Letter

Self-induced decay of intense laser pulse into a pair of surface plasmons

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(Received 1 April 2022; revised 1 July 2022; accepted 9 August 2022; published 22 August 2022)

We show theoretically that an intense femtosecond optical pulse incident normally on a metal surface tends to decay into a pair of counterpropagating surface plasmon polaritons (SPPs). The interference field heats the medium periodically, which causes a periodic permittivity perturbation and, in turn, resonantly amplifies the magnitudes of the SPPs. The instability growth time is 10–50 fs for typical metals at laser fluences of about $\sim 1\,\mathrm{J/cm^2}$, so this mechanism is important for understanding the dynamics of the optical pulse absorption under damaging conditions, and it is promising for the interpretation of laser-induced periodic surface structure formation in single- and few-pulse pumping regimes.

DOI: 10.1103/PhysRevB.106.L081408

I. INTRODUCTION

The phenomenon of laser-induced periodic surface structure (LIPSS) formation has been studied for more than 50 years since the first observation by Birnbaum in Ge and GaAs samples in 1965 [1]. Further, it was found that the irradiation of metals and dielectrics by normally incident short laser pulses of near-threshold fluence also leads to the appearance of periodic ripples on the surface (e.g., see recent review papers [2,3]). In addition to the fundamental interest, this effect attracts attention as a relatively simple and fast method of submicron material structuring. An orientation and a period of ripples are determined both by the material properties and the laser irradiation parameters; the detailed analysis of LIPSS characteristics can be found in Refs. [2–4]. Below we focus only on the case of metals and highly doped semiconductors, where the condition of surface plasmon-polariton existence is fulfilled; i.e., the real part of permittivity ε' is less than -1.

The key role of the SPP in the formation of LIPSS in metals has been proven in many experimental and theoretical papers [2–7]. The period of ripples is usually equal to the SPP period or twice smaller. It was also shown that the prestructuring of the surface for better coupling between the incident wave and SPP increases further growth of ripples [8,9].

General electromagnetic theory of LIPSS formation assisted by SPP excitation was developed by Sipe and coauthors in the early 1980s in a series of pioneering papers [5–7]. In the framework of this model the SPP (along with other diffraction waves) are generated on random surface irregularities and simultaneously interfere with the incident laser pulse. The resulting interference pattern has a period equal to the SPP wavelength which explains the period of LIPSS observed in metals experimentally. The mentioned series of papers by Sipe became the base for the developing of various more complicated and detailed models of LIPSS formation [10–13].

Further experiments showed that during the multipulse machining and the growth of LIPSS depth their period gradually decreases, which is probably a consequence of the efficient permittivity change in the damaged layer [14]. The key role of SPPs in surface structuring was also demonstrated for semiconductors [15,16]: In a strong laser field the density of excited charge carriers becomes enough for the medium metallization, so the existence of the SPP mode becomes possible. In the paper by Sakabe [17] the idea of parametric decay of the incident optical wave into SPP and idler photon was proposed, which seems to be an important step toward the explanation of LIPSS formation at high optical intensities; however, no specific nonlinear mechanism has been considered in Ref. [17], so that the probability of the decay process could not be estimated.

Plasma effects play a role in LIPSS not only in the case of metals or excited semiconductors, but also in the case of bulk dielectrics structuring [18–21]. In particular, the simplified numerical model developed in Ref. [19] demonstrated the effect of spontaneous self-organization during the multiphoton ionization of bulk glass leading to the formation of plasma stripes perpendicular to the laser pulse polarization. As for the applications, the papers [22–24] demonstrated that the laser structuring of many metals can be both high quality and fast (with only two laser pulses), so that the manufacturing speed achieves $1\,\mathrm{cm^2}/10\,\mathrm{s}$ even with a widespread laser system of mJ energy level.

