

# Second harmonic and drag current generation by an s-polarised wave rapidly heating a metal

S.G. Bezhanov, S.A. Uryupin

**Abstract.** The dependences of the second harmonic and drag current generation efficiency on the electron collision frequency, changing during a rapid heating of electrons and the lattice in a metal in the case of absorption of s-polarised femtosecond radiation, are established.

**Keywords:** second harmonic generation, femtosecond pulse, drag current, nonlinear response of a metal.

## 1. Introduction

Generation of laser radiation harmonics by conduction electrons is an efficient tool in studying the electron properties of metals [1, 2]. Along with the widely used methods for determining the physical parameters of the electron by the measured absorption coefficient [3] or of polarisation characteristics of reflected radiation [4], investigation of the generation properties of the fundamental frequency harmonics makes it possible to obtain additional information on the collision frequencies of electrons. The possibilities of this approach for determining the electron–electron collision frequency with an umklapp process were demonstrated in paper [5], which gave a theoretical description of the third harmonic generation during the heating of electrons in a metal by a femtosecond laser pulse. To further demonstrate the possibilities of studying nonequilibrium states of metals by the optical nonlinear response, in this paper we present a rather simple model of the influence of electron collisions on the second harmonic and drag current generation efficiency, which are produced by an s-polarised wave heating the electrons.

When the influence of the electron collisions can be neglected, the second harmonic generation was studied both in metals [6, 7] and in plasma [8]. The authors of paper [9] pointed out the necessity to take into account the interband transitions in studying the second harmonic generation with a rather high frequency. In this case, the influence of electron collisions was assumed insignificant. This consideration is justified by the fact that the generation efficiency of

harmonics of visible radiation in pure metals at temperatures lower than the room temperature is virtually independent of the small frequency of electron collisions.

The situation changes when metals interact with femtosecond laser pulses heating the electrons. The electron heating during the time shorter or of the order of a hundred of femtoseconds, when the energy transfer from the electrons to the lattice is still small, leads to the establishment of a nonequilibrium state in which the electron temperature  $T$  is although lower than the Fermi energy but much higher the lattice temperature  $T_{\text{lat}}$ . In this case, we deal with a substantial increase in the frequency  $\nu_{\text{ee}}$  of electron–electron collisions, including those proceeding with an umklapp process. Already at the electron temperature exceeding several thousand degrees and room temperature of the lattice, the frequency  $\nu_{\text{ee}}$  is comparable with or higher than the frequency  $\nu_{\text{eph}}$  of electron–phonon collisions. If  $T$  is of the order of one electronvolt,  $\nu_{\text{ee}}$  is comparable with the visible radiation frequency  $\omega$ . Under these conditions, the effect of collisions on a weak nonlinear response of the metal becomes dominant and there arises a need in a theory adequately describing the nonlinear optical properties of a nonequilibrium metal.

Note that at such high collision frequencies in the visible frequency range, we can restrict our consideration to the study of the regimes of normal and high-frequency skin effects for typical metals. Below, taking into account the electron collisions, we derive basic relations for the second harmonic generation efficiency of the s-polarised wave and the drag current produced by this wave. Based on the equations for the electron and lattice temperatures, we show that as the metal is heated, the radiation generation efficiency at the frequency  $2\omega$  decreases. On the contrary, the electron cooling due to the heat release from the skin layer is accompanied by an increase in the second harmonic generation efficiency. Similar dependences are established for the drag current as a function of the changing electron and lattice temperatures. We demonstrate how the second harmonic generation efficiency depends on the constants to be determined, which characterise the influence of the umklapp processes to the frequency of electron–electron collisions determining both the heat conductivity and conductance of the metal.

## 2. Basic equations. The field at the fundamental frequency

Consider the interaction of the s-polarised electromagnetic wave with the metal occupying the half-space  $z > 0$ . The electric field of the wave incident on the metal has the form

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$$\mathbf{E}_L(\mathbf{r}, t) = \frac{1}{2} \mathbf{E}_L \exp(-i\omega t + ikx \sin \theta + ikz \cos \theta) + \text{c.c.} \quad (1)$$

where  $\mathbf{E}_L = \{0, E_L, 0\}$  is electric field strength weakly changing at a distance  $2\pi/k$  during the time  $2\pi/\omega$ ;  $\omega$  is the frequency;  $k = \omega/c$  is the wave number;  $c$  is the speed of light;  $\theta$  is the angle between the direction of the wave propagation and the vector of the normal to the metal surface. The magnetic field of the incident wave is  $\mathbf{B}_L = \mathbf{E}_L \{-\cos \theta, 0, \sin \theta\}$  and has the same dependence (1) on the time and coordinate.

To describe the metal response to the action of field (1), we will use the hydrodynamics equations for the electron concentration  $n$  and the velocity  $\mathbf{u}$ :

$$\frac{\partial n}{\partial t} + \text{div}(n\mathbf{u}) = 0, \quad (2)$$

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\nabla)\mathbf{u} = -\frac{1}{nm}\nabla p - \nu\mathbf{u} + \frac{e}{m}\left(\mathbf{E} + \frac{1}{c}[\mathbf{u}, \mathbf{B}]\right), \quad (3)$$

where  $\nu(n, T)$  is the characteristic collision frequency depending on the concentration and temperature  $T$  of the electrons;  $e$  and  $m$  are the electron charge and mass;  $p = p(n, T)$  is the electron pressure;  $\mathbf{E} = \mathbf{E}(\mathbf{r}, t)$  and  $\mathbf{B} = \mathbf{B}(\mathbf{r}, t)$  are the electric and magnetic fields in the metal. These fields are described by Maxwell's equations

$$\text{rot } \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \quad (4)$$

$$\text{rot } \mathbf{B} = \frac{\varepsilon}{c} \frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c} \text{enu}, \quad (5)$$

where  $\varepsilon$  is the static dielectric constant caused by the coupled electrons and the lattice. Taking into account a periodic change in the time of the incident wave (1), the solutions of equations (2)–(5) should be naturally sought for in the form

$$F = F_0 + \frac{1}{2} \sum_{s=1}^{\infty} [F_s \exp(-is\omega t) + \text{c.c.}], \quad (6)$$

where  $F$  denotes  $n$ ,  $T$  or one of the vector components  $\mathbf{u}$ ,  $\mathbf{E}$  or  $\mathbf{B}$ . In expression (6), the functions  $F_0$  and  $F_s$  depend on  $x$  and  $z$  and slowly change in time:

$$\left| \frac{\partial \ln F_0}{\partial t} \right|, \left| \frac{\partial \ln F_s}{\partial t} \right| \ll s\omega, \quad s = 1, 2, \dots \quad (7)$$

The approximate account for this time dependence of  $F_0$  and  $F_s$  allows one, in particular, to describe slow switching on and switching off of the field  $E_L$  caused by the finite duration of the laser pulse as well as to describe the metal parameters. We will restrict our consideration by the analysis of the approximate solution of equations (2)–(5) with the accuracy to corrections quadratic in the field strength  $E_L$  (1). In this approximation, it is sufficient to retain only the terms with  $s \leq 2$  in expansion (6). In this case, being interested in harmonics with  $s = 1$ , we can neglect the perturbations of the electron temperature and concentrations quadratic in the field and perturbations of the pressure  $p$  and frequency  $\nu$  caused by them. The influence of the quasi-stationary field  $E_0$ , which is proportional to  $E_L^2$ , is also insignificant.

