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Harmonic generation upon mixing an s-polarised wave and a wave heating a metal

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***Abstract*. It is shown that when an s-polarised wave is mixed with a wave of the same frequency, which is incident along the normal to the metal surface, three frequency-doubled waves are generated. Two of them are p-polarised and one is s-polarised. The effect of the electron and lattice temperature evolution on the SHG efficiency is described quantitatively. An anomalously strong change in the generation efficiency of an s-polarised harmonic is found during temperature variations, which leads to a noticeable change in the electron collision frequency.**

Keywords: *second harmonic generation, wave mixing, femtosecond pulse, heating of metal electrons.*

1. Introduction

Many papers are devoted to investigation of the SHG by metal electrons (see, for example, [1 – 15]). In metals, harmonics are generated in a comparatively narrow skin-layer; therefore, the conversion coefficient of radiation with the frequency ω into radiation with the frequency 2ω is rather small. At the same time, in the simplest description within the framework of the perturbation theory the radiation flux density of the second harmonic is proportional to the square of the radiation flux density at the fundamental frequency ω. In this connection to achieve a noticeable SHG effect it is desirable to use radiation with a rather large intensity, which does not lead, however, to the damage of the metal surface. This radiation can be obtained with the help of femtosecond laser pulses. When ultrashort laser pulses are used already at a moderate radiation flux density of the fundamental wave, the SHG is accompanied by the electron heating leading to a change in the generation efficiency. The necessity to account for the electron and lattice heating during the SHG was demonstrated in papers [7, 9, 11 – 15]. The metal heating is accompanied by a noticeable increase in the electron – electron and electron – phonon collision frequencies. Under conditions of an efficient SHG, the electron collision frequencies can be comparable with the radiation frequency and even exceed it. The theoretical description of the SHG suitable at relatively large electron collision frequencies was performed in [15]. It is based on the simultaneous solution of Maxwell's equations, equations for the electron density and average electron velocity as well as equations for the electron and lattice temperatures. The authors of paper [15] described the influence of the metal heating on the SHG efficiency of an s-polarised wave.

In this paper, we develop the approach of paper [15] by presenting the SHG theory based on the same equations under conditions of mixing of an s-polarised wave and a heating wave (with the same frequency) incident normal on the metal surface. We show that when these waves are mixed, three waves with the frequency 2ω are generated: two p-polarised harmonics and one s-polarised harmonic. One of the p-polarised harmonics is emitted in the reflection direction of the s-polarised wave with the frequency ω. This harmonic appears due to the nonlinear current generation by an incident s-polarised wave. The second p-polarised harmonic is generated upon mixing the s-polarised wave with the wave heating a metal. This wave is emitted at the metal-surface angle exceeding the angle at which the initial s-polarised wave is reflected. At the same angle, one more second harmonic appearing upon mixing the same waves is emitted, the harmonic being s-polarised. This harmonic appears only when we take into account the electron collisions resulting in lifting the ss prohibition under studied conditions – generation of only p-polarised second harmonic upon irradiation of the metal surface by an s-polarised wave (see, for example, [10]).

We will show below that the electron heating due to absorption of the wave incident normally to the metal affects the generation efficiency of all the three harmonics. We also describe the conditions under which the rise of the electron or lattice temperature leads to a relatively small decrease in the generation efficiency of p-polarised harmonics while the electron cooling due to the heat transfer deep into the metal leads to an increase in their generation. On the contrary, the influence of the heating and subsequent cooling of the metal on the s-polarised harmonic generation is rather large and accompanied by the change in its generation efficiency by an order of magnitude. The latter property makes this harmonic a convenient tool for experimental investigation of electron collision frequencies in a thermodynamically non-equilibrium metal.

2. Model description of a metal

We will describe the conductivity dynamics of electrons by using the equation of motion in the form

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$$\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u}\nabla)\boldsymbol{u} = -\frac{1}{nm}\nabla p - \nu \boldsymbol{u} + \frac{e}{m}\left(\boldsymbol{E} + \frac{1}{c}[\boldsymbol{u}\boldsymbol{B}]\right), \tag{1}$$

where n is the conduction electron density; $\boldsymbol{u}$ is the velocity of their directed motion; e and m are the charge and mass usually close to their values for a free electron; c is the speed of light; $p = p(n, T)$ is the electron pressure; T is the electron temperature; $\boldsymbol{E} = \boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{B} = \boldsymbol{B}(\boldsymbol{r}, t)$ are the electric and magnetic fields in a metal; $\nu = \nu(n, T, T_{\rm lat})$ is the characteristic electron collision frequency depending on n, T, and the lattice temperature $T_{\rm lat}$. Assuming the collisions of the electrons with the impurities to be insignificant, we will represent the frequency ν as a sum of electron–phonon ($\nu_{\rm eph}$) and electron–electron ($\nu_{\rm ee}$) collision frequencies: $\nu = \nu_{\rm eph} + \nu_{\rm ee}$. If the electron distribution is strongly degenerate and $T_{\rm lat}$ is larger than the Debye temperature $\Theta_{\rm D}$, the approximation [16, 17]

$$\nu = \nu_{\rm eph}(T_0)\frac{T_{\rm lat}}{T_0} + a\,\frac{k_{\rm B}^2 T^2}{\hbar \varepsilon_{\rm F}} \tag{2}$$

is possible for ν, where $k_{\rm B}$ is the Boltzmann constant; $\hbar$ is Planck's constant; T_0 is the initial metal temperature in an equilibrium state; $\varepsilon_{\rm F}$ is the Fermi energy; a is the numerical factor whose quantity depends on the type of the energy-band structure of the metal.

The electric and magnetic fields in a metal are described by Maxwell's equations

$$\mathrm{rot}\boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}, \tag{3}$$

$$\mathrm{rot}\boldsymbol{B} = \frac{\varepsilon_0}{c}\frac{\partial \boldsymbol{E}}{\partial t} + \frac{4\pi}{c}\,en\boldsymbol{u}, \tag{4}$$

where ε_0 is the the static dielectric constant caused by the coupled electrons and the lattice.

