Nanofocusing of Optical Energy in Tapered Plasmonic Waveguides

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We predict theoretically that surface plasmon polaritons propagating toward the tip of a tapered plasmonic waveguide are slowed down and asymptotically stopped when they tend to the tip, never actually reaching it (the travel time to the tip is logarithmically divergent). This phenomenon causes accumulation of energy and giant local fields at the tip. There are various prospective applications in nano-optics and nanotechnology.

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The recent explosive progress in nano-optics has been based on the nanoscale local fields that are greatly enhanced due to resonant properties of metal nanosystems [1–4]. Among many dramatic such phenomena and applications, we mention the giant surface-enhanced Raman scattering (SERS) that allows for observation of single molecules [5–8]. Surface plasmon polaritons (SPP's) in nanostructured systems enable one to observe propagation, interference, and imaging on the nanoscale [9–16]. The strong nanolocalized optical fields induce many enhanced nonlinear-optical phenomena and have various prospective applications. The central problem of the nano-optics is the delivery and concentration (nanofocusing) of the optical radiation energy on the nanoscale, which is formidable because the wavelength of light is on the microscale, many orders of magnitude too large. Coupling laser radiation to the nanoscale through, e.g., tapered optical fibers [13] or by focusing on metal tips [7] leads to an enormous loss: only a miniscule part of the excitation energy is transferred to the nanoscale. Here we show that it is possible to focus and concentrate in three dimensions the optical radiation energy on the nanoscale without major losses. This can be done by exciting the surface plasmon polaritons (SPP's) propagating toward a tip of a tapered metal-nanowire surface-plasmonic waveguide. This propagation of SPP's causes their rapid adiabatic slowing down and asymptotic stopping. This phenomenon leads to a giant concentration of energy on the nanoscale. The SPP's are adiabatically transformed into localized surface plasmons (SP's) that are purely electric oscillations that can and do nanolocalize [17] leading to the three-dimensional (3D) nanofocusing.

To introduce this phenomenon of the rapid adiabatic nanofocusing in 3D, here we use the results of the actual computations that will be presented later in this Letter. In our example, the tapered nanoplasmonic waveguide is a silver cone in vacuum; its angle of opening is 0.04 rad, as shown in Fig. 1(a). The vacuum reduced wavelength of the excitation radiation is $\lambda = 100$ nm, which corresponds to red light of $\lambda = 630$ nm. The SPP's are efficiently excited at the wide end of the waveguide nanowire by using, e.g., grating or Kretschman [16] geometry and propagate to the tip as indicated. This propagation causes accumulation of the SPP energy at the tip and the corresponding increase of the local fields by more than 3 orders of magnitude. As shown in Fig. 1(b), the intensity of the local optical field is sharply concentrated in 3D in a nanolayer at the surface of the metal, which is a signature of SPP's. In this figure, as everywhere in this Letter, we show all lengths in units of λ, so the sizes range from the micro to nanoscale. The hot spot of local fields is created in a nanosize region at the very tip. If it were not a plasmonic nanowire waveguide, but the conventional tapered optical fiber supporting guided photonic modes, than there would be a cut off at some waveguide radius

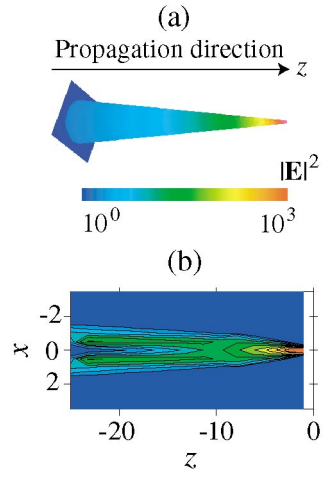

FIG. 1 (color). (a) Geometry of the nanoplasmonic waveguide. The propagation direction of the SPP's is indicated by the arrow. Intensity $I(\mathbf{r}) = |\mathbf{E}(\mathbf{r})|^2$ of the local fields relative to the excitation field is shown by color. The scale of the intensities is indicated by the color bar in the center. (b) Local electric field intensity $I(\mathbf{r})$ is shown in the longitudinal cross section of the system. The coordinates are indicated in the units of the reduced radiation wavelength in vacuum, $\lambda = 100$ nm. The radius of the waveguide gradually decreases from 50 to 2 nm.

beyond which the propagation is not possible [18]: the wave is reflected back with only a short evanescent tail in the forward direction; no field enhancement would occur, and only an exponentially small part of the incident energy would reach the tip.

