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On the influence of electron heat transport on generation of the third harmonic of laser radiation in a dense plasma skin layer

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***Abstract*.** **The flux density is determined for radiation emitted by a plasma at the tripled frequency of an ultrashort laser pulse, which produces weak high-frequency modulations of the electron temperature in the plasma skin layer. It is shown that heat removal from the skin layer can reduce high-frequency temperature modulations and decrease the nonlinear plasma response. The optimum conditions for the third harmonic generation are found.**

Keywords: *ultrashort pulse, generation of harmonics, heat transfer, skin effect.*

The interaction of ultrashort laser pulses with solids results in the production of a high-density plasma on their surface, whose boundaries remain sharp during the pulse action (see, for example, [1 – 4]). Due to a high electron density, the conditions are quite simply realised under which the interaction of the pulse with plasma occurs in the normal skin-effect regime [2, 4]. It is known that absorption and reflection of radiation in the case of normal skin effect are substantially determined by collisions of electrons with ions. At the same time, electron – ion collisions lead to the generation of odd harmonics of the high-frequency field [5]. The third harmonic generation of the field in a dense plasma skin layer caused by electron – ion collisions was considered in papers [6, 7]. The description of plasma emission at the tripled laser frequency proposed in [6, 7] can be applied when the emission frequency ω exceeds the frequency of electron – electron collisions ν_{ee}.

In this paper, we present the theory of third harmonic generation, which can be used for $\nu_{ee} \gg \omega$. Due to frequent electron – electron collisions, the electron distribution function in the skin layer remains close to the Maxwell function, which allows us to describe the third harmonic generation by using the equation for the electron temperature. Because this equation takes into account heat removal from the skin layer, the approach used below describes the influence of heat transfer on the third harmonic generation by applying, as in [8, 9], a system of coupled equations for the field and electron temperature. In [8, 9], the numerical solution was obtained for these equations and the emission spectrum of the plasma was found under conditions of the rapid heating of electrons in the skin layer irradiated by a laser pulse.

Unlike [8, 9], we obtained below the approximate analytic solution of the equations for the field and temperature, which is valid for comparatively low laser radiation densities and short pulses, when the laser pulse causes weak perturbations of the electron temperature in the skin layer. This analytic solution gives the explicit dependences of the third harmonic generation efficiency on the plasma and laser pulse parameters. We found that heat removal from the skin layer reduces the amplitude of high-frequency modulations of the temperature perturbation, resulting in the decrease in the plasma emission intensity at the tripled laser frequency. This effect is especially strong when the electron mean free path exceeds 6 % of the ratio of the speed of light to the electron plasma frequency. For lower mean free paths, the third harmonic generation is comparatively efficient.

Consider the interaction of an ultrashort laser pulse with the dense plasma having a sharp boundary and occupying the region $z > 0$. The assumption about the sharp boundary of the plasma means that the plasma expansion during its preparation and emission can be neglected. When studying optical effects caused by electrons in the skin layer, the plasma expansion can be neglected at times shorter than the ratio of the skin-layer depth to the velocity of hydrodynamic expansion of the plasma, which is of the order of the sound speed. In particular, when the normal skin effect is realised, the solid state plasma expansion can be neglected at times shorter or of the order of 100 fs. We assume that the laser pulse has a sharp leading edge, which arrives to the plasma boundary at the instant $t = 0$. Then, the electric field strength of the incident pulse is

$$\boldsymbol{E}_{\rm i}(z,t) = \boldsymbol{E}_{\rm p}\sin(\omega t - kz)\eta(\omega t - kz), \quad z < 0, \tag{1}$$

where $\boldsymbol{E}_{\rm p} = (E_{\rm p}, 0, 0)$; ω is the frequency; k is the wave number; $\omega = kc$; c is the speed of light; and $\eta(x)$ is the Heaviside function. The step switching of the pulse described by the Heaviside function simplifies the consideration but is not necessary. The matter is that of most interest are the properties that are independent of the switching effect. In the case of step pulse switching, these properties are realised at times $t > 1/\omega$. It is these properties that are studied in our paper. At the same time, the method of switching on and off an ultrashort laser pulse considerably determines the spectrum of incident and reflected radiation. In experiments, laser pulses are usually switched on and off comparatively smoothly. For example,