Collisions of the electrons lead to their heating and dissipation of the electromagnetic field energy. The absorption power is determined by the Joule heat $Q = \boldsymbol{j}\boldsymbol{E} = en\boldsymbol{u}\boldsymbol{E}$ released per unit time per unit volume. The inhomogeneous electron heating caused by the field absorption is accompanied by the heat transport and cooling of the electrons due to the energy transfer to the lattice. The electron-temperature equations, taking these processes into account, have the form [18, 19]

$$C_{\rm e}\frac{\partial T}{\partial t} - \mathrm{div}(\lambda \nabla T) = Q - G(T - T_{\rm lat}), \tag{5}$$

where $C_{\rm e} = \pi^2 k_{\rm B}^2 nT/2\varepsilon_{\rm F}$ is the heat capacity of the electrons; G is the constant of electron coupling with the lattice. In (5) the heat conductivity has the form $\lambda = C_{\rm e} v_{\rm F}^2/3\nu_\lambda$, where $v_{\rm F}$ is the Fermi velocity; ν_λ is the electron collision frequency determining the heat conductivity process. The effective electron–phonon and electron–electron collision frequencies entering $\nu_\lambda = \nu_{{\rm eph},\lambda} + \nu_{{\rm ee},\lambda}$ differ in the quantity from those determining the conductivity but have the same dependence on the lattice and electron temperatures:

$$\nu_\lambda = \nu_{{\rm eph},\lambda}(T_0)\frac{T_{\rm lat}}{T_0} + b\,\frac{k_{\rm B}^2 T^2}{\hbar \varepsilon_{\rm F}}, \tag{6}$$

where $b \neq a$. Finally, the lattice temperature variation is described by the equation

$$C_{\rm lat}\frac{\partial T_{\rm lat}}{\partial t} = G(T - T_{\rm lat}), \tag{7}$$

where $C_{\rm lat} \simeq 3k_{\rm B}N$ is the heat capacity of the lattice at $T_{\rm lat} > \Theta_{\rm D}$; N is the lattice atom density. The presented equations are basic for describing the properties of the non-equilibrium metal in the electromagnetic field at temperatures larger than the Debye temperature. In this case, the lattice temperature is assumed small compared to the melting temperature, while the electron temperature is assumed small compared to the degeneracy temperature.

3. Field penetration into a metal

Consider the interaction of the s-polarised electromagnetic wave with a metal occupying the half-space $z > 0$. We will represent the wave field in the form

$$\frac{1}{2}\,\boldsymbol{E}_{\rm las}\exp(-{\rm i}\omega t + {\rm i}\boldsymbol{k}_{\rm las}\boldsymbol{r}) + {\rm c.c.}, \tag{8}$$

where $\boldsymbol{E}_{\rm las} = (0, E_{\rm las}, 0)$; $E_{\rm las}$ is the electric field strength amplitude slowly varying in time $2\pi/\omega$; ω is the frequency; $\boldsymbol{k}_{\rm las} = (k_x, 0, k_z)$ is the wave vector. Taking into account the fact that the field amplitude of the incident wave (8) slowly changes in time, it is natural to search for the solution of equations (1)–(7) in the form

$$A = A_0 + \frac{1}{2}\sum_{s=1}^{\infty}[A_s\exp(-{\rm i}s\omega t) + {\rm c.c.}], \tag{9}$$

where A denotes n, T, $T_{\rm las}$ or one of the vector components $\boldsymbol{u}$, $\boldsymbol{E}$, or $\boldsymbol{B}$. In expression (9), the functions A_0 and A_s depend on x and z and slowly change in time,

$$\left|\frac{\partial \ln A_0}{\partial t}\right|,\ \left|\frac{\partial \ln A_s}{\partial t}\right| \ll s\omega, \quad s = 1, 2, \ldots. \tag{10}$$

Taking into account this inequality and in accordance with expression (8) for the magnetic field of the incident wave from expression (3), we have

$$\frac{1}{2}\,\boldsymbol{B}_{\rm las}\exp(-{\rm i}\omega t + {\rm i}\boldsymbol{k}_{\rm las}\boldsymbol{r}) + {\rm c.c.}, \tag{11}$$

$$\boldsymbol{B}_{\rm las} = (-k_z, 0, k_x)\frac{c}{\omega}\,E_{\rm las}.$$

Assuming that the characteristic velocity of the electrons in the field is small compared to their thermal velocity, we will restrict below our consideration to the approximate solution of equations (1)–(7) with an accuracy up to the terms quadratic in the field strength. In such an approximation it is sufficient to retain the terms with $s \leqslant 2$ in expansion (9). In this case, being interested in the change of the quantities at the frequency ω, we can neglect the electron temeperature and density perturbations quadratic in the field as well as pressure (p) and frequency (ν) perturbations caused by them. In the studied approximation for harmonics with $s = 1$ we have a system of linear equations. In the linear approximation over the external field, the field in the metal can be expressed as

$$\frac{1}{2}\boldsymbol{E}_1 \exp(-\mathrm{i}\omega t) + \text{c.c.}, \tag{12}$$

$$\boldsymbol{E}_1 = (0, E_1(z), 0)\exp(\mathrm{i}k_x x).$$

It follows that $\mathrm{div}\boldsymbol{E}_1 = 4\pi e n_1 = 0$, i.e. $n_1 = 0$, as well as $p_1 = 0$ and $v_1 = 0$. As a result, taking into account inequality (10) from linearised equation (1) we find

$$\boldsymbol{u}_1 = \frac{e}{m}\boldsymbol{E}_1 \frac{\mathrm{i}}{\omega + \mathrm{i}v_0}, \tag{13}$$

where v_0 depends on n_0 and T_0 – the electron density and temperature slowly varying in time. Accounting for inequality (10) and relation (13), we obtained from (3) and (4) the equation for the function $E_1(z)$:

$$\frac{\mathrm{d}^2 E_1(z)}{\mathrm{d}z^2} + \left[\frac{\omega^2}{c^2}\,\varepsilon(\omega) - k_x^2\right]E_1(z) = 0, \tag{14}$$

where

$$\varepsilon(\omega) = \varepsilon_1(\omega) + \mathrm{i}\varepsilon_2(\omega) = \varepsilon_0 - \frac{\omega_{\mathrm{pl}}^2}{\omega(\omega + \mathrm{i}v_0)} \tag{15}$$

is the dielectric constant of metal at the frequency ω; $\omega_{\mathrm{pl}} = (4\pi e^2 n_0/m)^{1/2}$ is the plasma frequency of the electrons. At $\omega_{\mathrm{pl}}^2 > (\omega^2 + v_0^2)\varepsilon_0$, the solution of Eqn (14) decreasing inside the metal has the form

$$E_1(z) = E_1(0)\exp[-\varkappa(\omega, k_x)z], \tag{16}$$

where

$$\varkappa(\omega, k_x) = \sqrt{k_x^2 - \frac{\omega^2\varepsilon(\omega)}{c^2}} = \varkappa_1(\omega, k_x) - \mathrm{i}\varkappa_2(\omega, k_x); \tag{17}$$

$$\varkappa_l(\omega, k_x) = \frac{1}{\sqrt{2}}\left\{\sqrt{\left[\frac{\omega^2\varepsilon_1(\omega)}{c^2} - k_x^2\right]^2 + \left[\frac{\omega^2\varepsilon_2(\omega)}{c^2}\right]^2}\right.$$

$$\left. -(-1)^l\left|\frac{\omega^2\varepsilon_1(\omega)}{c^2} - k_x^2\right|\right\}^{1/2}, \quad l = 1, 2. \tag{18}$$

The solution of form (16) takes place if the dependence $\varepsilon(\omega)$ at the scales $\sim \varkappa_l^{-1}(\omega)$ can be neglected (see details in [20, 21]). By using inequality (10) and expressions (12), (16), we find from expression (3) the magnetic field in the metal:

$$\frac{1}{2}\boldsymbol{B}_1 \exp(-\mathrm{i}\omega t) + \text{c.c.},$$

$$\boldsymbol{B}_1 = (-\mathrm{i}\varkappa(\omega, k_x), 0, k_x)\frac{c}{\omega}E_1(z)\exp(\mathrm{i}k_x x). \tag{19}$$

