a finite or disordered chain, one can find $\mathcal{D}_k(x_n, x_{n_0})$ by solving Eq. (1) numerically. However, in infinite ordered chains such that $\alpha_n = \alpha = \text{const}$ and $x_{n+1} - x_n = h = \text{const}$, the following analytic solution is obtained by Fourier transform:[17]

$$\mathcal{D}_k(x_n, 0) = \int_{-\pi/h}^{\pi/h} \frac{\exp(iqx_n)}{1/\alpha - S(k,q)} \frac{h \, dq}{2\pi}, \quad (9)$$

where $S(k,q)$ is the "dipole sum" given by

$$S(k,q) = 2 \sum_{n>0} G_k(0, x_n) \cos(qx_n). \quad (10)$$

Obviously, in infinite chains $\mathcal{D}_k(x_n, x_{n'}) = \mathcal{D}_k(x_n - x_{n'}, 0) = \mathcal{D}_k(x_{n'} - x_n, 0)$. Note that the dipole sum (10) is independent of material properties. It can be shown[14] that, for all values of parameters, $\text{Im } S(k,q) \geq -2k^3/3$. The equality holds when $q > k$. This is a manifestation of the fact that SPs with $q > k$ are nonradiating due to the light-cone constraint.[8] Nonradiating modes exist if $\pi/h > k$ or, equivalently, if $\lambda > 2h$. Obviously, these SPs do not couple to running waves but can be excited by a near-field probe. The dimensionless radiative relaxation parameter can be defined as $Q(k,q) = [\text{Im } S(k,q) + 2k^3/3]/(2k^3/3)$; this factor is identically zero for $q > k$.

III. DISPERSION RELATIONS AND RESONANT EXCITATION OF SP

In this section, we consider periodic chains with $a_n = a$, $\omega_{pn} = \omega_p$, $\omega_{Fn} = \omega_F$, $\gamma_n = \gamma$ and, consequently, with the constant polarizability $\alpha_n = \alpha$.
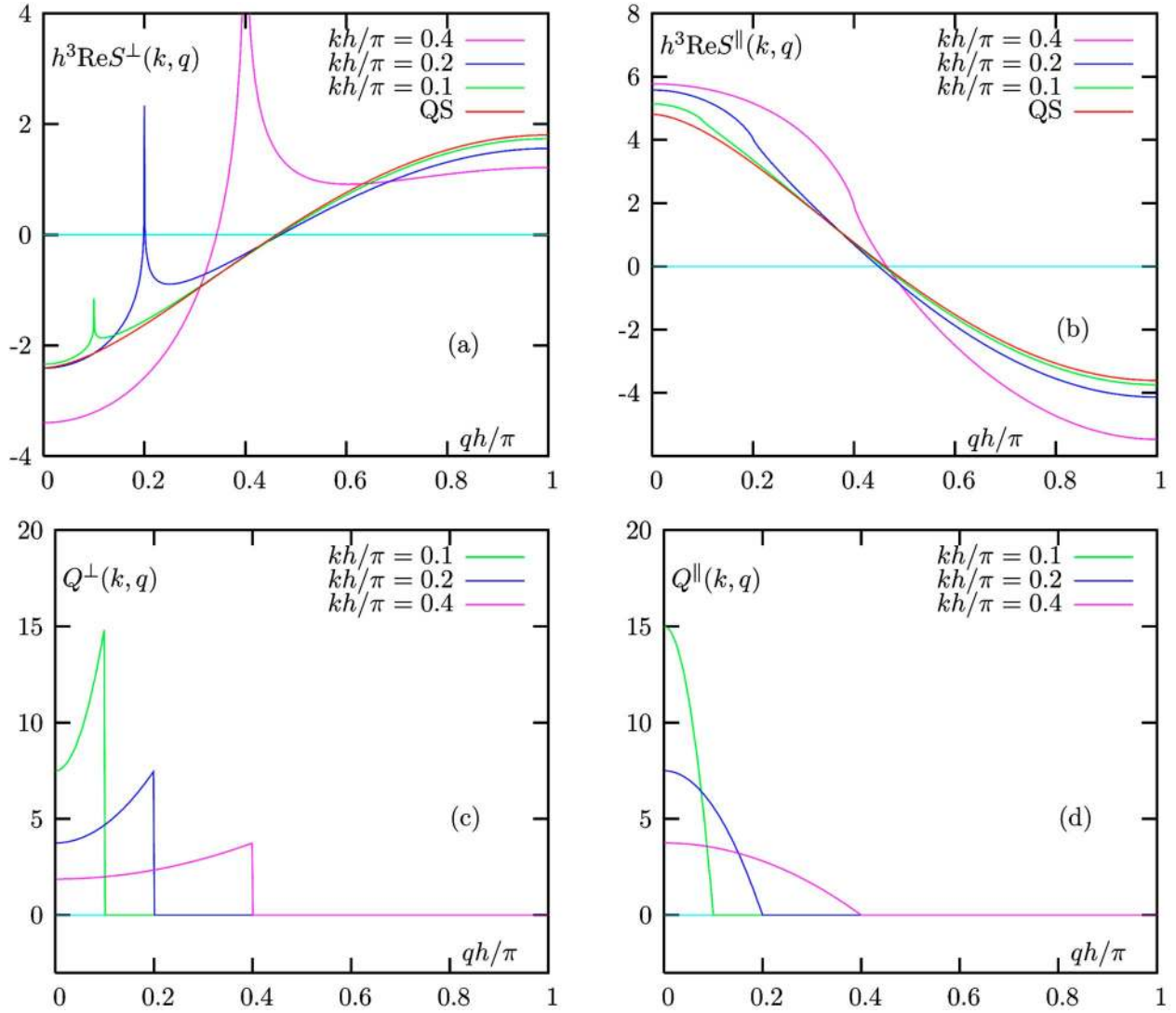
FIG. 1. (Color online) (a), (b) "Real parts" of the dipole sum $S(k,q)$ as a function of q, for different values of kh, as indicated. Sharp peaks corresponding to divergence of Re $S(k,q)$ at the point $q=k$ are not completely resolved. Quasistatic results obtained in the limit $k=0$ are shown by curves labeled "QS." Only half of the Brillouin zone is shown since $S(k,-q)=S(k,q)$. Polarization of SP is perpendicular (a) and parallel (b) to the chain. (c), (d) Dimensionless radiative relaxation parameter $Q(k,q)=[\text{Im } S(k,q)+2k^3/3]/(2k^3/3)$ as a function of q for the same sets of parameters as above. Polarization of SP is perpendicular (c) and parallel (d) to the chain. Quasistatic result is not shown since Q is not defined in the quasistatic limit.