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when the pulse switching is described by the function $\propto\exp[-(t/\tau_p)^2]$, where τ_p is the characteristic pulse duration, the effective spectral width of the pulse $\Delta\omega$ is $\sqrt{2}/\tau_p$. In this case, as shown in [8, 9], the influence of the pulse duration on the absorption coefficient is manifested only for $\tau_p \leqslant 1/\omega$, while the third harmonic in reflected radiation is distinctly observed against the background of the exponential tail of the broadened signal at the fundamental frequency.

For $t > 0$, the laser pulse is reflected from the surface $z = 0$ and partially penetrates into the plasma. The field of the reflected pulse has the form

$$\boldsymbol{E}_r(z,t) = \boldsymbol{E}_r(z+ct)\eta(z+ct), \quad z<0. \tag{2}$$

We assume that the pulse interacts with plasma under the conditions of normal skin effect, when the frequency ω is much lower than the electron–ion collision frequency ν and the mean free path of thermal electrons $l = v_T/\nu$ is much smaller than the skin-layer depth d (v_T is the thermal velocity of electrons). The evolution of the electron temperature $T = T(z,t)$ and the electric field strength $\boldsymbol{E}(z,t) = (E(z,t),0,0)$ in the case of normal skin effect in the plasma is described by a system of coupled equations [8, 9]

$$\frac{\partial^2}{\partial z^2}E(z,t) = \frac{4\pi}{c^2}\frac{\partial}{\partial t}[\sigma(z,t)E(z,t)], \tag{3}$$

$$\frac{3}{2}nk_B\frac{\partial}{\partial t}T(z,t) = \sigma(z,t)E^2(z,t) + \frac{\partial}{\partial z}\left[\lambda(z,t)\frac{\partial}{\partial z}T(z,t)\right], \tag{4}$$

where $\sigma(z,t) = [32/(2\pi)^{1/2}]e^2n/m\nu(z,t)$ is the plasma conductivity; e, m, and n are the electron charge, mass, and density, respectively; $\nu = \nu(z,t) = 4\pi Ze^4n\Lambda/\sqrt{m}\,[k_BT(z,t)]^{3/2}$; Z is the degree of ionisation of ions; k_B is the Boltzmann constant; Λ is the Coulomb logarithm; and $\lambda = \lambda(z,t) = [128/(2\pi)^{1/2}]\,nk_B^2T(z,t)/m\nu(z,t)$ is the Spitzer–Harm heat conductivity.

System of equations (3), (4) has the initial

$$E(z,t=0) = 0, \quad T(z,t=0) = T_0, \tag{5}$$

and boundary

$$E(z\to\infty,t) = 0, \quad T(z\to\infty,t) = T_0, \tag{6}$$

$$\frac{\partial}{\partial z}E(z,t)\bigg|_{z=0} = -2\frac{\omega}{c}E_p\cos\omega t, \quad \frac{\partial}{\partial z}T(z,t)\bigg|_{z=0} = 0, \tag{7}$$

conditions, where T_0 is the initial electron temperature. The boundary condition for $z = 0$ for the temperature derivative corresponds to the absence of a heat source on the plasma surface, while the boundary condition for the field derivative is the corollary of the continuity of the electric and magnetic fields for $z = 0$.