In this approximation, we have a system of linear equations to determine the harmonics with  $s = 1$ . In the linear approximation, the field  $\mathbf{E}_1$  proportional to the external field  $\mathbf{E}_L$  can be represented in the form  $\mathbf{E}_1 = \mathbf{E}_1(z) \exp(ikx \sin \theta)$ . It follows that  $\text{div } \mathbf{E}_1 = 4\pi en_1 = 0$ , i.e.  $n_1 = 0$  as well as that  $p_1 = 0$  and  $\nu_1 = 0$ . As a result, neglecting the weak change in the velocity  $\mathbf{u}_1$ , we find from linearized equation (3) that

$$\mathbf{u}_1 = \frac{e}{m} \mathbf{E}_1 \frac{1}{\omega + i\nu_0}, \quad (8)$$

where  $\nu_0$  depends on  $n_0$  and  $T_0$  (the electron concentration and temperature slowly varying in time). Taking into account inequality (7) and relation (8), we obtain from (4) and (5) the equation for the function  $E_1(z)$ :

$$\frac{d^2 E_1(z)}{dz^2} + k^2 [\varepsilon(\omega) - \sin^2 \theta] E_1(z) = 0, \quad (9)$$

where  $\varepsilon(s\omega) = \varepsilon'(s\omega) + i\varepsilon''(s\omega)$  is the permittivity at the frequency  $s\omega$ ;  $s = 1, 2, \dots$ ;

$$\varepsilon'(s\omega) = \varepsilon - \frac{\omega_L^2}{(s\omega)^2 + \nu_0^2}; \quad (10)$$

$$\varepsilon''(s\omega) = \frac{\nu_0 \omega_L^2}{s\omega[(s\omega)^2 + \nu_0^2]}; \quad (11)$$

$\omega_L = (4\pi e^2 n_0 / m)^{1/2}$  is the plasma frequency of the electrons. At  $\omega_L^2 > (\omega^2 + \nu_0^2)(\varepsilon - \sin^2 \theta)$ , when  $\varepsilon'(\omega) < \sin^2 \theta$ , the solution of equation (9) decreasing inside the metal depth has the form

$$E_1(z) = E_1(0) \exp[-\varkappa(\omega)z], \quad (12)$$

where the quantity  $\varkappa(\omega)$  is described by the relations

$$\varkappa(s\omega) = \varkappa_1(s\omega) + i\varkappa_2(s\omega) \text{sgn}[\varepsilon'(s\omega) - \sin^2 \theta], \quad (13)$$

$$\varkappa_l(s\omega) = \frac{s\omega}{c\sqrt{2}} \left[ \sqrt{[\varepsilon'(s\omega) - \sin^2 \theta]^2 + [\varepsilon''(s\omega)]^2} - (-1)^l |\varepsilon'(s\omega) - \sin^2 \theta| \right]^{1/2}, \quad l = 1, 2. \quad (14)$$

The solution (12), (13) takes place if the change in  $\varepsilon(\omega)$  at distances  $\varkappa_l^{-1}(\omega)$  can be neglected (see details in [4]). In accordance with equation (4), the magnetic field in the metal has two components:  $B_x(x, z, t)$  and  $B_z(x, z, t)$ . For the function  $B_x(x, z, t)$  used below in the linear approximation in the field  $E_L$  taking into account (7), we obtain from (4) and (12) the expression

$$B_x(x, z, t) = \frac{\varkappa(\omega)}{2ik} E_1(z) \exp(-i\omega t + ikx \sin \theta) + \text{c.c.} \quad (15)$$

Radiation with the frequency  $\omega$  is partially reflected from the metal. In the approximation linear in  $E_L$ , the field of the reflected s-polarised wave has the form

$$\mathbf{E}_r(\mathbf{r}, t) = \frac{1}{2} \mathbf{E}_L R \exp(-i\omega t + ikx \sin \theta - ikz \cos \theta) + \text{c.c.} \quad (16)$$

where  $R$  is the complex reflection coefficient. For the strength vector of the magnetic field of the reflected wave, we have the expression  $RE_L\{\cos\theta, 0, \sin\theta\}$  from (4) and (16). Using relations (1), (12), (16) and their corresponding expressions for the magnetic field, from the condition of the continuity of tangential components of the electric and magnetic fields on the metal surface we find the relation of  $E_1(0)$  with  $E_L$  and the complex reflection coefficient:

$$E_1(0) = \frac{2k \cos \theta}{k \cos \theta + i\kappa(\omega)} E_L, \quad (17)$$

$$R = \frac{k \cos \theta - i\kappa(\omega)}{k \cos \theta + i\kappa(\omega)}. \quad (18)$$

Relations (6), (12), and (17) completely determine the field in the metal at the frequency  $\omega$ .

In deriving relations (17), (18) in accordance with inequality (7), we assumed that the change in the field  $E_L$  and metal parameters during the time  $2\pi/\omega$  is negligibly small. In this case, we made no assumptions about the relation between the characteristic time of  $E_L$  variation and the times of variations in the quantities determining the dielectric constant in the metal. The absence of the mentioned restrictions makes it possible to use relations (17), (18) for the description of the influence of laser pulses with the finite duration of the order of  $\tau_p \gg 2\pi/\omega$  on the metal, which leads to a weak change in the metal parameters during the time  $2\pi/\omega$ . The insignificance of the field switching on/off effects at  $\tau_p \gg 2\pi/\omega$  for relations (17), (18) was demonstrated earlier in paper [10].