It follows from formula (9) that the wave numbers q of SP excitations that can propagate effectively in an infinite periodic chain are such that $1/\alpha \approx S(k,q)$. This condition must be satisfied for a range of q which is, in some sense, small. Indeed, there is no effective interaction in the chain if $1/\alpha - S(k,q) \approx$ const, in which case integration according to Eq. (9) yields[22] $d_n \propto \delta_{n,0}$. We now examine the conditions under which the denominator of Eq. (9) can become small. The dispersion relation in the usual sense, e.g., the dependence of the resonant SP frequency on its wave number is obtained by solving the equation $1/\alpha(\omega) - S(\omega/c,q)=0$. This approach was adopted, for example, in Refs. 7, 8, 10, and 18. Solution to the above equation depends on the model for $\epsilon(\omega)$ and may result in several branches of the complex function $\omega(q)$. Here we adopt a slightly different point of view. Namely, we note that for real frequencies ω and wave numbers q, the real

part of the denominator can change sign while the imaginary part is always non-negative. Physically, $\text{Re}[1/\alpha(\omega) - S(\omega/c,q)]$ can be interpreted as the generalized detuning from a resonance while $\text{Im}[1/\alpha(\omega) - S(\omega/c,q)]$ gives total (radiative and absorptive) losses. We thus define the resonance condition to be $\text{Re}[1/\alpha - S(k,q)]=0$ and view ω and q as independent purely real variables.

Plots of the dimensionless function $h^3 S(k,q)$ are shown in Fig. 1 for some typical sets of parameters and for two orthogonal polarizations of the SP [similar plots have also been shown in Ref. 20, where the function $S(k,q)$ was referred to as the dipolar self-energy]. Note that the imaginary part of $h^3 S(k,q)$ is related to radiative relaxation parameter $Q(k,q)$ [shown in Figs. 1(c) and 1(d)] by $h^3 \text{Im } S(k,q) = [2(kh)^3/3][Q(k,q)-1]$. Apart from the very narrow peaks appearing in the case of perpendicular polarization and cen-

tered at[23] $q=k$, the numerical values of $h^3|\text{Re } S|$ do not exceed, approximately, 6. On the other hand, according to Eq. (7), we have $h^3 \text{Re}(1/\alpha)=(h/a)^3[1-(\omega/\omega_F)^2]$. The dipole approximation is valid when $(h/a)^3 \gtrsim 64$. Therefore, if we stay within the range of parameters in which the dipole approximation is valid, the real part of the denominator in Eq. (9) is relatively small only if $\omega \approx \omega_F$, i.e., if ω is near the Frohlich frequency of an isolated sphere. This is the case that will be considered below.

Consider first oscillations polarized orthogonally to the chain and let $\omega=\omega_F$. The condition of resonant excitation of SP is then $\text{Re } S(k,q)=0$. It can be seen from Fig. 1(a) that, for sufficiently small values of the dimensionless parameter kh, there are two different values of q that satisfy the above resonance condition.

The first solution is $q=q_1 \approx 0.47\pi/h$ (for $kh=0.2\pi$). The value of q_1 depends only weakly on kh, as long as $kh \lesssim 0.2\pi$, and is approximately the same as in the quasistatic limit $kh=0$, in which case $q_1 \approx 0.46\pi/h$. We will refer to the SP with this wave number as the ordinary, or the quasistatic SP. This is because propagation of this SP is mediated by near-field interaction, while the far-field interaction is suppressed by destructive interference of waves radiated by different nanoparticles in the chain. Since the wave number of the ordinary SP is greater than k, it propagates without radiative losses. However, it may experience spatial decay due to absorption in metal. The characteristic exponential length of decay can be easily inferred from Eq. (9) by making the quasiparticle pole approximation. Namely, we approximate $S(k,q)$ as

$$S(k,q) \approx \text{Re } S(k,q_1) + (q-q_1) \left| \frac{\partial \text{Re } S(k,q)}{\partial q} \right|_{q=q_1} - i\frac{2k^3}{3}.$$

(11)

We then extend integration in Eq. (9) to the real axis and obtain the following characteristic exponential decay length l:

$$l = \frac{1}{\delta} \left| \frac{\partial \text{Re } S(k,q)}{\partial q} \right|_{q=q_1},$$

(12)

where

$$\delta = -\text{Im}(1/\alpha) - 2k^3/3$$

(13)

is a positive parameter characterizing the absorption strength of a nanosphere. In general, it can be shown that $\delta=0$ in nonabsorbing particles whose dielectric function $\epsilon(\omega)$ is purely real at the given frequency ω. For the Drude model adopted in this paper, we have $\delta=\omega\gamma/a^3\omega_F^2$. Thus, the ordinary SP can decay exponentially due to absorption in metal with the characteristic scale given by Eq. (12).

The second solution is obtained at $q=q_2 \approx k$, when $\text{Re } S(k,q)$ has a narrow sharp peak as a function of q (for fixed k).[24] This peak is explained by far-field interaction in an infinite chain.[14,17] It does not disappear in the limit $kh \to 0$, but becomes increasingly (super-exponentially) narrow.[17] Note that in the above limit, this peak appears as a singularity of zero integral weight which cannot be obtained

from the formal quasistatic approximation. We will refer to this SP as extraordinary, or nonquasistatic. It is mediated by the far-field interaction. The latter is important in the case of extraordinary SP because of the constructive interference of far field contributions from all nanoparticles arriving at a given one. Obviously, the extraordinary SP can be excited only in chains with $h < \lambda/2$, where λ is the wavelength of light in free space.

It is interesting to consider radiative losses of the extraordinary SP. As can be seen from Fig. 1(c), the factor $Q(k,q)$ is discontinuous at $q=k$: it is zero for $q > k$ but positive for $q < k$. Since the extraordinary SP has $q \approx k$, it can experience some radiative losses, although the exact law of its decay depends on parameters of the problem in a complicated manner. This is, in part, related to inapplicability of the quasiparticle pole approximation for evaluating the integral (9) in the vicinity of $q \approx q_2$.

In the case of oscillations polarized along the chain, the resonance condition can be satisfied only at $q=q_1 \approx 0.45\pi/h$. This is the wave number of an ordinary (quasistatic) SP which depends on k only weakly, as long as $kh \lesssim 0.2\pi$. However, the extraordinary (nonquasistatic) SP can be excited even for longitudinal oscillations. Mathematically, this can be explained by observing that $\partial \text{Re } S(k,q)/\partial q$ diverges at $q=k$ while $\partial \text{Im } S(k,q)/\partial q$ is discontinuous at $q=k$ and performing integration (9) by parts. However, the amplitude of the extraordinary SP is much smaller for longitudinal oscillations than for transverse oscillations; this will be illustrated numerically in the next section.