Absorption of the filed in the skin layer causes the heating of electrons. We assume that a change $\delta T = \delta T(z,t)$ in the initial temperature due to such heating is comparatively small:

$$\delta T \ll T_0. \tag{8}$$

Apart from condition (8), we also assume that the inequality

$$\left|\frac{\partial}{\partial t}\ln E\right| \gg \frac{3}{2}\left|\frac{\partial}{\partial t}\ln T\right| \simeq \frac{3}{2}\left|\frac{1}{T_0}\frac{\partial}{\partial t}\delta T\right| \tag{9}$$

is fulfilled, which means that the field in the skin layer changes faster than temperature. Then, the solution of the system of equations (3), (4) can be found by using the perturbation theory. In the first approximation, by neglecting a change in the electron temperature, we obtain from (3) a linear equation for the field in plasma, which satisfies boundary conditions (6), (7) and initial condition (5). The solution of the corresponding linear equation has the form

$$E_0(z,t) = 2\frac{d}{c}\omega E_p\int_0^{\omega t}\frac{d\tau}{(2\pi\tau)^{1/2}}\cos(\omega t-\tau) \times\exp\left(-\frac{z^2}{2d^2\tau}\right), \tag{10}$$

where

$$d = \frac{(2\pi)^{1/4}}{4}\frac{c}{\omega_L}\left(\frac{\nu_0}{\omega}\right)^{1/2}; \tag{11}$$

ν_0 is the electron–ion collision frequency at the temperature T_0; and ω_L is the Langmuir frequency. For $\omega t \gg 1$, we obtain approximately from (10)

$$E_0(z,t) \simeq \sqrt{2}\frac{d\omega}{c}E_p\exp\left(-\frac{z}{d}\right)\cos\left(\omega t - \frac{z}{d} - \frac{\pi}{4}\right). \tag{12}$$

It follows from (9) and (12) that solution (12) takes place if the electron temperature varies for the time exceeding $1/\omega$.

In turn, for a small temperature change, we obtain from (4) a linear equation of the type

$$\frac{3}{2}nk_B\frac{\partial}{\partial t}\delta T(z,t) - \lambda\frac{\partial^2}{\partial z^2}\delta T(z,t) = \sigma E_0^2(z,t), \tag{13}$$

$$\delta T(z,t=0) = 0, \quad \delta T(z\to\infty,t) = 0, \quad \frac{\partial}{\partial z}\delta T(z,t)\bigg|_{z=0} = 0, \tag{14}$$

where the conduction σ and the heat conductivity λ depend on temperature T_0. Equations (4) and (13) contain the electron density, which satisfies the equation of continuity $\partial n/\partial t + \mathrm{div}(\boldsymbol{j}/e) = 0$, where $\boldsymbol{j}$ is the current density. In the conditions under study, the current along the inhomogeneity direction is absent because the contribution to the current due to the temperature gradient, which is responsible for thermal diffusion, is compensated by the contribution to the current from an ambipolar electric field. The description of heat transfer within the framework of the Spitzer–Harm approach is based on this concept (see, for example, [10]). The nonzero component of the current density along the surface depends only on the coordinate z. As a result, we have $\mathrm{div}\boldsymbol{j} = 0$ or $\partial n/\partial t = 0$, which allows us to neglect the density perturbation. Note also that the relative perturbation of the electron density caused by high-frequency modulations of the pondermotive force in the case of the normal skin effect under study, which is neglected in our approach, is substantially weaker than the relative perturbation of the electron temperature due to their Joule heating described by Eqn (13). From (13) and (14), we find the temperature perturbation

$$\delta T(z,t) = \left(1+\frac{1}{\sqrt{3}}\right)\frac{2\omega}{nk_{\mathrm{B}}\pi\sqrt{\pi}}\int_0^t \mathrm{d}t'\int_0^\infty \frac{\mathrm{d}z'}{Ld[\omega(t-t')]^{1/2}}$$
$$\times\left[\frac{c}{2d\omega}E_0(z',t')\right]^2\left\{\exp\left[-\frac{3}{2}\left(2+\sqrt{3}\right)\frac{(z-z')^2}{L^2d^2\omega(t-t')}\right]\right.$$
$$\left.+\exp\left[-\frac{3}{2}\left(2+\sqrt{3}\right)\frac{(z+z')^2}{L^2d^2\omega(t-t')}\right]\right\}, \qquad (15)$$

where

$$L = \frac{64}{(2\pi)^{1/2}}\left(1+\sqrt{3}\right)\frac{\omega_{\mathrm{L}}}{c}\,l_0 \simeq 70 l_0\,\frac{\omega_{\mathrm{L}}}{c}; \qquad (16)$$

