

# Thermal ablation of an aluminium film upon absorption of a femtosecond laser pulse

S.G. Bezhanov, A.P. Kanavin, S.A. Uryupin

**Abstract.** We have found the time dependence of the ablation depth of aluminium irradiated by a femtosecond laser pulse. It is shown to what extent an increase in the radiation energy flux density leads to an increase in the quasi-stationary value of the ablation depth. By reducing the aluminium film thickness down to one hundred nanometres and less, the ablation depth significantly increases. At the same time, the quasi-stationary value of the ablation depth of a thin film is obtained due to the removal of heat from the focal spot region.

**Keywords:** femtosecond pulse, metal nanofilm, laser ablation, aluminium.

## 1. Introduction

Ablation of metals upon absorption of laser pulses has been studied experimentally and theoretically in many papers (see, e.g., [1–5] and review [6]). The essential difference in the ablation process of bulk samples under the action of femtosecond, picosecond and nanosecond pulse has been determined in [2, 3]. This difference arises due to changes in the ratio between the time of the pulse action and the times of the metal surface heating and cooling. Since the last two times depend on the size of the sample, we can expect changes in the ablation properties in quite small samples compared with those inherent in bulk samples. This fact was confirmed in the studies of ablation of thin films of metals [7–10]. In particular, a decrease in the radiation energy flux density, at which gold, nickel [7, 10] and copper films are efficiently ablated [10] with decreasing film thickness down to a micron and less, was experimentally observed.

The authors of [8] found threshold radiation energy flux densities  $I_{th}$  of 200-fs pulses at which gold and nickel films are damaged. For gold, the values of  $I_{th}$  increase with increasing film thickness  $L$  and reach saturation at  $L \approx 500$  nm. A similar dependence of  $I_{th}$  on  $L$  was found for nickel; however, the saturation in this case occurs only at  $L \gtrsim 50$  nm, which is associated with a more efficient transfer of energy from the electrons to the lattice [8]. A dependence of  $I_{th}$  on  $L$ , similar to

that from paper [8], was found in [9], where irradiation of a gold film by 600-fs pulses lead to a decrease in  $I_{th}$  at  $L \lesssim 100–200$  nm.

In this paper we have theoretically investigated the depth of thermal ablation of aluminium samples interacting with a relatively weak femtosecond pulse, which leads to the removal of a small number of atomic layers of the material. It is shown that with increasing time the ablation depth reaches a quasi-stationary value  $d_t$ , which is dependent on  $I_L$ , i.e. the energy flux density of incident radiation. The dependence of  $d_t$  on  $I_L$  is found. It has been demonstrated that the ablation depth increases with decreasing film thickness. In sufficiently thin films the establishment of a quasi-stationary value of the ablation depth becomes possible if the focusing of laser radiation is taken into account. We have found the dependences of  $d_t$  on the focal spot size, film thickness and aluminium parameters.

## 2. Absorption of radiation and heating of the metal

Consider the interaction of a laser pulse propagating along the normal to the film surface with a metal film, which occupies the space region  $0 < z < L$ . The laser radiation field in vacuum is given by

$$\mathbf{E}_{inc} = \mathbf{E}_L(t - z/c)\sin(\omega t - kz), \quad z < 0, \quad (1)$$

where  $\omega$  is the carrier frequency of laser radiation;  $k = \omega/c$  is the wavenumber;  $c$  is the speed of light; and  $\mathbf{E}_L(t) = [0, E_L(t), 0]$  is the pulse envelope weakly changing in time  $\sim 1/\omega$ . The electromagnetic field penetrates into the film, is reflected from the surface  $z = 0$  and passes into the region  $z > L$ . The field  $\mathbf{E}_m = [0, E_m(z, t), 0]$  penetrating inside has the form (see [11])

$$E_m(z, t) \simeq \frac{1}{2i} [\exp(i\omega t)F(z, -\omega) - \exp(-i\omega t)F(z, \omega)]E_L(t), \quad (2)$$

where

$$F(z, \omega) = \frac{2\omega \{ \omega \sinh[\kappa(z - L)] - i\kappa c \cosh[\kappa(z - L)] \}}{(\kappa^2 c^2 - \omega^2) \sinh(\kappa L) - 2i\omega \kappa c \cosh(\kappa L)}, \quad (3)$$

$$\kappa^2 = -\frac{\omega^2}{c^2} \varepsilon(\omega); \quad \varepsilon(\omega) = \varepsilon_0(\omega) - \frac{\omega_p^2}{\omega(\omega + i\nu)} = \varepsilon'(\omega) + i\varepsilon''(\omega); \quad (4)$$

$\varepsilon_0(\omega) = \varepsilon_0'(\omega) + i\varepsilon_0''(\omega)$  is the contribution to the dielectric constant from the bound electrons;  $\omega_p$  is the electron plasma frequency; and  $\nu$  is the collision frequency of electrons. Assuming that bound electrons pass to the conduction band

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Received 9 June 2015; revision received 26 November 2015  
Kvantovaya Elektronika 46 (2) 119–124 (2016)  
Translated by I.A. Ulitkin

upon absorption of the field, the power averaged over period  $2\pi/\omega$  and absorbed by the electrons can be expressed in the form [11]:

$$Q(z, t) \simeq \frac{c\varepsilon''(\omega)}{\omega |\varepsilon(\omega)|^2} I_L(t) \left| \frac{\partial F(z, \omega)}{\partial z} \right|^2, \quad (5)$$

where  $I_L(t) = cE_L^2(t)/(8\pi)$  is the energy flux density of the incident pulse.

