

# Nonlocal ponderomotive nonlinearity in plasmonics

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We analyze an inherent nonlinearity of surface plasmon polaritons at the interface of Fermi–Dirac metal plasma, stemming from the depletion of electron density in high-intensity regions. The derived optical nonlinear coefficients are comparable with the experimental values for metals. We calculate the dispersion relations for the nonlinear propagation of high-intensity surface plasmon polaritons, predicting a nonlinearity-induced cutoff and vanishing group velocity. © 2010 Optical Society of America  
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Plasmonic nanocircuits present promising solutions for on-chip interconnect and have gained considerable interest recently [1,2]. The subdiffraction light confinement by surface plasmon polaritons (SPPs) [3,4] paves the way for efficient nonlinear interactions and sensing [5]. Plasmonic structures are used as building blocks for metamaterials with a negative index of refraction [6]. In the linear regime, behavior of the SPP on a single metal–dielectric interface is well known [7,8], while the nonlinear regime of plasmonic waveguides [9,10] is still not fully explored. In general, metal nonlinearities may stem from several physical effects. In metal nanoparticles [11] the most significant contribution to the nonlinear response is attributed to the limited volume of the particle [12], while in bulk metals this effect does not exist [13]. However, bulk metals, which serve as the key medium for SPP guiding, are inherently nonlinear. These nonlinearities may induce dramatic changes in device performance [14] and should be considered in plasmonic circuitry modeling [15] and in the analysis of SPP localization [16], as well as for practical applications such as optical delays [17]. Previously studied nonlinear effects in bulk metals are related to the saturation of interband transitions [18] that are wavelength dependent, and hot-electron contribution [19] related electron–electron scattering rate change [20].

Here we present a nonlocal metal nonlinearity, originating from a collective ponderomotive interaction of charged particles, where the charge carriers are repelled from the high-field intensity region, making the dielectric constant  $\varepsilon$  average-intensity dependent. We develop a theoretical model for the ponderomotive nonlinearity and its effect on basic SPP propagation, resulting in intensity-dependent SPP light slowing and propagation cutoff. This ponderomotive nonlinearity, stemming from the equation of motion in metals, is present in plasmonics even far from interband transition wavelengths [21].

To model the dependence of metal dielectric coefficient  $\varepsilon_M$  on the field intensity, we calculate the modification of charge carrier density by the nonlocal ponderomotive (Gaponov–Miller) force given by [22–24]

$$\vec{F}_{PM}(\vec{r}) = -\frac{1}{m} \left( \frac{e}{\omega} \right)^2 (\vec{E}(\vec{r}) \times (\nabla \times \vec{E}(\vec{r})) + \vec{E}(\vec{r}) \cdot \nabla \vec{E}(\vec{r})), \quad (1)$$

where  $e$  and  $m$  are the electron charge and mass,  $\omega$  is the light angular frequency, and  $\vec{E}(\vec{r})$  is the local field amplitude. The force can be recast in a simpler form  $F_{PM} = -1/2m(e/\omega)^2 \nabla(|E(\vec{r})|^2)$ , and the corresponding ponderomotive potential is

$$\Phi_{PM}(\vec{r}) = e^2 |E(\vec{r})|^2 / 2m\omega^2. \quad (2)$$

The carrier density in dilute plasma physics is derived by applying Maxwell–Boltzmann distribution for the ponderomotive potential [25,26]. Here we apply the Fermi–Dirac electron distribution suitable for metals in quasi-equilibrium, resulting in the following carrier density:

$$n_{PM}(\vec{r}) = \frac{1}{3\pi^2} \left( \frac{2m}{\hbar^2} \right)^{3/2} (E_f - \Phi_{PM}(\vec{r}))^{3/2}, \quad (3)$$

where  $E_f$  is the Fermi energy assumed to be located deep within the band, such that zero temperature approximation for the distribution is applicable. The resulting intensity-dependent metal–dielectric constant is

$$\varepsilon_{PM}(|E(\vec{r})|^2) = 1 - \frac{e^2}{3\pi^2 \varepsilon_0 m \omega^2} \left( \frac{2m}{\hbar^2} \right)^{3/2} \times \left( E_f - \frac{e^2 |E(\vec{r})|^2}{2m\omega^2} \right)^{3/2}, \quad (4)$$

For realistic field intensities the leading term in Taylor series yields a Kerr-like nonlinearity:

$$\begin{aligned} \varepsilon_{PM}(|E(\vec{r})|^2) &= 1 - \left( \frac{\omega_p}{\omega} \right)^2 + \frac{3}{2} \left( \frac{\omega_p}{3\pi^2 \varepsilon_0 \hbar m e} \right)^{2/3} \frac{e^4}{\omega^4} |E(\vec{r})|^2 \\ &= \varepsilon_M + \chi_{PM} |E(\vec{r})|^2, \end{aligned} \quad (5)$$

where  $\omega_p$  is the plasma frequency for low intensity,  $\varepsilon_0$  is the vacuum permittivity,  $\varepsilon_M$  is the linear part of

the metal dielectric constant, and  $\chi_{PM}$  is the nonlinear ponderomotive susceptibility. This Kerr-like coefficient is highly dispersive ( $\sim 1/\omega^4$ ) and for telecom wavelengths is on the order of  $\chi_{PM} \sim 10^{-18} \text{ m}^2/\text{V}^2$ , comparable with that of nonlinear glasses.

