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Interaction of femtosecond pulses of p-polarised radiation with a rapidly heated metal

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***Abstract*. The effects of rapid nonuniform heating of electrons on reflection of a p-polarised pulse from the metal are quantitatively described. Under conditions of normal and high-frequency skin effects, the absorption coefficient and phase shift of the reflected wave are calculated numerically upon irradiation of a gold target.**

Keywords: *femtosecond pulse, nonuniform heating of electrons, reflection coefficient.*

1. Introduction

In modern experiments on interaction of femtosecond laser pulses with metals, the conditions under which the lattice remains relatively cold during the pulse action and electrons are heated during the time shorter than the time of heat removal from the skin layer, are relatively simply realised (see, for example, [1 – 6]). Under these conditions, the time-varying electron temperature $T_e(z, t)$ is significantly nonuniform in the skin layer and significantly exceeds the lattice temperature $T_{lat}(z, t)$. The nonuniformity of the electron temperature leads to nonuniformities in the electron collision frequency $\nu(z, t)$ and dielectric constant $\varepsilon(z, t)$ of a metal. The dependence of ε on the coordinates necessitates the revision of the optical properties of a metal, the description of which is usually based on the use of the Fresnel formulas [7]. In the case when the electron collision frequency ν is small compared with the laser frequency ω, the nonuniform part of the dielectric constant is less homogeneous and the perturbation theory can be used to construct an analytical solution of reflection and penetration of the field in the metal. This approximate description of the absorption coefficient and phase shift of the reflected wave is given in [8, 9].

However, opposite conditions are possible in the experiment: the electron collision frequency ν is comparable to the radiation frequency and can be even higher. For example, in the optical frequency range it takes place at an electron temperature of ~ 1 eV, while in the IR frequency range – at a much lower temperature. In this paper we consider the optical properties of a metal whose electrons are rapidly heated to a temperature at which $\nu \gtrsim \omega$. We study the typical experimental conditions, when the metal is irradiated by two laser pulses at the same fundamental frequency. It is assumed that the heating pulse is normally incident on a metal surface, and a weaker probe p-polarised pulse propagates at an angle θ to the normal. The electron and lattice temperatures are determined from the system of equations, which takes into account the nonuniform heating of the metal when the heating pulse energy is absorbed in the skin layer. In this case, the field equation takes into account the dependence of the dielectric constant ε on the coordinates arising due to the nonuniform heating of the electrons and lattice. The influence of metal heating resulting from absorption of a weak probe p-polarised pulse in the metal, is neglected. The field produced by a p-polarised pulse in the metal is also determined from Maxwell's equations, which take into account nonuniformity of ε. Then, the absorption coefficient A_p and the phase shift ϕ_p of the reflected p-polarised wave are found.

The complex reflection coefficient is calculated numerically for a gold target. It is shown that when the target is irradiated by a Cr : forsterite laser, the method of analytic description proposed in [9] yields values of A_p close to the results of numerical calculations. In contrast, when exposed to a CO_2 laser, the established change in A_p with time cannot be described either by approximate analytical formulas [9] or Fresnel formulas. Similar results were obtained for the phase shift ϕ_p of the reflected wave. The account for the nonuniformity of ε in the skin layer in describing the reflection of pulsed p-polarised radiation is as important as the reflection of s-polarised radiation [10].

2. Metal in a heating field

To describe the response of a metal to the effect of laser radiation with a frequency ω under conditions of a high-frequency skin effect, the dielectric constant can be represented in the form

$$\varepsilon = \varepsilon_0 - \frac{\omega_p^2}{\omega(\omega + i\nu)}, \qquad (1)$$

where $\varepsilon_0 = \varepsilon_0' + i\varepsilon_0''$ is the contribution of bound electrons and the lattice; ω_p is the plasma frequency. Note that in the case of a high-frequency skin effect, the electron mean free path is small compared with the thickness of the skin layer. Such an approximation of the dielectric constant is also possible in a normal skin effect. However, in this case, an

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expression that defines the static conductivity of the metal should be used for ν. In pure normal metals, the electron collision frequency ν in formula (1) is equal to the sum of the frequencies of collisions between electrons and phonons and electron–electron collisions, accompanied by the umklapp processes. At lattice temperatures $T_{\rm lat}$, higher than the Debye temperature $\Theta_{\rm D}$, the electron–phonon collision frequency $\nu_{\rm ep}$ is proportional to $T_{\rm lat}$. When the thermal energy of electrons $k_{\rm B}T_{\rm e}$ is smaller than the Fermi energy $\varepsilon_{\rm F}$, the electron–electron collision frequency $\nu_{\rm ee}$ is a quadratic function of the electron temperature [11]. Thus,

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

$$T_{\rm lat} \gg \Theta_{\rm D}, \quad k_{\rm B}T_{\rm e} \ll \varepsilon_{\rm F},$$

where $k_{\rm B}$ is the Boltzmann constant; $\hbar$ is Planck's constant; a is a numerical coefficient depending on the band structure of the metal; and T_0 the initial temperature of the metal.