and $l_0 = (k_{\mathrm{B}}T_0/m)^{1/2}/\nu_0$ is the mean free path for $T = T_0$. By using the field distribution (12) at times much longer than $1/\omega$, we find from (15) the temperature perturbation for $z = 0$:

$$\frac{\delta T(z=0,t)}{T_0} = \frac{I}{cnk_{\mathrm{B}}T_0}\left\{\frac{8}{\sqrt{\pi}}\left(1+\frac{1}{\sqrt{3}}\right)\frac{\sqrt{\omega t}}{L}\right.$$
$$\times\int_0^\infty \frac{\mathrm{d}x}{x^2}\left[1-(1+x)\mathrm{e}^{-x}\right]\exp\left[-\left(\frac{3+\sqrt{3}}{4L}\right)^2\frac{x^2}{\omega t}\right]$$
$$\left.-\frac{4\cos(2\omega t)}{3+6L/\left(3+\sqrt{3}\right)}\right\},\quad \omega t \gg 1, \qquad (17)$$

where $I = cE_{\mathrm{p}}^2/8\pi$ is the incident radiation flux density. Relation (17) allows one to write conditions (8), (9) in the explicit form. For a weak heat removal, when $L \ll 1$, at times satisfying inequalities

$$1 \ll \omega t \ll \frac{\left(3+\sqrt{3}\right)^2}{16L^2}, \qquad (18)$$

the perturbation of the electron temperature is small if

$$\frac{8I}{3cnk_{\mathrm{B}}T_0}\,\omega t \ll 1. \qquad (19)$$

Because $\omega t \gg 1$, condition (9) automatically follows from inequality (19). Inequality (19) imposes restrictions both on the radiation flux density and the pulse duration.

When the current density is sufficiently small,

$$I \ll cnk_{\mathrm{B}}T_0L^2\,\frac{2}{\left(1+\sqrt{3}\right)^2}, \qquad (20)$$

the time interval can be increased. Indeed, if

$$\omega t \gg \frac{\left(3+\sqrt{3}\right)^2}{16L^2}, \qquad (21)$$

the temperature perturbation remains small until the inequality

$$\frac{8I}{\sqrt{\pi}cnk_{\mathrm{B}}T_0}\left(1+\frac{1}{\sqrt{3}}\right)\frac{\sqrt{\omega t}}{L} \ll 1 \qquad (22)$$

is satisfied. Because $\omega t \gg 1$, condition (22) provides the fulfilment of inequality (9).

For a comparatively intense heat removal, $L \gg 1$. In this case, the ratio $\delta T/T_0$ remains small if inequality (22) is satisfied, in which $\omega t \gg 1$. Inequality (9) is satisfied automatically in this case.

The temperature perturbation leads to a change in the electric field in the skin layer. For small corrections to the field, we obtain from (3) the linear equation

$$\frac{\partial}{\partial t}\,\delta E(z,t) - \frac{1}{2}\,\omega d^2\,\frac{\partial^2}{\partial z^2}\,\delta E(z,t)$$
$$= \frac{3}{2}\,E_0(z,t)\,\frac{\partial}{\partial t}\left[\frac{\delta T(z,t)}{T_0}\right] \qquad (23)$$

with the boundary and initial conditions

$$\delta E(z,t=0)=0,\ \delta E(z\to\infty,t)=0,\ \frac{\partial}{\partial z}\,\delta E(z,t)\bigg|_{z=0}=0. \qquad (24)$$

The field $E_0(z,t)$ in the right-hand side of Eqn (23) is described by relation (10), while the temperature perturbation $\delta T(z,t)$ is described by relation (15). We find from (23) and (24)

$$\delta E(z,t) = \int_0^t \frac{\mathrm{d}t'}{[2\pi\omega(t-t')]^{1/2}}\int_0^\infty \frac{\mathrm{d}z'}{d}\left\{\exp\left[-\frac{(z-z')^2}{2d^2\omega(t-t')}\right]\right.$$
$$\left.+\exp\left[-\frac{(z+z')^2}{2d^2\omega(t-t')}\right]\right\}E_0(z',t')\,\frac{\partial}{\partial t'}\left[\frac{3\delta T(z',t')}{2T_0}\right]. \qquad (25)$$

