Femtosecond-pulse control in nonlinear plasmonic systems

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The collision of the two surface plasmon polariton pulses at the interface between a metal and a dielectric with cubic nonlinearity is investigated. We reveal the possibility of the reflection of a weak signal surface plasmon from the strong pump plasmon pulse resulting in the time delay and spectral shift in the signal plasmon pulse. Using such an interaction, one can control the propagation dynamics of the signal pulse via the modulation of the intensity of the pump pulse.

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I. INTRODUCTION

A. Pulse collision as a method of light-by-light control

Methods of light-by-light control are of great interest for various photonics applications [1,2]. Nowadays, a huge variety of methods of all-optical switching has been proposed including soliton interaction [3–5], usage of specially designed photonic crystals [6,7], and cavities [8,9]. Another method presented in Ref. [10] is based on the phenomenon analogous to the total internal reflection of light implemented in the time domain for the bulk optical pulses.

The idea of the method is the following. A high-power pump pulse induces the change in the refractive index in the dielectric (which can occur due to the various nonlinear mechanisms). A weak signal pulse at another frequency propagating with a different velocity due to the dispersion reaches the moving-induced inhomogeneity. If the refractive index variation is enough, the signal pulse can be "reflected" from the inhomogeneity, that means it can slow down and can continue traveling behind the pump pulse (or vice versa, depending on the sign of the group-velocity dispersion). Therefore, the relative position of the two pulses and the delay of the signal pulse can be controlled by the pump intensity.

The advantage of such a light-by-light control method is that it allows manipulating the signal pulse in a rather easy way. Such switching can be performed both in spatial and in temporal domains using media with quadratic, cubic, photorefractive, or thermal nonlinearity (see Refs. [11–13]). No phase-matching conditions are required to be satisfied. At the same time, for the experimental realization of the optical switching, a very fast nonlinear response together with a rather low threshold value of the pump intensity is needed. However, the nonlinear mechanisms mentioned above are either slow (for example, thermal or photorefractive nonlinearity) or require rather high pump intensities (i.e., cubic or quadratic nonlinearities). The implementation of such a method in plasmonic systems can maintain the advantages of a low threshold intensity and a high switching rate, however, it is quite a challenging task.

B. Plasmonic interface as a system for nonlinear interaction

Plasmonic systems are promising for the performance of light-by-light control at the nanoscale [14–16]. The main reason for using surface plasmon polariton (SPPs) instead of the bulk waves is the following. Due to the high energy concentration in the SPP wave near the metal-dielectric interface, plasmonic systems can provide more efficient methods of light control in contrast to ordinary crystals or optical fibers [17]. The efficiency of the various light-matter interactions increases, that leads to the amplification of different linear (e.g., Refs. [18–20]) and nonlinear effects (e.g., Refs. [21,22]).

Let us discuss the features of a plasmonic interface used for a nonlinear interaction of optical pulses. The bulk radiation can be transformed into the surface wave propagating along the metal-dielectric interface in several ways [23,24]. First of all, one can use a coupling prism in a scheme of attenuated total internal reflection with Otto or Kretschmann geometry. The second way, which is supposed to be more convenient for SPP pulse management, is to use the coupling diffraction gratings. The efficiency of the transformation from the bulk to the surface wave typically has an order of 10%. However, if only 10% of the energy is transformed into the surface wave, its intensity is several orders higher than the intensity of the bulk wave due to the high localization near the interface at a distance of about 1 μm in the optical frequency range.

Plasmonic systems are characterized by the high frequency dispersion. For a smooth metal-dielectric interface, the SPP propagation constant β is as follows:

$$\beta(\omega) = \frac{\omega}{c}\sqrt{\frac{\varepsilon_m(\omega)\varepsilon_d}{\varepsilon_m(\omega)+\varepsilon_d}}, \tag{1}$$

where ω denotes the frequency, c is the light speed in vacuum, and $\varepsilon_{d,m}$ is the dielectric permittivity of the metal or dielectric (index m or d, correspondingly). The experimental data on frequency dispersion of metal permittivity [25] can be approximated with a Drude-Lorentz model,

$$\varepsilon_m = \varepsilon_\infty - \frac{\omega_{pl}^2}{\omega^2 - i\gamma\omega}, \tag{2}$$

where ε_∞ is the asymptotic permittivity, ω_{pl} is the plasma frequency, and γ is the frequency of electron collisions. The dispersion of the dielectric is usually much smaller than the dispersion of the metal and can be neglected. For example, for silica glass, which is a material with cubic nonlinearity, one can use the Sellmeier equation to describe its permittivity from 0,21 to 6,7 μm with rather good accuracy [26]. In

