

Peculiar features of surface plasmon-polariton modulation instability in a metal film with varying thickness

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ABSTRACT

We report on the modulation instability of surface plasmon-polariton wave developed in a thin metal film with varying thickness. It is shown both analytically and numerically that the modulation instability effect can give rise to spatial redistribution and longitudinal localization of surface plasmon-polariton wave energy in subwavelength scale. The frequency, around which modulation instability of surface plasmon polariton wave can be observed, is determined. In this frequency range the propagation of a modulated plasmon wave in a film with increasing thickness is similar to the propagation of modulated light in a dispersion decreasing fiber, enabling generation of high repetition rate ultrashort pulse train.

Keywords: surface plasmon polaritons, nonlinear optics, modulation instability, subpicosecond pulses.

1. INTRODUCTION

Surface plasmon polaritons (SPPs), propagating bound oscillations of electrons and light at a metal surface, exhibit the capacity for subwavelength field confinement and enhancement that make them a promising photonic platform at the nanometer scale [1-3]. SPPs are attractive due to their potential for manufacturing optical chips comparable in size to electronic chip components. Novel light technologies based on plasmon structures can operate within submicron and nanometer ranges similar to electron technologies, thereby, contributing to the development of ultrafast optoelectronic and energy-efficient integrated information processing devices. The concentration of electromagnetic fields in the smallest possible volume enhances nonlinear optical effects such as harmonic generation and frequency mixing [4,5], self-phase modulation and plasmon-soliton formation [6-10], optically induced damping, and all-optical modulation [11,12].

Here, we study the modulation instability (MI) of SPPs propagating in a thin-film structure. MI is the fundamental effect caused by interplay between nonlinearity and dispersion and observed in many nonlinear media and nanostructures maintaining propagation of the localized waves [13-16]. The effects of MI in systems described by nonlinear Schrödinger equation (NLS) are well known, the best example is the propagation of a modulated wave in an optical fiber [17-19]. At the same time, similar processes are observed in optical resonators [20-22], in the space-time dynamics of a laser beam [23], during the formation of stable structures in waveguides, etc [24-26]. MI and its applications are intensively studied in different branches of laser physics, especially in fiber optics, due to their ability to control laser radiation. In particular, MI can be employed in generators of ultrashort pulse trains [27-29] that are of great demand. In this study, we demonstrate that MI of SPP wave propagating in a structure with a conductive film of subwavelength thickness can lead to a periodic localization of surface electromagnetic field resulting in generation of subpicosecond pulses. Unlike our previous works [8, 30] in this article the main attention will be paid to the propagation of SPP wave in a thin metal film with varying thickness. It will be shown that this approach is greatly promising for high repetition rate (~ 1 THz) ultrashort pulse trains generation necessary for wide range of applications.

2. NONLINEAR EQUATION FOR SPP WAVE PROPAGATION

Let us consider a quasi-monochromatic surface plasmon-polariton (SPP) wave propagating with the carrier frequency ω_0 and propagation constant β_0 in a layer structure. Taking into account the dependence of medium dielectric permittivity

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(DP) on the radiation intensity I , the function $\beta = \beta(\omega, I)$ can be expanded in the Taylor series in the vicinity of point $\omega = \omega_0, I = 0$:

$$\beta \approx \beta_0 + (\omega - \omega_0) \left. \frac{\partial \beta}{\partial \omega} \right|_{\omega_0, I=0} + \frac{1}{2} (\omega - \omega_0)^2 \left. \frac{\partial^2 \beta}{\partial \omega^2} \right|_{\omega_0, I=0} + \frac{1}{6} (\omega - \omega_0)^3 \left. \frac{\partial^3 \beta}{\partial \omega^3} \right|_{\omega_0, I=0} + I \left. \frac{\partial \beta}{\partial I} \right|_{\omega_0, I=0}. \quad (1)$$