According to the above relations, dielectric constant (1) is nonuniform due to the nonuniformity of the collision frequency ν (2). Nonuniformity of ν results from nonuniformities of the lattice ($T_{\rm lat}$) and electron ($T_{\rm e}$) temperatures, arising due to the relatively rapid heating of electrons and the lattice upon absorption of a femtosecond laser pulse in the skin layer. For typical metals, the time of the energy transfer from electrons to the lattice is several picoseconds; therefore, the lattice temperature remains smaller than the electron temperature during the entire action of the femtosecond pulse. To describe such a nonequilibrium state, use is made of a system of equations for the temperatures of electrons and the lattice. The equation for the temperature of electrons takes into account their heating due to absorption in the skin layer of the field produced by the heating pulse, cooling due to the transfer of energy to the lattice and the change in their temperature due to heat transfer from the skin layer into the metal [12]:

$$C_{\rm e}\frac{\partial T_{\rm e}}{\partial t} - \frac{\partial}{\partial z}\left(\lambda\frac{\partial T_{\rm e}}{\partial z}\right) = \frac{\omega_{\rm p}^2}{8\pi}\frac{\nu}{\omega^2+\nu^2}\,|\boldsymbol{E}_{\rm h}|^2 - G(T_{\rm e}-T_{\rm lat}), \tag{3}$$

where $C_{\rm e} \equiv C_{\rm e}(z,t) = \pi^2 N k_{\rm B}^2 T_{\rm e}/(2\varepsilon_{\rm F})$ is the heat capacity of electrons with the concentration N; $\lambda \equiv \lambda(z,t) = C_e v_{\rm F}^2 \times [3\nu_\lambda(z,t)]^{-1}$ is the thermal conductivity coefficient; $\boldsymbol{E}_{\rm h} \equiv \boldsymbol{E}_{\rm h}(z,t)$ is the complex amplitude of the absorbed field strength; G is the coupling constant of electrons to the lattice; $v_{\rm F}$ is the Fermi velocity; $\nu_\lambda(z,t)$ is the electron collision frequency, which determines λ. Effective frequencies of electron–phonon and electron–electron collisions in ν_λ differ from those which determine the conductivity, but have the same temperature dependence:

$$\nu_\lambda = \nu_{{\rm ep}\lambda}(T_0)\frac{T_{\rm lat}}{T_0} + b\,\frac{k_{\rm B}^2 T_{\rm e}^2}{\hbar\varepsilon_{\rm F}}, \tag{4}$$

where $a \neq b$. Note that in a simplified expression for the absorbed power in equation (3), at $\nu < \omega$ we should use for ν a quantity, which determines the high-frequency conductivity, and at $\nu > \omega$ – the quantity, which determines the static conductivity. A change in the temperature of the lattice is described by the equation [13], which takes into account its heating during the energy transfer from electrons:

$$C_{\rm lat}\frac{\partial T_{\rm lat}}{\partial t} = G(T_{\rm e}-T_{\rm lat}), \tag{5}$$

where $C_{\rm lat}$ is the heat capacity of the lattice, for which the estimate $C_{\rm lat} \simeq 3k_{\rm B}N_{\rm a}$ is possible at $T_{\rm lat} > \Theta_{\rm D}$; $N_{\rm a}$ is the concentration of lattice atoms. The lattice temperature is small compared with the melting temperature. The field in the metal is found from Maxwell's equations, which take into account the nonuniformity of the dielectric constant of the metal (1). For the field amplitude $E_{\rm h}$ slowly varying in time $2\pi/\omega$, we have the equation

$$\frac{{\rm d}^2 E_{\rm h}}{{\rm d}z^2} + k^2\varepsilon E_{\rm h} = 0, \quad z > 0 \tag{6}$$

with the boundary conditions

$$\left(\frac{1}{{\rm i}k}\frac{{\rm d}E_{\rm h}}{{\rm d}z} + E_{\rm h}\right)\Big|_{z=0} = 2E_{\rm pump}(t), \quad E_{\rm h}(z\to\infty,t) = 0, \tag{7}$$

where $k = \omega/c$; c is the speed of light; $E_{\rm pump}(t)$ is the real field strength amplitude, which varies only slightly during the time $2\pi/\omega$ and is related to the flux density of the heating pulse $I_{\rm pump}(t) = (c/8\pi)E_{\rm pump}^2(t)$. Note that small derivatives $E_{\rm h}$ over time are omitted in (6) and (7). The equations presented in this section form the basis for describing the behaviour of characteristics of a nonequilibrium metal in a heating electromagnetic field. It is assumed that the electron energy distribution can be approximated by the Fermi distribution, but with a temperature higher than the lattice temperature, and the concept of the lattice temperature is meaningful. The influence of electron emission is neglected, assuming that the electron temperature is much smaller than the work function.