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the region from 0,5 to 2 μm, its permittivity changes very slightly in comparison with metallic dispersion. The difference between the group velocities of the SPP waves at 1 and 1.2 μm wavelengths is 3% for smooth silica glass—gold interface, 14% for the 20-nm gold film surrounded by silica glass layers, and only 0.09% for the bulk wave in silica glass. This example illustrates that plasmonic systems have much stronger dispersion than the bulk crystals.

Very strong dissipation should be referred to the negative but intrinsic features of a plasmonic system. Nowadays, several plasmonic systems with gain have been proposed [27–29] to overcome the limits arising due to the strong SPP attenuation. In the absence of the external gain, the imaginary part of the propagation constant obtained from Eq. (1) can be used to estimate the propagation length of the SPP [30],

$$l_{\text{prop}} = \frac{1}{2|\text{Im}\,\beta|}. \tag{3}$$

Equation (3) shows that the propagation length grows with the increase in the wavelength, therefore, using infrared SPPs instead of the ones of the visible spectrum is reasonable. Typical values of the propagation lengths are assumed to be about 100 μm for a wavelength of 1 μm. Therefore, input and output gratings should be placed at this distance in order to allow the detection of the signal after the interaction.

II. THEORETICAL APPROACH TO SPP PULSE INTERACTION

Description of the SPP pulse propagation with a slowly varying amplitude

For a correct description of two SPP pulse collisions, we need to perform an accurate analysis of SPP pulse propagation along the interface between a metal (e.g., gold) and a dielectric with cubic nonlinearity taking into account dispersion, nonlinearity, and energy dissipation. For a weak signal SPP pulse, the nonlinearity can be neglected, whereas, the impact of the pump SPP on the dielectric permittivity should be taken into account (and vice versa for the pump SPP). The approach used for the description of the propagation and nonlinear interaction of the two plasmonic modes of the metal-dielectric structure is similar to the method presented for the waveguide modes of the optical fibers [31].

We start from the wave equation for the electric field $\vec{E}$ derived directly from Maxwell equations,

$$\Delta\vec{E} - \frac{1}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\vec{P}}{\partial t^2}, \tag{4}$$

where $\vec{P} = \vec{P}_L + \vec{P}_N$ is the polarization, $\vec{P}_L$ is its linear part, whereas, $\vec{P}_N = \vec{P}_{nl} + \vec{P}_{\text{ext}}$ corresponds to the component arising due to the nonlinearity and external impact of the pump SPP. The dispersion of the metal (located at $z < 0$) is treated in a usual way (see Ref. [31]), whereas, the dispersion of the dielectric is neglected for simplicity.

For the simplification of the solution of Eq. (4) for the two media matched by the boundary conditions, several assumptions are made.

First of all, we consider the addition to the linear polarization term $\vec{P}_N$ to be small in comparison with the linear one $\vec{P}_{\text{ext}}, \vec{P}_{nl} \ll \vec{P}_L$. Second, we consider the nonlinear response of the Kerr dielectric to be nearly instantaneous and the permittivity change caused by the external impact of the pump SPP to be slow on the scale of one period. Next, we focus our attention on femtosecond pulses of about 30-fs duration so that their spectra with the central frequency ω_0 have the width $\Delta\omega/\omega_0 \sim 0.1 \ll 1$. Finally, we neglect changes in the SPP transversal structure and its polarization due to the nonlinear impact during the propagation.