Despite the undoubted progress made in this field, the very initial stage of LIPSS formation along with the single-pulse experiments remains not fully understood from the theoretical point of view, which was discussed in detail in the review paper [2]. In a nutshell, we cannot expect enough efficient excitation of SPP on a random surface. Metal and semiconductor samples used in real experiments are rather smooth, and so the spectral measure of their surface roughness $b(\mathbf{k})$ introduced by Sipe [5] is very small in the resonant interval of wave numbers $\mathbf{k}$. For comparison, the influence of the surface prestructuring on LIPSS formation was proved experimentally in Refs. [8,9] for the depths of *resonant* gratings of 10 and

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66 nm, respectively. Another close example is the optical to terahertz conversion in metals: It was shown theoretically and experimentally that the significant influence of SPPs on the terahertz output takes places when the depth of a resonant grating is more than 20–30 nm [25,26]. Obviously, if we consider not a regular, but a random surface shape, it should have a much larger amplitude of irregularities to reach comparable SPP excitation efficiency. However, such rough samples are not used in the experiments on LIPSS formation. Also note that in recent theoretical papers (see [11–13] and many others) the efficiency of SPP generation is usually chosen as a constant free parameter.

In this Letter we are trying to fill the gap in the theory of surface plasmon excitation showing that the surface roughness is not necessarily the main source of SPPs, while it just creates the initial conditions for their nonlinear growth during the laser pulse action (the alternative sources of seed SPPs are thermal fluctuations of the electron density and the electromagnetic field). To describe it theoretically, we should take into account that the interference pattern of SPPs and the incident wave not only influence the further material melting, but also cause periodic heating of electrons, which changes local permittivity strongly.

Below we demonstrate that the discussed feedback mechanism leads to the instability of a flat wave reflection with respect to decay into a pair of counterpropagating SPPs. The instability growth time (estimated for the set of typical parameters of gold) may be as short as 10–50 fs for the incident fluences of about $1\,\mathrm{J/cm^2}$ used in real single-shot experiments. It means that during the femtosecond laser pulse action, the SPP magnitude has enough time to grow several orders higher than would be expected from the theory of linear transformation on random irregularities, which significantly change initial conditions for further heat redistribution, material melting, and ablation. Of course, the proposed decay mechanism should not be treated as an alternative model of LIPSS formation, while it just aims to clarify the very first electrodynamical stage of this process and, in particular, to answer the question of why the SPP magnitude is indeed enough for achieving such strong contrast in material heating.

II. SPPs EXCITATION ON THE PERMITTIVITY PERTURBATION

To describe the stage of ultrafast nonlinear decay analytically, we need to derive equations on the evolution of SPP magnitude explicitly, without introducing any phenomenological transformation coefficients.

Let us start by considering the influence of some permittivity perturbation $\delta\varepsilon$ on the reflection process. Since we assume that the perturbations are relatively small, the problem of the laser pulse diffraction can be linearized, so each Fourier component of the incident pulse and of $\delta\varepsilon$ can be taken into account independently. The geometry of the problem is shown in Fig. 1: A flat conductor with the permittivity ε ($\mathrm{Re}\,\varepsilon < -1$) occupies the region $x < 0$ and the optical wave with the wave number k_0 and frequency ω polarized along the z axis becomes incident normally at its surface. In the absence of any medium

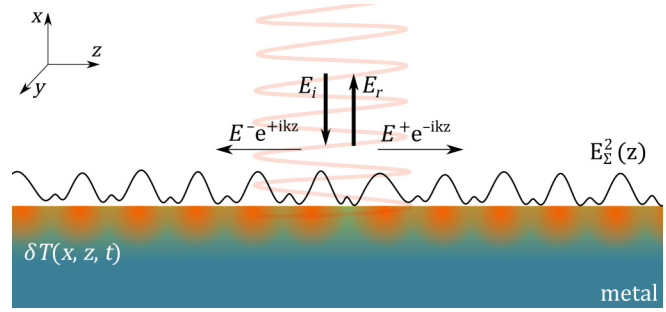

FIG. 1. Incident wave decay into a pair of SPPs. The resulting optical field intensity at the surface $\mathbf{E}_\Sigma^2(z)$ is shown by a black curve [also see Eq. (22)]. Periodic temperature perturbation inside the metal is shown by a color gradient.