3. Interaction of p-polarised radiation pulses

The structure of the field produced by a probe laser pulse depends on its polarisation. Consider the interaction of a laser pulse of p-polarised probe electromagnetic radiation with the metal heated by the pump pulse and occupying the half-space $z > 0$.

The magnetic field of the p-polarised wave has the form

$$\frac{1}{2}\,\boldsymbol{B}_{\rm i}\exp(-{\rm i}\omega t + {\rm i}\boldsymbol{k}_{\rm i}\boldsymbol{r}) + {\rm c.c.}, \quad z < 0, \tag{8}$$

where $\boldsymbol{B}_{\rm i} = (0, B_{\rm i}, 0)$ changes weakly over time $2\pi/\omega$; $\boldsymbol{k}_{\rm i} = k(\sin\theta, 0, \cos\theta)$; θ is the angle between the vector $\boldsymbol{k}_{\rm i}$ and the axis z (Fig. 1). The wave (8) is reflected from the surface $z = 0$ and penetrates into the metal. The field of the wave reflected in the direction $\boldsymbol{k}_{\rm r}$ ($|\boldsymbol{k}_{\rm i}| = |\boldsymbol{k}_{\rm r}| = k$) has the form

$$\frac{1}{2}\,\boldsymbol{B}_{\rm r}\exp(-{\rm i}\omega t + {\rm i}kx\sin\theta - {\rm i}kz\cos\theta) + {\rm c.c.}, \quad z < 0, \tag{9}$$

where $\boldsymbol{B}_{\rm r} = R_{\rm p}\boldsymbol{B}_{\rm i}$; $R_{\rm p}$ is the complex reflection coefficient. The field in the metal is expressed as

$$\frac{1}{2}\,\boldsymbol{B}\exp(-{\rm i}\omega t + {\rm i}kx\sin\theta) + {\rm c.c}, \quad z > 0, \tag{10}$$

$$\frac{1}{2}\boldsymbol{E}\exp(-\mathrm{i}\omega t+\mathrm{i}kx\sin\theta)+\mathrm{c.c},\quad z>0, \tag{11}$$

where

$$\boldsymbol{B}=(0,B(z,t),0)\equiv(0,B,0);$$

$$\boldsymbol{E}=\left((\mathrm{i}k\varepsilon)^{-1}\mathrm{d}B/\mathrm{d}z,0,-B\sin\theta/\varepsilon\right).$$

Functions $\boldsymbol{E}$, $\boldsymbol{B}_\mathrm{r}$, $\boldsymbol{B}$ vary weakly over time $2\pi/\omega$, due to a change in $\boldsymbol{B}_\mathrm{i}$ and dielectric constant (1) that evolves during heating and cooling of electrons and the lattice. In accordance with Maxwell's equations, the distribution $B(z,t)$ in a metal is described by the equation

$$\frac{\mathrm{d}}{\mathrm{d}z}\left(\frac{1}{\varepsilon}\frac{\mathrm{d}B}{\mathrm{d}z}\right)+k^2\left(1-\frac{\sin^2\theta}{\varepsilon}\right)B=0,\quad z>0. \tag{12}$$

The electric and magnetic fields are continuous on the surface $z=0$:

$$B_\mathrm{i}\cos\theta(1-R_\mathrm{p})=\frac{1}{\mathrm{i}k\varepsilon}\frac{\mathrm{d}B}{\mathrm{d}z}\bigg|_{z=0}, \tag{13}$$

$$B_\mathrm{i}(1+R_\mathrm{p})=B(z=0,t). \tag{14}$$

From (13), (14) we find the boundary condition on the metal surface

$$\left(\frac{1}{\mathrm{i}k\varepsilon}\frac{\mathrm{d}B}{\mathrm{d}z}+B\cos\theta\right)\bigg|_{z=0}=2B_\mathrm{i}\cos\theta \tag{15}$$

and the complex reflection coefficient

$$R_\mathrm{p}=-1+\frac{B(z=0,t)}{B_\mathrm{i}}\equiv r_\mathrm{p}\exp(\mathrm{i}\phi_\mathrm{p}), \tag{16}$$

where r_p is the absolute quantity of the reflection coefficient; ϕ_p is the phase shift of the reflected wave. The quantity r_p is related to the absorption coefficient A_p: $A_\mathrm{p}=1-r_\mathrm{p}^2$.

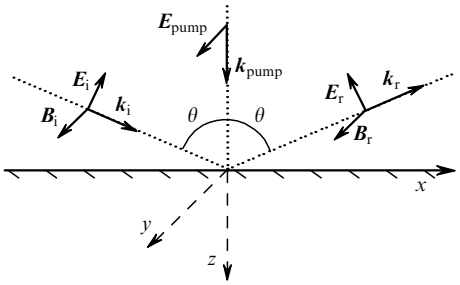

Figure 1. Scheme of interaction of pulses with a metal.