### 3. Second harmonic generation

Consider now the field at the second harmonic frequency  $2\omega$ . Taking into account the smallness of the derivative in time,  $|\partial \ln u_2 / \partial t|$  [see (7)], we find from (3) and (6)

$$(v_0 - 2i\omega)\mathbf{u}_2 = \frac{e}{m}\mathbf{E}_2 + \frac{e}{2mc}[\mathbf{u}_1, \mathbf{B}_1] - \frac{1}{2}(\mathbf{u}_1 \nabla)\mathbf{u}_1 - \frac{1}{n_0 m} \nabla p_2. \quad (19)$$

In expression (19), the pressure perturbation is proportional to the perturbation of the electron concentration  $n_2$  and temperature  $T_2$ , which depend on  $E_1^2$  – the square of the field at the frequency  $\omega$ . The quantity  $E_1^2$  is associated with  $u_1^2$  by relations (8), (12), and (17). If we designate  $p_2$  by  $u_1^2$ , we can see that under the discussed conditions of the normal or high-frequency skin effects, the term containing  $\nabla p_2$  is small compared to the term depending on  $[\mathbf{u}_1, \mathbf{B}_1]$ . Then, neglecting  $\nabla p_2$  and using (8) and the relation between  $\mathbf{B}_1$  and  $\mathbf{E}_1$  given by equation (4), we obtain from (19)

$$\mathbf{u}_2 = \frac{ie}{m(2\omega + i\nu_0)}\mathbf{E}_2 + \frac{ie^2}{4m^2\omega(\omega + i\nu_0)(2\omega + i\nu_0)}\nabla E_1^2, \quad (20)$$

which takes into account that  $(\mathbf{E}_1 \nabla)\mathbf{E}_1 = 0$ . According to (20), the energy density gradient of the field at the fundamental frequency  $\nabla E_1^2$  leads to the electron motion along the axes  $x$  and  $z$ . Due to this, the electromagnetic field at the frequency  $2\omega$  has two components of the electric field  $\mathbf{E}_2 = \{E_{2x}, 0, E_{2z}\}$  and one components of the magnetic field  $\mathbf{B}_2 = \{0, B_{2y}, 0\}$ . This configuration corresponds to the

p-polarised field at the frequency  $2\omega$ . In this case,  $B_{2y} = B_{2y}(z) \exp(i2kx \sin \theta)$  can be determined from (4)–(6), (20) using the equation

$$\frac{d^2 B_{2y}(z)}{dz^2} + 4k^2[\varepsilon(2\omega) - \sin^2 \theta]B_{2y}(z) = 0. \quad (21)$$

If the condition

$$\omega_L^2 > (4\omega^2 + \nu_0^2)(\varepsilon - \sin^2 \theta) \quad (22)$$

is fulfilled, the solution of equation (21) decreasing at  $z \rightarrow \infty$  has the form

$$B_{2y}(z) = B_{2y}(0) \exp[-\kappa(2\omega)z], \quad (23)$$

where  $\kappa(2\omega) = \kappa_1(2\omega) - i\kappa_2(2\omega)$  is given by expressions (13), (14) at  $s = 2$ . In deriving expression (23), we took into account inequality (7). Note that according to (13),  $\kappa_1(2\omega) > \kappa_2(2\omega) > 0$ . The quantity  $\kappa_1^{-1}(2\omega)$  determines the penetration depth of the magnetic field of the second harmonic into the metal. Under condition (22), the solution of type (23) corresponds to the nonuniform electromagnetic wave. Its amplitude decreases proportionally to  $\exp[-\kappa_1(2\omega)z]$ , while the wave vector  $\{2k \sin \theta, 0, \kappa_2(2\omega)\}$  is directed at an angle to the metal surface. If the influence of the collisions is insignificant,  $\kappa_2(2\omega) = 0$  and the wave propagates along the surface. Below, taking into account inequality (7) and relation (20), we obtain from (5), (6), and (23) the electric field components in the metal:

$$E_{2x}(z) = \frac{i}{2k\varepsilon(2\omega)} \left[ \kappa(2\omega)B_{2y}(0) e^{-\kappa(2\omega)z} + \frac{\omega_L^2}{c^2} \frac{eE_1^2(0) \sin \theta}{2m(\omega + i\nu_0)(2\omega + i\nu_0)} e^{-2\kappa(\omega)z} \right], \quad (24)$$

$$E_{2z}(z) = -\frac{1}{4k^2\varepsilon(2\omega)} \left[ 4k^2 \sin \theta B_{2y}(0) e^{-\kappa(2\omega)z} + \frac{\omega_L^2}{c^2} \frac{e\kappa(\omega)E_1^2(0)}{m(\omega + i\nu_0)(2\omega + i\nu_0)} e^{-2\kappa(\omega)z} \right], \quad (25)$$

where  $E_1(0)$  is related to the fundamental wave field  $E_L$  by (17). Unlike the magnetic field (23), the dependence of the electric field on the coordinate  $z$  is described by two different functions. In this case, the vortex part of the electric field, as the magnetic field (23), is proportional to  $\exp[-\kappa(2\omega)z]$ , while its potential part is proportional to  $\exp[-2\kappa(\omega)z]$  [see (20), (24), (25)].

The electromagnetic field at the frequency  $2\omega$  is emitted from the metal surface. It obeys Maxwell's equations in vacuum and has the form

$$\mathbf{B}_{2r}(\mathbf{r}, t) = \frac{1}{2}\mathbf{B}_2^r \exp(-2i\omega t + 2ikx \sin \theta - 2ikz \cos \theta) + \text{c.c.}, \quad (26)$$

where  $\mathbf{B}_2^r = \{0, B_{2y}^r, 0\}$ . The magnetic field has the same dependence on the coordinate and time, while its strength is  $\mathbf{E}_2^r = -B_{2y}^r\{\cos \theta, 0, \sin \theta\}$ . The tangential components of the electric and magnetic fields are continuous on the metal surface:

$$B_{2y}(0) = B_{2y}^r, \quad E_{2x}(0) = E_{2x}^r = -B_{2y}^r \cos \theta. \quad (27)$$

From relations (24), (27), we find the electromagnetic field of the wave at the frequency  $2\omega$ :

$$B_{2y}^r = B_{2y}(0) = -\frac{k^2[k \cos \theta + i\kappa(\omega)]^{-2}}{2k\varepsilon(2\omega) \cos \theta + i\kappa(2\omega)} \\ \times \frac{2i\omega_L^2 e E_L^2}{mc^2(\omega + iv_0)(2\omega + iv_0)} \sin \theta \cos^2 \theta. \quad (28)$$

In deriving (28), we used the relation of  $E_1(0)$  with  $E_L$  (17). In accordance with relations (23)–(27), the magnetic field (28) completely determines the electromagnetic field in the metal and vacuum. Note that according to relations (25), (28), and  $E_{2z}^r = -B_{2y}^r \sin \theta$ , the electric flux density  $\varepsilon E_{2z}$  normal to the metal surface changes jump-wise at  $z = 0$ . In addition, according to (20), (25), and (28), the  $z$  component of the current density is  $j_{2z} = en_0 u_{2z} \neq 0$  at  $z = 0$ . The appearance of these nonphysical properties is the consequence of the use of expression (20) for  $u_{2z}$ , obtained by solving hydrodynamic equations (2), (3). However, these inaccuracies in the hydrodynamic description do not lead to the change in expression (28) derived using only the expression for the velocity  $u_{2x}$  (20), which follows from the rigorous kinetic consideration under the discussed conditions of normal and high-frequency skin effects, when corrections caused by the thermal motion of the electrons can be neglected.