perturbations the electric field above and below the surface is described as the sum of three waves:

$$x > 0: \ \mathbf{E}_i(t, x, z) = \mathbf{z}_0 E_i \exp(i\omega t + ik_0 x), \tag{1}$$

$$x > 0: \ \mathbf{E}_r(t, x, z) = \mathbf{z}_0 E_r \exp(i\omega t - ik_0 x), \tag{2}$$

$$x < 0: \ \mathbf{E}_t(t, x, z) = \mathbf{z}_0 E_t \exp(i\omega t + \alpha x), \tag{3}$$

where $\mathbf{z}_0$ is the unit vector; $\alpha = \sqrt{-\varepsilon k_0^2}$; $\mathbf{E}_i$, $\mathbf{E}_r$, and $\mathbf{E}_t$ are the electric fields of the incident, reflected, and transmitted waves, respectively, with the magnitudes determined by the following Fresnel relations:

$$E_t = \frac{2E_i}{1-\sqrt{\varepsilon}}, \quad E_r = E_i \frac{1+\sqrt{\varepsilon}}{1-\sqrt{\varepsilon}}. \tag{4}$$

Now let us move to a permittivity perturbation of the harmonic form with an arbitrary wave number k_ε, so $\varepsilon = \varepsilon_0 + \delta\varepsilon(x, z, t)$ and

$$\delta\varepsilon(x, z, t) = \delta\tilde{\varepsilon}(x, t)\cos k_\varepsilon z, \tag{5}$$

where $\delta\tilde{\varepsilon}(x, t)$ is assumed to be a slowly varying function of time ($|\partial\delta\tilde{\varepsilon}/\partial t| \ll \omega\delta\tilde{\varepsilon}$) and the characteristic spatial scale of $\delta\tilde{\varepsilon}(x)$ is expected to be comparable with the optical skin layer. Note that an arbitrary phase of the perturbation $\sim \cos(k_\varepsilon z + \varphi)$ would not change further analysis. Evolution of $\delta\tilde{\varepsilon}$ due to the heating of electrons will be considered after solving the diffraction problem.

Substituting an inhomogeneous permittivity to the Maxwell equations, we obtain the following equation inside the conductor for the total magnetic field $\mathbf{H}$ and electric field $\mathbf{E}$:

$$\Delta\mathbf{H} - \frac{\varepsilon}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{H} = \frac{1}{c}\frac{\partial}{\partial t}[\nabla\varepsilon, \mathbf{E}], \tag{6}$$

where Δ and ∇ are the Laplace and nabla differential operators, respectively, and the square brackets denote the cross product. Using perturbation theory, we can separate diffraction fields in the explicit form representing the solution of Eq. (6) as a sum of unperturbed fields inside the medium $\mathbf{E}_t$ and $\mathbf{H}_t$ and the first-order fields $\mathbf{E}_1$ and $\mathbf{H}_1$: $\mathbf{E} = \mathbf{E}_t + \mathbf{E}_1$ and

$\mathbf{H} = \mathbf{H}_t + \mathbf{H}_1$. Neglecting second-order terms we obtain

$$\Delta\mathbf{H}_1 - \frac{\varepsilon_0}{c^2}\frac{\partial^2\mathbf{H}_1}{\partial t^2} = \frac{1}{c}\frac{\partial}{\partial t}[\nabla\delta\varepsilon, \mathbf{E}_t] + \frac{\delta\varepsilon}{c^2}\frac{\partial^2\mathbf{H}_t}{\partial t^2}, \quad (7)$$

which is a common wave equation with a source. In the chosen geometry both terms on the right side of Eq. (7) are directed along the y axis, so the magnetic field has only a y component: $\mathbf{H}_1 = H_1\mathbf{y}_0$. Since Eq. (7) is linear with respect to the electromagnetic fields and the permittivity varies relatively slowly, the frequency transformation effects are negligible and the diffraction fields can be represented as the harmonic waves $\sim e^{i\omega t}$, which leads to

$$\Delta H_1 + \varepsilon_0 k_0^2 H_1 = -ik_0 E_t e^{\alpha x}\frac{\partial\delta\varepsilon}{\partial x} - k_0^2\delta\varepsilon H_t e^{\alpha x}. \quad (8)$$