In equation (12) the dielectric constant varies with time due to electron heating by the heating pulse. Therefore, B and the complex reflection coefficient (16) also vary with time. To establish the time dependence of the dielectric constant ε, it is necessary to solve jointly the equations for the temperatures (3), (5) and for the field E_h (6) produced by the heating pulse. Next, using the function ε, we determine from equation (12) the field B, the absorption coefficient and phase shift of the reflected probe wave.

In the case of $\nu\ll\omega$, equation (12) admits an approximate analytic solution. In this case, with an accuracy up to terms of the second order of smallness in ν/ω, for the dielectric constant (1) we have

$$\varepsilon\simeq\varepsilon_1+\delta\varepsilon_1+\mathrm{i}\varepsilon_2, \tag{17}$$

where

$$\varepsilon_1=\varepsilon_0'-\frac{\omega_\mathrm{p}^2}{\omega^2};\quad \varepsilon_2=\varepsilon_0''+\frac{\nu\omega_\mathrm{p}^2}{\omega^3};\quad \delta\varepsilon_1=\frac{\nu^2\omega_\mathrm{p}^2}{\omega^4}. \tag{18}$$

In this approximation, following paper [9], we find from (12)–(16) the absorption coefficient

$$A_\mathrm{p}=\frac{2\cos\theta}{\sqrt{\sin^2\theta-\varepsilon_1}}\frac{2\sin^2\theta-\varepsilon_1}{\varepsilon_1^2\cos^2\theta-\varepsilon_1+\sin^2\theta}\langle\varepsilon_2\rangle \tag{19}$$

and the phase shift of the reflected wave

$$\begin{aligned}\tan\phi_\mathrm{p}=&\left(-\frac{2\varepsilon_1\cos\theta\sqrt{\sin^2\theta-\varepsilon_1}}{\varepsilon_1^2\cos^2\theta-\sin^2\theta+\varepsilon_1}\right)\\&\times\left\{1-\frac{\langle\varepsilon_2\rangle^2}{8\varepsilon_1^2}\left(\frac{2\sin^2\theta-\varepsilon_1}{\sin^2\theta-\varepsilon_1}\right)^2-\frac{\varepsilon_1^2\cos^2\theta+\sin^2\theta-\varepsilon_1}{\varepsilon_1^2\cos^2\theta-\sin^2\theta+\varepsilon_1}\right.\\&\times\left[\frac{\langle\delta\varepsilon_1\rangle}{2\varepsilon_1}\frac{2\sin^2\theta-\varepsilon_1}{\sin^2\theta-\varepsilon_1}+\frac{\langle\varepsilon_2^2\rangle}{2\varepsilon_1^2}\frac{\sin^2\theta}{\sin^2\theta-\varepsilon_1}\right.\\&\left.\left.-\frac{\langle\Xi_2^2\rangle}{4\varepsilon_1}\frac{2\sin^2\theta-\varepsilon_1}{(\sin^2\theta-\varepsilon_1)^2}\right]\right\}.\end{aligned} \tag{20}$$

In formulas (19), (20), we used the following notations:

$$\langle\varepsilon_2\rangle=\frac{2}{d}\int_0^\infty\mathrm{d}z\varepsilon_2(z)\exp\left(-\frac{2z}{d}\right), \tag{21}$$

$$\langle\delta\varepsilon_1\rangle=\frac{2}{d}\int_0^\infty\mathrm{d}z\delta\varepsilon_1(z)\exp\left(-\frac{2z}{d}\right), \tag{22}$$

$$\begin{aligned}\langle\Xi_2^2\rangle&=\frac{4}{d}\int_0^\infty\mathrm{d}z\exp\left(-\frac{2z}{d}\right)\Xi^2(z)\\&=\frac{4}{d^3}\int_0^\infty\mathrm{d}z\exp\left(-\frac{2z}{d}\right)\left[\int_0^z\mathrm{d}z'\varepsilon_2(z')\right]^2,\end{aligned} \tag{23}$$

$$\langle\varepsilon_2^2\rangle=\frac{2}{d}\int_0^\infty\mathrm{d}z\varepsilon_2^2(z)\exp\left(-\frac{2z}{d}\right), \tag{24}$$

where d is the effective depth of penetration of a p-polarised wave $[(kd)^{-2}=\sin^2\theta-\varepsilon_1>0]$.