The wave incident on the metal is partially reflected from it. In the conditions under study, the electric and magnetic fields of the reflected wave can be expressed as

$$\frac{1}{2}\boldsymbol{E}^{\mathrm{r}}\exp(-\mathrm{i}\omega t + \mathrm{i}k_x x - \mathrm{i}k_z z) + \text{c.c.},$$

$$\frac{1}{2}\boldsymbol{B}^{\mathrm{r}}\exp(-\mathrm{i}\omega t + \mathrm{i}k_x x - \mathrm{i}k_z z) + \text{c.c.}, \tag{20}$$

$$\boldsymbol{E}^{\mathrm{r}} = \boldsymbol{E}_{\mathrm{las}}R_{\mathrm{s}}(\omega, \boldsymbol{k}), \quad \boldsymbol{B}^{r} = (k_z, 0, k_x)\frac{c}{\omega}E_{\mathrm{las}}R_{\mathrm{s}}(\omega, \boldsymbol{k}), \tag{21}$$

where $R_{\mathrm{s}}(\omega, \boldsymbol{k})$ is the complex reflectivity of the s-wave having the frequency ω and propagating in the direction of the $\boldsymbol{k}$ vector. According to the definition

$$E_1(0) = F_{\mathrm{s}}(\omega, \boldsymbol{k})E_{\mathrm{las}} \tag{22}$$

we will introduce the complex coefficient of the s-wave penetration, $F_{\mathrm{s}}(\omega, \boldsymbol{k})$. The relation $F_{\mathrm{s}}(\omega, \boldsymbol{k}) = 1 + R_{\mathrm{s}}(\omega, \boldsymbol{k})$ follows from the continuity condition of tangential electric field components. Taking into account the continuity of the tangential component of the magnetic field from (8), (11), (12), (16), and (19)–(22), we find the explicit expression for the s-wave penetration coefficient:

$$F_{\mathrm{s}}(\omega, \boldsymbol{k}) = \frac{2k_z}{k_z + \mathrm{i}\varkappa(\omega, k_x)}. \tag{23}$$

Relations (12), (19) and (22), (23) completely determine the field in the metal at the frequency ω.

The above-described peculiarities of the s-wave penetration and reflection allow us to write the field in the metal in a more general case when the second wave incident normally together with the s-wave affects it. Let us assume that the total field of two waves has the form

$$\frac{1}{2}\boldsymbol{E}_{\mathrm{las}}\exp(-\mathrm{i}\omega t + \mathrm{i}\boldsymbol{k}_{\mathrm{las}}\boldsymbol{r})$$

$$+\frac{1}{2}\boldsymbol{E}_{\mathrm{p}}\exp(-\mathrm{i}\omega\boldsymbol{t} + \mathrm{i}\boldsymbol{k}_{\mathrm{p}}\boldsymbol{r}) + \text{c.c.}, \tag{24}$$

where $\boldsymbol{E}_{\mathrm{p}} = (E_{\mathrm{p}}\cos\phi, E_{\mathrm{p}}\sin\phi, 0)$; $\boldsymbol{E}_{\mathrm{las}} = (0, E_{\mathrm{las}}, 0)$; $\boldsymbol{k}_{\mathrm{las}} = (k\sin\theta, 0, k\cos\theta)$; $\boldsymbol{k}_{\mathrm{p}} = (0, 0, k)$. Then, in the linear approximation with respect to the intensity of the interacting waves, in accordance with the above-mentioned, we have

$$\boldsymbol{E}_1 = \boldsymbol{E}(\boldsymbol{k}_{\mathrm{las}}) + \boldsymbol{E}(\boldsymbol{k}_{\mathrm{p}}), \quad \boldsymbol{B}_1 = -\mathrm{i}\frac{c}{\omega}\,\mathrm{rot}\boldsymbol{E}_1, \tag{25}$$

$$\boldsymbol{E}(\boldsymbol{k}_{\mathrm{las}}) = F_{\mathrm{s}}(\omega, \boldsymbol{k}_{\mathrm{las}})\boldsymbol{E}_{\mathrm{las}}\exp[\mathrm{i}kx\sin\theta - \varkappa(\omega, k\sin\theta)z],$$

$$\boldsymbol{E}(\boldsymbol{k}_{\mathrm{p}}) = F_{\mathrm{s}}(\omega, \boldsymbol{k}_{\mathrm{p}})\boldsymbol{E}_{\mathrm{p}}\exp[-\varkappa(\omega, 0)z] \tag{26}$$

from (3), (12), (16), and (22). These relations are basic for calculating the nonlinear current in the metal.

4. Nonlinear current and susceptibility

Consider the electron dynamics at the second harmonic frequency 2ω, using the motion equation (1). According to inequality (10), in describing the oscillations of the electrons with the frequency 2ω the time derivative $|\partial \ln u_2/\partial t|$ can be neglected. In addition, under conditions of normal and high-frequency skin effect discussed in this paper, we can neglect the weak influence of ∇p_2. Taking into account these simplifications and using relation (9), we derive from Eqn (1)

$$(v_0 - 2\mathrm{i}\omega)\boldsymbol{u}_2 = \frac{c}{m}\boldsymbol{E}_2 + \frac{e}{2mc}[\boldsymbol{u}_1\boldsymbol{B}_1] - \frac{1}{2}(\boldsymbol{u}_1\nabla)\boldsymbol{u}_1. \tag{27}$$

With allowance for relations (13) and (25), we find from (27) a slow current-density envelope at the second harmonic frequency:

$$\boldsymbol{j}_2 = en\boldsymbol{u}_2 = \frac{\mathrm{i}ne^2}{m(2\omega + \mathrm{i}\nu_0)}\,\boldsymbol{E}_2 + \boldsymbol{j}_{\mathrm{nl}}, \tag{28}$$

where the nonlinear current is

$$\boldsymbol{j}_{\mathrm{nl}} = \frac{\mathrm{i}ne^3}{4m^2(\omega + \mathrm{i}\nu_0)^2(2\omega + \mathrm{i}\nu_0)}\,\nabla \boldsymbol{E}_1^2 - \frac{ne^3}{2m^2(\omega + \mathrm{i}\nu_0)^2(2\omega + \mathrm{i}\nu_0)}\,\frac{v_0}{\omega}\,[\boldsymbol{E}_1 \mathrm{rot}\boldsymbol{E}_1]. \tag{29}$$