The right side of Eq. (8) contains only two spatial harmonics, $e^{\pm ik_\varepsilon z}$. Taking this into account and writing the magnetic field $H_1(x, z, t)$ as

$$H_1(x, z, t) = H_1^+(x)e^{i\omega t - ik_\varepsilon z} + H_1^-(x)e^{i\omega t + ik_\varepsilon z}, \quad (9)$$

we come to similar equations for both spatial harmonics $H_1^\pm$ (which meets the requirements of symmetry $z \to -z$):

$$\frac{\partial^2 H_1^\pm}{\partial x^2} - k_\varepsilon^2 H_1^\pm + \varepsilon_0 k_0^2 H_1^\pm = -\frac{ik_0 E_t e^{\alpha x}}{2}\frac{\partial\delta\tilde{\varepsilon}(x)}{\partial x} - \frac{k_0^2\delta\tilde{\varepsilon}(x)}{2}H_t e^{\alpha x}. \quad (10)$$

Equation (10) can be solved analytically for some model profiles $\delta\tilde{\varepsilon}(x)$; from further analysis it follows that the specific form of this function influences the effect we focus on insignificantly. Assuming that $\delta\tilde{\varepsilon}$ should decrease with depth we set it as $\delta\tilde{\varepsilon}(x) = \delta\tilde{\varepsilon}\, e^{gx}$ and obtain the following solution for the magnetic field:

$$x < 0: \; H_1^\pm = Ae^{\alpha_2 x} + \Psi(x), \quad (11)$$

where A is an arbitrary constant, $\alpha_2 = \sqrt{k_\varepsilon^2 - \varepsilon_0 k_0^2}$, and

$$\Psi(x) = -\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}e^{(g+\alpha)x}. \quad (12)$$

The magnetic field in free space $x > 0$ also consists of two spatial harmonics, $H_1^\pm(x)e^{i\omega t \mp ik_\varepsilon z}$, which, according to the Helmholtz equation, gives

$$x > 0: \; H_1^\pm(x) = Ce^{-\alpha_1 x}, \quad (13)$$

where $\alpha_1 = \sqrt{k_\varepsilon^2 - k_0^2}$ and C is another arbitrary constant. Note that an imaginary value of α_1 ($k_\varepsilon < k_0$) does not contradict our consideration and corresponds to the case of propagating scattered waves in the upper half space, but the chosen notation is more natural for the analysis of localized plasmon modes.

To find both constants A and C, the boundary conditions should be taken into account. First, the amplitudes of tangential electric fields of both harmonics $E_z^\pm$ can be found from the Maxwell equations:

$$x < 0: \; ik_0\varepsilon E_z^\pm = \frac{\partial H_1^\pm(x)}{\partial x}, \quad (14)$$

$$x > 0: \; ik_0 E_z^\pm = \frac{\partial H_1^\pm(x)}{\partial x}. \quad (15)$$

Using the conditions of the magnetic and electric field continuity at the boundary $x = 0$, we find:

$$A = \frac{\varepsilon_0\alpha_1 + g + \alpha}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}, \quad (16)$$

$$C = \frac{g + \alpha - \alpha_2}{\alpha_2 + \varepsilon_0\alpha_1}\frac{ik_0 g - k_0^2\sqrt{\varepsilon_0}}{(g+\alpha)^2 - k_\varepsilon^2 + \varepsilon_0 k_0^2}\frac{E_i\delta\tilde{\varepsilon}}{(1-\sqrt{\varepsilon_0})}. \quad (17)$$

Both of the obtained expressions (16) and (17) have the same resonant denominator $D = \alpha_2 + \varepsilon_0\alpha_1$, which reaches the minimal value when the wave number of permittivity perturbation k_ε coincides with the real part of the SPP wave number:

$$k_\varepsilon = k_0\sqrt{\frac{\varepsilon_0'}{1+\varepsilon_0'}}, \quad (18)$$

where $\varepsilon_0 = \varepsilon_0' + i\varepsilon_0''$. Note that near the resonant point $H_1^\pm(x=0) \cong A \cong C \gg \Psi(x=0)$.

Equations (16) and (17) give only the stationary amplitude of SPP generated by a continuous monochromatic incident wave and limited by the absorption effects ($\mathrm{Im}[D] \propto i\varepsilon_0''$). To describe the dynamics of SPP excitation by a femtosecond laser pulse, one needs to consider the spectral properties of Eqs. (16) and (17), for which some specific model of the medium permittivity $\varepsilon(\omega)$ should be introduced. Since the aim of this Letter is to demonstrate fundamental feasibility of spontaneous decay, we use the simplest Drude model:

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega - i\nu)} \cong -\frac{\omega_p^2}{\omega^2} - i\frac{\nu\omega_p^2}{\omega^3}, \quad (19)$$

where ω_p is the plasma frequency and ν is the scattering rate of electrons. Here we assume that $\omega_p \gg \omega$ and $\omega \gg \nu$ or, at least, $\omega \gtrsim \nu$. These conditions are satisfied for many metals in the IR and some part of the visible spectrum.