Finally, we note, that when intersphere separations become smaller than the radii, higher-order multipole resonances can be excited.[25–28] In this case, resonant excitation of SP can become possible even at frequencies which are far from the Frohlich resonance of an isolated sphere, e.g., in the IR part of the spectrum.

IV. PROPAGATION IN FINITE ORDERED CHAINS

We now turn to propagation of SP in ordered chains of finite length N. We, however, emphasize that the finite size effects play a very minor role in the computations shown below. Citrin[19] has studied dispersion relations in finite chains and has found that the infinite-chain limit is reached at $N \approx 10$ (although we anticipate that longer chains are needed for accurate description of the extraordinary SP). In this and following sections, we work with chains of $N \gtrsim 1000$. In this limit, propagation of both ordinary and extraordinary SP is not much different from the case of infinite chains. In particular, we have verified numerically that the Green's function $\mathcal{D}_k(x_n, x_{501})$ (for $kh=0.2\pi$) in a chain of $N=1001$ particles does not differ in any significant way from that in an infinite chain, except for values of n very close to either end of the finite chain. The Green's function $G_k(x_n, x_1)$ (here $n_0=1$ is the end point of the finite chain) differed by a trivial factor in finite and infinite chains (results not shown). However, proper numerical evaluation of integral (9) required very fine discretization of q and was a more demanding and less stable procedure than direct numerical solution of the system of equations (8).
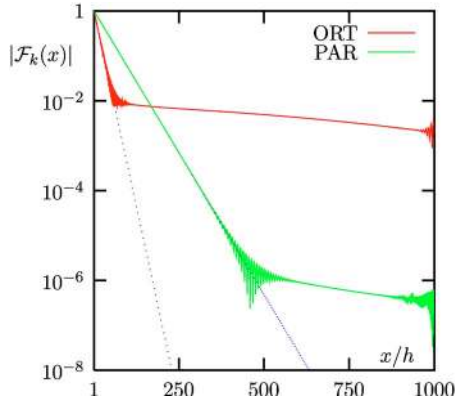
FIG. 2. (Color online) Propagation of a SP in an ordered chain of $N=1000$ nanospheres for orthogonal (ORT) and parallel (PAR) polarization of oscillations with respect to the chain. Parameters: $\omega=\omega_F$, $\gamma/\omega_F=0.002$, $\lambda=10h$, $h=4a$.

In this section, we take $a_n=a=$const, $\alpha_n=\alpha=$const and $x_n=nh$, $n=1,\dots,N$. We also assume that $kh=0.2\pi$ and $h=4a$. Practically, this can be realized for silver particles in a transparent host matrix with refractive index of approximately $n=1.4$ so that the wavelength at the Frohlich frequency is $\lambda_F=2\pi c/\omega_F\approx400$ nm, the chain spacing is $h=40$ nm$=0.1\lambda_F$ and the sphere radius is $a=10$ nm$=h/4$. The dipole approximation is very accurate for this set of parameters. We then obtain the SP Green's function $\mathcal{D}_k(x_n,x_1)$ by solving Eq. (8) numerically.

The absolute value of the normalized SP Green's function

$$\mathcal{F}_k(x_n)=\frac{\mathcal{D}_k(x_n,x_1)}{\mathcal{D}_k(x_1,x_1)} \qquad (14)$$

in a chain of $N=1000$ nanospheres is shown in Fig. 2 as a function of x (sampled at $x=x_n$) for two orthogonal polarizations. Here the frequency of SP was taken to be exactly equal to the Frohlich frequency ω_F and the Drude relaxation con-

stant was $\gamma=0.002\omega_F$. It can be seen from the figure that two different SP are excited in the system. The first is the ordinary (quasistatic) SP that decays exponentially as $\exp(-x/l)$, where l is defined by Eq. (12). Corresponding asymptotes are shown by dotted lines. Note that the quasistatic SP in a finite chain (with the point of excitation coinciding with one of the chain ends) is very well described by the exponential decay formula, even though the latter was obtained for infinite chains. When the amplitude of the ordinary SP becomes sufficiently small, there is a crossover to the extraordinary SP. The decay rate of the extraordinary SP is much slower. We have also confirmed by inspecting the real and imaginary parts of $\mathcal{D}_k(x_n,x_1)$ (data not shown) that it oscillates at the spatial frequency corresponding to the ordinary SP in the fast-decaying segments of the curves shown in Fig. 2 and with the spatial frequency that corresponds to the extraordinary SP in the slow-decaying segments.

Mathematically, the relatively slow decay of the extraordinary SP can be understood as follows. First, note that the exponential decay of the ordinary SP is, in fact, the result of superposition of an infinite number of plane waves whose wave numbers are in the interval $\Delta q\propto\delta$. The corresponding wave packet decays spatially on scales $l\propto1/\Delta q$. However, in the case of extraordinary SP, Δq cannot be defined since the corresponding resonance is non-Lorentzian. It can be, however, stated that the extraordinary SP is a superposition of plane waves whose wave numbers are very close to k. Since the wave numbers can still slightly deviate from k, some spatial decay at large distances can still occur.