Note that relation (20) follows from formula (42) [9], if in simplifying the latter for the function $b(z)$ from [9], use is made of expression (34) [9] in which a minus sign instead of a plus sign is in front of the third term containing derivatives $\delta\varepsilon_1'(z)$ and $\varepsilon_2'(z)$. Correction of the sign in formula (34) from paper [9] leads to a change in expression (50) from [9], obtained under the assumption that in equation (3) we can neglect heat removal from the skin layer and the transfer of energy from the electrons to the lattice. Under these conditions, when $\omega\gg\nu$ and $kd\ll1$, we have from (2), (3) and (7) [9]

$$\frac{v(z,t)}{v(t_0)} = \exp\left[\alpha \exp\left(-\frac{2z}{d}\right)\right],$$
$$\alpha \equiv \alpha(t) = \frac{16ad^2\omega_p^2}{\pi^2 N\hbar c^3}\int_{-\infty}^{t} dt' I(t'). \quad (25)$$

Taking into account relations (18) and (25), we find from (21)–(24)

$$\langle \varepsilon_2 \rangle = \varepsilon_2(T_0)\frac{1}{\alpha}(e^{\alpha}-1), \quad \langle \varepsilon_2^2 \rangle = \varepsilon_2^2(T_0)\frac{1}{2\alpha}(e^{2\alpha}-1),$$
$$\langle \delta\varepsilon_1 \rangle = \delta\varepsilon_1(T_0)\frac{1}{2\alpha}(e^{2\alpha}-1), \quad (26)$$
$$\langle \Xi_2^2 \rangle = \varepsilon_2^2(T_0)\frac{1}{\alpha}\int_0^{\alpha}\frac{dx}{x}(e^{2x}-e^{x}),$$

where $\varepsilon_2(T_0)$ and $\delta\varepsilon_1(T_0)$ are the values of ε_2 and $\delta\varepsilon_1$ at temperature T_0. Substituting expressions (26) into (20), we arrive at a modified formula (50) from [9]. In this case, Fig. 3 from paper [9] also changes. However, these quantitative changes do not affect the main statement in [9] about the inapplicability of Fresnel formulas under conditions of nonuniform heating of electrons in the skin layer.

4. Physical parameters of gold

To further describe the interaction of femtosecond pulses with a target made of gold, we will give data from some experiments and reference books. According to [14], the density N of conduction electrons with the effective mass $m = 0.91 \times 10^{-27}$ g [15] is 5.9×10^{22} cm^{-3}. In this case, the Fermi energy $F = 5.5$ eV, the Fermi velocity $v_F \simeq 1.4 \times 10^8$ cm s^{-1} [16], and the plasma frequency $\omega_p = 1.37 \times 10^{16}$ s^{-1}. At room temperature, $T_0 = 300$ K, the frequencies of collisions between electrons and phonons, $\nu_{ep}(T_0) \simeq 0.93 \times 10^{14}$ s^{-1} [15] and $\nu_{ep\lambda}(T_0) \simeq 3.7 \times 10^{13}$ s^{-1} [17], are significantly higher than the frequencies of electron–electron collisions. The heat capacity of the lattice is $C_{lat} \simeq 2.5 \times 10^7$ erg K^{-1} cm^{-3} [18], and the electron–lattice coupling constant is $G \simeq 3.5 \times 10^{10}$ W K^{-1} cm^{-3} [19].

According to [10], the calculations of the phase shift of the reflected wave and the absorption coefficient are very sensitive to the value $\varepsilon_0 = \varepsilon_0' + i\varepsilon_0''$. To determine ε_0' and ε_0'', we use the experimental data [15] obtained at $\omega \gg \nu$. It follows from relation (1) that

$$\varepsilon = \varepsilon_0' - \frac{\omega_p^2}{\omega^2+\nu^2} + i\left[\varepsilon_0'' + \frac{\nu\omega_p^2}{\omega(\omega^2+\nu^2)}\right]. \quad (27)$$

Paper [15] lists the values of the real and imaginary parts of the refractive index $n_T + ik_T = \sqrt{\varepsilon}$ measured for gold with 99.99 % purity at room temperature. For the real and imaginary parts of the dielectric constant, determined by the lattice and bound electrons, we have the expressions

$$\varepsilon_0' = n_T^2 - k_T^2 + \frac{\omega_p^2}{\omega^2+\nu^2}, \quad (28)$$
$$\varepsilon_0'' = 2n_T k_T - \frac{\nu\omega_p^2}{\omega(\omega^2+\nu^2)}. \quad (29)$$

The results of calculations by formulas (28) and (29) are shown in Fig. 2. It should be noted that in [15] the experimental data are presented in the wavelength range 0.8–8 μm. This allows for relatively accurate calculations under the irradiation of gold by a ~ 1.25-μm Cr : forsterite laser. For longer wavelengths the data are not available. However, at $\omega_p^2 \gg \omega^2 + \nu^2$ the contribution of ε_0 to the dielectric constant (1) is small. The latter makes it possible, in particular, to discuss the impact of IR radiation on gold, without sufficient information about the value of ε_0.