Considering a quasimonochromatic incident laser pulse with the central frequency ω_0 which satisfies the resonant condition (18), we calculate the amplitudes (16) and (17) at some shifted frequency $\omega = \omega_0 + \delta\omega$:

$$A(\omega) = C(\omega) = -\frac{\omega_0}{\delta\omega - i\nu\omega_0^2/2\omega_p^2}\frac{\omega_0^4}{4\omega_p^4}E_i(\omega)\delta\tilde{\varepsilon}. \quad (20)$$

Here we assumed that the spatial profile of the perturbation $\delta\tilde{\varepsilon}(x)$ is proportional to $E_z^\pm(x)E_t(x)$, so that $g = \alpha + \alpha_2 \cong 2\omega_p/c$. Below, we show that this is valid in the case of quite fast interactions, before the diffusion significantly changes the heat distribution inside the metal. In the time domain, Eq. (20) can be rewritten as an equation for the envelopes:

$$\left(\frac{\partial}{\partial t} + \nu\frac{\omega_0^2}{2\omega_p^2}\right)\tilde{E}_z^\pm(t) = -\frac{\omega_0^6}{4\omega_p^5}\tilde{E}_i(t)\delta\tilde{\varepsilon}, \quad (21)$$

where $\tilde{E}_z^\pm(t)$ and $\tilde{E}_i^\pm(t)$ are slowly varying time envelopes of SPP and of the incident pulse, respectively. The growth rate of the SPP is proportional to the incident field magnitude and to the current permittivity perturbation; the absorption of SPP takes time $\tau_a = \nu^{-1}\frac{2\omega_p^2}{\omega_0^2}$ which coincides with the well-known expressions for the SPP propagation length [27]. Thus, we obtained the equation of resonant excitation of counterpropagating SPP on given permittivity perturbation $\delta\varepsilon(x, z)$. Below

we demonstrate the existence of positive feedback, so that the presence of the SPP should increase the magnitude of $\delta\varepsilon$.

III. GROWTH OF THE PERMITTIVITY PERTURBATION

The total electric field inside the metal is the sum of the electric fields of the transmitted wave and SPP. The electric field of the SPP under the surface is almost tangential, so the total field $\mathbf{E}_\Sigma$ can be expressed as

$$x<0:\ \mathbf{E}_\Sigma = \mathrm{Re}\,(E_t e^{\alpha x} + 2E_z^{\pm} e^{\alpha_2 x}\cos k_\varepsilon z) e^{i\omega t}\mathbf{z}_0. \quad (22)$$

Let us consider the perturbation of permittivity caused by the periodically modulated electric field (22). Suppose that the medium is a metal with high free electron density n_e which is expected to change relatively weakly during the laser pulse action. At damaging fluences the electronic temperature reaches several eV which leads to a several times increase of the collision frequency. For example, in Ref. [28], where the ultrafast heating of gold was studied, it was found that at incident laser fluences of about 0.5–1 J/cm^2, the density of free electrons increases only 30%–50% while the collision frequency increases more than an order of magnitude.

In the general case, comparable contributions to the increase of total electron collision frequency ν are made by electron-electron and electron-phonon scattering processes, which are mainly determined by the electronic and crystal lattice temperatures, respectively. For the sake of simplicity, here we use a linear approximation for the dependence of ν on the absorbed laser pulse energy per one electron, W_e:

$$\nu(W_e) = \nu_0 + \xi\frac{W_e}{\hbar}, \quad (23)$$

where $\hbar$ is Planck's constant, ν_0 is the initial collision frequency (for example, at room temperature), and ξ is a dimensionless constant, which is expected to be of the order of 1 in the simplest theoretical model of a Fermi liquid [29]. From recent experimental data on ultrafast heating of gold we can find that $\xi \approx 0.5$ [28].

