Giant Surface-Plasmon-Induced Drag Effect in Metal Nanowires

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Here, for the first time we predict a giant surface-plasmon-induced drag-effect rectification (SPIDER), which exists under conditions of the extreme nanoplasmonic confinement. In nanowires, this giant SPIDER generates rectified THz potential differences up to 10 V and extremely strong electric fields up to $\sim 10^5$–10^6 V/cm. The giant SPIDER is an ultrafast effect whose bandwidth for nanometric wires is $\sim$20 THz. It opens up a new field of ultraintense THz nanooptics with wide potential applications in nanotechnology and nanoscience, including microelectronics, nanoplasmonics, and biomedicine.

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The fact that electromagnetic fields exert mechanical forces on matter is well known and has found applications in atomic physics [1,2], bio- and nanotechnology [3–5], and in picosecond photodetectors based on the photon drag effect (PDE) [6]. The semiconductor PDE detectors have proved to be very practical for relatively fast detection of picosecond pulses in a wide frequency range spanning from THz to infrared. It has been proposed to use enhanced fields in phonon-polariton silicon carbide structures for laser particle accelerators [7]. There have been experimental investigations of the PDE in metal nanofilms that are thicker than the skin depth [8,9]. Metals support surface plasmon polaritons (SPPs) [10] that exert forces on electrons causing an SPP-enhanced PDE [8]. However, it is typically rather small, with the induced potential differences in the mV range. This modest enhancement of the PDE in these experiments was due to relatively slow variation of SPP fields in space, and the momentum transferred to the electrons was correspondingly small. A drag effect under the conditions of strong nanoplasmonic confinement, when the SPP localization radius is less than the skin depth ($\sim$25 nm), has not been studied or exploited theoretically or experimentally.

In this Letter, we predict a giant surface-plasmon-induced drag-effect rectification (SPIDER) in metal nanowires, which is fast, with response on the femtosecond time scale. We show that the ultrashort, nanolocalized SPP pulses exert forces on electrons in the nanowires, inducing giant THz electromotive force (emf) in the SPP propagation direction. We have found that in thin ($\sim$5 nm radius) wires this emf can reach $\sim$10 V, with nanolocalized THz fields as high as $\sim$1 MV/cm. Such THz fields have previously been generated in the far zone [11], where they produce nonperturbative effects [12], but not on the nanoscale. In contrast, the SPIDER can serve as a nanosource of high THz fields. We also study dynamics of the SPIDER for short SPP pulses and suggest that adiabatically tapered nanowires [13] can be used as broadband nanophotodetectors with extremely fast response due to the femtosecond momentum-relaxation times in metals [14]. The nature of the giant enhancement of the SPIDER is novel in nanoplasmonics: it is not the enhancement of the optical fields *per se* (the maximum magnitude of the local fields is limited by the breakdown at the metal surface that occurs for fields $\sim$1 V/Å) but the very high *gradients* of these fields. The SPIDER is ultrafast because it is a nonresonant, wide-band effect.

Consider a metal nanostructure with a propagating SPP pulse. The SPP field produces polarization $\mathbf{P}$, charges with macroscopic density $\rho = -\mathrm{div}\mathbf{P}$, current density $\mathbf{j} = \partial\mathbf{P}/\partial t$, and the surface charge density $\sigma = \mathbf{Pn}$ at the surface of the metal, where $\mathbf{n}$ is the normal to the surface pointing outward. We do not consider systems with optical magnetism, whose introduction at optical frequency is problematic [15,16]. Therefore we set $\mathbf{B} = \mathbf{H}$, which precludes the existence of surface currents. Using Eqs. (1)–(5) of the supplementary material [17], we obtain the following general expression for the total force:

$$\mathbf{F} = \int_V \left[\mathrm{grad}(\mathbf{P}^c \cdot \mathbf{E}) + \frac{1}{c}\frac{\partial(\mathbf{P}\times\mathbf{B})}{\partial t} \right] dV, \qquad (1)$$

where superscript "c" implies that the differentiation does not apply to the labeled vector. This result is of fundamental importance for processes involving interaction of nanoplasmonic fields with metal electrons. Equation (1) is valid for a wide range of problems with a general material equation $\mathbf{P} = \hat{\chi}\mathbf{E}$, including those where operator $\hat{\chi}$ describes anisotropic or nonlocal media. The first term in Eq. (1) is related to the force acting on a point dipole moment [18]. The second term in Eq. (1) is the Abraham force. In a monochromatic field, this force averaged over the period of oscillations is zero, but in the field of a pulsed excitation it has a finite magnitude.