In accordance with the definition $\boldsymbol{j}_{\mathrm{nl}} = \partial \boldsymbol{P}^{\mathrm{nl}}/\partial t$, the nonlinear polarisation at the frequency 2ω

$$P_i^{\mathrm{nl}}(2\omega, \boldsymbol{k}_1, \boldsymbol{k}_2) = \chi_{ijk}(2\omega, \boldsymbol{k}_1, \boldsymbol{k}_2) E_j(\boldsymbol{k}_1) E_k(\boldsymbol{k}_2) \tag{30}$$

corresponds to the nonlinear current (29). Here, $\chi_{ijk}(2\omega, \boldsymbol{k}_1, \boldsymbol{k}_2)$ is the nonlinear susceptibility tensor. According to (30) and (29), the components of the nonlinear susceptibility tensor corresponding to the field (25), (26) and determining the tangential components of the nonlinear polarisation vector have the form

$$\chi_{xyy}(2\omega, \boldsymbol{k}_{\mathrm{las}}, \boldsymbol{k}_{\mathrm{las}}) = \chi_{xyy}(2\omega, \boldsymbol{k}_{\mathrm{p}}, \boldsymbol{k}_{\mathrm{las}}) = -\frac{\mathrm{i}e\omega_{\mathrm{pl}}^2 \sin\theta}{16\pi mc\omega(\omega + \mathrm{i}\nu_0)(2\omega + \mathrm{i}\nu_0)}, \tag{31}$$

$$\chi_{yxy}(2\omega, \boldsymbol{k}_{\mathrm{p}}, \boldsymbol{k}_{\mathrm{las}}) = -\frac{\mathrm{i}\nu_0}{\omega + \mathrm{i}\nu_0}\,\chi_{xyy}(2\omega, \boldsymbol{k}_{\mathrm{p}}, \boldsymbol{k}_{\mathrm{las}}) = -\frac{e\omega_{\mathrm{pl}}^2 \nu_0 \sin\theta}{16\pi mc\omega(\omega + \mathrm{i}\nu_0)^2(2\omega + \mathrm{i}\nu_0)}. \tag{32}$$

Expressions (26) and (30)–(32) determine the polarisation of the metal at the doubled frequency and allow us to consider the SHG.

5. Radiation field produced by induced polarisation

Assume that in metal there exists induced polarisation

$$\boldsymbol{P}_{\mathrm{st}}(\Omega, q_x, \xi) = \frac{1}{2}\,\boldsymbol{P}_{\mathrm{st}} \times \exp(-\mathrm{i}\Omega t + \mathrm{i}q_x x - \xi z) + \mathrm{c.c}, \quad z > 0, \tag{33}$$

where Ω is the frequency of polarisation variation; q_x is the wave vector component along the x axis; $1/\xi$ is the characteristic dimension of the polarisation localisation region. We will first consider radiation by a metal of an s-polarised wave whose electric field is directed along the y axis. Then, outside the metal the electromagnetic field of the emitted wave has the form

$$\frac{1}{2}\,\boldsymbol{E}_{\mathrm{rad}} \exp(-\mathrm{i}\Omega t + \mathrm{i}q_x x - \mathrm{i}q_z z) + \mathrm{c.c.}, \quad \boldsymbol{E}_{\mathrm{rad}} = (0,1,0)E_{\mathrm{rad}}, \quad z < 0, \tag{34}$$

$$\boldsymbol{B}_{\mathrm{rad}} = (q_z, 0, q_x)\,\frac{c}{\Omega}\,E_{\mathrm{rad}}, \tag{35}$$

where $\Omega = qc$; $q = \sqrt{q_x^2 + q_z^2}$. As before, the electromagnetic field in the metal is described by equations (3), (4), but now the right-hand side of equation (4) contains the induced-polarisation source:

$$\mathrm{rot}\boldsymbol{B} = \frac{\varepsilon_0}{c}\,\frac{\partial \boldsymbol{E}}{\partial t} + \frac{4\pi}{c}\,en\boldsymbol{u} + \frac{4\pi}{c}\,\frac{\partial \boldsymbol{P}_{\mathrm{st}}(\Omega, q_x, \xi)}{\partial t}. \tag{36}$$

We will seek the electric field in the metal in the form

$$\frac{1}{2}\,\boldsymbol{E}(z)\exp(-\mathrm{i}\Omega t + \mathrm{i}q_x x) + \mathrm{c.c.}, \quad \boldsymbol{E}(z) = (0, E_y(z), 0), \quad z > 0. \tag{37}$$

Because, when the s-polarised wave is emitted, $\mathrm{div}\boldsymbol{E} = \partial E_y/\partial y = 0$, we have

$$\frac{\mathrm{d}^2 E_y(z)}{\mathrm{d}z^2} - \varkappa^2(\Omega, q_x) E_y(z) = -\frac{4\pi}{c^2}\,\Omega^2 P_{\mathrm{st},y} \exp(-\xi z) \tag{38}$$

from Maxwell's equations for the $E_y(z)$ function. Here, $\varkappa = \varkappa(\Omega, q_x)$ is described by expressions (17) and (18), valid at $\omega_{\mathrm{pl}}^2 > (\Omega^2 + \nu_0^2)(\varepsilon_0 - q_x^2/q^2)$. The solution of the nonuniform equation (38) decreasing at $z \to \infty$ has the form:

$$E_y(z) = \left[E_y(0) + \frac{4\pi\Omega^2}{c^2(\xi^2 - \varkappa^2)}\,P_{\mathrm{st},y}\right]\exp(-\varkappa z) - \frac{4\pi\Omega^2}{c^2(\xi^2 - \varkappa^2)}\,P_{\mathrm{st},y}\exp(-\xi z). \tag{39}$$

The magnetic field

$$\frac{1}{2}\,\boldsymbol{B}(z)\exp(-\mathrm{i}\Omega t + \mathrm{i}q_x x) + \mathrm{c.c.}, \quad \boldsymbol{B}(z) = \frac{c}{\Omega}\,[\mathrm{i}E_y'(z), 0, q_x E_y(z)], \quad z > 0 \tag{40}$$

corresponds to the electric field described by expressions (37) and (39).

Using the continuity condition of the tangential electric-field components on the metal surface, we find from expression (34), (35), (37), (39), and (40) the relationship for the electric field strength of the emitted wave with the y components of induced polarisation:

$$E_{\mathrm{rad}} = E_y(0) = \Phi_{\mathrm{s}}(\Omega, q_x, \xi) P_{\mathrm{st},y}, \tag{41}$$

where we introduced the notation for the complex radiation coefficient:

$$\Phi_{\mathrm{s}}(\Omega, q_x, \xi) = \frac{4\mathrm{i}\pi}{[\xi + \varkappa(\Omega, q_x)][q_x + \mathrm{i}\varkappa(\Omega, q_x)]}\,\frac{\Omega^2}{c^2}. \tag{42}$$

Consider now radiation of a p-polarised wave. In this case, the magnetic field of the emitted wave has only one component along the y axis:

$$\frac{1}{2}\,\boldsymbol{B}_{\mathrm{rad}}\exp(-\mathrm{i}\Omega t + \mathrm{i}q_x x - \mathrm{i}q_z z) + \mathrm{c.c.}, \quad \boldsymbol{B}_{\mathrm{rad}} = (0,1,0)B_{\mathrm{rad}}, \quad z < 0, \tag{43}$$

$$\boldsymbol{E}_{\mathrm{rad}} = -(q_z, 0, q_x)\,\frac{c}{\Omega}\,B_{\mathrm{rad}}. \tag{44}$$