Using

$$\Omega = \omega - \omega_0, \quad K = \beta - \beta_0,$$

expression (1) reduces to a nonlinear dispersion relation between the frequency and propagation constant:

$$K = v_g^{-1} \Omega + \frac{\beta_2}{2} \Omega^2 + \frac{\beta_3}{6} \Omega^3 + \gamma I, \quad (2)$$

where

$$v_g^{-1} = \left. \frac{\partial \beta}{\partial \omega} \right|_{\omega_0, I=0} \quad (3)$$

is the parameter determined by the group velocity,

$$\beta_2 = \left. \frac{\partial^2 \beta}{\partial \omega^2} \right|_{\omega_0, I=0}, \quad \beta_3 = \left. \frac{\partial^3 \beta}{\partial \omega^3} \right|_{\omega_0, I=0} \quad (4 \text{ a,b})$$

are the group velocity dispersion (GVD) and third-order dispersion (TOD) and

$$\gamma = \left. \frac{\partial \beta}{\partial I} \right|_{\omega_0, I=0} = v_g^{-1} \left. \frac{\partial \omega}{\partial I} \right|_{\omega_0, I=0} \quad (5)$$

is the parameter of Kerr nonlinearity at $\omega = \omega_0$.

In the approximation of slowly varying complex amplitudes, the quasi-monochromatic SPP wave field can be expressed as $A(x, z, t) \exp[i(\beta_0 x - \omega_0 t)]$, where the parameters β_0 and ω_0 are real values. Oscillation periods of the wave packet envelope $A(x, z, t)$ in time and space (coordinate x) are determined by Ω and K with $|\Omega| \ll \omega_0, |K| \ll \beta_0$.

With

$$\frac{\partial A}{\partial t} = -i\Omega A, \quad \frac{\partial A}{\partial x} = iKA,$$

the complex amplitude $A(x, z, t)$ satisfies the nonlinear Schrödinger equation

$$\frac{\partial A}{\partial x} + v_g^{-1} \frac{\partial A}{\partial t} - \frac{i\beta_2}{2} \frac{\partial^2 A}{\partial t^2} + \frac{\beta_3}{6} \frac{\partial^3 A}{\partial t^3} + i\gamma |A|^2 A = 0, \quad (6)$$

where $|A|^2 = I$ is the wave intensity. It is known that Eq. (6) describes the rapid growth of small time-periodic perturbations of the steady-state solution. In the nonlinear wave theory, this phenomenon is referred to as modulation instability (MI) [13-16]. To generate MI, the Kerr nonlinearity and GVD have to be of opposite signs ($\gamma\beta_2 < 0$).

3. MODULATION INSTABILITY

The dispersion and nonlinear parameters (3) - (5) for SPP waves in a conductive film can be calculated using the dispersion relation [31, 32]

$$\exp(-2q_3h) = \frac{q_3\varepsilon_1 + q_1\varepsilon_3}{q_3\varepsilon_1 - q_1\varepsilon_3} \frac{q_3\varepsilon_2 + q_2\varepsilon_3}{q_3\varepsilon_2 - q_2\varepsilon_3}, \quad (7)$$

where $q_j = \sqrt{\beta^2 - k_0^2\varepsilon_j} \in \Re$ is the attenuation constants (transverse components of imaginary wave vectors in a conductive layer and dielectrics), $k_0 = \omega/c$, c is the speed of light in vacuum (Fig. 1).

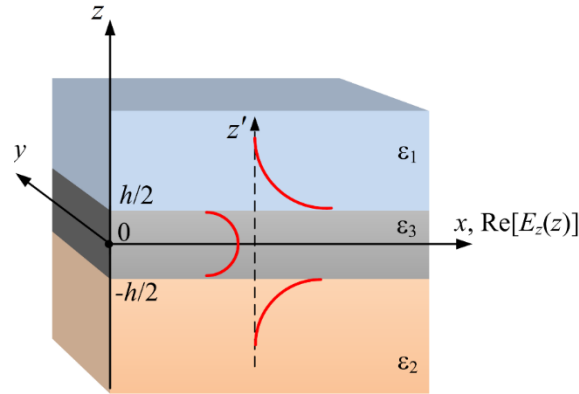


Figure 1. Schematic of the structure.