According to relations (16) and (26), the waves reflected with the frequency  $\omega$  and generated with the frequency  $2\omega$  propagate in the same direction specified by the unit vector  $\mathbf{n} = \{\sin \theta, 0, -\cos \theta\}$ . The Poynting vector averaged over the period  $\pi/\omega$ , which describes radiation at the frequency  $2\omega$ , has the form  $\mathbf{S}(2\omega) = \mathbf{n}I(2\omega)$ , where  $I(2\omega) = (c/8\pi)|B_{2y}^r|^2$  is the radiation flux density. The ratio of  $I(2\omega)$  to  $I(\omega) = (c/8\pi)E_L^2$ , the flux density at the fundamental frequency, yields the second harmonic generation efficiency:  $\eta(2\omega) = I(2\omega)/I(\omega)$ . According to this definition of  $\eta(2\omega)$ , we find from (28)

$$\eta(2\omega) = \left(\frac{2eE_L}{mc\omega}\right)^2 \frac{\omega_L^4 k^6 \sin^2 \theta \cos^4 \theta}{(\omega^2 + v_0^2)(4\omega^2 + v_0^2)} \\ \times \{[k \cos \theta + \kappa_2(\omega)]^2 + \kappa_1^2(\omega)\}^{-2} \\ \times \{[2k\varepsilon'(2\omega) \cos \theta + \kappa_2(2\omega)]^2 \\ + [2k\varepsilon''(2\omega) \cos \theta + \kappa_1(2\omega)]^2\}^{-1}. \quad (29)$$

If the collisions are insignificant and  $v_0 = 0$ , the efficiency (29) is four times smaller than that presented in paper [6]. This difference is caused by the use of the unconventional definition of the field strength in [6]. At  $v_0 = 0$ , the results of paper [6] follow from the relations [8] determining the radiation field at the frequency  $2\omega$ .

At  $\omega \gg v_0$ , relation (29) allows generalisation to the case when it is necessary to take into account the interband transitions. According to paper [7], in (29) and expressions (13), (14) determining  $\kappa_i(s\omega)$ , it is needed to change  $\varepsilon(s\omega)$  by  $\varepsilon(s\omega) + \delta\varepsilon(s\omega)$ , where

$$\delta\varepsilon(s\omega) = -\frac{\omega_L^2}{nms^2\omega^2} \sum_{k,b,b'} \frac{\langle b\mathbf{k}|\mathbf{p}|b'\mathbf{k}\rangle \langle b'\mathbf{k}|\mathbf{p}|b\mathbf{k}\rangle}{(E_{b'\mathbf{k}} - E_{b\mathbf{k}} - \hbar s\omega - i\delta)} \\ \times [f_F(E_{b'\mathbf{k}}) - f_F(E_{b\mathbf{k}})]; \quad (30)$$

$|b\mathbf{k}\rangle$  is the Bloch function;  $\mathbf{p}$  is the momentum operator;  $k$  is the quasi-momentum;  $\hbar$  is Planck's constant;  $E_{b\mathbf{k}}$  is the electron energy in the band  $b$ ;  $f_F(E_{b\mathbf{k}})$  is the Fermi distribution;  $\delta > 0$  is a small correction determining the trip around the pole. In addition, it is necessary to multiply expression (29) by  $|\varepsilon(2\omega) + \delta\varepsilon(2\omega) - 1|[\varepsilon(2\omega) - 1]^{-1}|^2$ .

Let us discuss the peculiarities of second harmonic generation under conditions when the inequality inverse to (22) is fulfilled:

$$\omega_L^2 < (4\omega^2 + v_0^2)(\varepsilon - \sin^2 \theta), \quad (31)$$

but, as before,  $\omega_L^2 > (\omega^2 + v_0^2)(\varepsilon - \sin^2 \theta)$ . Under these conditions,  $\varepsilon'(2\omega) > \sin^2 \theta$  and the solution of equation (21) has the form [cf. (23)]

$$B_{2y} = B_{2y}(0) \exp[i\kappa_1(2\omega)z - \kappa_2(2\omega)z]. \quad (32)$$

In the absence of dissipation due to electron collisions,  $\kappa_2(2\omega) = 0$  and this solution corresponds to the wave with the frequency  $2\omega$  and the wave vector  $\{2k \sin \theta, 0, \kappa_1(2\omega)\}$  propagating inside the metal. Because of the dissipation caused by collisions, this wave decays at a distance  $\sim \kappa_2^{-1}(2\omega)$ , and its electric field (23) has two components described by expressions (24), (25), if we replace in them  $\kappa(2\omega)$  by  $-i\kappa(2\omega)$ . As before, this wave is partially emitted into vacuum. The field in the vacuum is found from the conditions of continuity of the tangential components (27). In this case, the magnetic field in vacuum and on the metal surface is described by expression (28) in which it is necessary to replace  $\kappa(2\omega)$  by  $-i\kappa(2\omega)$ . As in the case of lower frequencies [see (22)], under condition (31) radiation at the frequency  $2\omega$  propagates in vacuum along the vector  $\mathbf{n}$ , and its generation efficiency is given by the relation [cf. (29)]

$$\eta(2\omega) = \left(\frac{2eE_L}{mc\omega}\right)^2 \frac{\omega_L^4 k^6 \sin^2 \theta \cos^4 \theta}{(\omega^2 + v_0^2)(4\omega^2 + v_0^2)} \\ \times \{[k \cos \theta + \kappa_2(\omega)]^2 + \kappa_1^2(\omega)\}^{-2} \\ \times \{[2k\varepsilon'(2\omega) \cos \theta + \kappa_1(2\omega)]^2 \\ + [2k\varepsilon''(2\omega) \cos \theta + \kappa_2(2\omega)]^2\}^{-1}. \quad (33)$$