We will seek the magnetic field in the metal in the form

$$\frac{1}{2}\,\boldsymbol{B}(z)\exp(-\mathrm{i}\Omega t+\mathrm{i}q_x x)+\text{c.c.},$$
$$\boldsymbol{B}(z)=(0,B_y(z),0),\quad z>0. \tag{45}$$

Let us eliminate the polarisation component $P_{\mathrm{st},y}(\Omega,q_x,\xi)$, which is a source of an s-polarised wave, from equation (36). We will restrict our consideration to the conditions when two other components of the polarisation vector $P_{\mathrm{st},y}(\Omega,q_x,\xi)$ meet the condition $[\mathrm{rot}\,\boldsymbol{P}_{\mathrm{st}}(\Omega,q_x,\xi)]_y=0$. Then, from expressions (1), (3), and (36) we derive

$$\frac{\mathrm{d}^2B_y(z)}{\mathrm{d}z^2}-\varkappa^2(\Omega,q_x)B_y(z)=0. \tag{46}$$

The solution of this equation decreasing at $z\to\infty$ has the form

$$B_y(z)=B_y(0)\exp(-\varkappa z)=B_{\mathrm{rad}}\exp(-\varkappa z). \tag{47}$$

The electric field is derived from equations (1), (36) and is described by the expressions

$$\frac{1}{2}\,\boldsymbol{E}(z)\exp(-\mathrm{i}\Omega t+\mathrm{i}q_x x)+\text{c.c.},$$
$$\boldsymbol{E}(z)=(E_x(z),0,E_z(z)),\quad z>0, \tag{48}$$

$$E_x(z)=\frac{\mathrm{i}c\varkappa}{\Omega\varepsilon(\Omega)}\,B_y(z)-\frac{4\pi}{\varepsilon(\Omega)}\,P_{\mathrm{st},x}\exp(-\xi z),$$
$$E_z(z)=\frac{cq_x}{\Omega\varepsilon(\Omega)}\,B_y(z)-\frac{4\pi}{\varepsilon(\Omega)}\,P_{\mathrm{st},z}\exp(-\xi z). \tag{49}$$

From the continuity condition E_x on the metal surface with the allowance for relations (45), (47) and (49) we find the magnetic field of the emitted wave

$$B_{\mathrm{rad}}=E_{\mathrm{rad}}=\Phi_{\mathrm{p}}(\Omega,q_x,\xi)P_{\mathrm{st},x}, \tag{50}$$

where

$$\Phi_{\mathrm{p}}(\Omega,q_x,\xi)=\frac{4\pi}{c}\,\frac{\Omega}{\varepsilon(\Omega)q_z+\mathrm{i}\varkappa(\Omega,q_x)} \tag{51}$$

is the complex radiation coefficient of a p-wave. Relations (42) and (51) allow us to find fields of the s- and p-waves emitted by a metal with induced polarisation.

6. Radiation at the second harmonic frequency

Consider now the waves emitted by a metal irradiated by an s-polarised wave incident at an arbitrary angle and by a wave incident normally with the same frequency. When these waves interact together in the metal, there appears induced polarisation at the sum frequency $2\omega=\omega+\omega$, which is described by relations (30)–(32). The induced-polarisation vector has three components. The component P_z^{nl} normal to the metal surface does not lead to generation. On the contrary, tangential components P_x^{nl} and P_y^{nl} are the sources of p- and s-polarised waves. According to relations (43), (47), (50), and (51), the quantities $P_x^{\mathrm{nl}}(2\omega,\boldsymbol{k}_{\mathrm{las}},\boldsymbol{k}_{\mathrm{las}})$ and $P_x^{\mathrm{nl}}(2\omega,\boldsymbol{k}_{\mathrm{p}},\boldsymbol{k}_{\mathrm{las}})$ determining polarisation along the x axis are the sources of two p-polarised waves with the frequency 2ω and the wave vectors $\boldsymbol{k}'=2k(\sin\theta,0,-\cos\theta)$ and $\boldsymbol{k}''=k(\sin\theta,0,\sqrt{4-\sin^2\theta})$, respectively. The wave with the wave vector $\boldsymbol{k}'$ propagates in the same direction as the reflected s-polarised wave at the frequency ω, i.e. $\theta'=\theta$, where θ' is the angle between $\boldsymbol{n}'=\boldsymbol{k}'/2k$ and $\boldsymbol{n}$ – the vector of the normal to the metal surface. The magnetic field of the p-polarised wave emitted in the direction of the $\boldsymbol{n}'$ vector has the form

$$\frac{1}{2}\,\boldsymbol{B}^{\mathrm{p}}(2\omega,\boldsymbol{k}')\exp(-\mathrm{i}2\omega t+\mathrm{i}\boldsymbol{k}'\boldsymbol{r})+\text{c.c.},$$
$$\boldsymbol{B}^{\mathrm{p}}(2\omega,\boldsymbol{k}')=(0,B_y^{\mathrm{p}}(2\omega,\boldsymbol{k}'),0),\quad z>0. \tag{52}$$

In accordance with relations (24)–(26), (30), (43), and (50), the nonzero y component of the magnetic field has the form

$$B_y^{\mathrm{p}}(2\omega,\boldsymbol{k}')=\Phi_{\mathrm{p}}(2\omega,2k_x,2\varkappa(\omega,k_x))$$
$$\times F_{\mathrm{s}}^2(\omega,\boldsymbol{k}_{\mathrm{las}})\chi_{xyy}(2\omega,\boldsymbol{k}_{\mathrm{las}},\boldsymbol{k}_{\mathrm{las}})E_{\mathrm{las}}^2, \tag{53}$$

where $\chi_{xyy}(2\omega,\boldsymbol{k}_{\mathrm{las}},\boldsymbol{k}_{\mathrm{las}})$ is the nonlinear susceptibility (31); $F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{las}})$ is the complex penetration coefficient (23); $\Phi_{\mathrm{p}}(2\omega,2k_x,2\varkappa(\omega,k_x))$ is the complex radiation coefficient of the p-wave (51). Expression (53) corresponds to the conditions under which the frequency of the generated wave satisfies the inequality

$$\omega_{\mathrm{pl}}^2>(4\omega^2+\nu_0^2)(\varepsilon_0-\sin^2\theta). \tag{54}$$

Generalisation to the case of higher frequencies 2ω is trivial (see, for example, [15]). The electromagnetic wave (52), (53) appears due to the nonlinear response of the metal to the action of the s-polarised wave. The Poynting vector $\boldsymbol{S}=(c/4\pi)[\boldsymbol{EB}]$:

$$\boldsymbol{S}^{\mathrm{p}}(2\omega,\boldsymbol{k}')=\frac{32\pi k^4e^2\omega_{\mathrm{pl}}^4\sin^2\theta\cos^4\theta}{m^2c^5(\omega^2+\nu_0^2)(4\omega^2+\nu_0^2)}\,\boldsymbol{n}'I_{\mathrm{las}}^2$$
$$\times\frac{1}{\{[k\cos\theta+\varkappa_2(\omega,k\sin\theta)]^2+\varkappa_2^2(\omega,k\sin\theta)\}^2}$$
$$\times\{[2k\varepsilon_1(2\omega)\cos\theta+\varkappa_2(2\omega,2k\sin\theta)]^2$$
$$+[2k\varepsilon_2(2\omega)\cos\theta+\varkappa_1(2\omega,2k\sin\theta)]^2\}^{-1} \tag{55}$$

corresponds to the wave (52), (53). Here, $I_{\mathrm{las}}=(c/8\pi)E_{\mathrm{las}}^2$ is the flux density of the s-polarised wave incident on the metal.