As a conductive film, we assume a metal (e.g., silver) film with the dielectric constant (DC) obtained as:

$$\varepsilon_3 = \varepsilon_L + \varepsilon_{NL}(I), \quad (8)$$

where

$$\varepsilon_L = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2} \quad (9)$$

is the linear part of the dielectric permittivity (DP) recorded using the Drude model, ε_∞ is DP high-frequency component, ω_p is the plasma frequency of electron gas in metal,

$$\varepsilon_{NL} = \alpha I \quad (10)$$

is the DP nonlinear part that is dependent on the field intensity I , $\alpha \approx 2n_L n_{NL}$, n_L and n_{NL} are the linear and nonlinear parts of the refractive index $n_3(I) = n_L + n_{NL}I$ [33]. The nonlinear DP coefficients can be obtained using the table values of the third order nonlinear susceptibility $\chi^{(3)}$. For silver, the parameters included in expression (9) (with corrections made for interband transitions using the experimental data [34]) in the optical range are $\varepsilon_\infty = 4.1$, $\omega_p = 13.3 \cdot 10^{15} \text{ s}^{-1}$. In the optical spectrum range, the silver films of several nanometers thick have the third-order nonlinear susceptibility $\chi^{(3)} \approx 2.5 \cdot 10^{-8} \text{ esu}$ [35]. The linear refractive indices for radiation frequencies varying in a wide range are summarized in [34].

The dispersion relations (in the linear approximation $\alpha = 0$) for silver films of various thickness on a dielectric substrate in vacuum ($\varepsilon_1 = 1, \varepsilon_2 = 4.8$) are plotted in Fig. 2(a). A curvature of the dispersion curves demonstrates the signs of dispersion parameters (3) and (4,a) for considered structure. The parameter v_g^{-1} inverse to the group velocity is positive on the lower dispersion branch, but it is negative within a wide range on the upper branch. The GVD parameter is maintained positive in both cases, since its positive value increases with ω on the lower branch, while on the upper branch an increase of negative v_g^{-1} in the absolute value corresponds to the frequency decrease in a wide range of β .

For noble metals $\alpha > 0$, thus the Kerr nonlinearity γ (5) is positive on the low-frequency SPP branch and is negative on the high-frequency dispersive SPP branch in a wide range of β . The analysis shows that there is a frequency range, where the signs of the GVD and Kerr nonlinearity parameter are opposite (e.g., around $\omega_0 = 5.8891 \cdot 10^{15} \text{ s}^{-1}$), making this domain available for modulation instability. Hereinafter, we will focus on the domain around this frequency.

The GVD and TOD obtained by numerical differentiation of dispersion relations (7) for $\omega = \omega_0$ as a function of the film thickness h are given in Fig. 2(b). The inset shows similar dependence for the inverted group velocity v_g^{-1} . At the selected frequency, the propagation constant β varies from $6.5 \cdot 10^7 \text{ m}^{-1}$ (for the thickest film $h = 25 \text{ nm}$) to $4.4 \cdot 10^8 \text{ m}^{-1}$ (for the thinnest film $h = 9 \text{ nm}$). One can see that the SPP wave dispersion parameters exhibit a strong dependence on the film thickness h (increase by several orders of magnitude per 10 nm of thickness), while the high GVD and TOD values, typical for thin films, quickly decrease with increasing thickness. Also, with an increase of h , v_g^{-1} and γ proportional to it rapidly decrease. At $\omega = \omega_0$, the MI condition $\gamma\beta_2 < 0$ is fulfilled, i.e. for a given frequency domain, any small time-periodic modulation of the steady-state SPP wave increases with a certain increment dependent on both the wave intensity I and modulation frequency Ω [14].