The description of the metal heating can be based on the equation for the electron  $T_e(z, t)$  and lattice  $T_{\text{lat}}(z, t)$  temperatures:

$$C_e(z, t) \frac{\partial}{\partial t} T_e(z, t) = Q(z, t) + \frac{\partial}{\partial z} \left[ \lambda(z, t) \frac{\partial}{\partial z} T_e(z, t) \right] - G(z, t) [T_e(z, t) - T_{\text{lat}}(z, t)], \quad (6)$$

$$C_{\text{lat}} \frac{\partial}{\partial t} T_{\text{lat}}(z, t) = G(z, t) [T_e(z, t) - T_{\text{lat}}(z, t)], \quad (7)$$

where  $C_e(z, t)$  is the electron heat capacity;  $\lambda(z, t)$  is the thermal conductivity;  $G(z, t)$  is the parameter describing the energy transfer from the electrons to the lattice; and  $C_{\text{lat}}$  is the heat capacity of the lattice.

Equations (6) and (7) should be supplemented with initial and boundary conditions:

$$T_e(z, t \rightarrow -\infty) = T_{\text{lat}}(z, t \rightarrow -\infty) = T_0, \quad (8)$$

$$\lambda(z, t) \frac{\partial}{\partial z} T_e(z, t) \Big|_{z=0} = -\lambda(z, t) \frac{\partial}{\partial z} T_e(z, t) \Big|_{z=L} \simeq 0. \quad (9)$$

At the same time, the heat loss due to thermal emission of electrons is considered negligible [12].

The properties of the evolution of temperatures depend on the explicit form of the functions  $C_e(z, t)$ ,  $G(z, t)$  and  $\lambda(z, t)$ , which are different for different substances. Below we restrict our consideration to the study of the heating of aluminium, whose plasma frequency is  $\omega_p = 1.91 \times 10^{16} \text{ s}^{-1}$  [13]. We assume that the film of thickness  $L$  is irradiated by a laser pulse focused into a spot of radius  $R$  and having a frequency  $\omega \simeq 2.35 \times 10^{15} \text{ s}^{-1}$ . Then, the time dependence of the energy flux density has the form:  $I_L(t) = I_L \exp(-t^2/t_p^2)$ , where  $t_p = \tau/2 \sqrt{\ln 2}$ , and  $\tau = 100 \text{ fs}$  is a characteristic pulse duration. The contribution (corresponding to a given frequency  $\omega$ ) to the dielectric constant from the bound electrons is  $\varepsilon_0(\omega) = 8.5 + 41i$ , which corresponds to the absorption coefficient of 0.14 if heating is neglected [13, 14]. For the heat capacity of the aluminium lattice we use  $C_{\text{lat}} = 2.4 \times 10^7 \text{ erg cm}^{-3} \text{ K}^{-1}$ . In accordance with the data of Ref. [15], at  $T_e < 6 \times 10^4 \text{ K}$  the electronic heat capacity of aluminium is approximated by the expression

$$C_e(z, t) = C_{\text{Al}} T_e(z, t) \frac{1 + 0.153[10^{-4} T_e(z, t)]}{1 + 0.065[10^{-4} T_e(z, t)]^2}, \quad (10)$$

where  $C_{\text{Al}} \simeq 9.1 \times 10^2 \text{ erg cm}^{-3} \text{ K}^{-2}$ , and the temperature is measured in Kelvins. In turn, taking into account the data of Ref. [16], obtained at  $T_e(z, t) < 2 \times 10^4 \text{ K}$ , the parameter  $G$  can be found by using the approximation:

$$G(z, t) = G_{\text{Al}}$$

$$\times \left\{ 1 - \frac{0.0009}{[10^{-4} T_e(z, t)]^4 + 0.0030} + 0.0028[10^{-4} T_e(z, t)] \right\}, \quad (11)$$

where  $G_{\text{Al}} = 3.5 \times 10^{18} \text{ erg s}^{-1} \text{ K}^{-1} \text{ cm}^{-3}$ . Under the conditions described hereinafter, the electron temperature exceeds 2 eV for several hundred femtoseconds in a sufficiently thin (of thickness 20–30 nm) surface layer, and therefore possible errors due to calculation using expression (11) are unimportant. The thermal conductivity in equation (6) has the form