We apply Eq. (1) to describe the SPIDER. For certainty, consider a metal nanowire with radius R and dielectric susceptibility $\chi(\omega) = \chi' + i\chi''$, which is oriented along the z axis and embedded into a dielectric with a dielectric permittivity of ε_d. This wire propagates an SPP pulse,

which can be excited by external sources using, e.g., the effect of adiabatic compression [13,19].

In the case of extreme nanoplasmonic confinement ($R \ll l_s$, where l_s is the skin depth), R becomes the only relevant quantity of the dimensionality of length [13]. Therefore there is scaling of all magnitudes in R. The SPP-wave power $\mathcal{P}$ scales as $\mathcal{P} \sim E^2 R^2 v_g$, where $v_g \sim \omega R$ is the SPP group velocity. The SPIDER-induced potential difference [electromotive force (emf)] $\mathcal{E}$ is proportional to the pressure produced by force (1), $\mathcal{E} \sim F/R^2$. The propagation length of the SPP $l_p \sim RQ$, where Q is the SPP figure of merit, independent of R. These arguments allow us to predict scaling of the SPIDER force F, emf $\mathcal{E}$, and the electric field due to SPIDER E_R (which for femtosecond SPP pulses possesses THz frequencies): $F \propto \mathcal{P}R^{-1}$, $\mathcal{E} \propto \mathcal{P}R^{-3}$, $E_R \propto \mathcal{P}R^{-4}$, $E_{mR} \propto R^{-1}$. We have also indicated the scaling of the maximum rectified field E_{mR} (at the maximum tolerable power $\mathcal{P}_m$). The scaling implies that all the effects caused by the SPIDER increase with decreasing the wire radius as its powers. This enhancement is not resonant and therefore has bandwidth comparable to that of the entire optical spectrum. The scaling describes only the dependence on R. There are also prefactors describing the dependence on dielectric permittivities, frequency, etc. They take into account an additional enhancement close to the SP resonant frequency, which is multiplicative. Below in this Letter we show that this scaling is reproduced by the theory results.

The SPPs are transverse magnetic (TM) modes, and their complex fields have the form

$$\mathbf{E} = A(t')(\tilde{E}_z \hat{\mathbf{z}} + \tilde{E}_\rho \hat{\boldsymbol{\rho}}) e^{i(kz-\omega t)}, \quad \mathbf{H} = A(t')\tilde{H}_\varphi \hat{\boldsymbol{\varphi}} e^{i(kz-\omega t)}, \tag{2}$$

where $t' = t - z/v_g$, and v_g is the SPP group velocity at the pulse carrier frequency ω, k is the SPP wave number [13]. Note that $Q = k'/k''$, where $k' = \mathrm{Re}k$, and $k'' = \mathrm{Im}k$; $l_p = 1/(2k'')$. The total power flowing through the plane $z = 0$ at the moment t is $\mathcal{P}(t) = 2\pi \int_0^\infty \bar{S}_z(\mathbf{r}, t)|_{z=0} \rho d\rho$, where $\bar{\mathbf{S}}(\mathbf{r}, t) = (c/8\pi)\mathrm{Re}[\mathbf{E} \times \mathbf{H}^*]$ is the Pointing vector averaged over SPP period. Considering the azimuthally symmetric (TM_0) modes, functions $\tilde{E}_z$, $\tilde{E}_\rho$, and $\tilde{H}_\varphi$ depend only on radius ρ. We normalize them for real amplitude $A(t)$ to satisfy a relation $A^2(t) = \mathcal{P}(t)$. The disregard of the group velocity dispersion in Eq. (2) is valid for pulses with duration of tens of femtoseconds and greater, and frequencies not too close to the SP resonance.

The momentum transferred from the SPPs to the electrons implies forces exerted on them. The density of these forces are given by Eqs. (1) and (2) of the supplementary material [17]. These forces lead to an emf, i.e., optical rectification, which for femtosecond SPP pulses results in the emf in the THz frequency range. Since the electron momentum-relaxation time is on the scale of femtoseconds, electrons come to a local equilibrium in the process of this rectification. Therefore, we will employ the hydrodynamic approximation, for which the pressure p and electrostatic potential ϕ satisfy an equation $p + ne\phi =$ const, where e is electron charge, and n is electron density. From this equation, we can find the emf $\mathcal{E} = \Delta\phi$, which is the total change of potential in the direction of SPP propagation (the z direction), $\mathcal{E} = -\Delta p/(ne)$, where $\Delta p = \bar{F}_z/(\pi R^2)$ is the corresponding change of the pressure. Here, $\bar{F}_z$ is z component of the force (1) averaged over the period of SPP oscillations.