The electric field of the SPP, therefore, can be found in the following form:

$$\vec{E}(\vec{r},t) = \tfrac{1}{2}[\vec{F}(z)A(x,t)\exp(i\beta_0 x - i\omega_0 t) + \text{c.c.}], \tag{5}$$

where $\vec{F}(z)$ describes the polarization and structure of the SPP at the central frequency ω_0, $A(x,t)$ is the slowly varying function of x, and β_0 is the propagation constant at ω_0 frequency corresponding to the linear case. The profile and polarization of the SPP for the smooth metal-dielectric interface without any perturbation or nonlinearity is well known

$$\vec{F}(z) = F_0\exp(-\gamma_j|z|)\{1,0,i\,\text{sgn}(z)\beta_0/\gamma_j\}. \tag{6}$$

Using the perturbation theory, we obtain the following change in the propagation constant in the first order ($\beta = \beta_0 + \Delta\beta$):

$$\Delta\beta = \frac{k_0^2}{2\beta_0}\frac{\int_0^{+\infty}\Delta\varepsilon_d(z)|F(z)|^2dz}{\int_{-\infty}^{+\infty}|F(z)|^2dz}. \tag{7}$$

Slight changes in the SPP profiles $\vec{F}$ caused by the induced inhomogeneity and small radiation losses are neglected in our consideration for simplicity.

Making the expansion of the propagation constant,

$$\beta = \beta_0 + u_0^{-1}(\omega-\omega_0) + D(\omega-\omega_0)^2 + \Delta\beta, \tag{8}$$

we derive the equations describing SPP dynamics where $u_0 = (\frac{\partial\beta}{\partial\omega}|_{\omega_0})^{-1}$ is the group velocity and $D = \frac{1}{2}\frac{\partial^2\beta}{\partial\omega^2}|_{\omega_0}$ is the group-velocity dispersion coefficient.

The equation for the slow amplitude of the pump SPP pulse in the time domain A_p has the form

$$\frac{\partial A_p}{\partial x} + iD_p\frac{\partial^2 A_p}{\partial\tau^2} - i\,\Delta\beta_p A_p + \Gamma_p A_p = 0, \tag{9}$$

where time coordinate $\tau = t - x/u_p$ is associated with the group velocity of the pump pulse u_p, the propagation constant change $\Delta\beta_p$ is calculated using Eq. (7) with $\Delta\varepsilon(z) = \chi^{(3)}|\vec{F}_p(z)A_p|^2$, and Γ_p is the imaginary part of the propagation constant.

For the amplitude of the signal SPP pulse of a different frequency, we get

$$\frac{\partial A_s}{\partial x} + \nu\frac{\partial A_s}{\partial\tau} + iD_s\frac{\partial^2 A}{\partial\tau^2} - i\,\Delta\beta_s A_s + \Gamma_s A_s = 0, \tag{10}$$

where $\nu = 1/u_p - 1/u_s$ is the group-velocity mismatch between the two pulses and $\Delta\beta_s$ is calculated according to Eq. (7) with $\Delta\varepsilon(z) = \chi^{(3)}|\vec{F}_p(z)A_p|^2$.

Further estimations, performed for the gold plasmonic system, reveal that the typical values of the nonlinear length associated with the self-action of the SPP $l_{nl} = \Delta\beta^{-1}$ as well as the dispersion length $l_{\text{disp}} = 0.25T_{p,s}^2/D_j$ associated with the pulse broadening processes exceed the propagation length of SPP at least twice for both the signal and the pump SPP pulses. Therefore, for the pump SPP pulse, the phenomenon of self-action due to the cubic nonlinearity and dispersion spreading can be neglected. Thus, we can consider the inhomogeneity induced by the pump SPP pulse to have a constant profile. Losses that are present due to the metallic absorption cause the intensity decay of about 40% in the middle of the interface between gratings.

Another simplification that can be performed is neglecting the dispersion of the coefficient D so that D_s is considered to be equal to D_p, and, moreover, any frequency changes in the signal SPP occurring due to the nonlinear interaction are not supposed to change its value. The direct calculation of the propagation constant compared with the value obtained with Eq. (8) with constant D shows less than 0.1% difference in these two values for 1-1.2-μm wavelengths as far as the propagation constant itself experiences about a 25% variation. This allows us to describe group-velocity dispersion analytically: $u(\omega)^{-1} = u_p^{-1} + 2D(\omega - \omega_p)$.