It was suggested earlier [14] that the propation of SPP's toward the tip can produce energy concentration. It was also noted [19] that at the cut-off point, the guided photonic modes of an optical fiber could couple to the plasmonic modes of its metal coating causing further transfer of optical energy to the tip. However, no role of the adiabatic slowing down and stopping of SPP's was previously elucidated. It is feasible that the observed [15] high efficiency of a metal tip on aperture probe is due to the proposed effect of the adiabatic accumulation.

The physical reason that the nanoplasmonic waveguide is an efficient energy concentrator can be inferred from Fig. 2(a). Both the phase and group velocity of SPP's asymptotically tend to zero toward the nanotip. Consequently, the SPP's are slowed down and adiabatically stopped at $z \to 0$, which leads to their accumulation at the tip. Correspondingly, in Fig. 2(b) the local optical field is oscillating in space with progressively decreasing wavelength and its amplitude increasing by more than an order of magnitude. The highest enhancement is in fact limited only by the minimum tip size that can be considered on the basis of continuous electrodynamics. Importantly, being adiabatic to prevent the back reflection and 3D scattering, this process should be as rapid as possible to prevent losses in the metal.

The theory considers a nanoplasmonic waveguide that consists of a metal nanowire whose axis coincides with the coordinate z axis and whose dielectric function $\varepsilon_m(\omega)$ is uniform in space, where ω is the optical excitation frequency. The radius $R(z)$ of this nanowire is a smooth function of z and is assumed to decrease from microscale for z large negative to a nanoscale size at $z \to 0$, as discussed above, see Fig. 1(a). This wire is surrounded by a dielectric medium with dielectric constant ε_d. Using the smoothness of dependence $R(z)$, we will employ the eikonal approximation [20] also called Wentzel-Kramers-Brillouin (WKB) or quasiclassical approximation in quantum mechanics [21].

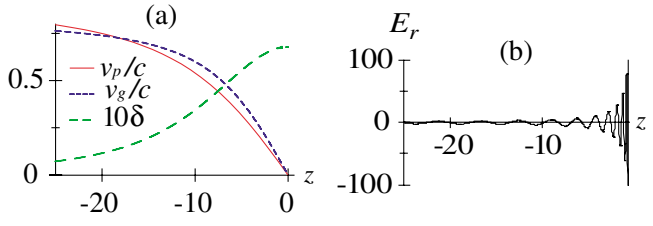

FIG. 2 (color). (a) Phase velocity v_p, group velocity v_g, and adiabatic parameter δ (scaled by a factor of 10) are shown as functions of the coordinate along the nanoplasmonic waveguide. (b) Radial optical electric field at the surface of the metal-nanowire waveguide in the units of the excitation field against the coordinate (in the propagation direction).

We consider an axially uniform SPP mode that is a TM wave whose magnetic field has the ϕ polarization, and electric field $\mathbf{E}$ has both transverse (radial) component E_r and longitudinal component E_z. In the eikonal (WKB) approximation, this field has the form

$$\mathbf{E}(\mathbf{r}, z, t) = \mathbf{E}_0(\mathbf{r}) A(z) \exp[ik_0 \varphi(\mathbf{r}) - i\omega t] \qquad (1)$$

