Nonlocal plasmon excitation in metallic nanostructures

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We investigate the excitation of electrostatic wake fields in metallic nanostructures (nanowires) due to the propagation of a short electron pulse. For that purpose, a dispersive (nonlocal) dielectric response of the system is considered, accounting for the conning along the transverse direction of the wire, which generalizes previous results presented in the literature. We discuss the stability conditions of wake fields and show that the underling mechanism can be useful to investigate new sources of radiation in the extreme-ultraviolet range.

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Experimental techniques[1] have been developed for the fabrication of metallic nanostructures (nanowires) of the order of 10 nm or less, recently receiving a considerable amount of attention from the scientific community. Nanowires, compared to other low-dimensional systems, have two conned directions, still leaving one unconned direction for electrical conduction. Due to their unique density of states, such systems are expected to exhibit significantly different optical, electrical, and magnetic properties from their bulk three-dimensional (3D) crystalline counterparts, especially in the limit of small diameters. Even if the number of electrons involved in the relevant features is high, and therefore a continuum description is expected to be adequate, the current models still lack completeness. One of the most striking cases is related to the dielectric response. It was experimentally shown that anomalous absorption can occur in thin metal films[2] due to the excitation of plasmons.[3] Liu *et al.*[4] have recently approached this problem for arbitrarily shaped nanostructures, although they neglected the wave-number dependence in their model. Recently, McMahon *et al.*[5] considered the effects of the nonlocal response by adding dispersion terms (proportional to the wave vector $\mathbf{k}$) in the Drude dielectric function of the bulk (conduction) electrons, as described by

$$\epsilon(\mathbf{k},\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma) - v_F^2 k^2}, \tag{1}$$

where $\epsilon_\infty(\approx 1)$ is the value for $\omega \to \infty$, ω_p and v_F stand for the plasma frequency and the electron Fermi velocity, respectively, and γ represents the electron collision frequency. However, it is well known that further quantum-mechanical effects participate in the dielectric response of electrons at nanoscales. In Ref. 6, Manfredi and Haas have shown that quantum diffraction ($\sim k^4$) plays a significant role in the dispersion of collective modes and instabilities, which, therefore, are expected to play an important role in nanowires as well. In particular, such quantum effects become important when the thermal wavelength is comparable to the typical dimensions of the system.

In this Brief Report, we extend the dispersive Drude model in Refs. 5 and 6 by taking into account the finiteness of the system along the transverse direction in quasi-one-dimensional nanostructures (nanowires). We then apply our result to investigate the excitation of wake fields due to the propagation of a finite electron pulse. It is shown that the features of the wake fields are intrinsically connected with the dispersive response of the system. We suggest that, due to the competition between two spatial scales (say classical and quantum), the excitation of wake fields in such materials can also lead to the emission of radiation in the extreme ultraviolet (XUV) range of frequency.

We start from a set of quantum hydrodynamic (QHD) equations,

$$\frac{\partial n}{\partial t} + \nabla \cdot (n\mathbf{u}) = \mathbf{0}, \tag{2}$$

$$\left(\frac{\partial}{\partial t} + i\gamma\right)\mathbf{u} + \mathbf{u}\cdot\nabla\mathbf{u} = \frac{e}{m_e}\nabla\phi - \frac{\nabla P_F}{m_e n} + \frac{\mathbf{F}_Q}{m_e}, \tag{3}$$

$$\nabla^2\phi = \frac{e}{\epsilon_0}(n - n_0), \tag{4}$$

where n, $\mathbf{u}$, and ϕ are the electron mean density, the electron velocity, and the electrostatic potential, respectively. The closure of the system is established via an equation of state for the electrons of the conduction band,

$$P_F = \frac{m_e v_F^2}{3n_0^2} n^3, \tag{5}$$

with m_e and e standing for the electron mass and charge. The last term in Eq. (3) corresponds to the so-called quantum (or Bohm) force and casts the effects of the quantum diffraction,

$$\mathbf{F}_Q = \frac{\hbar^2}{2m_e}\nabla\left(\frac{\nabla^2\sqrt{n}}{\sqrt{n}}\right). \tag{6}$$

Equations (2)–(6) have also been used to model superdense astrophysical bodies[7] (i.e., the interior of Jupiter and massive white dwarfs, magnetars, and neutron stars, etc.), intense laser-solid density plasma experiments,[8] and ultrasmall electronic devices,[9,10] carbon nanotubes,[11] and quantum diodes.[12] Quantum hydrodynamic models have also revealed important features occurring in superfluidity[13] and superconductivity.[14]