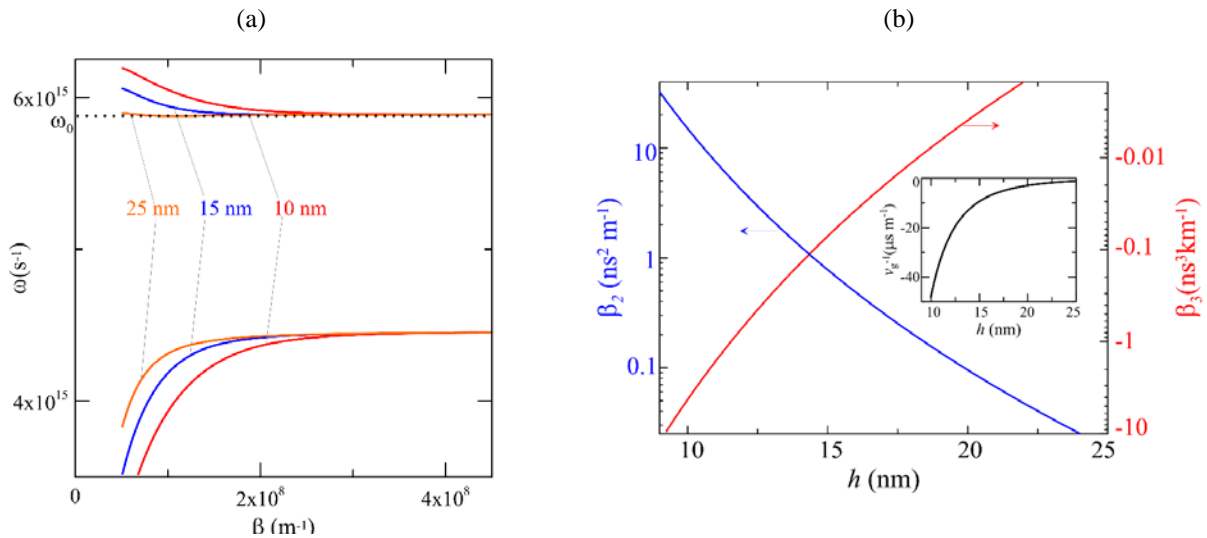


Figure 2. (a) Dispersion relations for SPP for silver film with different thickness ($h = 10, 15, 25 \text{ nm}$) on a dielectric in vacuum ($\epsilon_1 = 1, \epsilon_2 = 4.8$). (b) GVD (blue curve) and TOD (red curve) as a function of thickness of silver film on dielectric in vacuum at $\omega_0 = 5.8891 \cdot 10^{15} \text{ s}^{-1}$.

The modulation gain for small periodic perturbations is determined by the GVD β_2 and Kerr nonlinearity γ

$$g_m(\Omega) = |\beta_2 \Omega| \sqrt{\Omega_c^2 - \Omega^2}, \quad (11)$$

where $\Omega_c = 2 \sqrt{\frac{|\gamma|}{|\beta_2|} I}$ is the maximal frequency of MI. A strong dependence of the dispersion parameters on the film thickness makes the modulation gain change drastically with h . Fig. 3 shows the modulation gain (11) for different thickness of a silver film at the initial intensity of modulated wave $I = 10^{-2} \text{ W } \mu\text{m}^{-2}$. One can see that the modulation gain is large enough to ensure decay of modulated wave into individual pulses while it propagates through the length of hundreds nm.

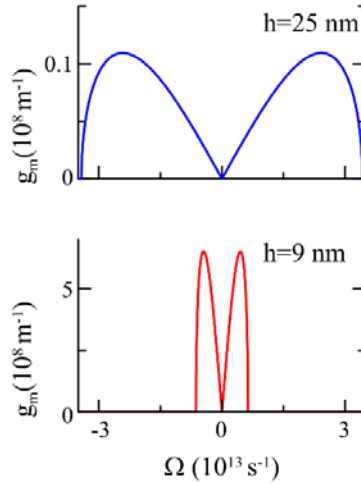


Figure 3. Modulation gain (11) calculated for different film thickness h . The initial intensity of modulated wave is $I_0 = 0.01 \text{ W } \mu\text{m}^{-2}$.

4. MODULATION INSTABILITY OF SPP WAVES IN A FILM OF INCREASING THICKNESS

The performed analysis allows us to study SPP MI in a wedge-shaped metal film. This problem is similar to MI of light propagating in a waveguide with the parameters varying along the waveguide length [28, 36, 37]. Due to MI, the initially modulated wave decays into a train of pulses, which in turn, again gather into a continuous modulated wave, and so on. [15, 38]. When MI is generated in the waveguide with the GVD decreasing along its length [28, 39], the MI frequency bandwidth $(-\Omega_c, \Omega_c)$ and the modulation gain peak frequency $\Omega_{\max} = \Omega_c / \sqrt{2}$ increase along the waveguide. Due to the GVD decrease along the waveguide, the periodicity of the pulse train generation and decay is violated.

The discussed modulation regime can be obtained in a plasmon waveguide with increasing thickness and, consequently, decreasing GVD. The structure can be designed to provide a constant plasmon mode area during SPP wave propagation. In other words, a decrease of the SPP wave intensity is compensated by a decrease of the film thickness. We also should add that the problem of SPP loss compensation is extremely important and requires the development of special methods for its solution [40, 41] Noteworthy, the presented mechanism can also be used to compensate for the SPP wave losses. Further, we will demonstrate that the structures with a length less than 1–2 hundred nm can be used to observe MI. In such a scale, the SPP attenuation is negligible and the intensity decrease can be entirely compensated by a film width reduction.