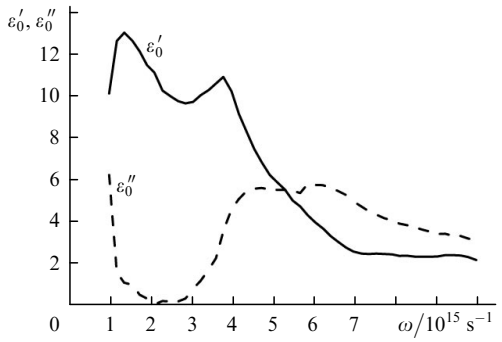

Figure 2. Frequency dependence of the real (ε_0') and imaginary (ε_0'') parts of the dielectric constant ε_0 of gold.

5. Results of numerical calculations

Let us dwell on the numerical solution of the equations for fields and temperatures when the metal is irradiated by the pump pulse whose radiation flux density varies with time according to the law $I_{pump}(t) = I_{pump}\exp(-t^2/t_p^2)$, and the parameter t_p is related to the pulse duration τ_p, as determined by FWHM of the function $I_{pump}(t)$, by the expression $\tau_p = 2t_p \ln 2$. Figures 3–5 shows results of the numerical solutions of equations (3), (5), (6) and (12) corresponding to the effect of such a pulse. The calculation was performed for the heating pulse of a Cr : forsterite laser with the frequency $\omega = 1.5 \times 10^{15}$ s^{-1}. The heating pulse parameters were as follows: $I_{pump} = 10^{13}$ W cm^{-2}, $t_p = 18$ fs ($\tau_p = 30$ fs). A probe pulse of the same laser is incident on the metal at an angle $\theta = \pi/4$ to the normal. For gold, according to Fig. 2, we have $\varepsilon_0' \simeq 11$, $\varepsilon_0'' \simeq 1.17$. In accordance with the results of paper [12], we have chosen the constants $a = 1$, $b = 2$. The initial temperatures of the lattice and electrons are considered equal: $T_e(z, t \to -\infty) = T_{lat}(z, t \to -\infty) = T_0 = 300$ K.

Figure 3 shows the collision frequency as well as the temperatures of electrons and the lattice on the metal surface as functions of time. The electron temperature increases to $\sim 1.5 \times 10^4$ K and then decreases monotonically. At the same time interval, the lattice temperature increases monotonically up to 330 K. In the course of the pulse action the electron collision frequency changes mainly due to changes in temperature. According to Fig. 3 the ratio ν/ω is still relatively small, but increases six-fold compared with the initial values. The resulting nonuniformity of the dielectric constant leads to differences in the absorption coefficient and phase shift of the reflected wave from their values A_{pF} and ϕ_{pF} calculated from the Fresnel formulas:

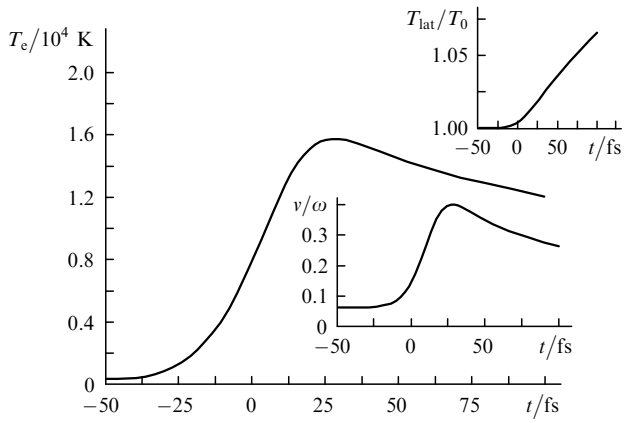

Figure 3. Time evolution of the electron temperature $T_e = T_e(z = 0, t)$ on the surface of a metal during its heating by radiation of a Cr : forsterite laser. The insets show the time dependences of the lattice temperature $T_{lat} = T_{lat}(z = 0, t)$ and the frequency of collisions between electrons $\nu = \nu(z = 0, t)$.

$$R_{pF} = -1 + \frac{2k\varepsilon\cos\theta}{k\varepsilon\cos\theta + i\kappa} \equiv r_{pF}\exp(i\phi_{pF}),$$
$$A_{pF} = 1 - r_{pF}^2, \tag{30}$$

where ε and $\kappa = \kappa_1 - i\kappa_2 = k\sqrt{\sin^2\theta - \varepsilon}$ are found using the values of the temperature on the metal surface. Because under these conditions the ratio ν/ω is small, the results of the calculation can be described using approximate relations (19)–(24).