We now consider a cylindrical nanowire of radius a and length $L \gg a$, in such a way that the system can be regarded as quasi-one-dimensional along the longitudinal direction. In that case, we decompose the Laplacian operator, which can be written as $\nabla^2 = \nabla_\perp^2 + \partial^2/\partial_z^2$. Any relevant quantity

$\Psi(=n,u,\phi)$ present in the above equations can therefore be decomposed as

$$\Psi(r,\theta,z,t)=\sum_{l,m}\Psi_{l,m}(z,t)J_m(k_{\perp;l,m}r)\exp(im\theta), \tag{7}$$

where l and m are integers ($|l|\leqslant m$), $k_{\perp;l,m}=\alpha_{l,m}/a$ is the transverse wave number, and $\alpha_{l,m}$ stands for the lth zero of the Bessel function of order m. In the present work, we consider the low-lying modes $l=1$ and $m=0$ only, without any loss of generality but for the sake of definiteness (for simplicity, we denote the transverse wave vector as $k_\perp$ in the remainder of the paper). After linearization, Eqs. (2)–(4) can be expressed in the following form:

$$\frac{\partial n_{l,m}}{\partial t}+n_0\frac{\partial u_{l,m}}{\partial z}=0, \tag{8}$$

$$\left(\frac{\partial}{\partial t}+i\gamma\right)u_{l,m}=\frac{e}{m_e}\frac{\partial\phi_{l,m}}{\partial z}-\frac{v_F^2}{n_0}\frac{\partial n_{l,m}}{\partial z}+\frac{\hbar^2}{4m_e^2n_0}\frac{\partial^3 n_{l,m}}{\partial z^3}, \tag{9}$$

$$\left(\frac{\partial^2}{\partial_z^2}-k_\perp^2\right)\phi_{l,m}=\frac{e}{\varepsilon_0}(n_{l,m}). \tag{10}$$

After Fourier transforming Eqs. (8) and (9) and using the constitutive relation $\mathbf{D}(\mathbf{k},\omega)=\epsilon_0\epsilon(\mathbf{k},\omega)\mathbf{E}(\mathbf{k},\omega)$, we can easily derive the dielectric function for the conduction band,

$$\epsilon(\mathbf{k},\omega)=1-\frac{k^2}{k^2+k_\perp^2}\frac{\omega_p^2}{\omega(\omega+i\gamma)-v_F^2k^2+h^2k^4/4m^2}. \tag{11}$$

This expression describes a nonlocal, dispersive dielectric response of the system, where the finiteness of the system along the transverse direction ($\sim k_\perp^2$) is taken into account and can also be used to described low-frequency electron waves in quantum plasmas.[15] In the limit of very-low-frequency waves, a coupling is possible between plasmons (or electron phonons) and the usual lattice phonons. Such a coupling, however, is a second-order (nonlinear) effect, smaller than the effect introduced on the electrons, and therefore without any major consequence on the results of the present paper. Nonetheless, we consider it worthwhile to unravel its physics in a future work. In the limit where the transverse finiteness is negligible, i.e., $L\sim a$, one easily recovers the Drude model in Ref. 6 or its truncated version in Eq. (1).

We now examine the consequences of a propagation of an electron pulse in a dispersive medium with dielectric response given by Eq. (11). In that case, the electron density is given by $n=n_0+n_{l,m}+N_{l,m}$, where $N_{l,m}$ is the electron pulse. Plugging into Eq. (10),

$$\left(\frac{\partial^2}{\partial z^2}-k_\perp^2\right)\phi_{l,m}=\frac{e}{\epsilon_0}(n_{l,m}+N_{l,m}), \tag{12}$$

and assuming a typical situation where the collision frequency is negligible, $\gamma\ll\omega_p$,[16] Eqs. (8), (9), and (12) easily yield

$$\left(\frac{\partial^2}{\partial t^2}+\omega_p^2-v_F^2\frac{\partial^2}{\partial z^2}+\frac{\hbar^2}{4m_e^2}\frac{\partial^4}{\partial z^4}\right)\left(\frac{\partial^2}{\partial z^2}-k_\perp^2\right)\times n_{l,m}+k_\perp^2\omega_p^2n_{l,m}=-\omega_p^2\frac{\partial^2}{\partial z^2}N_{l,m}. \tag{13}$$