The second p-polarised wave with the wave vector $\boldsymbol{k}''$ propagates in the direction $\boldsymbol{n}''=\boldsymbol{k}''/2k$ at the angle θ'' to the normal vector $\boldsymbol{n}$ smaller than θ':

$$\tan\theta''=\frac{\sin\theta}{\sqrt{4-\sin^2\theta}}<\tan\theta',\quad\theta''<\theta'. \tag{56}$$

This wave appears due to the nonlinear mixing of the s-polarised wave with the wave vector $\boldsymbol{k}_{\mathrm{las}}=(k_x,0,k_z)$ and the wave with $\boldsymbol{k}_{\mathrm{p}}=(0,0,k)$ having the same frequency but incident along the normal to the metal surface. The magnetic field of this wave is described by expression (52) which contains $\boldsymbol{k}''$' instead of $\boldsymbol{k}'$, while the y component of the magnetic field has the form

$$B_y^{\mathrm{p}}(2\omega,\boldsymbol{k}'') = \Phi_{\mathrm{p}}(2\omega,k_x,\varkappa(\omega,k_x)+\varkappa(\omega,0))$$

$$\times F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{las}})F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{p}})\chi_{xyy}(2\omega,\boldsymbol{k}_{\mathrm{p}},\boldsymbol{k}_{\mathrm{las}})E_{\mathrm{las}}e_{\mathrm{p}}\sin\phi, \qquad (57)$$

where the frequency 2ω satisfies inequality (54). Expression (57) describes the simplest mixing of two waves emerging when they have components along the y axis. The latter takes place at $\sin\phi \neq 0$ or $\phi \neq 0,\pi$. By using expressions (23), (31), and (51), we find from (57) the Poynting vector

$$\boldsymbol{S}^{\mathrm{p}}(2\omega,\boldsymbol{k}'') = \frac{32\pi k^4 e^2\omega_{\mathrm{las}}^4\sin^2\theta\cos^2\theta\sin^2\phi}{m^2c^5(\omega^2+\nu_0^2)(4\omega^2+\nu_0^2)}\,\boldsymbol{n}''I_{\mathrm{las}}I_{\mathrm{p}}$$

$$\times\frac{1}{[k\cos\theta+\varkappa_2(\omega,k\sin\theta)]^2+\varkappa_1^2(\omega,k\sin\theta)} \qquad (58)$$

$$\times\frac{1}{[k+\varkappa_2(\omega,0)]^2+\varkappa_1^2(\omega,0)}\left\{\left[k\varepsilon_1(2\omega)\sqrt{4-\sin^2\theta}\right.\right.$$

$$\left.\left.+\varkappa_2(2\omega,k\sin\theta)\right]^2+\left[k\varepsilon_2(2\omega)\sqrt{4-\sin^2\theta}+\varkappa_1(2\omega,k\sin\theta)\right]^2\right\}^{-1},$$

where $I_{\mathrm{p}} = (c/8\pi)E_{\mathrm{p}}^2$ is the flux density of the wave incident on the metal normally to its surface. The origin of radiation of both p-polarised harmonics has the same physical reason – the presence of the nonlinear polarisation along the x axis, proportional to the field energy density gradient in the metal [see (29)]. Therefore, in expressions (55) and (58) the dependences of the factors in the product at the flux densities from the metal parameters and mixed waves are qualitatively similar. Generation of only p-polarised second harmonic is retained during the mixing of two waves, if the electric field of the wave incident normally onto the metal is collinear to the s-polarised wave field.

However, the situation changes when the electric fields of the mixed waves are not collinear. Then, if the normally incident wave has the electric field component $E_{\mathrm{p},x} = E_{\mathrm{p}}\cos\phi$, orthogonal to the electric field of the s-polarised wave $E_{\mathrm{las}} = (0,E_{\mathrm{las}},0)$, due to the simultaneous action of $E_{\mathrm{p},x}$ and the magnetic field component (produced by an s-wave) normal to the metal surface, there appears a periodic motion of electrons with a doubled frequency in the direction of the electric field of the s-wave, collinear to the metal surface. One more surface-collinear nonlinear polarisation component P_y^{nl}, which is a source of the s-polarised second harmonic, is induced in the metal skin-layer. In this case, due to the mixing of the waves under study, there appears the possibility to lift the ss-prohibition. According to expressions (24)–(26), (30), (34), and (41), the electric field of the s-polarised second harmonic has the form

$$\frac{1}{2}\,\boldsymbol{E}^{\mathrm{s}}(2\omega,\boldsymbol{k}'')\exp(-\mathrm{i}2\omega t+\mathrm{i}\boldsymbol{k}''\boldsymbol{r})+\mathrm{c.c.},$$

$$\boldsymbol{E}^{\mathrm{s}}(2\omega,\boldsymbol{k}'') = (0,E_y^{\mathrm{s}}(2\omega,\boldsymbol{k}''),0), \quad z>0, \qquad (59)$$

$$E_y^{\mathrm{s}}(2\omega,\boldsymbol{k}'') = \Phi_{\mathrm{s}}(2\omega,k_x,\varkappa(\omega,k_x)+\varkappa(\omega,0))F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{las}})$$

$$\times F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{p}})\chi_{yxy}(2\omega,\boldsymbol{k}_{\mathrm{p}},\boldsymbol{k}_{\mathrm{las}})E_{\mathrm{las}}E_{\mathrm{p}}\cos\phi. \qquad (60)$$

As before, inequality (54) is assumed fulfilled. Then, using explicit expressions for F_{s} (23), χ_{yxy} (32), and Φ_{s} (42), we find the Poynting vector of the s-polarised second harmonic

$$\boldsymbol{S}^{\mathrm{s}}(2\omega,\boldsymbol{k}'') = \frac{128\pi k^6 e^2 v_0^2\omega_{\mathrm{pl}}^4\sin^2\theta\cos^2\theta\cos^2\phi}{m^2c^5(\omega^2+\nu_0^2)^2(4\omega^2+\nu_0^2)}$$

$$\times\boldsymbol{n}''I_{\mathrm{las}}I_{\mathrm{p}}\left\{\left[k\sqrt{4-\sin^2\theta}+\varkappa_2(2\omega,k\sin\theta)\right]^2\right.$$

$$\left.+\varkappa_1^2(2\omega,k\sin\theta)\right\}^{-1}\{[\varkappa_1(\omega,0)+\varkappa_1(\omega,k\sin\theta)$$

$$+\varkappa_1(2\omega,k\sin\theta)]^2+[\varkappa_2(\omega,0)+\varkappa_2(\omega,k\sin\theta)$$

$$+\varkappa_2(2\omega,k\sin\theta)]^2\}^{-1}\{[k\cos\theta+\varkappa_2(\omega,k\sin\theta)]^2$$

$$+\varkappa_1^2(\omega,k\sin\theta)\}^{-1}\{[k+\varkappa_2(\omega,0)]^2+\varkappa_1^2(\omega,0)\}^{-1}. \qquad (61)$$