At  $\varepsilon'(2\omega) = \sin^2 \theta$ ,  $\kappa_1(2\omega) = \kappa_2(2\omega)$ , relations (29) and (33) coincide. If  $\varepsilon'(2\omega) > \sin^2 \theta$ , the denominators containing the functions  $\kappa_1(2\omega)$  and  $\kappa_2(2\omega)$  in expressions (29) and (33) differ by the quantity  $4k[\kappa_1(2\omega) - \kappa_2(2\omega)][\varepsilon'(2\omega) - \varepsilon''(2\omega)] \times \cos \theta$ . Because  $\kappa_1(2\omega) = \kappa_2(2\omega)$ , the denominator in (33) is greater if  $\kappa_1(2\omega) > \kappa_2(2\omega)$ , and, vice versa, the denominator is smaller if  $\varepsilon'(2\omega) < \varepsilon''(2\omega)$ . In particular, at a comparatively low dissipation due to collisions, when  $\varepsilon'(2\omega) > \varepsilon''(2\omega)$ , the comparison of expressions (33) and (29) allows one to make a conclusion about the relative decrease in the generation efficiency of radiation into vacuum at the frequency  $2\omega$ . The latter is caused by the fact that at  $\varepsilon'(2\omega) > \sin^2 \theta$ , the wave propagating inside the

metal carries away more energy of the fundamental wave than the wave localised at the surface at  $\varepsilon'(2\omega) < \sin^2\theta$  [see (22), (23)]. If  $\omega \gg v_0$ , similarly to (29), expression (33) also allows generalisation to the case when interband transitions are significant. To this end, according to paper [7], it is sufficient to introduce in (33) the same changes as in expression (29).

#### 4. Drag current

In the approximation quadratic in the field strength  $\mathbf{E}_L$  (1), along with the second harmonic generation there appears a direction motion of electrons with the velocity  $\mathbf{u}_0$ , slowly varying within  $2\pi/\omega$ . Using relations (6), (8), (12), (14), (17) and equations (3), (4), we obtain the equation describing the slow evolution of the velocity  $\mathbf{u}_0$ :

$$\begin{aligned} \frac{\partial \mathbf{u}_0}{\partial t} + v_0 \mathbf{u}_0 &= \frac{e}{m} \mathbf{E}_0 + \frac{2e^2 E_L^2}{m^2(\omega^2 + v_0^2)} \\ &\times \frac{k^2 \cos^2 \theta}{[k \cos \theta + \varkappa_2(\omega)]^2 + \varkappa_1^2(\omega)} \\ &\times \left\{ \left[ \varkappa_1(\omega) + \frac{v_0}{\omega} \varkappa_2(\omega) \right] \mathbf{e}_z + \left( k \frac{v_0}{\omega} \sin \theta \right) \mathbf{e}_x \right\} \\ &\times \exp[-2\varkappa_1(\omega)z], \end{aligned} \quad (34)$$

where  $\mathbf{e}_x$  and  $\mathbf{e}_z$  are the unit vectors along the axes  $x$  and  $z$ . After the time of the order of the inverse frequency of electron collisions ( $\sim v_0^{-1}$ ), the quasi-stationary velocity  $\mathbf{u}_0$  is established. The presence of the current along the normal to the metal surface would lead to charge accumulation. Therefore, the equality  $j_z = en_0 u_{0z} = 0$  should be fulfilled. The appearance of the  $z$  component of the quasi-stationary field

$$\begin{aligned} E_{0z} &= -\frac{2eE_L^2}{m(\omega^2 + v_0^2)} \frac{k^2 \cos^2 \theta [\varkappa_1(\omega) + \varkappa_2(\omega)v_0/\omega]}{[k \cos \theta + \varkappa_2(\omega)]^2 + \varkappa_1^2(\omega)} \\ &\times \exp[-2\varkappa_1(\omega)z] \end{aligned} \quad (35)$$

ensures the vanishing velocity  $u_{0z}$ . On the contrary, the current density  $\mathbf{j}_0 = en_0 u_{0x} \mathbf{e}_x$  along the metal surface is not zero. At  $v_0 t \gg 1$  from (34), we find  $u_{0x}$  and  $\mathbf{j}_0 = \{j_0, 0, 0\}$ , where  $j_0 = \sigma_0 E_{0x} + j_d$ :

$$\begin{aligned} j_d &= \frac{eE_L^2 k^2}{2\pi mc} \frac{\omega_L^2}{\omega^2 + v_0^2} \frac{\sin \theta \cos^2 \theta}{[k \cos \theta + \varkappa_2(\omega)]^2 + \varkappa_1^2(\omega)} \\ &\times \exp[-2\varkappa_1(\omega)z] \end{aligned} \quad (36)$$

is the drag current density;  $\sigma_0 = \omega_L^2/(4\pi v_0)$  is the conductivity. If  $E_{0x} = 0$ , then  $j_0 = j_d$ . The drag current (36) produces a quasi-stationary magnetic field, which is directed along the metal surface:  $\mathbf{B}_0 = \{0, B_0, 0\}$ , where

$$\begin{aligned} B_0(z) &= \frac{eE_L^2 k^2}{mc^2 \varkappa_1(\omega)} \frac{\omega_L^2}{\omega^2 + v_0^2} \\ &\times \frac{\sin \theta \cos^2 \theta}{[k \cos \theta + \varkappa_2(\omega)]^2 + \varkappa_1^2(\omega)} \exp[-2\varkappa_1(\omega)z]. \end{aligned} \quad (37)$$

Outside the metal, the magnetic field is uniform and equal to  $B_0(z=0)$ . The expressions for the drag current density and the quasi-stationary magnetic field strength have a very simple form at  $\omega_L \gg \sqrt{\varepsilon}\omega$  and  $\omega \gg v_0$ , when  $j_d \simeq [4eI/(mc^2)] \sin \theta \cos^2 \theta$ ,  $B_0(z=0) \simeq [8\pi eI/(\omega_L mc^2)] \sin \theta \cos^2 \theta$ , where  $I = cE_L^2/8\pi$  is the energy flux density of the incident wave. By assuming that  $E_{0x} = 0$ , we will estimate the drift motion velocity  $u_{0x}$  and the magnetic field strength for gold when  $n_0 \simeq 5.9 \times 10^{22} \text{ cm}^{-3}$ ,  $m \simeq 10^{-27} \text{ g}$ ,  $\omega_L \simeq 1.4 \times 10^{16} \text{ s}^{-1}$ . Then, at the radiation flux density  $I \simeq 10^{13} \text{ W cm}^{-1}$ , we find  $u_{0x} \simeq 8 \times 10^3 \text{ cm s}^{-1}$ ,  $B_0(z=0) \simeq 80 \text{ Gs}$ .