III. INTERACTION OF THE SIGNAL SPP PULSE WITH THE INDUCED INHOMOGENEITY

A. Reflection of the signal SPP pulse

In order to investigate the propagation dynamics of the signal SPP pulse in the presence of the inhomogeneity induced by the pump SPP pulse, we apply the eikonal method and trajectory approach to the description of the propagation of the signal SPP center [10]. We consider both signal and pump pulses to have Gaussian profiles. The equation for the SPP center trajectory is the following:

$$\frac{\partial \tau_s}{\partial x} = \pm\sqrt{\nu^2 - 4D\,\Delta\beta_s(\tau_s)}. \tag{11}$$

As shown below, the most dramatic influence of the pump SPP-induced inhomogeneity is associated with $D\,\Delta\beta > 0$. The dispersion of the SPP waves is determined mainly by the metallic permittivity so that the sign of the D coefficient is always positive for the smooth metal-dielectric interface. So, if the cubic dielectric possesses the focusing nonlinearity ($\chi^{(3)} > 0$), the trajectory of the signal SPP pulse can experience the reflection from the inhomogeneity if the absolute value of the propagation constant variation is above the threshold,

$$\Delta\beta_t(I_p) = \frac{\nu^2}{4D}. \tag{12}$$

Actually, such a simple geometro-optical theory predicts that there are three regimes of the propagation of the signal SPP (see Fig. 1). The corresponding dynamics of the pump and the signal SPPs is illustrated in Fig. 2 for the different inhomogeneity values.

Figure 2(a) depicts the propagation of the pump SPP in a spatiotemporal domain. One can see that the duration of the pump pulse remains almost constant during the propagation since the distance is much smaller than the characteristic values of its dispersion length associated with the visible pulse broadening. The black dashed lines depict the temporal boundaries of the pump SPP. The position of the time coordinate $\tau = 0$ is selected to coincide with the center of the pump SPP. At the same time, it is obvious that the pump SPP experiences the significant attenuation. Due to the pump SPP damping, the threshold intensity is higher than predicted by Eq. (12). If the interaction of the pump and the signal SPPs occurs at the distance of $x = 0.5l_{\text{damp}}$, then the threshold value can approximately be calculated as $\Delta\beta_t(I_p) \approx \exp(0.5)\nu^2/4D$.

If the intensity of the pump SPP is below the threshold value (12), the induced inhomogeneity is very weak and practically does not influence the signal SPP dynamics. Figure 2(b) illustrates the propagation of the signal SPP in the presence of a very weak inhomogeneity induced by the pump SPP. Due to the dispersion of the plasmonic structure, the signal SPP of the shorter wavelength that initially was launched before the pump (with the center located at $\tau = -30$ fs) propagates slower so that, finally, it comes after the pump (case 1 in Fig. 1).

If the intensity of the pump SPP is equal to the threshold value (as the propagation distance is limited, the discussed dynamics can be observed for a range of inhomogeneity magnitudes that are close to the threshold value), the signal SPP is attracted to the pump center [see Fig. 2(c) and case 2 in Fig. 1]. The inhomogeneity acts like a trap for the signal SPP pulse and accelerates it so that the resulting delay between these two pulses is nearly zero. In Fig. 2(c), it is shown that, at the threshold pump intensity, practically the whole outcome signal pulse is localized inside the temporal domain of the pump pulse.

In the case of pump intensity higher than the threshold value, the signal SPP is reflected from the inhomogeneity (case 3 in Fig. 1). The reflection actually means that the signal SPP pulse continues propagation in the initial direction, but it changes its group velocity so that the sign of the group-velocity mismatch also changes. Note that the value and the sign of the group-velocity mismatch determine the tilt angle of the SPP trajectory on the (τ,z) plane. Therefore, the induced inhomogeneity causes the acceleration of the slower SPP of the initially shorter wavelength and the spectral redshift of its central frequency too so that it keeps propagating before the pump. In the case of such a reflection, the center of the signal SPP remains in the region of $\tau < 0$ [see Fig. 2(d)], and the order of the outcome pulses is unchanged in contrast to the low-pump case 1.