Figure 4 shows the time dependences of the absorption coefficient, corresponding to the numerical solution of equations (3), (5), (6) and (12), to the calculation by Fresnel formulas (30) and expression (19). According to Fig. 4 the Fresnel formulas give a higher value of the absorption coefficient (at a maximum absorption – by 60 %). In contrast to [8, 9], expression (18) for ε_2 contains a small supplement ε_0'' to the imaginary part ε_0. At the initial stage of heating, when $\nu/\omega \simeq 0.06$, retention of ε_0'' improves the accuracy of the A_p calculation by 20 %. At a peak temperature $\nu/\omega \simeq 0.4$ and the contribution of ε_0'' is 3 %. One can see from Fig. 4 that the dotted curve corresponding to the calculation by formula (19) almost merges with the solid curve illustrating the results of a more accurate numerical calculation. Figure 4 also presents the calculations of the phase shift $\Delta\phi(t) = \phi_p(t) - \phi_p(t \to -\infty)$ of the reflected waves. One can see that formula (20) describes the phase shift with sufficient accuracy. It can be used for processing the experimental data, if $-\varepsilon_1 \gg \max[1, \langle\varepsilon_2\rangle, \langle\delta\varepsilon_1\rangle]$. At $-\varepsilon_1 \gg \langle\varepsilon_2\rangle$, expression (19) also yields good accuracy for the absorption coefficient. These inequalities are fulfilled if the radiation frequency ω is much smaller than the electron plasma frequency ω_p but significantly higher than the frequency of collisions. Note that in this frequency range, small changes in the phase shift are very sensitive to the accuracy of the quantities ε_0' and ε_0''. The latter should be considered when analysing the experimental data on the phase shift.

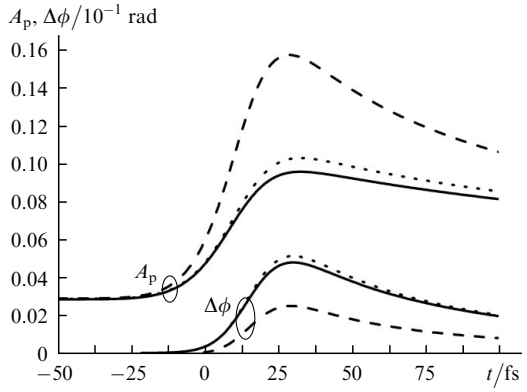

Figure 4. Time evolution of the absorption coefficient A_p and phase shift $\Delta\phi$ of the reflected wave upon irradiation of a gold target by pulses from a Cr : forsterite laser. Solid curves – the numerical solution of equations (3), (5), (6) and (12); dashed curves – the calculation by Fresnel formulas (30); dotted curves – the calculation by approximate formulas (19), (20).

The inequality $\nu \ll \omega$ may be violated. In particular, this is possible when a femtosecond pulse from a CO_2 laser with the frequency $\omega \simeq 1.8 \times 10^{14}$ s^{-1} interacts with a gold target, thereby heating the electrons. In the process of heating, there occurs a transition from the condition $\nu \ll \omega$ to the opposite one. At $\nu \ll \omega$ relations (17), (18) are valid for the dielectric constant, while at $\nu \gg \omega$ we have [cf. expression (1)]

$$\varepsilon \simeq \varepsilon_0 + i\frac{\omega_p^2}{\omega\nu_\lambda}. \tag{31}$$

In accordance with the Wiedemann–Franz law, when writing formulas (31) it is assumed that the conductivity and thermal conductivity depend on the same collision frequency ν_λ (4). Since in transition from small to large collision frequencies $\nu_\lambda \sim \nu \sim \omega$, then approximation for ε is possible

$$\varepsilon \simeq \varepsilon_0 - \frac{\omega_p^2}{\omega^2 + \nu\nu_\lambda} + i\nu\frac{\omega_p^2}{\omega(\omega^2 + \nu\nu_\lambda)}. \tag{32}$$

Using relation (32), the form of the first term in the right-hand side of equation (3) is modified. Now to describe the heating due to absorption of the alternating field it is necessary to use the expression

$$\frac{\omega_p^2}{8\pi}\frac{\nu}{\omega^2 + \nu\nu_\lambda}|\boldsymbol{E}_h|^2. \tag{33}$$

Relations (32) (33) introduce the largest error at $\omega^2 \sim \nu\nu_\lambda$. However, if the time interval in which $\omega^2 \sim \nu\nu_\lambda$ is much smaller than the laser pulse duration, the effect of this error on the dependences given below in the region $\nu_\lambda > \omega$ is largely weakened.

The collision frequencies ν (2) and ν_λ (4) entering formulas (32), (33) depend on the parameters a and b. This dependence manifests itself at electron temperatures exceeding several thousand kelvins, when electron–electron collisions make the main contribution to ν (2) and ν_λ (4). Because the parameter b is known with insufficient accuracy, in the case of electron heating and reflectance of CO_2 laser radiation, calculations were performed for several b, close to the value established in [12] in processing the experimental data of paper [4].