#### 5. Electron and lattice heating

The characteristic frequency of electron collisions  $\nu_0 = \nu_0(n_0, T_0)$  determining the field in the skin layer (12), (13), (17), the efficiency of the second harmonic generation (29), (33), and the drag current density (36) depends on their concentration  $n_0$  and temperature  $T_0$ . Usually, in normal metals, the collision frequency  $\nu_0$  is equal to the sum of frequencies of electron collisions with impurities ( $\nu_{ei}$ ), phonons ( $\nu_{eph}$ ), and with each other ( $\nu_{ee}$ ):  $\nu_0 = \nu_{ei} + \nu_{eph} + \nu_{ee}$ . We will neglect the weak dependence of  $\nu_{ei}$  on the electron temperature. At temperatures above the Debye temperature  $\Theta_D$ , the frequency of electron–phonon collisions depends on temperature  $T_0$ , which can be significantly higher than  $T_{lat}$  during the electron heating by a femtosecond laser pulse. Under the conditions of a strongly degenerate electron distribution [11, 12], we have

$$\nu_{ee} = a(k_B T_0)^2 / (\hbar \varepsilon_F), \quad (38)$$

where  $\varepsilon_F$  is the Fermi energy;  $a$  is the numerical factor depending on the type of the band structure of the metal. Relation (38) also takes place in the case when umklapp processes are significant [11]. In describing rapidly varying processes for which  $\hbar\omega \gtrsim 2\pi k_B T_0$ , one should take into account the increase in the frequency of electron–electron collisions by  $1 + [\hbar\omega/(2\pi k_B T_0)]^2$  times [13]. The term proportional to  $\omega^2$  enters additively altered expression (38) and in the approximation under study is independent of the electron temperature. This makes it possible to assume that the term containing  $\omega^2$  leads to an additive contribution to  $\nu_{ei}$ , and to use expression (38) for  $\nu_{ee}$  at high frequencies. For example, for gold with  $\varepsilon_F \simeq 5.5 \text{ eV}$  at  $\omega \simeq 1.5 \times 10^{15} \text{ s}^{-1}$ , the additive contribution to  $\nu_{ei}$  is  $\sim 10^{13} \text{ s}^{-1}$  if  $a \sim 1$ . Note that this contribution is an order of magnitude smaller than the frequency of the electron–phonon collisions  $\nu_{eph}$ , which is equal to  $\sim 0.93 \times 10^{14} \text{ s}^{-1}$  at room temperature of the lattice.

The electron collisions lead to dissipation of the high-frequency field in the skin layer. The absorption power is determined by the Joule heat  $Q$  released per unit time in a unit volume of the electron subsystem:

$$\begin{aligned} Q(z) &= \frac{\omega_L^2}{8\pi} \frac{v_0}{\omega^2 + v_0^2} |E_1|^2 = \frac{4}{c} I \frac{v_0 \omega_L^2}{\omega^2 + v_0^2} \\ &\times \frac{k^2 \cos^2 \theta \exp[-2\varkappa_1(\omega)z]}{[k \cos \theta + \varkappa_2(\omega)]^2 + \varkappa_1^2(\omega)}. \end{aligned} \quad (39)$$

Absorption of the field in the skin layer is the reason of a nonuniform electron heating. At the initial heating stage, the

spatial scale of the temperature nonuniformity is comparable with the skin layer dimensions, and, strictly speaking, the above-used expressions for the field at frequencies  $\omega$  and  $2\omega$  should be revised similarly to that in paper [4] for Fresnel formulae. At the same time, at the beginning of heating the difference between  $T_0$  and  $T_{\text{lat}}$  is not large and the nonuniform frequency  $\nu_{\text{ee}}$  is small compared to the virtually uniform frequency of electron–phonon collisions. Therefore, unless  $\nu_{\text{eph}} \gtrsim \nu_{\text{ee}}$ , the nonuniformity can be not taken into account approximately in the description of the fields in the skin layer. If  $\nu_{\text{ee}} \gtrsim \nu_{\text{eph}}$  but  $\nu_0 \simeq \nu_{\text{ee}} < \omega$ , expression (39) also yields a sufficient accuracy in the case when the frequency  $\nu_{\text{ee}}$  determining  $\nu_0$  changes in the depth of the skin-layer. This property of expression (39) is caused by the fact that the change in the effective skin-layer thickness during heating, proportional to  $\kappa_1^{-1}(\omega)$ , is comparatively small, if  $\nu_{\text{ee}}$  is two–three times smaller than  $\omega$ . Further, expression (39) is used under those conditions when the corrections quadratic in  $\nu_0/\omega \sim \nu_{\text{ee}}/\omega$  can be neglected in the expression for  $\varepsilon(\omega)$ . The electron cooling is caused by the heat release from the skin layer and the energy transfer in the lattice. The equation for the temperature taking into account the above processes has the form [14–16]

$$C_e \frac{\partial T_0}{\partial t} + \frac{\partial q}{\partial z} = Q(z) - G(T_0 - T_{\text{lat}}), \quad (40)$$

where  $C_e = \pi^2 k_B^2 n_0 T_0 / (2\varepsilon_F)$  is the heat capacity of electrons;  $G$  is the coupling constant of electrons with the lattice. Note that the use of the temperature equation is justified at times greater than the relaxation time of the electron energy, which is of the order of a picoseconds at room temperature. Therefore, at the initial heating stage, it is not applicable. However, at small times,  $\nu_{\text{ee}} < \nu_{\text{eph}}$  and the errors appearing when equation (40) is used, do not affect substantially the optical nonlinear properties of the metal. By the time when  $\nu_{\text{ee}} \gtrsim \nu_{\text{eph}}$ , the relaxation time of the electron energy is no more than 10 fs, and equation (40) is quite suitable to describe the next slower temperature evolution. The density of the heat  $q$  transferred by the electrons is proportional to the temperature gradient:

$$q = -\lambda \frac{\partial T_0}{\partial z}, \quad (41)$$

where the heat conductivity is  $\lambda = C_e v_F^2 / (3\nu_\lambda)$ , which depends on the total frequency  $\nu_\lambda = \nu_{\lambda\text{ei}} + \nu_{\lambda\text{eph}} + \nu_{\lambda\text{ee}}$  of electron collisions with the impurities ( $\nu_{\lambda\text{ei}}$ ), phonons ( $\nu_{\lambda\text{eph}}$ ), and with each other ( $\nu_{\lambda\text{ee}}$ ), and  $v_F$  is the Fermi velocity. The effective collision frequencies determining the heat conductivity differ from those, which are responsible for the high-frequency and quasi-static conductivity. Note that in the above described model of the metal conductivity and the second harmonic generation efficiency, the difference in the frequencies determining the high-frequency and quasi-static conductivities was neglected. If necessary, this difference can be taken into account by introducing into the theory additional parameters determining the distinction of characteristic frequencies from  $\nu_{\text{ei}}$ ,  $\nu_{\text{eph}}$  and  $\nu_{\text{ee}}$ , for example, at  $\omega < \nu_0$ . Returning to the discussion of the frequencies determining the heat conductivity, note that for  $\nu_{\lambda\text{eph}}$  and  $\nu_{\lambda\text{ee}}$ , we also deal with the same dependences on the lattice and electron temperatures, namely:  $\nu_{\lambda\text{eph}} \sim k_B T_{\text{lat}} / \hbar$  and  $\nu_{\lambda\text{ee}} = b(k_B T_0)^2 / (\hbar \varepsilon_F)$ , where

$b \neq a$ . The change in the lattice temperature is described by the equation [14–16]

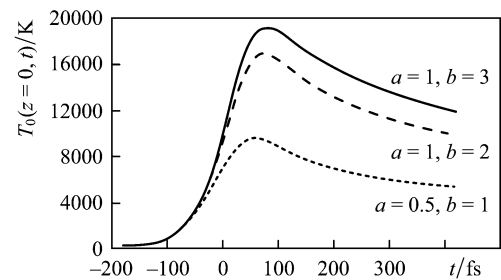
$$C_{\text{lat}} \frac{\partial T_{\text{lat}}}{\partial t} = G(T_0 - T_{\text{lat}}), \quad (42)$$

where  $C_{\text{lat}}$  is the heat capacity of the lattice for which at  $T_{\text{lat}} > \Theta_D$  the estimate  $C_{\text{lat}} \simeq 3k_B N$  is possible;  $N$  is the concentration of atoms in the lattice. The system of equations (40)–(42) allows us to study the influence of the comparatively low evolution of electron and lattice temperatures on the second harmonic generation efficiency.

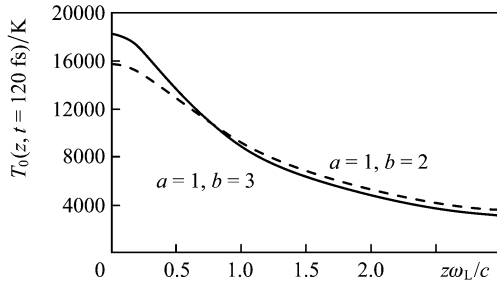
## 6. Numerical solution of temperature equations

Consider the properties of weakly nonlinear response of the metal to the effect of the femtosecond pulse heating the electrons. Let the density of the laser radiation flux change in time according to the law  $I(t) = I \exp(-t^2/\tau_p^2)$ , where the time  $\tau_p$  characterising the pulse duration is much greater than the period  $2\pi/\omega$  corresponding to the fundamental frequency of the pulse:  $\tau_p \gg 2\pi/\omega$ . Consider, as an example, the evolution of the electron and lattice temperatures and the evolution of the second harmonic generation efficiency caused by a change in  $T_0$  and  $T_{\text{lat}}$  in pure gold for which  $\varepsilon_F \simeq 5.5$  eV,  $n_0 \simeq N \simeq 5.9 \times 10^{22}$  cm $^{-3}$ ,  $\omega_L \simeq 1.4 \times 10^{16}$  s $^{-1}$ ,  $\nu_{\text{eph}} \simeq 0.93 \times 10^{14}$  s $^{-1}$ ,  $\nu_{\lambda\text{eph}} \simeq 3.7 \times 10^{13}$  c $^{-1}$ ,  $G = 3.5 \times 10^{10}$  W K $^{-1}$  cm $^{-3}$ ,  $C_{\text{lat}} \simeq 2.4 \times 10^7$  erg K $^{-1}$ , while the scattering of electrons on the impurities can be neglected. The initial temperatures of electrons and lattice are the same:  $T_0 = T_{\text{lat}} = 300$  K. The presented values of  $\nu_{\text{eph}}$  and  $\nu_{\lambda\text{eph}}$  correspond to this temperature  $T_{\text{lat}}$ . We used the following parameters of the laser pulse:  $\omega = 1.5 \times 10^{15}$  s $^{-1}$ ,  $\tau_p \simeq 60$  fs,  $I \simeq 10^{13}$  W cm $^{-2}$ . These values of  $\tau_p$  and  $I$  are typical of the experiment. Note that at these parameters of the pulse and metal, the characteristic electron velocity in the laser pulse field is small compared to their thermal velocity, which makes it possible to use the above-stated theory in which the effect of the alternating field is taken into account in the approximation quadratic in the field strength.

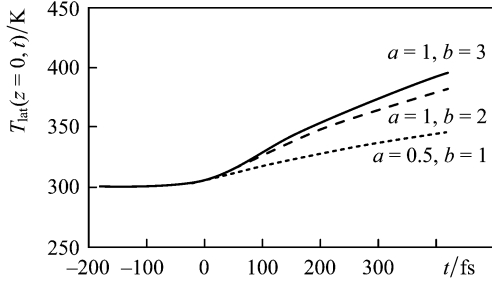
Figures 1–3 show the numerical solution of equations (40), (42) obtained for the mentioned parameters. Figure 1 presents the time dependences of the electron temperature  $T_0(z=0, t)$  on the metal surface. The function  $T_0(z=0, t)$  first increases, achieves a maximum, and then monotonically decreases. The shape of the function  $T_0(z=0, t)$  significantly depends on the parameters  $a$  and  $b$  determining the frequencies  $\nu_{\text{ee}}$  and  $\nu_{\lambda\text{ee}}$ , respectively. The larger  $a$ , the stronger the



**Figure 1.** Temporal changes in the electron temperature on the gold surface. Calculations are performed for a laser pulse with the frequency  $\omega = 1.5 \times 10^{15}$  s $^{-1}$ , the flux density  $10^{13}$  W cm $^{-2}$ , and the characteristic switching-on time  $\tau_p = 60$  fs.