When the incident laser pulse is acting on metal, the equation of electron heating has the following form:

$$\frac{\partial W_e}{\partial t} = \nu\frac{e^2|\mathbf{E}_\Sigma|^2}{2m\omega_0} \cong \nu\frac{e^2}{2m\omega_0}\left|E_t^2 e^{2\alpha x} + 4E_t E_z^{\pm} e^{(\alpha+\alpha_2)x}\cos k_\varepsilon z\right|, \quad (24)$$

where e is the elementary charge; it is also taken into account that E_t and $E_z^{\pm}$ are in phase in the time domain according to Eq. (21) and that $E_z^{\pm} \ll E_t$. From Eq. (24) it follows that the presence of the SPP field leads to the periodic spatial modulation of the heating source, $\sim e^{(\alpha+\alpha_2)x}\cos k_\varepsilon z$, which causes the same modulation of the collision frequency (23) and, finally, of metal permittivity (19). This proves the presence of positive feedback between the SPP excitation and $\delta\varepsilon$ growth.

From Eqs. (21) and (24), using definitions (19) and (23), we obtain the equation describing nonlinear growth of SPP magnitude:

$$\frac{\partial^2\tilde{E}_z^{\pm}}{\partial t^2} + \nu\frac{\omega_0^2}{2\omega_p^2}\frac{\partial\tilde{E}_z^{\pm}}{\partial t} = \xi\nu\frac{\omega_0^4}{\omega_p^4}\frac{E_i^2 e^2}{\hbar m\omega_0^2}\tilde{E}_z^{\pm}. \quad (25)$$

Initially, when the collision frequency perturbation is relatively small ($\nu_0 \gg \xi W_e/\hbar$), the SPP magnitude increases exponentially as $\tilde{E}_z^{\pm} = \tilde{E}_{z,0}^{\pm} e^{\Gamma_0 t}$, where the increment is given by

$$\Gamma_0 = \frac{\omega_0^2}{\omega_p^2}\sqrt{\xi\nu_0\frac{E_i^2 e^2}{\hbar m\omega_0^2}}. \quad (26)$$

For the parameters of gold ($\xi = 0.5$, $\varepsilon_0 \cong -26 - 1.85i$) and a 100 fs laser pulse of 800 nm central wavelength and fluence of 1 J/cm^2, we found $\Gamma^{-1} \cong 50$ fs. With the overall heating of electrons in the skin layer, collision frequency ν increases more than 10 times [28], so the characteristic time of SPP growth decreases down to 10–15 fs. It means that during the action of an intense femtosecond laser pulse the magnitude of SPP will increase 10^1–10^3 times due to the described instability.

Here we should note that the heat transport effects neglected in Eq. (24) will gradually change the permittivity profile $\delta\tilde{\varepsilon}(x)$ at the characteristic timescale of 200–300 fs (heat diffusion along the z axis is several orders slower). This slightly decreases the instability increment. On the other hand, heat diffusivity also decreases with an increase of electronic temperature, which enhances the temperature difference between the neighboring near-surface regions. The influence of this effect on heat deposition and LIPSS formation was studied in Ref. [11]. Anyway, we emphasize that the discussed instability development accompanied by the periodic heating of electrons is significantly faster than the crystal lattice heating and melting; thus the laser pulse decay should be treated as the first stage of surface structuring, taking place before any matter redistribution.

Depending on the excitation conditions and on the material, the SPP can be either fully absorbed during the laser pulse action or exist for several hundred femtoseconds after pumping. If the absorption time τ_a is less than the laser pulse duration, the distribution of deposited energy is described by Eq. (24), so the model predicts the appearance of LIPSS with the period $2\pi/k_\varepsilon$. If τ_a is significantly longer, then the SPP will be absorbed mostly after the laser pulse reflection, so the absorption profile will follow the SPP standing wave intensity $\propto \cos^2 k_\varepsilon z$ and the LIPSS period is expected to be about π/k_ε.