where $\mathbf{r}$ is a two-dimensional (2D) vector in the xy plane, $k_0 = 1/\lambda$, and $A(z)$ is a slow-varying preexponential factor, to be determined later in this Letter. From the Maxwell equations, using the corresponding boundary conditions at the interface, for the SPP guided mode, we find the eikonal as $\varphi = k_0 \int n(z) dz$, where $n(z)$ is the effective surface index of the plasmonic waveguide at a point z, which is determined by the equation

$$\frac{\varepsilon_m}{\kappa_m} \frac{I_1(k_0 \kappa_m R)}{I_0(k_0 \kappa_m R)} + \frac{\varepsilon_d}{\kappa_d} \frac{K_1(k_0 \kappa_d R)}{K_0(k_0 \kappa_d R)} = 0, \qquad (2)$$

where I_p and K_p $(p = 0, 1)$ are the modified Bessel functions; the complex decrements of the field in the metal and dielectric are: $\kappa_m = \sqrt{n^2 - \varepsilon_m}$ and $\kappa_d = \sqrt{n^2 - \varepsilon_d}$. This equation determines n as a function of the local wire radius R, which together with the grading dependence $R = R(z)$ defines the required effective index $n(z)$. Under the conventional plasmonic condition $\mathrm{Re}\varepsilon_m < -\varepsilon_d$, Eq. (2) has nearly real solutions corresponding to the propagating SPP's. For a thick wire $(k_0 R \gg 1)$, the solution is, understandably, the same as for the flat surface, $n = \sqrt{\varepsilon_m \varepsilon_d / (\varepsilon_m + \varepsilon_d)}$. For a thin, nanoscale-radius wire $(k_0 R \ll 1)$ with logarithmic precision, we have

$$n(R) \approx \frac{1}{k_0 R} \sqrt{-\frac{2\varepsilon_d}{\varepsilon_m}} \left[\ln \sqrt{-\frac{4\varepsilon_m}{\varepsilon_d}} - \gamma \right]^{-1}, \qquad (3)$$

where $\gamma \approx 0.57721$ is the Euler constant. Note that at the tip $n \to \infty$, and SPP's do asymptotically stop, i.e., both the phase velocity $v_p = c/n$ and group velocity $v_g = c[d(n\omega)/d\omega]^{-1}$ tend to zero $\propto k_0 R$ for $k_0 R \to 0$. The point $R = 0$ (or $z = 0$) is an essential singularity. The time to reach this point $\propto \int n(R) dR \propto -\ln(k_0 R) \to \infty$ diverges logarithmically. In this regard, the present one-dimensional (1D) wire geometry principally differs from the layered 2D geometry where the stopping of surface plasmons occurs at a regular, finite point [22].

The eikonal parameter (also called WKB or adiabatic parameter) is defined as $\delta = |R' d(k_0 n)^{-1}/dR|$, where $R' = dR/dz$ is the wire grading. For the applicability of the eikonal (WKB) approximation, it necessary and sufficient that $\delta \ll 1$. At the nanoscale tip of the wire, which is the critical site for the adiabaticity (eikonal approximation applicability), from Eq. (3) we obtain

$$\delta \approx |R'\sqrt{-\frac{\varepsilon_m}{2\varepsilon_d}}[\ln\sqrt{-\frac{4\varepsilon_m}{\varepsilon_d}} - \gamma]|.$$

Thus, δ stays finite at the tip and can be made small enough by choosing sufficiently small grading R', so the eikonal approximation is valid for the entire wire waveguide, including the stopping point at the tip. This conclusion does not rely or significantly depend on $\mathrm{Im}\varepsilon_m$ (the optical losses in the system), in contrast with the two-dimensional waveguides in Ref. [22].

Returning to our example, we consider a conic nanowire of silver with $R' = -0.02$, see Fig. 1(a). In Fig. 2(a), along with the phase velocity v_p and group velocity v_g, we show also the adiabatic (WKB) parameter δ. It is of principal importance that this adiabatic parameter does stay finite and small ($\delta \leq 0.07$) throughout the entire system, ensuring the global applicability of the eikonal approximation, including the essentially singular point at $z = 0$.