**Figure 2.** Electron temperature profile at the instant  $t = 120$  fs.



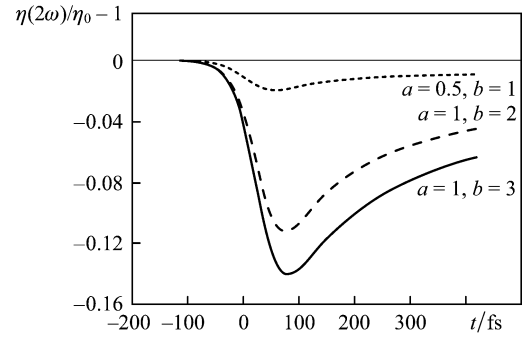
**Figure 3.** Time dependence of the lattice temperature on the gold surface. The calculation parameters are the same as in Figs 1 and 2.

electron heating. An increase in  $b$  leads to a decrease in the heat conductivity coefficient and to a decrease in the cooling rate of electrons in the skin layer. The latter is well seen in Fig. 2 presenting the temperature profile  $T_0(z, t)$  at the instant  $t = 120$  fs. At  $t = 120$  fs, the temperature  $T_0$  is close to the maximum values equal to  $\sim 2 \times 10^4$  K (see Fig. 1). In this case, it is nonuniform over the skin-layer thickness. However, the maximum difference of the skin-layer thickness  $\varkappa_1^{-1}(\omega)$  from  $c/\omega_L$  is no more than 10% during the entire action of the laser pulse and the nonuniformity  $T_0$  does not lead to significant changes in the field in the skin layer.

The behaviour of the lattice temperature on the metal surface  $T_{\text{lat}}(z=0, t)$  is shown in Fig.3 for the same parameters  $a$  and  $b$ . One can see that at small  $t$ , a monotonic increase in the lattice temperature takes place. At the selected parameters of the laser pulse,  $T_{\text{lat}}(z=0, t)$  increases only by 30%, which does not lead to a change in the crystal lattice during the pulse action.

## 7. Effect of heating on the second harmonic generation and drag current

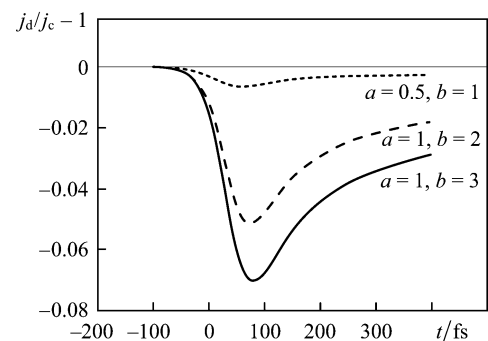
The temporal change in the electron and lattice temperatures in the skin layer is accompanied by a change in the collision frequencies of electrons with phonons and with each other. The efficiency of the second harmonic generation changes with varying  $v_0$ . Figure 4 presents the time dependence of the function  $\eta(2\omega)/\eta_0 - 1$ , where  $\eta_0$  is the radiation efficiency at  $\tau_p \rightarrow \infty$  for initial temperatures  $T_0(t \rightarrow -\infty)$  and  $T_{\text{lat}}(t \rightarrow -\infty)$ . One can see that the electron heating is accompanied by a decrease in the second harmonic generation efficiency, and during cooling, the function  $\eta(2\omega)/\eta_0 - 1$  monotonically increases. The minimal generation efficiency takes place at those instants when the electron temperature is close to the maximum. The shape of the curves in Fig. 4 substantially depends on the parameters  $a$  and  $b$ . This means that by measuring the radiation flux



**Figure 4.** Temporal evolution of the relative efficiency of the second harmonic generation for gold. The calculation parameters are the same as in Figs 1–3.

density at the frequency  $2\omega$  at different instant as well as by measuring the reflection coefficient at the fundamental frequency  $\omega$  (see details in [1, 3]), we can obtain additional information on the frequency of electron collisions. Note that in numerical calculations, the radiation frequency was assumed equal to the generation frequency of a Cr : forsterite laser. In this case, the photon energy of the second harmonic is  $2\hbar\omega \simeq 1.8$  eV, which is smaller than the band gap for gold, close to 2.5 eV. According to [17], for the mentioned energy and room temperature, the contribution to the dielectric constant from the interband transitions is comparatively small. If this condition is violated at high electron temperatures, the accuracy in determining the parameters  $a$  and  $b$  for gold can be increased by using radiation sources with somewhat lower frequencies. For example, an erbium femtosecond laser for which  $2\hbar\omega \simeq 1.4$  eV.

During the metal heating, the drag current density (36) also changes. When the radiation flux density changes over time, relation (36) for  $v_0 t \gg 1$  can be obtained if  $v_0 \tau_p \gg 1$  and temperatures of electrons and lattice vary slowly during the time  $\sim v_0^{-1}$ . The peculiarities of the evolution of the drag current density due to the metal heating is demonstrated in Fig. 5, which presents the functions  $\Delta J(t) = j_d/j_c - 1$  ( $j_c$  is the current density at  $\tau_p \rightarrow \infty$  and the metal temperature at the instant of the laser pulse action). One can see that during the metal heating, the function  $\Delta J(t)$  first decreases, achieved a minimum, and then monotonically increases up to zero. The relative decrease in the drag current is caused by an increase in the frequency of electron collisions. For the selected pulse parameters and the times under study, the change in  $\Delta J(t)$  is mainly caused by the frequency evolution



**Figure 5.** Relative drag current density on the gold surface as a function of time. The calculation parameters are the same as in Figs 1–3.

of the electron–electron collisions. The weak heating of the lattice is manifested at the final stage of laser pulse action and leads to an increase in the relaxation time of the drag current density to the initial value. The dip in the curve  $\Delta J(t)$  the deeper, the greater the electron heating. Therefore, the curve in Fig. 5 corresponding to the parameters  $a = 1$  and  $b = 3$  lies noticeably lower than the curve obtained at  $a = 0.5$  and  $b = 1$ . One can see from expression (36) and Fig. 5 that to generate large drag currents it is necessary to deal with pure metals and to minimise the heating of the electron and the lattice.

## 8. Conclusions

In this paper, we have presented the theory of second harmonic and drag current generation by a femtosecond pulse of s-polarised radiation heating the metal. The theory takes into account the possibility of a significant increase in the frequencies of electron–electron and electron–phonon collisions in the metal skin layer. We have demonstrated how important it is to consecutively describe the dynamics of the electron and lattice temperatures in order to obtain reliable values of the radiation intensity of the metal at the double frequency and the drag current. The theory developed can be used for interpreting and planning experiments on the interaction of femtosecond moderate-intensity laser pulses with metals and serves as a basis for obtaining information on the frequency of electron collisions. Under modern conditions, the utility of the stated theory consists in the fact that the number of the experiments studying a weakly nonlinear response of the metal with the electron temperature, greater than the lattice temperature, continue to increase.

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