As it follows from Eq. (21), τ_a decreases with the increase of collision frequency during the electron heating (τ_a changes approximately from 370 to 30 fs for the parameters of gold listed above). This will limit the exponential growth. According to Eqs. (21) and (19), the saturated magnitude of the SPP does not depend on ν when $\nu \gg \nu_0$ and can be expressed as

$$\tilde{E}_{z,\,\mathrm{sat}}^{\pm} = i\frac{\omega_0}{2\omega_p}\tilde{E}_i \cong E_t/4. \quad (27)$$

Thus, in the saturation regime the internal electric field given by Eq. (22) varies 3 times along the z axis (from $0.5E_t$ to $1.5E_t$), so the electric field intensity $|\mathbf{E}_\Sigma|^2$ changes by almost an order of magnitude. Strictly speaking, this takes us beyond the limits of full applicability of the perturbation theory, so that Eq. (27) should be treated as an estimation proving that the magnitude of the SPP electric field can be comparable to the magnitude of the transmitted electric field inside the medium E_t. Nevertheless, the regime of saturation seems to be the most probable in the experiments on single-pulse LIPSS formation where the electrons in the skin layer are heated up

to several eV and the collision frequency becomes comparable to ω_0.

Diffraction of SPPs on the permittivity grating $\delta\varepsilon(z)$ also shortens their lifetime. Due to this effect SPPs are transformed to the reflected optical wave propagating normally to the surface. The corresponding lifetime can be found by solving the similar (more precisely, inverse) problem relative to the problem of direct transformation into the SPP solved above [Eqs. (5)–(21)]. According to these calculations, the diffraction lifetime can be estimated as $t_{\text{diff}} \cong 16(-\varepsilon_0)^{7/2}/\omega\delta\varepsilon^2$, which gives ∼3–4 ps for the parameters discussed above, so under the chosen conditions the effect is negligible.

IV. CONCLUSION

To conclude, we showed that normal reflection of an intense laser pulse from a metal surface is an unstable process. Excitation of seed surface plasmon polaritons at any random permittivity perturbations or surface irregularities leads to the formation of a periodic interference pattern and, consequently, to the periodic heating of the medium and permittivity modulation. In turn, this periodic modulation of permittivity happens to be resonant for further enhancement of conversion of the incident wave into the pair of counterpropagating SPPs. This positive feedback leads to several-order growth of SPP magnitude during the action of the femtosecond laser pulse. Regardless of the nature of the initial excitation of the SPPs, nonlinear (instable) growth of their magnitude should take place at the femtosecond timescale if the incident fluence is about $1\,\text{J/cm}^2$ and larger.

The described instability seems to be important for understanding LIPSS formation in the single-pulse or few-pulse regime (when the theoretical models based on the interpulse feedback cannot interpret the appearance of LIPSS satisfactorily). The resulting magnitude of the SPP standing wave and the contrast in local laser intensities directly influence the initial conditions for crystal lattice heating, melting, and ablation. For the parameters of gold, the instability becomes faster than the typical femtosecond laser pulse duration at fluences of about ~ 0.5–$1\,\text{J/cm}^2$ which is the experimentally observed threshold of single-pulse LIPSS formation [12,13]. As it follows from the developed theory, spontaneous growth of the SPP due to the thermal nonlinearity saturates when the magnitude of the SPP standing wave reaches $\sim 1/2$ of the transmitted electric field inside the metal. In contrast to previous studies, fast exponential growth and saturation of the SPP make the presented model less sensitive to the amplitude of initial surface roughness. Notice that the laser-induced formation of periodic ripples inside the glass was recently interpreted in Refs. [19,30] as a consequence of the instability of a flat wave reflection from an ionized dielectric layer.

In the case of few-picosecond laser pulses (or longer) the discussed mechanism of decay into SPPs is still relevant; however, the crystal lattice heating and heat diffusion should be taken into account explicitly. The total electron collision frequency is also expected to increase periodically, but the dominant contribution should come from the electron-phonon scattering rate which grows linearly with the lattice temperature $\nu_{e-ph} \propto T_l$.

ACKNOWLEDGMENTS

The initial analysis of the ultrafast nonlinear phenomena in metals and the conceptualization were carried out in the framework of the project of the Ministry of Science and Higher Education of the Russian Federation (Project No. 075-15-2020-790). Development of the analytical theory was supported by the Foundation for the Development of Theoretical Physics and Mathematics "BASIS." The author also thanks V. A. Mironov and V. B. Gildenburg for fruitful discussions.

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