The conclusion we can make so far is that the ordinary SP experiences exponential decay along the chain due to Ohmic losses in metal. This decay is very accurately described by the quasiparticle pole approximation. The extraordinary SP has, initially, much smaller amplitude than the ordinary one. This is because the peaks in Fig. 1 are very narrow. The quasiparticle pole approximation is invalid for the extraordinary SP and its decay is much less affected by Ohmic losses. As a result, the extraordinary SP decays at a much slower
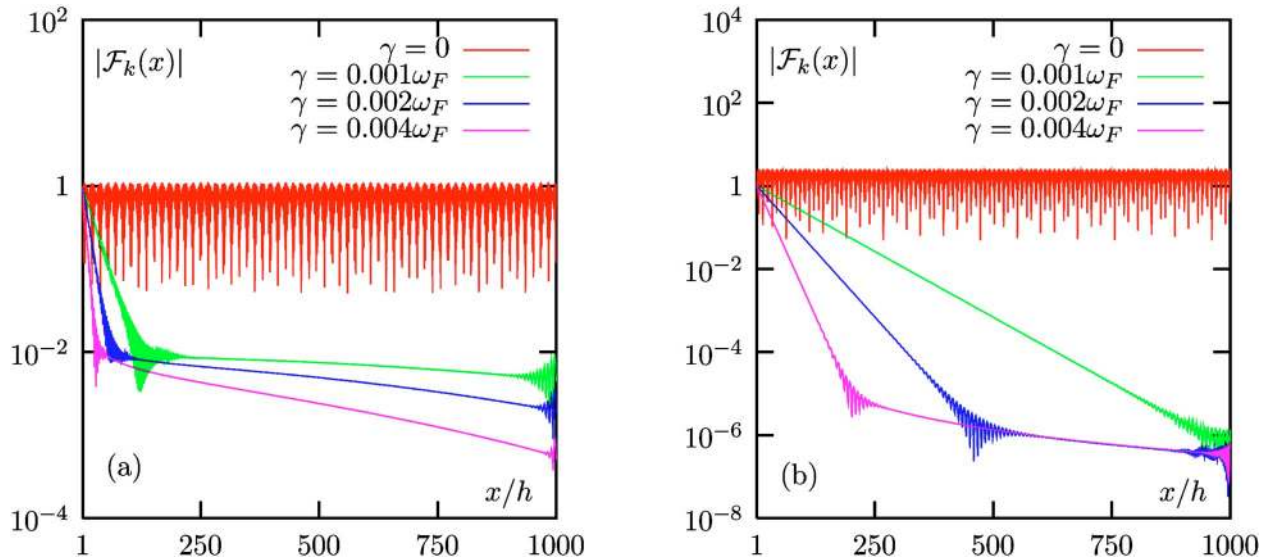
FIG. 3. (Color online) Same as in Fig. 3 for different ratios γ/ω_F and for SP polarized orthogonally (a) and parallelly (b) to the chain.
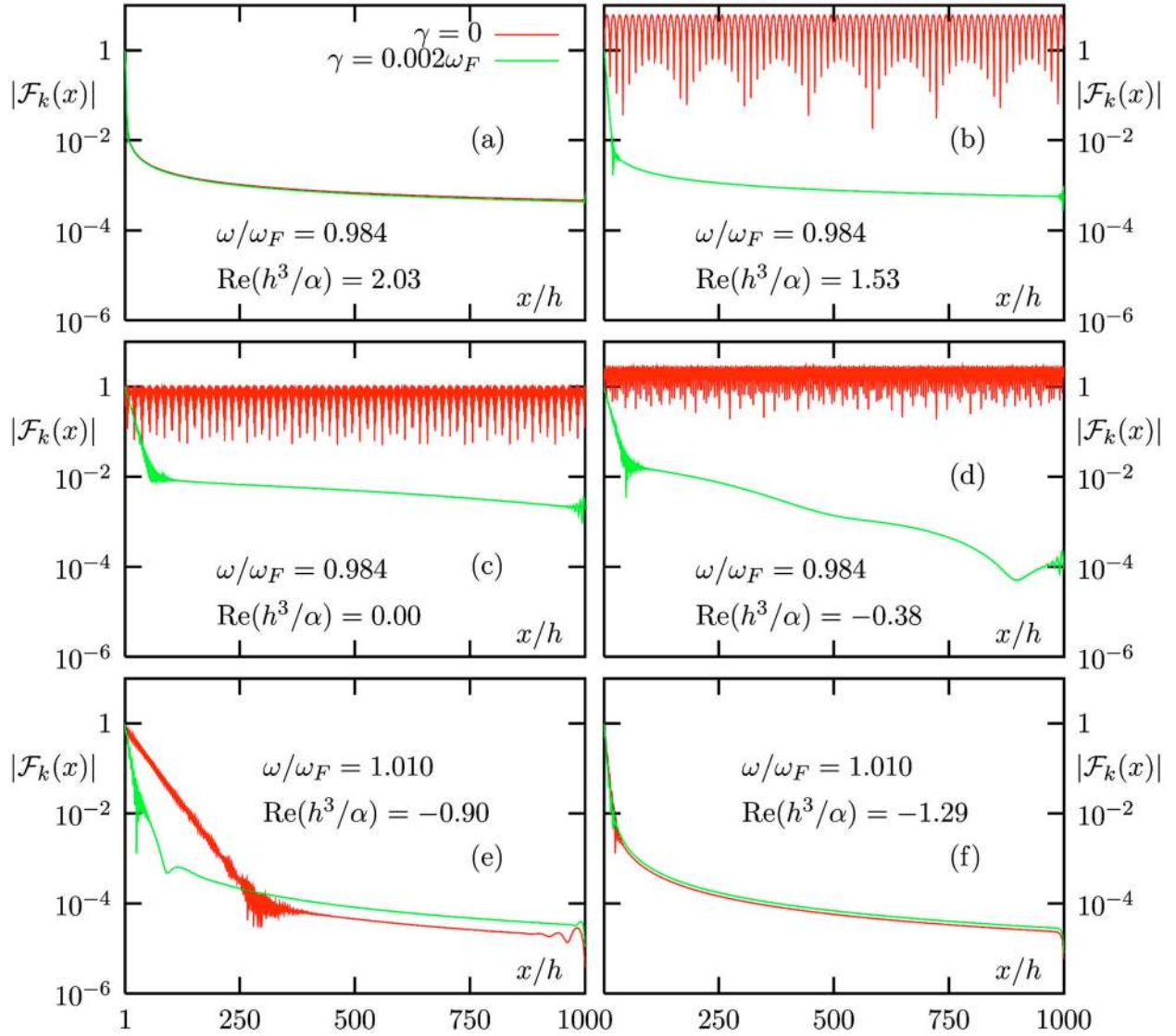
FIG. 4. (Color online) Same as in Fig. 3 for different ratios ω/ω_F and γ/ω_F, as indicated, and for SP polarized orthogonally to the chain.

rate and, at sufficiently large propagation distance, begins to dominate. We also note that the extraordinary SP can be excited even for longitudinally polarized SP, although its amplitude is smaller by some four orders of magnitude than for the case of transverse oscillations.

In Fig. 3, we illustrate the influence of Ohmic losses on SP propagation. Here we plot $|\mathcal{F}_k(x)|$ as a function of x (sampled at $x=x_n$) for $\omega=\omega_F$ and different values of the ratio γ/ω_F. First, in the absence of absorption ($\gamma=0$), the ordinary SP propagates along the chain without decay. Once we introduce absorption, the ordinary SP decays exponentially with the characteristic length scale l given by Eq. (12). Note that, for the specific metal permeability model (6), $l \propto \omega_F/\gamma$. Some dependence of the rate of decay of the extraordinary SP on the ratio γ/ω_F is visible in the case of orthogonal polarization [Fig. 3(a)]. However, when the polarization is longitudinal [Fig. 3(b)], decay of the extraordinary SP is dominated by radiative losses. In particular, the slow-decaying segments of the curves for $\gamma/\omega_F=0.002$ and $\gamma/\omega_F=0.004$ in Fig. 3(b) coincide with high precision.