We study the nonlinear propagation effects due to the ponderomotive potential in the most basic structure supporting SPPs, namely, a single metal-dielectric interface. The nonlinear dispersion relations were extracted by several methods. The first is a crude effective index approach, using the nonlinear term for the SPP dispersion:

$$\beta = \sqrt{\frac{\varepsilon_D \varepsilon_{PM}}{\varepsilon_D + \varepsilon_{PM}}}, \quad (6)$$

where  $\varepsilon_D$  is the dielectric constant of the substrate, substituting the nonlinear  $\varepsilon_{PM}$  from Eq. (5). Equation (6) does not represent the accurate nonlinear dispersion; however, it yields a qualitative description, which is refined by the exact quantitative models below. An intuitive explanation of the nonlinear effects is related to the electron depletion in high-intensity regions near the metal-air interface. As a result of the depletion, the metal-dielectric constant near the interface approaches the critical value of  $\varepsilon_{PM} = -\varepsilon_D$ , increasing the effective index (Fig. 1 dashed red curve). Since SPP power propagation directions in the dielectric and in metal are opposite, a corresponding nonlinear SPP cutoff is reached when the intensity-induced mode reshaping results in exactly equal, but opposite, power flow in metal and dielectric.

The second modeling method consistently extracts the nonlinear dispersion relations from the appropriate nonlinear Maxwell's equations and boundary conditions taking into consideration the full spatial dependence of the field, however, without deriving the latter explicitly [27,28]. We assert here that

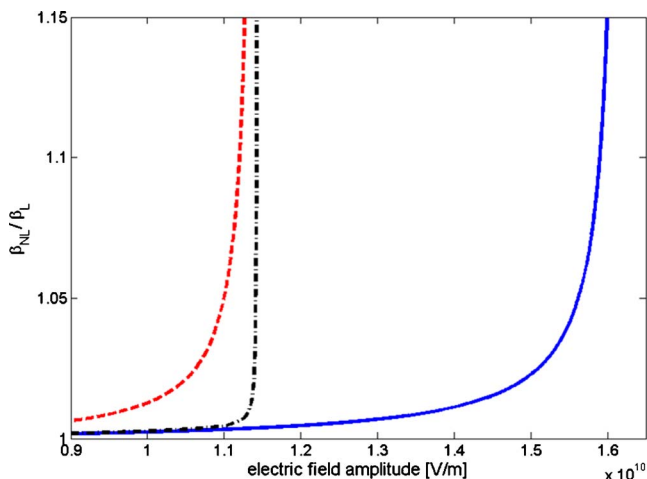


Fig. 1. (Color online) Nonlinear effective index  $\beta_{NL}$ , normalized by the low-intensity linear effective index  $\beta_L$  versus the electric field magnitude in the metal at the air-gold interface for wavelength of  $1.5 \mu\text{m}$  calculated by: effective index method (dashed red curve), scalar dispersion equation (dashed-dotted black curve) and the full vectorial method (solid blue curve).

$E_z(x) \gg E_x(x)$ , such that propagation of a single dominant component  $E$  field is considered. The resulting nonlinear dispersion relation is

$$\frac{\varepsilon_D}{\sqrt{\beta^2 - \varepsilon_D}} + \sqrt{(\beta^2 - \varepsilon_M) - \frac{1}{2}\chi_{PM}E_{z0}^2} \times \frac{(\varepsilon_M + \chi_{PM}|E_z(0)|^2)}{\beta^2 - (\varepsilon_M + \chi_{PM}|E_z(0)|^2)} = 0, \quad (7)$$

where  $E_z(0)$  is the magnitude of the electrical field phasor on the interface. This method is expected to be inaccurate near the modal cutoff point—where the magnitude of both field components is comparable; however, below this point it agrees with the following exact model (Fig. 1).

Finally, a full vectorial method was applied [29] (beyond the approximation of  $E_z(x) \gg E_x(x)$ ), yielding the exact dispersion relation as coupled nonlinear equations:

$$\left[ \varepsilon_M + \frac{\varepsilon_D^2}{\beta^2 - \varepsilon_D} \right] E_{z0}^2 - \varepsilon_D \beta \frac{E_z(0)E_x(0)}{\sqrt{\beta^2 - \varepsilon_D}} - \frac{1}{2}\chi_{PM}E_x^4(0) + \frac{1}{2}\chi_{PM}E_z^4(0) = 0,$$

$$\beta \varepsilon_D \frac{E_z(0)}{\sqrt{\beta^2 - \varepsilon_D}} = (\varepsilon_M + \chi_{PM}E_x^2(0) + \chi_{PM}E_z^2(0))E_x(0), \quad (8)$$

where  $\beta$  and  $E_x(0)$  are the respective effective index and  $x$  component of the electrical field phasor amplitude on the interface, while  $E_z(0)$  is related to a surface intensity parameter by  $E_{av}^2 = E_x^2(0) + E_z^2(0)$ .