Assuming that the electron pulse propagates with velocity $\mathbf{v}_0=v_0\hat{\mathbf{z}}$, we introduce the axial Lagrange coordinate $\zeta=z-v_0t$ to get

$$\left\{\frac{\partial^2}{\partial\tau^2}+(V^2-1)\frac{\partial^2}{\partial\xi^2}-2V\frac{\partial^2}{\partial\tau\partial\xi}+1+\frac{H^2}{4}\frac{\partial^4}{\partial\xi^4}\right\}\times\left(\frac{\partial^2}{\partial\xi^2}-K_{\perp l,m}^2\right)\tilde{n}_{l,m}+K_{\perp l,m}^2\tilde{n}_{l,m} \tag{14}$$

$$=-\frac{\partial^2}{\partial\xi^2}\tilde{N}_{l,m}, \tag{15}$$

where $\xi=\zeta/\lambda_F$, $\tau=\omega_pt$, $\tilde{N}_{l,m}=N_{l,m}/n_0$, $K_\perp=k_\perp\lambda_F$, and $V=v_0/v_F$ are dimensionless variables. Here, $H=\hbar\omega_p/(2k_BT_F)$ is a dimensionless quantum. For short electron pulses, $\omega_{pe}^{-1}\gg\tau$, where τ is the typical duration of the pulse, an electrostatic wake field is expected to be excited (for Au nanowires, the plasma frequency ω_{pe}^{-1} is of the order of a few femtoseconds). The stationary solution in the moving frame, $(\partial/\partial\tau\to0)$, corresponds to the case in which the pulse propagates with negligible deformation and can be easily solved. In that case, we may write Eq. (14) as

$$\left(K_a^2\frac{\partial^2}{\partial\xi^2}+\frac{H^2}{4}\frac{\partial^4}{\partial\xi^4}+K_b^4\right)\tilde{n}_{l,m}(\xi)=-\tilde{N}_{l,m}(\xi), \tag{16}$$

where $K_a^2=V^2-1-H^2K_\perp^2/4$ and $K_b^4=1+K_\perp^2(1-V^2)$. Taking the Fourier transform of Eq. (16), one obtains

$$\hat{n}_{l,m}(K)=-\hat{N}_{l,m}(K)\hat{G}(K), \tag{17}$$

where $\hat{G}(K)$ is the Fourier-transformed Green function and can be expressed as

$$\hat{G}(K)=\frac{1}{(K^2-K_+^2)(K^2-K_-^2)}, \tag{18}$$

with

$$K_\pm^2=\frac{K_a^2\pm\sqrt{K_a^4-H^2K_b^4}}{H^2/2}. \tag{19}$$

We notice that the existence of two spacial frequencies $K_\pm$ is a strong consequence of the quantum diffraction: in the limit $\hbar\to0$, only one modulation frequency would be present [see Eq. (15)]. The solution to Eq. (16) is therefore readily obtained via the convolution theorem

$$\tilde{n}_{l,m}(\xi)=\int_{-\infty}^{\infty}\tilde{N}_{l,m}(\xi_0)G(\xi-\xi_0)d\xi_0, \tag{20}$$

where $G(\xi-\xi_0)$ is the inverse Fourier transform of $G(K)$, which leads to the following solution to the density perturbation created by a short electron pulse moving along the axis of the nanowire:

$$\tilde{n}_{l,m}(\xi)=\frac{1}{K_+^2-K_-^2}\int_{-\infty}^{\infty}d\xi_0\left(\frac{\sin K_+(\xi-\xi_0)}{K_+}-\frac{\sin K_-(\xi-\xi_0)}{K_-}\right)\Theta(\xi_0-\xi)\tilde{N}_{l,m}(\xi_0), \tag{21}$$

with $\Theta(x)$ representing the step function. We can use $\tilde{N}_{l,m}(\xi_0)=N_0\exp(-\xi_0^2/\sigma^2)$ to describe a Gaussian electron pulse of width σ. The latter equation describes the excitation

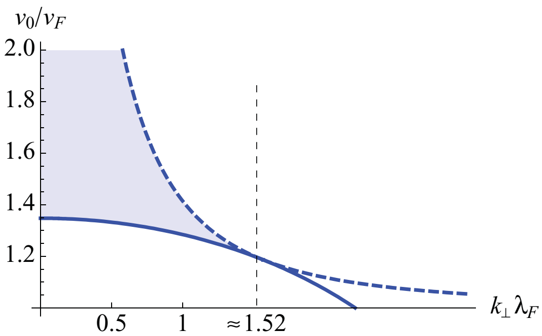

FIG. 1. (Color online) Wake-field stability diagram for Au nanowires ($H \approx 0.817$) as a consequence of the finiteness of the system along the transverse direction. The two curves coincide at $k_\perp^{\rm cr} \approx 1.52/\lambda_F$ (vertical line). The shadowed area is stable.

of a wake field when a short pulse is set to propagate in a metallic nanowire.