Total force $\bar{F}_z$ is composed of three forces [see Eq. (6) of the supplementary material [17]]: the SPP pressure, striction, and Abraham force $\bar{F}_z = f_z^{\mathrm{pr}} + f_z^{\mathrm{st}} + f_z^A$. These forces result in three terms of the SPIDER emf

$$\mathcal{E} = R_H \left(\frac{\mathcal{F}(t)}{A_{\mathrm{pr}}} + \frac{\mathcal{P}(t)}{A_{\mathrm{st}}} + \frac{\mathcal{F}'(t)}{cL_A} \right), \tag{3}$$

where $R_H = -1/(ecn)$ is the Hall constant, and coefficients A_{pr}, A_{st}, and L_A and $\mathcal{F}(t)$ are defined by Eqs. (7)–(10) of the supplementary material [17]. The Abraham force contribution [the third term in Eq. (3)] is small under a condition $Q \ll (\lambda\!\!\!^{-}/R)^2 \omega\tau$, where $\lambda\!\!\!^{-} = c/\omega$ is the reduced wave length in vacuum, and τ is the pulse duration. This condition is well satisfied for the parameters used in this Letter. We will consider two limiting cases pertaining to Eq. (3): a regime of long pulses ($\tau \gg t_p$) and a regime of short pulses ($\tau \ll t_p$), where $t_p \sim Q/\omega$ is the SPP dissipation time. All computations will be made for a silver [20] nanowire embedded in vacuum ($\varepsilon_d = 1$).

Consider first the long-pulse regime where $\mathcal{F}(t) \approx \mathcal{P}(t)$ [see Eq. (7) of the supplementary material [17]]. In such a case, emf $\mathcal{E}(t)$ follows the pulse-envelope time dependence $\mathcal{P}(t)$. The calculated emf $\mathcal{E}$ as a function of frequency ω and wire radius R is displayed in Fig. 1(a). In contrast to dielectric media, the SPP pressure and striction contributions to the emf have the same sign since $\chi' < 0$. These two contributions are equal if a condition $\chi'' Q = -\chi'$ is satisfied. The black solid line represents this condition; to the left of this line the pressure dominates the SPIDER, and to the right the striction force dominates. This is understandable because close to the SP resonance of the wire (at ≈ 3.7 eV), the gradient of the SPP intensity increases due to the high loss: the striction force is of a gradient nature, dominating therefore. Similarly, with the decrease of R, the intensity gradient increases due to the increased confinement, which also leads to the relative increase of the striction with respect to the pressure force, as we clearly can see from this and other panels of Fig. 1. An increase of the SPIDER at the SP resonance can be seen as the red region in Fig. 1(a) and peaks in Figs. 1(c) and 1(e).

The relative magnitude of the SPIDER emf $\mathcal{E}/\mathcal{P}$ is illustrated in Fig. 1(b) as a function of the wire radius R for a frequency of $\hbar\omega = 2.9$ eV. The SPIDER is gigantically enhanced for strong nanoplasmonic confinement: by 4 orders of magnitude when R decreases from 100 to 5 nm at the same SPP power. There is a pronounced scaling $\mathcal{E}/\mathcal{P} \propto R^{-3}$ at $R \lesssim l_s$, in accord with the discussion following Eq. (1).