Next, we study SP propagation for different values of the ratio ω/ω_F. As noted above, we assume the parameters $kh=\omega h/c=0.2\pi$ and $h/a=4$ to be fixed. Thus, ω/ω_F can vary either due to a change in ω_F or due to a simultaneous change in ω, h, and a such that $\omega h=$const and $h/a=$const. It follows from Fig. 1(a) that, for the selected set of parameters, the ordinary plasmon can be excited for $-0.89 < h^3 \text{Re}(1/\alpha) < 1.56$. This corresponds to ω/ω_F lying in the interval $0.988 < \omega/\omega_F < 1.007$. In Fig. 4, we illustrate propagation of SP excitations for some values of ω/ω_F inside this interval, exactly at the lower and upper bounds of this interval, and slightly outside of the interval. Results are shown for two values of absorption strength: $\gamma/\omega_F=0$ and $\gamma/\omega_F=0.002$.

First, consider the two cases when ω/ω_F is outside of the interval where the ordinary SP can be excited: $\omega/\omega_F=0.984$ and $\omega/\omega_F=1.010$ [correspondingly, $h^3 \text{Re}(1/\alpha)=2.03$ and $h^3 \text{Re}(1/\alpha)=-1.29$], shown in Figs. 4(a) and 4(f). In the case $\omega/\omega_F=0.984$, the ordinary SP exhibits very fast
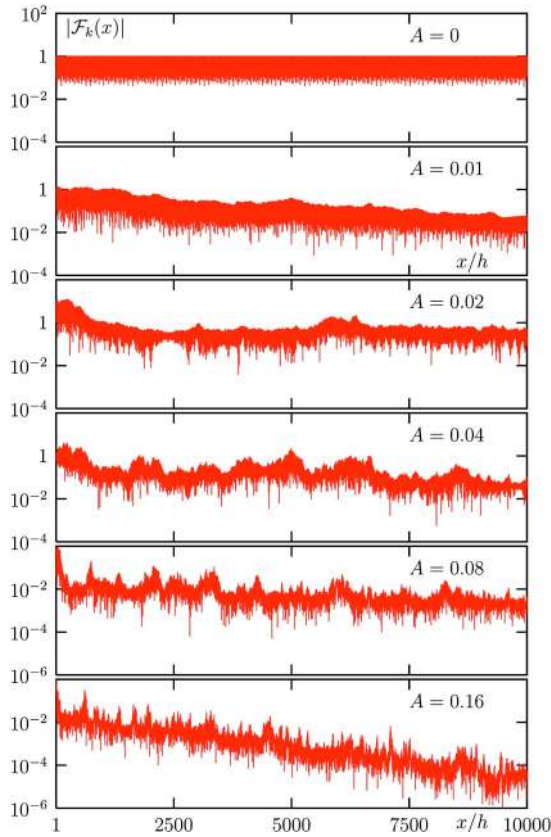
FIG. 5. (Color online) Propagation of SP in a chain of $N=10\,000$ nonabsorbing ($\gamma=0$) nanospheres for different levels of off-diagonal disorder A, as indicated. SP polarization is orthogonal to the chain. Other parameters: $kh=0.2\pi$, $\omega/\omega_F=1$, $h/a=4$.

spatial decay, which is characteristic for the noninteracting limit when $G_k(n,n_0) \propto \delta_{n,n_0}$. After the initial decay of the ordinary SP, the extraordinary SP becomes dominating. The extraordinary SP decays slowly by means of radiative losses. It is interesting to note that decay of the extraordinary SP is almost unaffected by Ohmic losses in metal, although a noticeable dependence on γ appears if we further increase the parameter γ/ω_F by the factor of 10 (data not shown). A qualitatively similar behavior is obtained at $\omega/\omega_F=1.010$. However, the decay of extraordinary SP in this case is a little faster. Paradoxically, the curve corresponding to $\gamma/\omega_F=0.002$ is slightly higher than the curve corresponding to $\gamma=0$ in Fig. 4(f) [similar peculiarity is seen in Fig. 4(e)].

When the ratio ω/ω_F is inside the interval where the ordinary SP can be excited [Figs. 4(c) and 4(d)], SP propagation is strongly influenced by absorptive losses. In the absence of such losses, the ordinary SP propagates along the chain indefinitely and dominates the extraordinary SP. However, in the presence of even small absorption, the ordinary SP decays exponentially so that, at sufficiently large propagation distances, the extraordinary SP starts to dominate. A qualitatively similar picture is also obtained for the borderline case $\omega/\omega_F=0.984$ [Fig. 4(b)]. In the second borderline case [Fig. 4(e)], SP propagation is more complicated. The wave numbers of both ordinary and extraordinary SPs in this case are close to k, so that both can experience radiative decay, as is evident in the case of zero absorption.
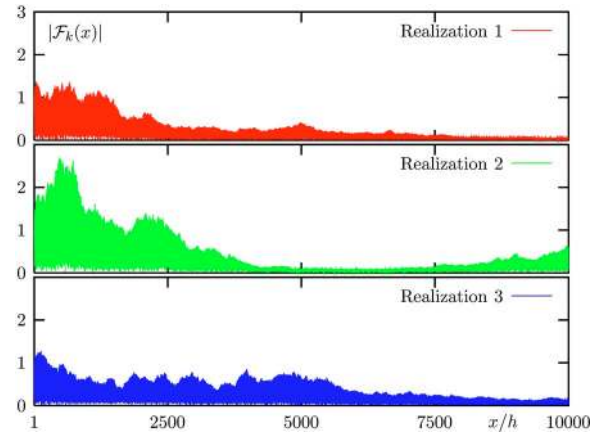

FIG. 6. (Color online) Propagation of SPs in a chain with off-diagonal disorder of amplitude $A=0.01$ for different random realizations of disorder. The other parameters are the same as in Fig. 5.

To conclude this section, we note that propagation of SP excitations in long periodic chains can be characterized by exponential decay. This decay is caused either by absorptive or by radiative losses. At small propagation distances, energy is transported by the ordinary SP excitation, if the ordinary SP can be excited [e.g., if $\text{Re}(1/\alpha)$ is inside the appropriate interval]. However, at sufficiently large propagation distances, there is a cross over to transport by means of the extraordinary SP. In this case, propagation is mediated by far-zone interaction and is characterized by slow, radiative decay which is affected by absorptive losses only weakly.