According to (61), the s-polarised harmonic is emitted in the direction of the vector $\boldsymbol{n}''$, i.e., at an angle θ'', which is closer to the normal vector than θ – the angle of the fundamental s-polarised wave reflection. The flux density of the s-polarised harmonic (61) is proportional to the square of the electron collision frequency. Within the framework of the proposed description of the nonlinear response of the metal without regard for the electron collisions, the effect of the ss-prohibition lifting is absent. This SHG property points at the new possibility of measuring the effective electron collision frequencies.

7. Effect of electron heating on harmonic generation

Electron collisions lead to electromagnetic field absorption. According to (9), (12), (13), the density of the power absorbed per unit volume is

$$Q = \frac{1}{4}\,en\boldsymbol{u}_1^*\boldsymbol{E}_1+\mathrm{c.c.} = \frac{\omega_{\mathrm{pl}}^2}{8\pi}\,\frac{v_0}{\omega^2+\nu_0^2}|E_1|^2. \qquad (62)$$

When two fields of type (24) interact, the square of the electric field moduli has the form

$$|E_1|^2 = |E_{\mathrm{las}}|^2|F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{las}})|^2\exp[-2\varkappa_1(\omega,k\sin\theta)z]$$

$$+|E_{\mathrm{p}}|^2|F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{p}})|^2\exp[-2\varkappa_1(\omega,0)z]+(\boldsymbol{E}_{\mathrm{las}}\boldsymbol{E}_{\mathrm{p}}^*)F_{\mathrm{s}}(\omega,\boldsymbol{k})$$

$$\times F_{\mathrm{s}}^*(\omega,\boldsymbol{k}_{\mathrm{p}})\exp[\mathrm{i}kx\sin\theta-\varkappa(\omega,k\sin\theta)z-\varkappa^*(\omega,0)z]$$

$$+(\boldsymbol{E}_{\mathrm{las}}^*\boldsymbol{E}_{\mathrm{p}})F_{\mathrm{s}}^*(\omega,\boldsymbol{k}_{\mathrm{las}})F_{\mathrm{s}}(\omega,\boldsymbol{k}_{\mathrm{p}})$$

$$\times\exp[-\mathrm{i}kx\sin\theta-\varkappa^*(\omega,k\sin\theta)z-\varkappa(\omega,0)z]. \qquad (63)$$

According to (62), (63), simultaneous absorption of the fields of type (24) results in electron heating inhomogeneous along the metal surface, with the period $2\pi/k\sin\theta$ along the x axis. This heating leads to the inhomogeneity along the surface of the effective dielectric constant of the metal. Due to this there appears the necessity to construct a modified theory of field penetration into a metal and, as a result, to revise the heating properties. To avoid this complication of the theory, we will restrict our consideration to the discussion of the heating properties due to the absorption of the wave incident normally to the metal surface and will neglect the absorption of the s-polarised wave, assuming the

inequality $|\boldsymbol{E}(\boldsymbol{k}_{las})| \ll |\boldsymbol{E}(\boldsymbol{k}_p)|$ to be fulfilled. Note that this restriction is not required if $(\boldsymbol{E}^*_{las}\boldsymbol{E}_p) = 0$.

Taking the above-said into account in considering the electron heating we have

$$Q \simeq \frac{1}{c}\frac{v_0\omega_{pl}^2}{\omega^2+v_0^2}\frac{4k^2}{[k+\varkappa_2(\omega,0)]^2+\varkappa_1^2(\omega,0)}$$

$$\times I_p \exp[-2\varkappa_1(\omega,0)z]. \quad (64)$$

Relations (2), (6), (64) and expressions (5), (7) make it possible to consider the change in the electron and lattice temperatures due to absorption of a normally incident laser pulse. We assume that the flux density of the laser pulse I_p heating the electrons changes in time according to the law $I_p(t) = I_p \exp(-t^2/\tau_{pulse}^2)$, where the time τ_{pulse} characterising the pulse duration is much larger than the period $2\pi/\omega$ corresponding to the fundamental frequency of the pulse. Consider the numerical solution of expressions (5), (7) when the pulse interacts with a gold target. For gold $\varepsilon_F \simeq 5.5$ eV, $n_0 \simeq N \simeq 5.9\times10^{22}$ cm^{-3}, $\omega_{pl} \simeq 1.4\times10^{16}$ s^{-1}, $G = 3.5\times10^{10}$ W K^{-1} cm^{-3}, $C_{lat} \simeq 2.4\times10^7$ erg K^{-1}. The initial electron and lattice temperatures are assumed identical: $T_0 = T_{lat} = 300$ K. In this case, $\nu_{eph} \simeq 0.93\times10^{14}$ s^{-1} and $\nu_{eph,\lambda} \simeq 3.7\times10^{13}$ s^{-1}. The laser pulse parameters are as follows: $\omega = 1.5\times10^{15}$ s^{-1}, $\tau_{pulse} \simeq 60$ fs, $I_p \simeq 5\times10^{12}$ W cm^{-2}. These values are typical of the experiment and realised when using a Cr:forsterite laser. Note that with this frequency ω, we can neglect the contribution of interband transitions to the dielectric constant of gold and use simple relation (15). The numerical solution of equations (5), (7) corresponding to the mentioned parameters of gold and the heating pulse is presented in Figs 1 and 2, respectively. The explicit form of the solution depends on the parameters a and b determining the electron–electron collision frequencies. Figure 1 presents the time dependence of the electron temperature $T_0(z=0,t)$ on the metal surface. The solid curve in Fig. 1 corresponds to $a=1$, $b=2$, while the dashed curve – to $a=0.5$, $b=1$. The larger the parameter a, the stronger the electron heating. An increase in b leads to a decrease in the heat conductivity and to a slower cooling of the electrons in the skin-layer. The function $T_0(z=0,t)$ first increases, achieves a maximum and then monotonically decreases. According to Fig. 2, the lattice temperature on the metal surface, $T_{lat}(z=0,t)$ monotonically increases at short times. The two curves in Fig. 2 correspond to the same values of the parameters a and b as in Fig. 1. For the given laser pulse

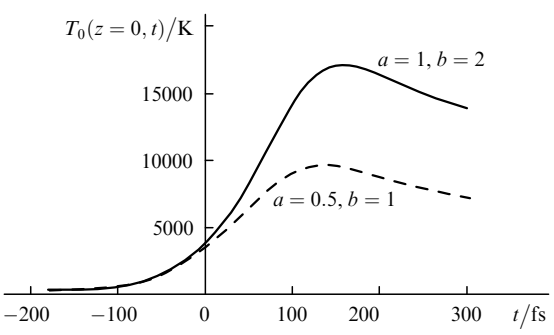

Figure 1. Time dependences of the electron temperature on the gold surface.