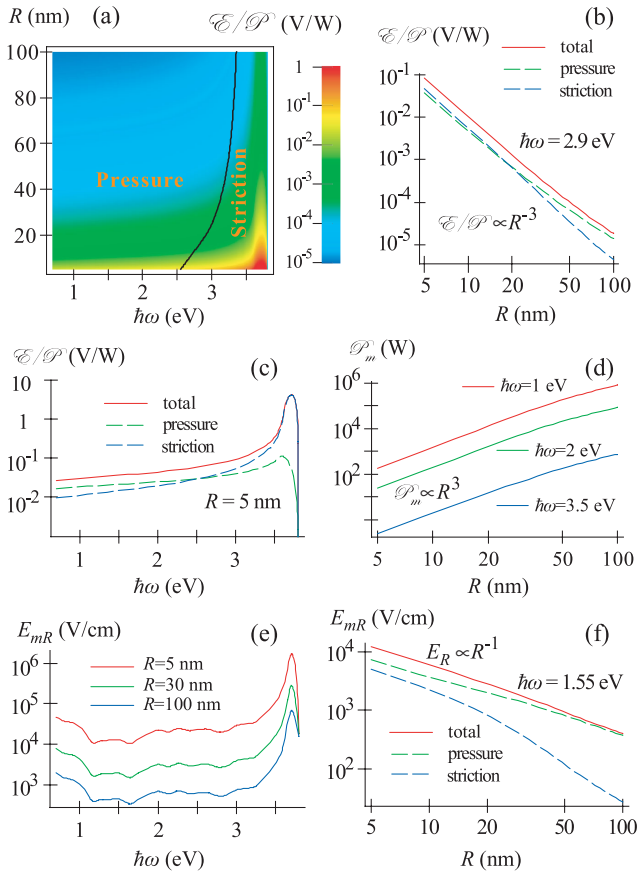

FIG. 1 (color). SPIDER for long SPP pulses: emf and rectified field dependence on the frequency $\hbar\omega$ and wire radius R. Note the logarithmic scale for the magnitude of the effect. (a) Dependence of the SPIDER emf per unit SPP power $\mathcal{E}/\mathcal{P}$ on wire radius and frequency. The black solid curve indicates the parameters at which SPP pressure is equal to striction. The magnitude of the effect is denoted by the color-coding bar. (b) Dependence of SPIDER magnitude $\mathcal{E}/\mathcal{P}$ on wire radius R per unit power of the SPP wave (solid red curve). The contributions of the pressure and striction to the total magnitude of SPIDER are shown by the dashed curves. (c) Dependence of SPIDER magnitude per unit power $\mathcal{E}/\mathcal{P}$ on frequency ω for $R =$ 5 nm. (d) Maximum power that a wire can tolerate $\mathcal{P}_m$ as a function of wire radius R for different frequencies ω. (e) The maximum SPIDER field E_{mR} (for the maximum tolerable power $\mathcal{P}_m$) as a function of frequency for three wire radii $R = 5$, 30, and 100 nm. (f) The maximum SPIDER field E_{mR} (for the maximum tolerable power $\mathcal{P}_m$) as a function of the wire radius R for frequency $\hbar\omega = 1.55$ eV.

The spectral dependence of the relative SPIDER emf, $\mathcal{E}/\mathcal{P}$, for a wire of the smallest radius considered, $R =$ 5 nm, is depicted in Fig. 1(c) [21]. Importantly, the magnitude of the emf in this case is very large, from 0.01 to 10 V/W in the entire optical range, with a pronounced resonance at the SP frequency. This large magnitude shows that the SPIDER can be used for the photodetection on the nanoscale, i.e., in the role that previously was deemed only possible for semiconductors. In this sense, it belongs to the area of the active nanoplasmonics [22].

By classification of nonlinear optics, the SPIDER is a second-order nonlinear effect: $\mathcal{E} \propto \mathcal{P}$. The maximum achievable magnitude of the emf is determined by the maximum $\mathcal{P}$ that the wire can tolerate. This we estimate setting the optical field E at the surface of the wire equal to 1 V/Å [23,24]. For fields significantly higher than this, there will be massive ionization and damage of the metal surface. We plot in Fig. 1(d) this maximum intensity as a function of the wire radius for three SPP frequencies. Note a very good scaling $\mathcal{P}_m \propto R^3$ for a strong nanoplasmonic confinement, i.e., $R \lesssim l_s$. The values of the $\mathcal{P}_m$ in Fig. 1(d) in comparison to the data of Figs. 1(a)–1(c) show that the gigantic values of the SPIDER emf $\mathcal{E} \sim 10$ V are realistically achievable, which are many orders of magnitude greater than observed previously [8,9].

One of the most important for applications properties of the giant SPIDER is high local electric field E_R in the vicinity of the nanowire. Such a field (averaged over the SPP decay length l_p) can be found as $E_R = \mathcal{E}/l_p$. We display the maximum achievable rectified field E_{mR} (at the propagating SPP power of $\mathcal{P}_m$) in Figs. 1(e) and 1(f). As we see from panel (e), the spectral dependence of the SPIDER THz field is very similar for all wire sizes, but the magnitude of this field is much greater for the 5 nm wire: $E_{mR} \sim 10^5$–10^6 V/cm. The nanolocalized THz fields of such a magnitude will excite a wealth of nonlinear THz responses on the nanoscale.

Consider dynamics of the emf response to SPP pulses that differ in duration τ with respect to the SPP pulse dissipation time t_p. The latter is displayed in Fig. 2(a) as a function of the frequency ω. As we can see, time t_p is in the range from 10 to 150 fs. The temporal dependencies of the emf in comparison with the power $\mathcal{P}$ of the SPP pulses for various pulse durations is illustrated in Figs. 2(b)–2(d). For a relatively long pulse ($\tau = 1$ ps $\gg t_p$) shown in Fig. 2(b), the shape of the emf $\mathcal{E}(t)$ repeats that of the power $\mathcal{P}(t)$. This relatively long, picosecond response, nevertheless, corresponds to a 1 THz bandwidth. Note that the amplitude of the emf is very large, ~10 V.