We note that exponential decay in ordered chains, if exists, is not caused by Anderson localization, since we have not, so far, introduced disorder into the system. For example, as was discussed in Sec. III, a linear superposition of delocalized plane wave modes of the form (9) can exhibit exponential decay with the characteristic length (12). The irreversible exponential decay is, in fact, obtained because the delocalized modes form a truly continuous spectrum (are indexed by a continuous variable q). Of course, any superposition of *discrete* delocalized modes would result in Poincare recurrences.
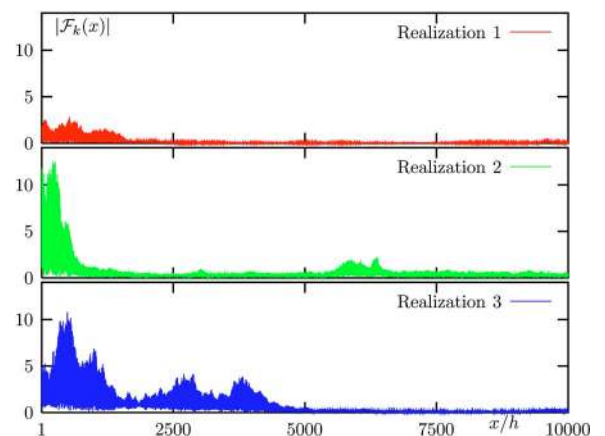

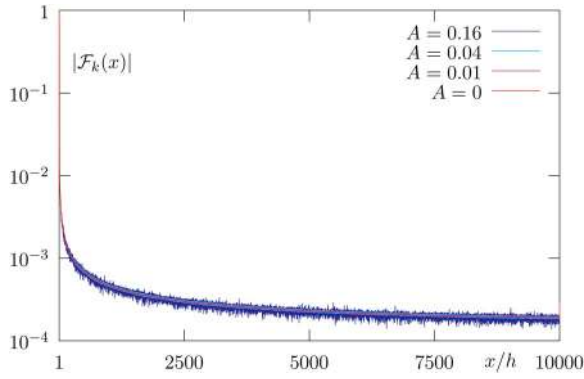FIG. 7. (Color online) Same as in Fig. 6, but for $A=0.02$.

FIG. 8. (Color online) Same as in Fig. 5 but for $\omega/\omega_F = 0.984$ and for different levels A of off-diagonal disorder, as indicated. Note that the curves for $A=0$, and $A=0.01$ and $A=0.04$ are indistinguishable. (In the black-and-white version of this figure, curves with $A=0.01$ and $A=0.04$ are not shown; the curve with $A=0.16$ is drawn through every tenth data point in order to ensure visual distinguishability of curves.)

V. PROPAGATION IN DISORDERED CHAINS

The ordinary (quasistatic) SP propagates in ordered chains without radiative losses due to the perfect periodicity of the lattice. However, once this periodicity is broken, the quasistatic SP can experience radiative losses and spatial decay even in the absence of absorption. Dependence of the radiative quality of SP modes in finite one-dimensional chains on disorder strength was studied in Ref. 8. It was shown that position disorder tends to decrease the radiative quality factor of initially nonradiating ("bound") modes. In this section we study how the disorder influences propagation of the SP along the chain and take a separate look at the ordinary and extraordinary SPs. We also consider two types of disorder: off-diagonal and diagonal. Off-diagonal disorder is disorder in particle positions where all particles are identical. Diagonal disorder arises due to differences in particle properties, even if the particle positions are perfectly ordered.

The exponential decay due to Ohmic losses in the material can mask the effects of disorder. Therefore, we assume in this section that the nanospheres are nonabsorbing, i.e., set $\gamma_n = 0$. Physically, absence of absorption can be realized by embedding the chain of nanoparticles in a transparent dielectric host medium with positive gain.[12,29–32] Such medium has dielectric permeability ϵ_h with positive real and negative imaginary parts. The gain can be tuned so that the effective permeability of inclusions ϵ/ϵ_h is purely real and negative. We also work in the regime $kh = 0.2\pi$, $h = 4a$.

A. Off-diagonal disorder

Off-diagonal disorder is disorder in particle position. It is called "off-diagonal" because it affects only off-diagonal elements of the interaction matrix $G_k(x_n, x_{n'})$. In the simulations shown below, coordinates of particles in a disordered chain were taken to be $x_n = h(n + \xi_n)$ where ξ_n is a random variable evenly distributed in the interval $[-A, A]$. The random numbers ξ_n are taken to be mathematically independent. Therefore, the disorder is uncorrelated.
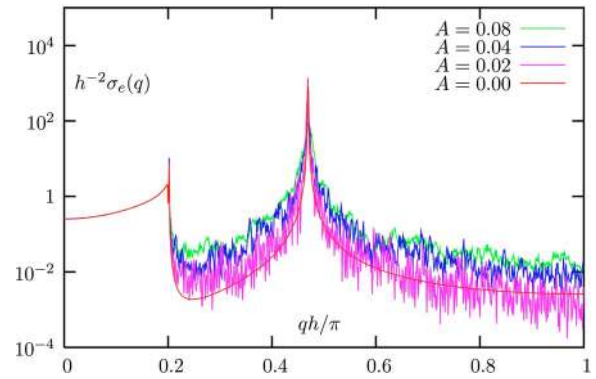

FIG. 9. (Color online) Specific extinction σ_e as a function of lateral wave number of incident wave, q, for different levels of off-diagonal disorder A. (In the black-and-white version of this figure, only curves with $A=0.02$ and $A=0.04$ are not shown.)

The first example concerns propagation in an nonabsorbing chain of $N=10\,000$ nanospheres excited exactly at the Frohlich frequency $\omega = \omega_F$. Numerical results for different levels of disorder are illustrated in Fig. 5 where we plot $|\mathcal{F}_k(x)|$ as a function of x (sampled at $x = x_n$).

One obvious conclusion that can be made from inspection of Fig. 5 is that disorder causes spatial decay of SP. Since the system has no absorption, energy is lost to radiation. However, the exact law of decay strongly depends on particular realization of disorder. In Figs. 6 and 7, we plot the function $|\mathcal{F}_k(x)|$ for $A=0.01$ and $A=0.02$, respectively, and for three different realizations of disorder (without the use of logarithmic scale). Giant fluctuations in the amplitude of transmitted SP are quite apparent. In all cases, after some initial growth, the amplitude decays, although some random recurrences (due to re-excitation) can take place. However, the amplitude $|\mathcal{F}_k(x)|$ at a given site $x = x_n$ strongly depends on realization of disorder and can be very far from its ensemble average, even at very large propagation distances. Therefore, the function $\mathcal{F}_k(x)$ appears to be not self-averaging. In particular, any particular realization of the intensity $\mathcal{I}_k(x) = |\mathcal{F}_k(x)|^2$ does not satisfy either the radiative transport equation or the diffusion equation. The absence of transport and self-averaging is consistent with the conjecture of Anderson localization of SP. However, a more definitive conclusion about the possibility of Anderson localization in the system would require application of a quantitative criterion, such as the inverse participation ratio for dipolar eigenmodes.[33] We note that the localization discussed here is an interference (radiative) effect which is different from localization of quasistatic polarization modes studied in Refs. 34–36.