Stable electrostatic wake fields can be excited provided the inequalities $K_a^2 > 0$, $K_b^4 > 0$, and $K_a^4 > H^2 K_b^4$ are simultaneously verified. These conditions constrain the velocity of the electron pulse as follows:

$$\left(1 + H - \frac{H^2 K_\perp^2}{4}\right)^{1/2} < V < \left(\frac{1 + K_{\perp l,m}^2}{K_\perp^2}\right)^{1/2}. \quad (22)$$

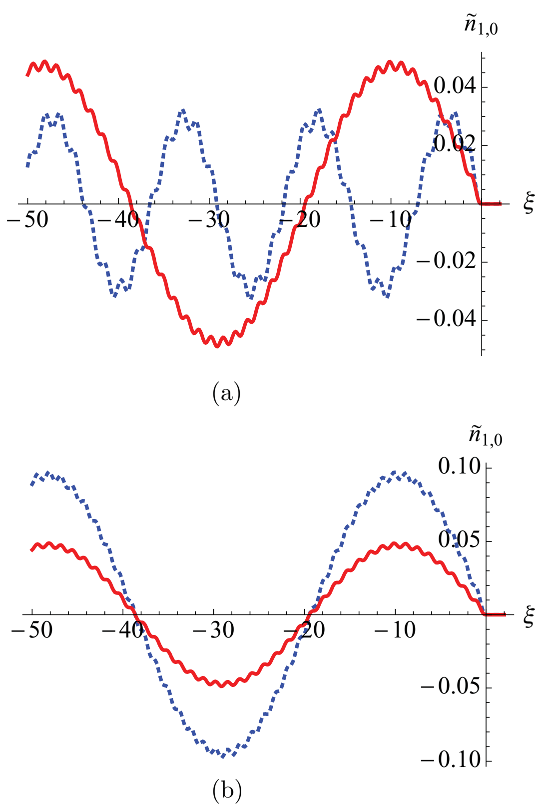

FIG. 2. (Color online) The excitation of a wake field due to a short electron pulse as function of (a) the velocity of the pulse, $v_0 = 1.85 v_F$, blue dashed line (light gray dashed line), and $v_0 = 2.21 v_F$, red solid line (dark gray solid line), for a width of $\sigma = 0.1\lambda_F$; (b) the width of the electron pulse, $\sigma = 0.1\lambda_F$, blue dashed line (light gray dashed line), and $\sigma = 0.2\lambda_F$, red solid line (dark gray solid line), obtained for $v_0 = 2.21 v_F$.

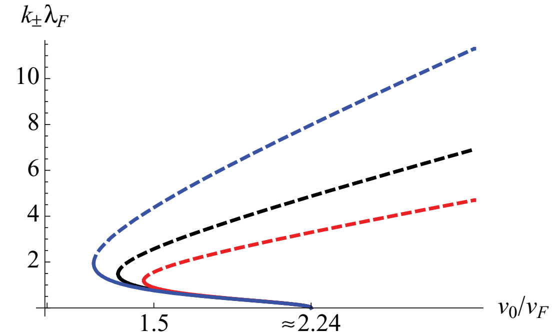

FIG. 3. (Color online) Wave numbers k_-, solid lines, and k_+, dashed lines, for different values of the quantum parameter H, plotted for $k_\perp = 0.5\lambda_F$: $H = 0.51$, blue line (light gray line); $H = 0.817$, black line (black line); and $H = 1.2$, red line (dark gray line). The lower cutoff corresponds to the upper limit in Eq. (22) to the pulse velocity v_0, which is independent of H. For $k_\perp \approx 2.41/a$, we have $v_0 \approx 2.24 v_F$.