For a much shorter, $\tau = 40$ fs, SPP pulse and the same 5 nm wire, as shown in Fig. 2(c), there is a small broadening and delay of the emf response $\mathcal{E}(t)$ with respect to $\mathcal{P}(t)$. This broadening is due to the pressure force that decays exponentially for long times, as Eq. (7) of the supplementary material [17] suggests, and the broken blue line in the figure indicates. However, this delay and broadening are not large. The frequency-response bandwidth of this wire is very large, ≈20 THz, which is characteristic of the extreme nanoplasmonic confinement. The amplitude of the SPIDER emf is also very large, $\mathcal{E} \sim 10$ V.

For a much thicker nanowire of $R = 30$ nm (weak plasmonic confinement case) and $\hbar\omega = 1.2$ eV, illustrated in Fig. 2(d), the SPP decay time becomes much longer ($t_p =$ 150 fs). This leads to a very significant delay and temporal broadening of the emf response with a pronounced exponential part due to the pressure forces shown by the broken blue line. These behavior is due to the much longer SPP

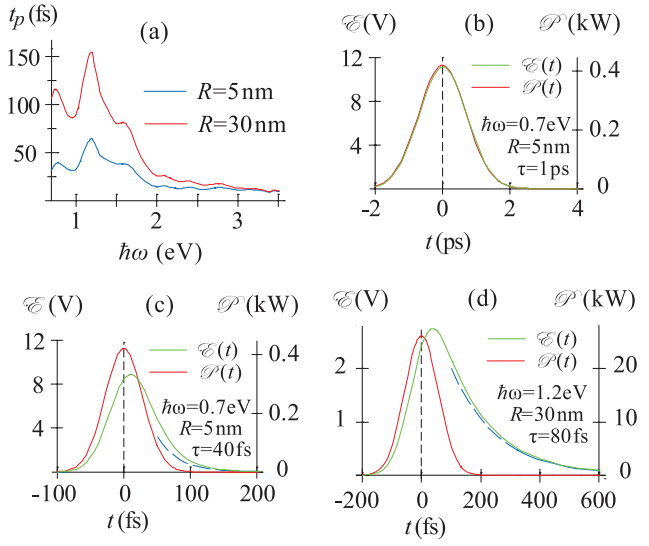

FIG. 2 (color). SPIDER created by ultrashort SPP pulses: fast femtosecond emf response. (a) The dependence of the SPP lifetime $t_p = l_p/v_g$ on the frequency $\hbar\omega$ for $R = 5$ nm and $R = 30$ nm. (b) The time dependence of the emf $\mathcal{E}(t)$ (green line, left scale) and input power $\mathcal{P}(t)$ (red line, right scale). Dashed blue line: exponential tail. The pulse duration is $\tau = 1$ ps $\gg t_p \approx 30$ fs and the emf closely follows the SPP pulse dynamics. (c) The same for much shorter pulse with $\tau = 40$ fs. (d) Emf induced by the short pulse in nanowire with $R = 30$ nm with frequency $\hbar\omega = 1.2$ eV. The emf response is broadened, since $\tau = 80$ fs, while $t_p \approx 150$ fs.

lifetimes for the weak confinement where a significant fraction of the SPP energy propagates in the dielectric (vacuum). Nevertheless, the emf response bandwidth is still very large, on the order of 5 THz, and its amplitude is also very large, $\mathcal{E} \gtrsim 1$ V.

In conclusion, the ultrafast giant SPIDER in metal nanowires excited by ultrashort SPP pulses is predicted in this Letter to generate a gigantic emf up to ~10 V for the SPP waves of realistic and tolerable amplitudes. The SPIDER enhancement is mostly nonresonant and is due to nanoplasmonic confinement. Because of its nonresonant nature, the SPIDER is an extremely fast effect: frequency bandwidth of the generated THz fields is realistically 5–20 THz. Because of high longitudinal localization of the SPP waves in the case of the strong nanoplasmonic confinement, the SPIDER generates very high local THz electric fields at the metal surface, $E_R \sim 10^5$–10^6 V/cm. Such fields are capable of inducing strongly nonlinear responses, including dissociation of molecules. Among possible applications of the giant SPIDER are rectification and detection of the nanoscale femtosecond optical fields, coupling of nanoplasmonic elements to semiconductor devices, nonlinear THz spectroscopy on the nanoscale of chemical and biological nanoobjects for biomedicine, etc.

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