The situation is complicated by the presence of two types of SP excitations. One can argue that localization properties of these two types of SPs might be different. To investigate this possibility, we perform two types of numerical experiments. First, we repeat simulations illustrated in Fig. 5 but for $\omega/\omega_F = 0.948$. In this case, the ordinary SP is not excited in ordered chains [see Fig. 4(a)]. Results are shown in Fig. 8. It can be concluded from the figure that off-diagonal disorder does not result in additional decay of the extraordinary SP. In fact, the curves with $A=0$, $A=0.1$, and $A=0.04$ are indistin-

guishable. This should be contrasted with the case $\omega/\omega_F=1$, when, at the level of disorder $A=0.02$, spatial decay is already well manifested. The conclusion one can make is that the extraordinary SP excitations do not experience localization due to uncorrelated off-diagonal disorder in the chain. Therefore, spatial decay seen in Figs. 5 and 7 is decay of the ordinary SP. At very large propagation distances, $\mathcal{F}_k(x)$ continues to oscillate around the baseline of the extraordinary SP.

The second numerical experiment that can elucidate the influence of disorder on ordinary and extraordinary SPs is simulation of an extinction measurement when the chain is excited, instead of a near-field tip, by a plane wave $E_0 \exp(iqx)$. Namely, we will look at the dependence of the specific (per one nanosphere) extinction cross section σ_e on q. There are two different experimental setups that can be used to measure $\sigma_e(q)$. When q is in the interval $0 \leq q < k$, this experiment can be carried out simply by varying the angle between the incident beam and the chain. However, values $q > k$ are not accessible in this experiment. In this case, the chain can be placed on a dielectric substrate and excited by evanescent wave originating due to the total internal reflection of the incident beam. The maximum longitudinal wave number of the evanescent wave is nk, where $n > 1$ is the refractive index of the substrate. We note that it is not realistically possible to access all wave numbers up to $q = \pi/h$ in this way because, in the particular case $kh = 0.2\pi$, this would require the refractive index $n=5$. Refractive indices of such magnitude are not achievable in the optical range. However, in a numerical simulation, we can assume that a hypothetical transparent substrate with $n=5$ exists. In addition, all wave numbers q in the first Brillouin zone of the lattice can be accessible for a different choice of parameters (particularly, for larger values of kh).

The specific extinction $\sigma_e(q)$ is given by the following formula:

$$\sigma_e(q) = \frac{4\pi k}{N|E_0|^2} \sum_{n=1}^{N} E_0^* \exp(-iqx_n)d_n, \qquad (15)$$

where d_n is the solution to Eq. (1) with the right-hand side $E_n = E_0 \exp(iqx_n)$. In Fig. 9, we plot the dimensionless quantity $h^{-2}\sigma_e(q)$ as a function of q for transverse oscillations in partially disordered nonabsorbing chains. Two peaks are clearly visible in the spectrum. The first peak at $q=q_1 \approx 0.47\pi/h$ corresponds to excitation of the ordinary SP. The second peak at $q=q_2 \approx k$ corresponds to excitation of the extraordinary SP. In infinite, periodic and nonabsorbing chains, the spectrum has a simple pole at[37] $q=q_1$. In the finite chain with $N=10\,000$, the singularity is replaced by a very sharp maximum. Introduction of even slight disorder tends to further broaden and randomize this peak. Obviously, this broadening, as well as the randomization of the spectrum in the vicinity of $q=q_1$, results in spatial decay of the ordinary SP excited by a near-field tip. However, the peak corresponding to the extraordinary SP is almost unaffected by disorder. Consequently, the extraordinary SP does not experience localization and related spatial decay when off-diagonal disorder is introduced into the system.
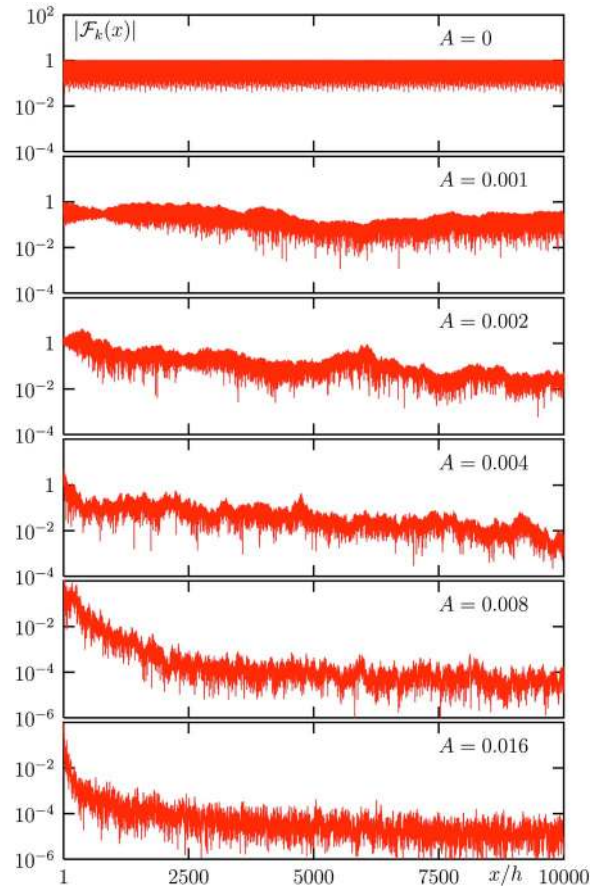

FIG. 10. (Color online) Same as in Fig. 5, but for different levels A of *diagonal* disorder.

The physical reason why the extraordinary SP is not affected by disorder is quite straightforward. This SP is mediated by electromagnetic waves in the far (radiation) zone that arrive at a given nanosphere from all other nanospheres. The synchronism condition (that all these secondary waves arrive in phase) is not affected by disorder as long as the displace-
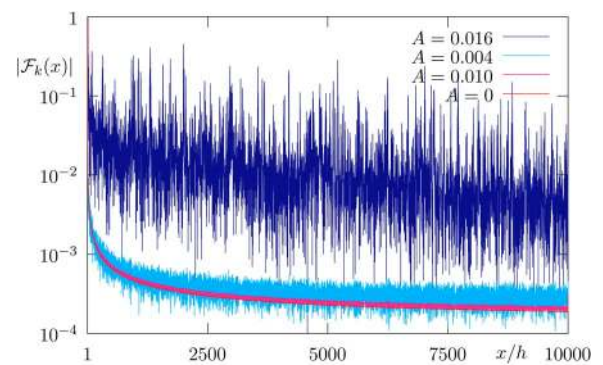