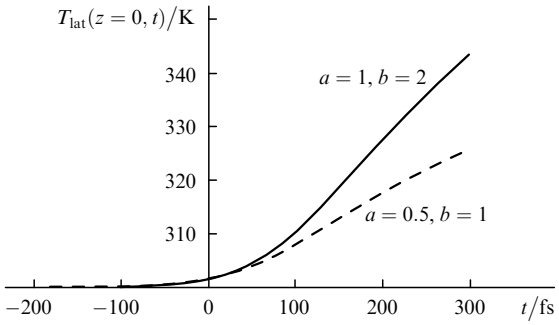

Figure 2. Time dependences of the lattice temperature on the gold surface.

parameters, $T_{lat}(z=0,t)$ increases only by 10 % – 15 %, which does not lead to deformation of the crystal lattice during the pulse action.

The temporal variation of the electron and lattice temperatures is accompanied by a change in the electron collision frequency ν. In the course of ν evolution, the harmonic generation efficiency changes. We will describe these changes by introducing the functions $H_{s,p}(2\omega,\boldsymbol{k})$:

$$\begin{aligned}\boldsymbol{S}^p(2\omega,\boldsymbol{k}') &= \boldsymbol{n}' I_{las}^2 H_p(2\omega,\boldsymbol{k}'),\\ \boldsymbol{S}^p(2\omega,\boldsymbol{k}'') &= \boldsymbol{n}'' I_{las} I_p H_p(2\omega,\boldsymbol{k}''),\end{aligned} \quad (65)$$

$$\boldsymbol{S}^s(2\omega,\boldsymbol{k}'') = \boldsymbol{n}'' I_{las} I_p H_s(2\omega,\boldsymbol{k}''). \quad (66)$$

Figure 3 presents the time dependences of the functions $H_p(2\omega,\boldsymbol{k}')/H_{p0}(2\omega,\boldsymbol{k}')-1$ and $H_p(2\omega,\boldsymbol{k}'')/H_{p0}(2\omega,\boldsymbol{k}'')-1$ demonstrating the change in the generation efficiency of p-polarised harmonics described by expressions (55) and (58). The quantities $H_{p0}(2\omega,\boldsymbol{k}')$ и $H_{p0}(2\omega,\boldsymbol{k}'')$ correspond to the values of the functions at $T_0 = T_{lat} = 300$ K. The curves were plotted for the case, when the angle of incidence of the s-polarised wave, θ, is equal to $\pi/4$. The angle ϕ is also assumed equal to $\pi/4$. According to Fig. 3 the electron and lattice heating is accompanied by a decrease in the harmonic generation efficiency. On the contrary, electron cooling leads to its increase. The relative minimum of the generation efficiency takes place at those times when the electron temperature is close to its maximum. As the temperature profiles in Figs 1 and 2, the shape of the curves in Fig. 3 noticeably depends on the parameters a and b. At $I_{las} \simeq 10^{12}$ W cm^{-2}, when the influence of the s-polarised wave on the electron heating can be neglected, the absolute

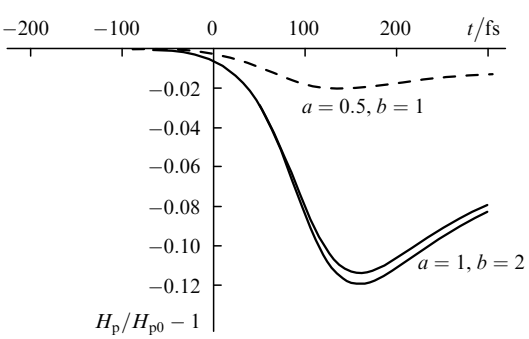

Figure 3. Temporal evolution of the relative generation efficiencies of p-polarised waves at the frequency 2ω.

values of the radiation flux densities (55) and (58) at the second harmonic frequency in the order of the quantity are equal to $\sim 10^2$ W cm^{-2}. Note that the difference in the curves (Fig. 3) corresponding to harmonic generation in the directions $\boldsymbol{k}'$ and $\boldsymbol{k}''$, is insignificant. At $a = 1$ and $b = 2$, this difference is no more than 0.5 %, and at $a = 0.5$ and $b = 1$ – no more than 0.1 % and the curves in Fig. 3 are indiscernible.

The generation efficiency of the s-polarised harmonic (61) emitted in the direction of the $\boldsymbol{n}''$ vector changes quite differently. The function $H_s(2\omega, \boldsymbol{k}'')$, according to Fig. 4, increases with increasing electron and lattice temperature and then decreases due to a relatively rapid cooling of the electrons. Because $H_s(2\omega, \boldsymbol{k}'')$ (61), (66) is proportional to the square of the collision frequency, its quantity changes significantly during the heating and the subsequent cooling of the electrons. In particular, in the conditions under study, the effect of the electron heating leads to a change in the flux density of the s-polarised harmonic from $\sim 0.5 \times 10^{-2}$ to $\sim 2 \times 10^{-1}$ W cm^{-2} at $a = 1$ and $b = 2$. Under the same conditions, as is seen from Fig. 3, the flux density of the p-polarised harmonic changes by several percent only. Such a sharp dependence of the flux density of the s-polarised harmonic on the electron heating makes this radiation a convenient tool in investigations of the hot electron kinetics.

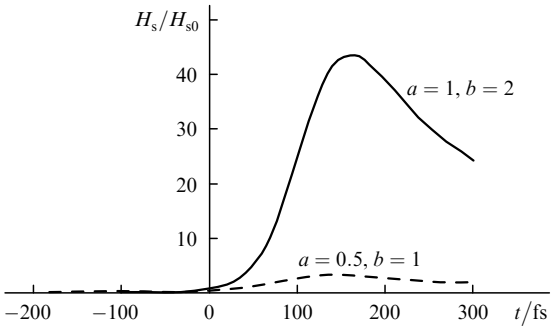

Figure 4. Temporal evolution of the relative generation efficiency of the s-polarised second harmonic.

8. Conclusions

The above-proposed comparatively simple theory of the second harmonic generation makes it possible to see the abundance of the outcomes of the nonlinear theory of optical properties of metals which appear upon wave mixing. Appearance of differently polarised harmonics propagating at various angles expands the possibilities of obtaining information on the properties of a nonequilibrium metal. In particular, investigation of the s-polarised harmonic generation could help to study experimentally the hot electron kinetics. At the same time, the further development of the theory of the nonlinear response of metals irradiated by several femtosecond pulses allows one to anticipate the prediction of new nonlinear optical effects.

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