The SPP electric fields are found from the Maxwell equations in eikonal (WKB) approximation in the form:

$$\begin{aligned} E_z(r, z) &= \theta(R - r) I_0(k_0\kappa_m r) + \theta(r - R) B K_0(k_0\kappa_d r), \\ z) &= \theta(R - r) i\frac{n}{\kappa_m} I_1(k_0\kappa_m r) \\ &\quad + \theta(r - R) i\frac{n}{\kappa_d} B K_1(k_0\kappa_d r), \end{aligned} \tag{4}$$

$E_z($

where $B = I_0(k_0\kappa_m R)/K_0(k_0\kappa_m R)$, and $\theta(\ldots)$ denotes the Heaviside θ function. To determine the preexponential $A(z)$ in Eq. (1), we use the energy flux conservation in terms of the Pointing vector integrated over the normal (xy) plane, obtaining

$$A \propto \mathrm{Re}\left[\frac{n^*\varepsilon_m^*}{|\kappa_m|^2}\left|K_0(k_0\kappa_d R)\right|^2 \int_0^R \left|I_1(k_0\kappa_m r)\right|^2 r dr + \frac{n^*\varepsilon_d^*}{|\kappa_d|^2}\left|I_0(k_0\kappa_m R)\right|^2 \int_R^\infty \left|K_1(k_0\kappa_d r)\right|^2 r dr\right]^{-\frac{1}{2}}, \tag{5}$$

where all the spatially varying quantities, n, κ_m, and κ_d, are functions of local radius R of the wire, as originally given by Eq. (2). The required dependence $A = A(z)$ is obtained by substituting the grading relation $R = R(z)$. We indicate only the proportionality of A: the total scale of A is undetermined by the equations and is defined by the total power of the propagating SPP wave. This completes the eikonal (WKB) solution.

The intensity of the optical electric fields has already been discussed in conjunction with Fig. 1 where they are shown on the logarithmic scale. For this example and below, we set the minimum radius of the wire to be $R_{\min} = 0.02\lambda = 2$ nm to avoid effects of the spatial dispersion of the dielectric response that are important at shorter distances, cf. Refs. [23,24], and the maximum radius (at $z = -25\lambda = -2.5\mu$) to be $R_{\max} = 0.5\lambda =$ 50 nm. In Fig. 3, we display the amplitudes of the local optical fields in the cross section of the system for the normal and longitudinal (with respect to the to the axis) components of the optical electric field. In Fig. 3, far from the tip of the nanoplasmonic waveguide, the optical electric field is mostly transverse, extending in vacuum to distances $\sim\lambda$ where most of the SPP field is propagating. The longitudinal field in the metal is very small, proportional to a factor of $|\varepsilon_d/\varepsilon_m| \ll 1$, as should be from the boundary conditions. Therefore, the guide itself is clearly seen in panel (a) as the acute triangular region of low fields. As SPP's move toward the tip, the SPP fields start to localize at the metal surface, and simultaneously, their wavelength is progressively reducing and amplitude growing. Because the very tip is not included, the singularity point of the fields does not show in these figures. Even with this truncation, the field magnitudes grow significantly at small $|z|$. The transverse x component grows by an order of magnitude as the SPP's approach the tip of the guide, while the longitudinal z component, which is very small far from the tip, grows relatively much stronger. Close to the tip, both these components are of the same order of magnitude, as is expected for the localized excitations. This growth in magnitude is concurrent with the energy localization in 3D and the significant reduction of the wavelength, which are due to the dramatic slowing down of the SPP's. Note the SPP's in Fig. 3 are not standing but running waves; the fields shown represent an instantaneous snapshot of these waves.