Using these estimations, we can apply the generalized nonlinear Schrödinger equation to simulate propagation of a modulated SPP wave

$$\frac{\partial A}{\partial x} + v_g^{-1}(x) \frac{\partial A}{\partial t} - \frac{i\beta_2(x)}{2} \frac{\partial^2 A}{\partial t^2} + \frac{\beta_3(x)}{6} \frac{\partial^3 A}{\partial t^3} + i\gamma(x) \left(|A|^2 - \tau_R \frac{\partial |A|^2}{\partial t} \right) A = 0. \quad (12)$$

In contrast to Eq. (6), the coefficients in Eq. (12) vary along the waveguide. Besides, the stimulated Raman scattering (SRS) is now taken into account. The SRS provided by the THz ultrashort pulse train transfers energy from high-frequency spectral components to low-frequency ones. The taken value of parameter $\tau_R \approx 10^{-15} \text{ s}$, describing the intensity of this process, is typical for materials with a "fast" electronic nonlinearity [42]. A modulated wave $A(t, 0) = \sqrt{I_0} (0.99 + 0.01 \cdot \cos(2\pi\nu_m t))$ with intensity $I_0 = 10^{-2} \text{ W } \mu\text{m}^{-2}$ is used as the initial conditions.

Fig. 4 shows the evolution of intensity $I = |A|^2$ for SPP wave modulated by the frequency $\nu_m = 0.44 \text{ THz}$, as it is described by (12) with periodic boundary conditions. One can see that tenfold increase of the SPP wave peak intensity is recorded after the propagation over $\sim 100 \text{ nm}$. The process has been modeled at various frequencies of the initial modulation. It is found that the periodic modulation induced by the frequencies $\Omega/2\pi = \nu_m = 0.3 - 1.5 \text{ THz}$ having high

modulation gain at the initial phase exhibit effective compression, thus converting to the pulse train. The modulation gain for frequencies beyond this range (0.3 – 1.5 THz) is relatively low.

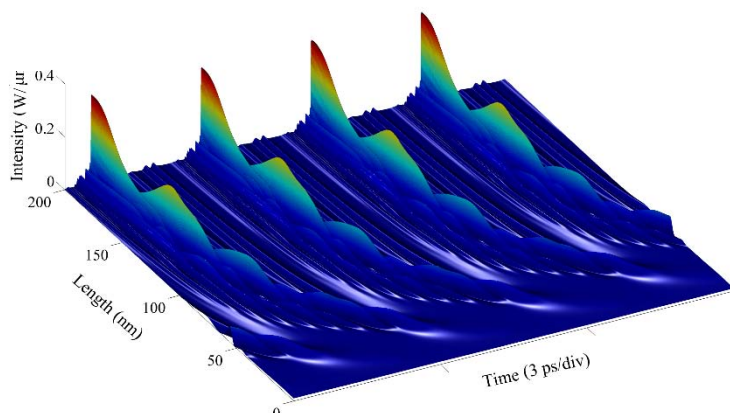


Fig. 4. Evolution of a plasmon wave with modulation frequency $\nu_m = 0.44$ THz and initial intensity $I_0 = 0.01$ W μm^{-2} in a silver film with a linearly increasing thickness.

5. CONCLUSION

SPP wave propagating in a thin metal film on a dielectric has been considered. Using standard approximations, we have derived the NLSE propagation equation taking into account the Kerr phase shift of propagating SPP wave. Based on the dispersion relation, the SPP wave dispersion and nonlinear characteristics are obtained as well as their dependences on metal film thickness.

For the frequency range defined in the manuscript, propagation of a modulated SPP wave in a film with increasing thickness is considered. We have shown its similarity to the propagation of modulated light in a dispersion decreasing nonlinear waveguide, enabling generation of high repetition rate ultrashort pulse train. Propagation of a modulated SPP wave in a film with linearly increasing thickness has been numerically simulated.

The proposed concept is greatly promising for development of compact generators of high repetition rate (~ 1 THz) ultrashort (~ 100 fs) pulse trains and applicable for multiple tasks of modern optoelectronics, including development of master clocks, frequency comb generators, and etc.

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