FIG. 11. (Color online) Same as in Fig. 10 but for $\omega/\omega_F = 0.984$ and for different levels A of off-diagonal disorder, as indicated. Note that the curves for $A=0$ and $A=0.01$ are indistinguishable. (In the black-and-white version of this figure, the curve with $A=0.010$ is not shown; the curves with $A=0.004$ and $A=0.016$ are drawn through every tenth data point in order to ensure visual distinguishability of curves.)
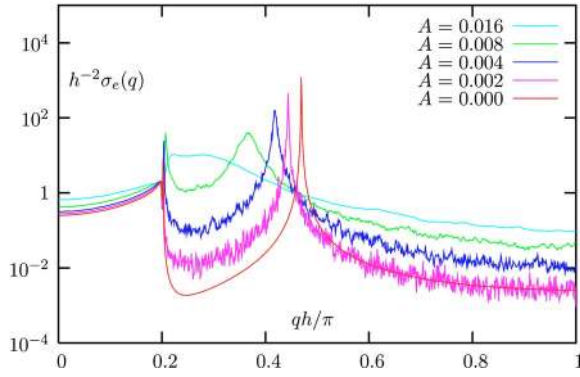
FIG. 12. (Color online) Specific extinction σ_e as a function of lateral wave number of incident wave q for different levels of off-diagonal disorder A.

ment amplitudes ξ_n are small compared to the wavelength. In the numerical examples of this section, wavelength is ten times larger than the interparticle spacing, which is, in turn, much larger than the displacement amplitudes. Additionally, effects of disorder are expected to be averaged if the amplitudes ξ_n are mathematically independent because the scattered field at a given nanosphere is a sum of very large number of secondary waves. In contrast, the ordinary SP is mediated by near-field interactions which are very sensitive to even slight displacements of nanospheres. In addition, the scattered field at a given nanosphere (when the ordinary SP propagates in the chain) is a rapidly converging sum of secondary fields, so that only a few terms in this sum are important and no effective averaging takes place.

B. Diagonal disorder

Diagonal disorder is disorder in the properties of nanospheres. There are several possibilities for introducing such disorder.

First, the spheres can be polydisperse, i.e., have different radii a_n. This is, however, not a truly diagonal disorder. Indeed, polarizability of nth nanosphere can be written in this case as $f_n\langle\alpha\rangle$ where $f_n=(a_n/\langle a\rangle)^3$ and $\langle a\rangle$ is the average radius [note that the radiative correction to $1/\alpha_n$ can be included into the dipole sum $S(k,q)$, so that this analysis remains valid even when this correction is important]. The factors f_n are all positive definite which allows one to introduce a simple transformation of Eq. (1) which removes the diagonal disorder.[38] Then the disorder becomes effectively off-diagonal. More importantly, one can retain a well-defined spectral parameter of the theory, $1/\langle\alpha\rangle$. We, therefore, do not consider polydispersity in this section.

The second possibility is variation of the absorptive parameter γ. This effect that can be practically important. Yet, we are interested in propagation in the absence of Ohmic losses and, therefore, set $\gamma_n=0$.

The third, and the most fundamental, reason for diagonal disorder is variation of the Frohlich frequency of nanospheres. Namely, we take the Frohlich frequency of the nth particle to be $\omega_{Fn}=\langle\omega_F\rangle(1+\xi_n)$, where ξ_n are statistically independent random variables evenly distributed in the interval

$[-A,A]$ and $\omega/\langle\omega_F\rangle=1$. The factors f_n in this case are no longer positive definite and the transformation of Ref. 38 cannot be applied. The most profound consequence of introducing the diagonal disorder is that the spectral parameter such as $1/\alpha$ is no longer well defined. As long as the disorder amplitude is relatively small, one can view $1/\langle\alpha\rangle$ as an approximate spectral parameter. However, as the amplitude of disorder increases, this approach becomes invalid. We have seen in Sec. IV that variation of the ratio ω/ω_F in the interval $0.988<\omega/\omega_F<1.007$ can result in dramatic changes in the way an SP excitation propagates along the chain. For ω/ω_F outside of this interval, ordinary SP could not be effectively excited. However, in Sec. IV, variation of ω/ω_F applied to all nanospheres simultaneously. We now introduce random uncorrelated variation of this ratio for each individual nanosphere. The effects of such disorder are difficult to predict theoretically. We can, however, expect that these effects become dramatic for $A\gtrsim0.01$ since, in this case, the ratio ω/ω_{Fn} can be outside of the interval $[0.988,1.007]$.

We first show in Fig. 10 the same dependencies as in Fig. 5 but for different levels of *diagonal* disorder. The results appear to be, qualitatively, quite similar, although in the case $A=0.016$, there is no visible trend for $x/h>3000$. We then investigate whether the extraordinary SP remains insensitive to diagonal disorder. Data analogous to those shown in Fig. 8, but for diagonal disorder, are presented in Fig. 11. It can be seen that the influence of diagonal disorder on the extraordinary SP is stronger than that of the off-diagonal disorder. When the disorder amplitude A exceeds 0.01, the influence becomes quite dramatic. Paradoxically, at $A=0.016$, the average decay rate is much slower than for $A=0.008$, although the amplitude of fluctuations is much larger.

To see why this happens, we look at the extinction spectra $\sigma_e(q)$. These are plotted in Fig. 12. The fundamental difference between the off-diagonal and diagonal disorder is clearly revealed by comparing this figure to Fig. 9. Namely, the effect of diagonal disorder is not only to broaden and randomize the peak at $q=q_1$, but also to shift it towards the peak corresponding to the extraordinary SP. At sufficiently large levels of disorder ($A\gtrsim0.016$), the separate peaks disappear and a broad structure emerges. At this point, ordinary and extraordinary SP can no longer be distinguished. Correspondingly, the ordinary and extraordinary SP are effectively mixed in the $A=0.016$ curve in Fig. 11, while for smaller amplitudes of the diagonal disorder, the extraordinary SP is excited predominantly.

VI. SUMMARY

We have considered surface plasmon (SP) propagation in a linear chain of metal nanoparticles. Computer simulations reveal the existence of two types of plasmons: ordinary (quasistatic) and extraordinary (nonquasistatic) 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. The ordinary SP can not radiate into the far zone in perfectly periodic chains because its wave number is larger than the wave number $k=\omega/c$ 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. Its excitation is possible due to the existence of the non-Lorentzian optical resonance in the chain introduced in Ref. 17. The extraordinary SP 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. Note that the rate of spatial decay of the extraordinary SP can not be understood by studying the radiative relaxation parameter $Q(k,q)$ shown in Fig. 1. This is because the quasiparticle

pole approximation, which was previously used in a similar context[20] is inaccurate for the extraordinary SP. This plasmonic excitation can be used to guide energy or information in all-optical integrated photonic systems.

We have also considered the effects of disorder and localization of the ordinary and extraordinary SPs. Results of numerical 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 in the paper.

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*Email address: vmarkel@mail.med.upenn.edu

†Email address: asarychev@ethertronics.com

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[24] Strictly speaking, there may be two solutions rather than one, since the curve can cross zero at two separate points. This happens in Fig. 1(a) for $kh \lesssim 0.2\pi$. However, since the peaks are exponentially narrow, the physical effect of exciting two SP with slightly different wave numbers is not expected to be physically observable.

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