In Fig. 4, we show the spatial behavior of relative intensity $I(\mathbf{r})$ and energy density [20] $W(\mathbf{r}) = \{d[\omega\varepsilon(\mathbf{r}, \omega)]/d\omega\}|\mathbf{E}|^2$ of the local optical electric field. The intensity grows by more than three and energy density by 4 orders of magnitude at the tip. If these fields were used to induce SERS, it would be enhanced by seven to 8 orders of magnitude. The further enhancement of SERS

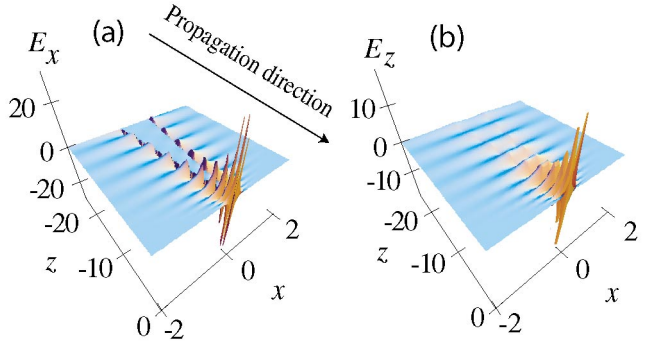

FIG. 3 (color). Snapshot of instanteneous fields (at some arbitrary moment $t = 0$): Normal component E_x (a) and longitudinal component E_z (b) of the local optical electric field are shown in the longitudinal cross section (xz) plane of the system. The fields are in the units of the far-zone (excitation) field.

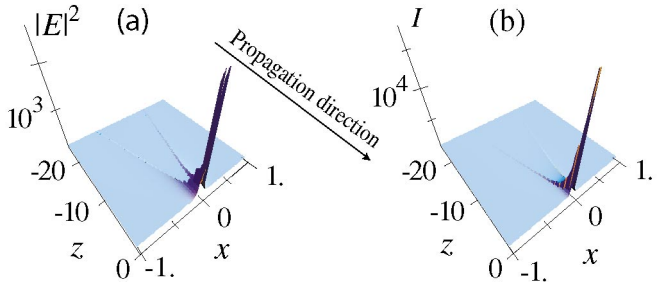

FIG. 4 (color). Mean (time averaged) intensity $I(\mathbf{r})$ (a) and the energy density $W(\mathbf{r})$ (b) of the local optical electric field in the xy plane of the system. The magnitudes are relative to those of the excitation wave.

by many orders of magnitude can be achieved by positioning a resonant nanolens [25] at the tip of the nanoplasmonic waveguide. Note that $|\mathbf{E}|^2$ is concentrated at the outer surface of the metal nanowire, with the exponential decay far from this surface. In contrast, a metal nanoparticle excited by an external field would produce dipolar local fields where $|\mathbf{E}|^2 \propto r^{-6}$. At the same time, $I(\mathbf{r})$ is significantly larger and localized inside the metal plasmonic waveguide where most of the SPP energy is propagating due to the large value of $d[\omega\varepsilon(\mathbf{r}, \omega)]/d\omega$ for metals.

To briefly conclude, we introduce a phenomenon of rapid adiabatic nanofocusing in 3D. This phenomenon is at the foundation of the proposed high-efficiency coupling of the far-field radiation to the near-field zone where the 3D energy concentration occurs at the tip of a smoothly tapered metal nanoplasmonic waveguide. This causes the local field increase by 3 orders of magnitude in intensity and four orders in energy density. The stopping of SPP's is asymptotic, i.e., they need logarithmically divergent time to reach the tip, which mathematically is the point of an essential singularity. Similar phenomena are likely to exist for a hollow tapered waveguides, in particular, subwavelength holes; this is of interest for the enhanced transmission phenomena [26]. The rapid adiabatic nanofocusing promises to find various applications in nano-optics and nanotechnology where greatly enhanced local optical fields are required, in particular, for probing, spectroscopy, detection, and modification on the nanoscale in physics, chemistry, biology, electrical engineering, etc.

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