The field perturbation $\delta E(z=0,t)$ on the plasma boundary determines corrections to the reflected-signal field $\delta E_{\mathrm{r}}(z=0,t) = \delta E(z=0,t)$. Consider the field perturbation $\delta E(z=0,t)$ for $\omega t \gg 1$. At such times, we can calculate integrals over t' and z' in (25) by using the expressions for $E_0(z',t')$ and $\delta T(z',t')$ corresponding to the instants $\omega t' \gg 1$. In addition, by considering generation at the frequency 3ω, we retain in (15) only the terms that vary with the frequency 2ω for $\omega t \gg 1$. As a result, we find from (12), (15), and (25) the field strength at the frequency 3ω for $z = 0$:

$$\delta E(z=0,t) = -\frac{2\sqrt{2}}{3\left(1+\sqrt{3}\right)}\,\frac{d\omega}{c}\,E_{\mathrm{p}}\,\frac{I}{cnk_{\mathrm{B}}T_0}\,F(L)$$
$$\times\cos\left(3\omega t-\frac{\pi}{4}\right). \qquad (26)$$

Here, the function $F(L)$ describes the influence of heat removal from the skin layer on the third harmonic generation:

$$F(L) = \left[1+\frac{2L^2}{3\left(1+\sqrt{3}\right)(1+L)}\right]^{-1}. \qquad (27)$$

The electric and magnetic field strengths in vacuum at the frequency 3ω are the same, while the radiation flux density is $\boldsymbol{I}_{\mathrm{r}}(3\omega) = (0, 0, -I_{\mathrm{r}}(3\omega))$ and is independent of the coordinate:

$$I_{\mathrm{r}}(3\omega) = \int_0^{T(3\omega)} \frac{c\mathrm{d}t}{4\pi T(3\omega)}\,[\delta E_{\mathrm{r}}(z=0,t)]^2, \qquad (28)$$

where $T(3\omega) = 2\pi/3\omega$. In accordance with the definition $I_r(3\omega) = I\eta(3\omega)$, we find from relations (26)–(28) the third harmonic generation efficiency

$$\eta(3\omega) = \frac{4}{9(2+\sqrt{3})}\left(\frac{\omega d}{c}\right)^2\left(\frac{I}{cnk_B T_0}\right)^2 F^2(L). \qquad (29)$$

If the electron mean free path is small so that $L \ll 4$ or $l_0 \ll 0.06c/\omega_L$, then $F(L) \simeq 1$. In this case, the function $\eta(3\omega)$ (29) depends on the plasma and radiation parameters as the function $\eta_k(3\omega)$ obtained in [6] by using the approximate solution of the kinetic equation for the electron distribution function. In [6], the third harmonic generation was studied by assuming that electron–electron collisions weakly affect the electron motion in the high-frequency field of a laser pulse. Such an assumption is justified if the electron–electron collision frequency $\nu_{ee} = \nu/Z$ is lower than ω. On the contrary, if $\nu_{ee} \gg \omega$, frequent electron–electron collisions maintain the electron distribution close to the Maxwell distribution, and the description based on the use of equations for the electron temperature can be applied. Due to frequent electron–electron collisions, the third harmonic generation efficiency $\eta(3\omega)$ (29) proves to be lower by a factor of $(315\pi/512)^2 \simeq 3.7$ than the efficiency $\eta_k(3\omega)$ found within the framework of kinetic approach [6].