We consider the concrete case of Au nanowires,[6] for which the electron density is $n_0 = 5.85 \times 10^{22}$ cm^{-3} and the Fermi temperature is $T_F = 63\,736.8$ K. In that case, we obtain $\omega_p \sim 1.54$ eV, $V_F \sim 1.39 \times 10^6$ m/s, and $\lambda_F \sim 10.2$ nm.

The stability diagram of the wake field is plotted in Fig. 1 for $H \sim 0.817$. The critical value of the wave number, above which dynamical instability occurs, strongly depends on the quantum parameter H (the ratio of the plasmon to the Fermi energies) and is defined as $k_\perp^{\rm cr} = \sqrt{2/H} \sim 1.52/\lambda_F$. Stable wake-field solutions for different sets of parameters are illustrated in Fig. 2. It is observed that the quantum oscillations, of periodicity k_+, are enhanced for very short pulses, i.e., $k_+\sigma \lesssim 1$. The pulse velocity v_0 also plays a role in the amplitude of the wake field, as it is related with

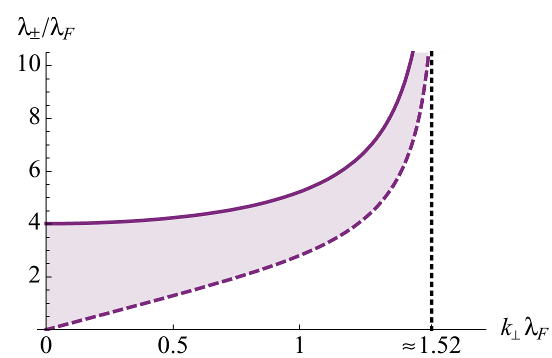

FIG. 4. (Color online) Accessible wavelength range (shadowed area) for the radiation emitted by the wake field as a function of the transverse radius of the nanowire ($k_\perp \approx 2.41/a$, see discussion in the text). The lower and upper curves correspond to the boundaries of stable wake fields in Eq. (22). The values fit the XUV wavelength range.

the value of k_- and k_+. The width of the pulse also seems to change the amplitude of the wake field, and that can be easily understood by noticing that Eq. (21) provides a factor of the form $\exp(-k_\pm\sigma)$, which damps the amplitude of the perturbation for long pulses. The difference between the modes $k_\pm$ can be enhanced by changing the value of the quantum parameter H (see Fig. 3). In practice, this simply corresponds to choosing a different metal, since H only depends on the nature of the sample.

The excitation of wake fields may also open the door for the investigation of possible new sources of radiation in nanoscale devices. The electron current created by the wake field emits radiation at frequencies $\omega_\pm \sim ck_\pm$. In Fig. 4, we evaluate the range of frequencies generated by a short pulse propagating in a nanowire by plugging the stability condition (22) into the definition of k_+ and k_-. For the special case of Au nanowires mentioned earlier, typical experiments are performed with cylinders of radius $a \sim 5$ nm, which corresponds to a transverse cutoff $k_\perp \sim 0.5/\lambda_F$. Stable structures can therefore modulate electrons at wavelengths of $\lambda \sim 10$–40 nm. We stress that the narrowness of the spectrum strongly depends on the value of $k_\perp$, as can be readily observed in Eq. (22): in the limit $k_\perp \to 0$, no upper limit would exist for the beam velocity v_0, which would broaden the spectrum. Also, the emission range can be broader or narrower provided that the value of H is decreased of increased, respectively (see Fig. 3). When properly amplified, these signals can be used to produce radiation in the XUV range. This may be particularly interesting in experiments where high-sensitivity, low-power XUV radiation is needed.

To summarize, we have extended the usual hydrodynamic description of metallic nanowires and derived an expression for the nonlocal (dispersive) dielectric constant accounting for finite-size effects in the transverse direction. This dispersive dielectric response of the system is then considered to investigate the excitation of wake fields due to the propagation of a short electron pulse. We show that stable wake-field generation is expected to occur for a reasonable set of experimentally accessible parameters. It was also shown that the competition between the classical and quantum behaviors of the system provides two scales of electronic modulation (k_+ and k_-), which strongly depend on the finiteness condition along the radial direction (a similar result without conning was obtained in Ref. 17). Finally, we investigate a possible application of wake-field excitation in nanowires as a narrow source of radiation in the XUV range. With the available state-of-the-art technology, very short pulses can be generated also at low power (using, e.g., lasers diodes[18]), allowing wake fields to be studied outside the traditional areas of plasma physics and, in particular, the investigation of wake-field phenomena in the context of nanotechnology.

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