The results of calculations of the evolution of the temperatures, collision frequency, absorption coefficient and phase shift of the reflected waves are given below for the pulse duration $t_p = 60$ fs and flux density $I_{pump} = 6 \times 10^{12}$ W cm^{-2}. The characteristics of gold are the same as above. Figure 5 shows the time dependences of temperatures of electrons and the lattice. They are qualitatively

similar to those presented in Fig. 3. However, because the radiation frequency of a CO_2 laser is approximately eight times smaller than the radiation frequency of a Cr : forsterite laser, the ratio $\sqrt{\nu\nu_\lambda}/\omega$ in the process of electron heating reaches the values that are higher than unity (Fig. 5). For the maximum heating, $\sqrt{\nu\nu_\lambda}/\omega \simeq 4$. At $\sqrt{\nu\nu_\lambda} \gtrsim \omega$, approximate formulas (19), (20) are inapplicable. The Fresnel formulas are also inapplicable, because the medium is inhomogeneous. In this connection, Fig. 6 shows only the results of numerical calculations for $a = 1$ and $b = 1$, 1.5 and 2. The quantity a is defined in [12] from the data on radiation absorption in gold [4] with greater accuracy than b; therefore, the calculations are performed for $a = 1$. According to Fig. 6, the absorption coefficient the greater the larger the parameter b. The dependence of A_p on b manifests itself most strongly at relatively long times, when the electron temperature decreases due to heat removal from the skin layer. In contrast, at short times if b doubles, then A_p varies only slightly. In particular, the maximum values of A_p corresponding $b = 1$ and 2 differ only by 20 %. Relative changes in the phase $\Delta\phi(t)$ are more sensitive to changes in b. One can see from Fig. 6 that a two-fold increase in b is accompanied by an increase in $\Delta\phi$ in the maximum by about 2.5 times. Therefore, by measuring $\Delta\phi$, we can determine b with relatively good accuracy if reliable data on other quantities is available. The parameter a, as in [12], is easier to determine from the measurements of A_p. At the same time, the description of A_p and $\Delta\phi$ should be naturally based on the above description of the field structure in the skin layer.

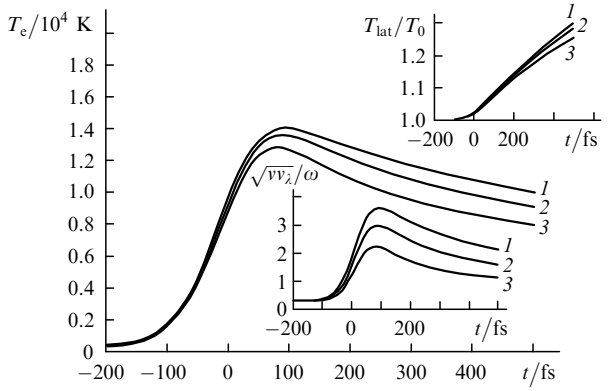

Figure 5. Time evolution of the electron temperature $T_e = T_e(z = 0, t)$ on the surface of a metal during its heating by radiation of a CO_2 laser for $b = 2$ (*1*), 1.5 (*2*) and 1 (*3*). The insets show the time dependences of the lattice temperature $T_{lat} = T_{lat}(z = 0, t)$ and the characteristic frequency of collisions between electrons $\sqrt{\nu\nu_\lambda} = \sqrt{\nu(z = 0, t)\nu_\lambda(z = 0, t)}$.

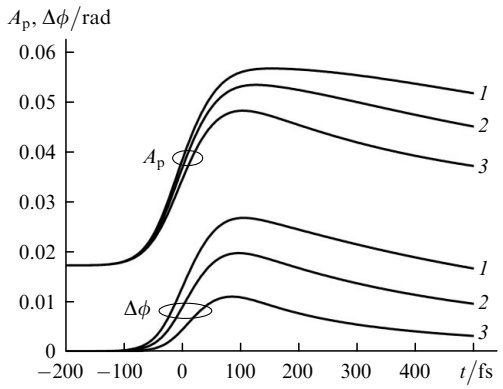

Figure 6. Time evolution of the absorption coefficient A_p and the phase shift $\Delta\phi$ of the reflected wave upon irradiation of a gold target by pulses from a CO_2 laser for $b = 2$ (*1*), 1.5 (*2*) and 1 (*3*).

6. Conclusions

Thus, we have demonstrated the need for a consistent description of the field distribution in the skin layer in the study of reflection and absorption of radiation by the metal whose electrons are heated nonuniformly upon absorption of radiation of a femtosecond laser pulse. The nonuniformity of the dielectric constant resulting from the rapid heating of the metal leads to significant changes both in the absorption coefficient and phase shift of the reflected wave. Given the relatively small heating of electrons previously proposed in [9], an approximate analytic description of the optical properties of the nonequilibrium metal is consistent with a more precise numerical calculation. If the electron heating leads to a strong nonuniformity of the dielectric constant, then the description of the optical properties of the nonequilibrium metal necessitates consistent numerical calculations of the field as well as of electron and lattice temperatures.

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