If $l_0 \gg 0.06c/\omega_L$, then $L \gg 4$, and we obtain approximately from (27)

$$F^2(L) \simeq \frac{9(2+\sqrt{3})}{2L^2} = \left(\frac{3\sqrt{\pi}c}{64\sqrt{2}l_0\omega_L}\right)^2 \simeq \left(\frac{0.06c}{l_0\omega_L}\right)^2 \ll 1. \qquad (30)$$

According to (30), for $l_0 \gg 0.06c/\omega_L$, the relative efficiency of third harmonic generation decreases, which is characterised by the coefficient $F^2(L) \ll 1$. The function $\eta(3\omega)$ (29) decreases due to heat removal from the skin layer, which results in a decrease in high-frequency modulations of the temperature perturbation producing generation at the frequency 3ω. One can see from (30) that the generation efficiency decreases with increasing mean free path. Recall, however, that expressions (29) and (30) were obtained assuming the classical heat transfer according to the Spitzer–Harm law. This assumption is justified if the mean free path is much smaller than the spatial scale of temperature perturbation inhomogeneities, which, under conditions under study, is comparable to the skin-layer thickness d (11), i.e., $l_0 \ll d = (\pi/128)^{1/4}(\nu_0/\omega)^{1/2}c/\omega_L \simeq 0.16(\nu_0/\omega)^{1/2}c/\omega_L$, where $\nu_0 \gg \omega$.

The mechanisms of the influence of heat transfer on the function $\eta(3\omega)$ (29) established above allow one to determine the optimum conditions for generation of the third harmonic of an ultrashort laser pulse. Let us present the example of such conditions. We assume for estimates that the plasma consists of fully ionised beryllium atoms with $Z = 4$ and electrons with the density $n = 5 \times 10^{23}$ cm^{-3} and has the temperature $T_0 = 100$ eV. In such plasma, $\nu_0 \sim 0.5\omega_L \simeq 2 \times 10^{16}$ s^{-1}, $l_0 \sim 3 \times 10^{-8}$ cm, $L \sim 3$, and $F(L) \sim 0.7$. We assume that the fundamental frequency ω of the laser pulse is 2×10^{15} s^{-1} and its duration is $\sim 2\pi/\omega \sim 3$ fs. Under these conditions, $\nu_0 \gg \omega$ and $d_0 \sim 10^{-6}$ cm $\gg l_0$. The electron temperature perturbation remains small during the pulse action if, according to inequality (22), the radiation flux density satisfies the condition $I/cnk_B T_0 \leqslant 0.15$, or $I \leqslant 4 \times 10^{16}$ W cm^{-2}. For the maximum possible flux density, we find from (29) that $\eta(3\omega) \sim 6 \times 10^{-6}$, which corresponds to the flux density at the frequency 3ω emitted from the plasma equal to $I_r(3\omega) \sim 2 \times 10^{11}$ W cm^{-2}.

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References

1. Price D.F., More R.M., Walling R.S., Guethlein G., Shepherd R.L., Stewart R.E., White W. E. *Phys. Rev. Lett.*, **75**, 252 (1995).
2. Losev L.L., Soskov V.I. *Kvantovaya Elektron.*, **25**, 467 (1998) [*Quantum Electron.*, **28**, 454 (1998)].
3. Schnurer M., Nolte R., Rousse A., Grillon G., Cheriaux G., Kalashnikov M.P., Nickles P.V., Sandner W. *Phys. Rev. E*, **61**, 4394 (2000).
4. Losev L.L., Soskov V.I. *Kvantovaya Elektron.*, **30**, 901 (2000) [*Quantum Electron.*, **30**, 901 (2000)].
5. Silin V.P. *Zh. Eksp. Teor. Fiz.* **47**, 2254 (1964).
6. Ferrante G., Zarcone M., Uryupin S.A. *Phys. Lett. A*, **315**, 378 (2003).
7. Ferrante G., Zarcone M., Uryupin S.A. *Phys. Rev. E*, **70**, 016403 (2004).
8. Isakov V.A., Kanavin A.P., Uryupin S.A. *J. Russ. Laser Res.*, **25**, 401 (2004).
9. Isakov V.A., Kanavin A.P., Uryupin S.A. *Laser Part. Beams*, **23** (3) (2005).
10. Lifshits E.M., Pitaevskii L.P. *Physical Kinetics* (New York: Pergamon Press, 1981; Moscow: Nauka, 1979).