A femtosecond laser pulse can be treated as a sum of the monochromatic waves, the dynamics of which is determined by Eq. (11). Therefore, the reflection coefficient (the amount of energy that is carried by the reflected components) can be

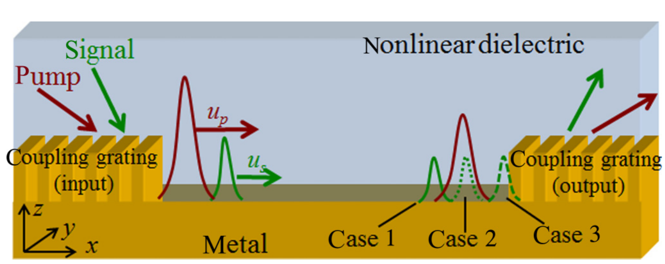

FIG. 1. (Color online) The schematic of the SPP pulse collision.

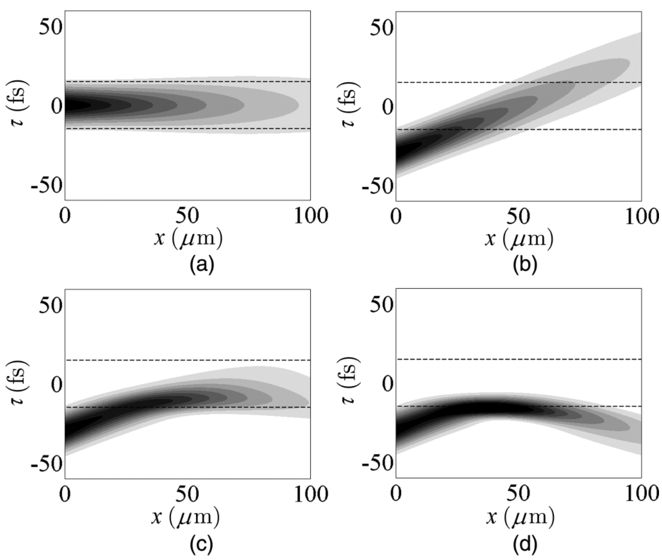

FIG. 2. Dynamics of (a) the pump and (b)–(d) the signal SPP pulses. (b) corresponds to the case of $\Delta\beta_s < \Delta\beta_t$, (c) corresponds to the case of $\Delta\beta_s = \Delta\beta_t$, and (d) corresponds to the case of $\Delta\beta_s > \Delta\beta_t$. The black dashed lines depict the temporal boundaries of the pump SPP.

calculated for the Gaussian pulses in the form

$$R = \frac{1}{2} + \frac{1}{2}\Phi\left[\frac{T_s}{2\sqrt{2D}}(\sqrt{\Delta\beta_{\max}} - \sqrt{\Delta\beta_t})\right], \tag{13}$$

where T_s is the duration of the signal SPP and $\Delta\beta_{\max}$ is the maximal value of the propagation constant change. Figure 3 illustrates the dependence of the reflection coefficient on the value of the induced inhomogeneity. If the intensity of the pump corresponds to near-threshold values, around half of its spectral components should be transmitted, whereas, the other part should be reflected (analogous to the beam splitting in the case of spatial total internal reflection, see Ref. [21]). Meanwhile, in Fig. 2, the splitting of these components it not seen because the propagation distance is limited to a rather short value. The relative amount of the signal SPP energy that comes out together with the pump is shown in Fig. 3.