The effective indices calculated by the three methods are depicted in Fig. 1 versus the field magnitude ( $E_z(0)$ ) on a gold-air interface at  $\lambda = 1.55 \mu\text{m}$ . All the effective indices exhibit an intensity cutoff, while the first two approximated methods underestimate the intensity required for the cutoff. The group-velocity reduction, evident from the full-fledged calculation of the nonlinear dispersion relations in Fig. 2, is shown explicitly as the group index in Fig. 3. The fields required for the effects described here are relatively high; however, for high fields and depleted metal plasma the SPP resonance becomes much closer to the telecom wavelength range making the normal and the tangential components of the electric field comparable with similar values on both sides of the interface—metal and air. Such field magnitudes ( $\sim 10^{10} \text{ V/m}$ ), although close to the ionization damage threshold, were recently used to predict the spectral phase interferometry for the direct electric-field reconstruction effect [30] and achieved experimentally in bow-tie antenna configuration for high-harmonic generation [31]. It was also shown [32] that the thermal damage threshold intensity in bulk material is much higher than that of small particles [31]. The proposed experiments in this regime should

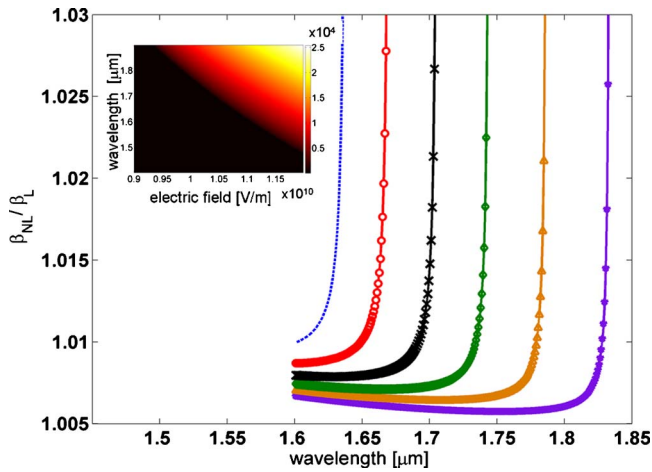


Fig. 2. (Color online) Nonlinear dispersion relation of a single-surface SPP on air-gold interface at different interface electric field amplitudes: dashed blue, 12 GV/m; red circles, 11.5 GV/m; black crosses, 11 GV/m; green diamonds, 10.5 GV/m; brown triangles, 10 GV/m; purple stars, 9.5 GV/m. The inset is the nonlinear effective index normalized by the linear one versus the wavelength and field amplitude.

be performed with low-duty-cycle short-pulse lasers in order to avoid any material damage.

In conclusion, we have analyzed a metal nonlinearity due to the ponderomotive force, which repels charge carriers from high-field intensity regions and introduced its effect on the metal susceptibility, which can be described approximately as a dispersive Kerr-like effect, with magnitudes similar to that of nonlinear glasses. The propagation of a single-surface nonlinear SPP was studied, and the exact nonlinear dispersion curve was derived analytically. The cutoff and slow-light features of the nonlinear dispersion were explained.

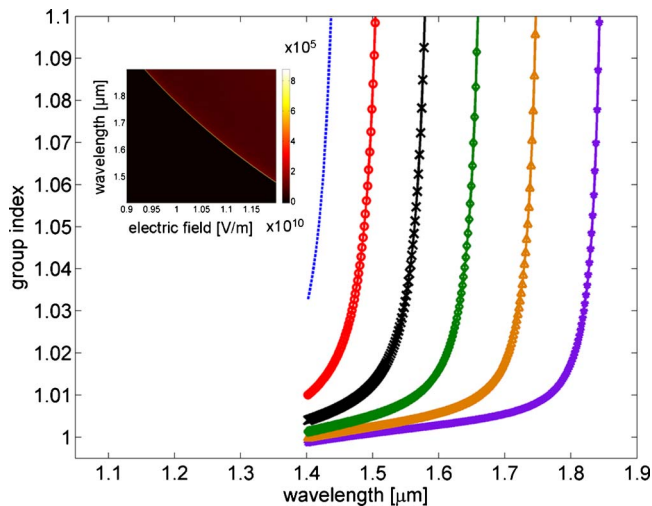


Fig. 3. (Color online) Group index versus wavelength at different electric field amplitudes: dashed blue, 12 GV/m; red circles, 11.5 GV/m; black crosses, 11 GV/m; green diamonds, 10.5 GV/m; brown triangles, 10 GV/m; purple stars, 9.5 GV/m. The inset is the nonlinear group index normalized by the linear one versus the wavelength and field amplitude.

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