The interaction between the two pulses can change not only their order (see Fig. 2, in the absence of the inhomogeneity,

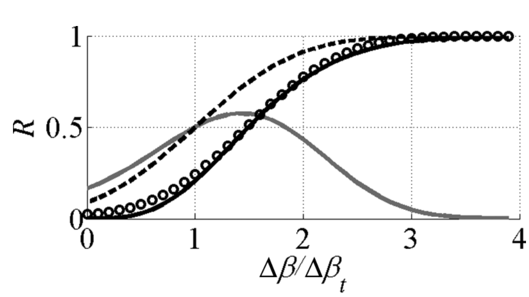

FIG. 3. The dependence of the signal SPP reflection coefficient on the value of the induced inhomogeneity: results of the numerical simulations (circles), analytical calculations in the absence (black dashed line) and in the presence (black solid line) of the attenuation. The gray line indicates the amount of energy inside the pump pulse.

the signal SPP pulse that initially propagated before the pump finally got behind, whereas, the presence of the pump-induced inhomogeneity accelerated it), but also their relative delay.

B. Frequency shift of the signal SPP

The reflection of the signal SPP from the pump-induced inhomogeneity leads to the group-velocity change that occurs due to the frequency shift of the signal SPP. This shift is analogous to the Doppler shift in the theory of relativity that is experienced by the light reflected from a moving mirror. In our case, the role of such a mirror is played by the induced inhomogeneity moving with the pump group velocity. Using Eq. (11) and dispersion model (8), we can obtain the following equation for the shift of the signal SPP central frequency acquired during the propagation:

$$\Delta\omega_s(\tau_s) = (\omega_p - \omega_s)\left(1 \mp \sqrt{1 + \frac{\Delta\beta(\tau_s)}{\Delta\beta_t}}\right). \tag{14}$$

Notice that, in dispersion model (8), $\omega_p - \omega_s = \nu/D$. Therefore, the resulting spectral shift depends on the initial frequency difference between the signal and the pump SPP and on the value of the inhomogeneity induced by the pump. The maximal shift is obtained in the case when the interaction between pulses is completed after passing the propagation distance and they move separately. In this case, the signal SPP frequency experiences a shift of $2(\omega_p - \omega_s)$, so the difference between the signal and the pump SPP frequencies changes its sign but keeps its value.

IV. EXPERIMENTAL ESTIMATIONS FOR SPP INTERACTION IN CUBIC MEDIA

For making some experimental estimations, we need to take practical limitations for the plasmonic systems into account. First of all, the interaction should occur at the scale of the damping length, which is about 100 μm for infrared surface plasmons. It requires a pulse duration of about 30 fs and a minimal group-velocity mismatch value of $\nu \sim 0.6$ fs/μm. On the other hand, the maximal value of the inhomogeneity induced by the pump SPP in the dielectric, typically, is about $\Delta n \sim 5 \times 10^{-3}$.

The dispersion of surface plasmon polariton determines the threshold value of $\Delta\beta_t$. The dispersion of the smooth dielectric—gold interface is not high enough to be used for such an interaction as far as $D = 0.3$ fs$^2/\mu$m requires the threshold value of $\Delta\beta_t/\beta \sim 3\%$. However, if we choose the structured plasmonic system that can be a gold film sandwiched between two dielectrics or perforated plasmonic crystal, the dispersion coefficient is higher. For example, the gold layer of 20 nm, sandwiched between two dielectric layers, has the dispersion coefficient $D = 2$ fs/μm^2 so $\Delta\beta_t/\beta \sim 0.4\%$. In this system, the central wavelength of the signal SPP pulse should be shifted from the pump at a distance of about 0.1 μm to achieve the group-velocity mismatch value of $\nu \sim 0.6$ fs/μm.

V. CONCLUSION

We analyzed the collision of two SPP femtosecond pulses propagating along the nonlinear plasmonic interface. Using both spectral and trajectory approaches, we revealed the conditions under which the weak signal SPP pulse is reflected from the moving inhomogeneity of the permittivity induced in the dielectric with cubic nonlinearity by the pump SPP pulse. We show that the reflection results in the change in the delay between the two pulses and the spectral shift acquired by the signal SPP pulse. Our estimations shows that such effects are possible in structured plasmonic systems, such as sandwich structures and plasmonic crystals. Therefore, ultrafast all-optical control of pulse dynamics can be performed in the nonlinear plasmonic systems at extremely small scales.

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