$$\lambda(z, t) = \frac{C_e(z, t) \bar{v}_s^2}{3\nu(z, t)}, \quad (12)$$

where  $\bar{v}_s^2 = [3v_F^2/5 + 3k_B T_e(z, t)/m]$  is the mean square of the velocity of electrons [17];  $v_F = 2.05 \times 10^8 \text{ cm s}^{-1}$  is the Fermi velocity;  $k_B$  is the Boltzmann constant; and  $m$  is the effective electron mass. The electron collision frequency  $\nu(z, t)$ , which determines the thermal conductivity, is equal to the sum of the electron–phonon collision frequency  $\nu_{\text{ep}}$ , depending on the lattice temperature, and electron–electron collision frequency  $\nu_{\text{ee}}$ , depending on the temperature of the electrons:

$$\nu(z, t) = \frac{3}{5} \nu_{\text{ep}}(T_{\text{lat}}(z, t)) + \nu_{\text{ee}}(T_e(z, t)).$$

The introduction of the factor 3/5 is due to difference of  $\bar{v}^2$  from  $v_F^2$  at low temperatures. When the lattice temperature exceeds the Debye temperature, the frequency of the electron–phonon collisions can be approximated as follows:

$$\nu_{\text{ep}}(T_{\text{lat}}(z, t)) = \nu_{\text{ep}}(T_0) \frac{T_{\text{lat}}(z, t)}{T_0}, \quad (13)$$

where  $\nu_{\text{ep}}(T_0) = 9.4 \times 10^{13} \text{ s}^{-1}$  is the frequency of collisions between electrons and phonons at room ( $T_0 = 300 \text{ K}$ ) temperature [13]. This approximation is justified, even though the Debye temperature for aluminium is 430 K. The fact is that due to rapid heating under the conditions to be discussed below, the lattice temperature reaches  $\sim 430 \text{ K}$  for several tens of femtoseconds. Usually, at electron temperatures much lower than the degeneracy temperature, there is a quadratic dependence of  $\nu_{\text{ee}}$  on  $T_e$ . However, as  $T_e$  approaches  $\mathcal{E}_F/k_B$ , where  $\mathcal{E}_F = 11.7 \text{ eV}$  is the Fermi energy, the impact of the band structure and lifting of the degeneracy lead to a significant change in the quadratic dependence [18]. The results obtained in [18] are approximated with good accuracy by the formula

$$\nu_{\text{ee}}[T_e(z, t)] \simeq b \frac{\mathcal{E}_F}{\hbar} \frac{[k_B T_e(z, t)/\mathcal{E}_F]^2}{1 + \alpha [k_B T_e(z, t)/\mathcal{E}_F]^2}, \quad (14)$$

where  $\hbar$  is the Planck constant;  $b = 1.7$ ; and  $\alpha = 15$ . It is this dependence of  $\nu_{\text{ee}}$  on  $T_e$  that is used below to solve equation (6).

The power absorbed by conduction electrons (5) also depends on the frequency of collisions. If  $\nu \ll \omega$ , formula (4) includes the frequency  $\nu = \nu_{\text{opt}}$ , describing the interaction of electrons with a high-frequency field. The frequency  $\nu_{\text{opt}}$  differs from  $\nu(z, t)$ , which enters the thermal conductivity coefficient (12), and is determined by the constants  $\nu_{\text{ep}}^{\text{opt}}(T_0) \neq \nu_{\text{ep}}(T_0)$  and  $a \neq b$  (see, e.g., [19, 20], where  $\alpha = 0$  under low heating conditions). However, in the frequency range discussed below at the initial stage of heating, absorption by bound electrons determined by  $\varepsilon_0''(\omega)$  is

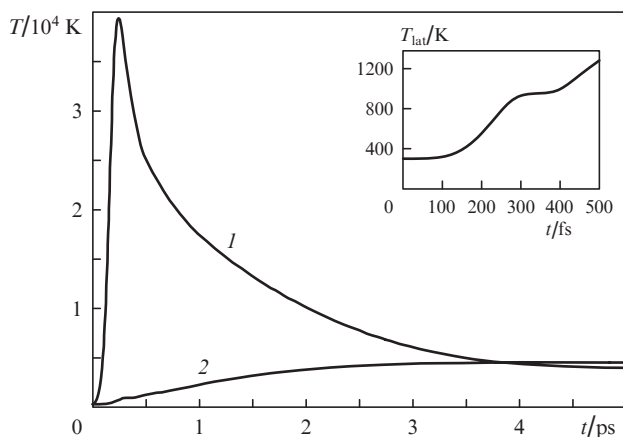
almost an order of magnitude higher than that due to collisions of conduction electrons determined by  $v_{\text{opt}}$ . This makes insignificant the allowance for the differences between  $v$  and  $v_{\text{opt}}$  at this stage of heating. If the electron collision frequency becomes on the order of  $\omega$  due to heating, then we can use formulas (13), (14). These expressions for the collision frequencies used in the solution of equations (6), (7).

If the temperature  $T_{\text{lat}}(z, t)$  reaches the melting temperature of aluminium  $T_m = 930$  K, the lattice is destroyed. In the numerical solution, melting is taken into account by stopping the temperature growth when the  $T_{\text{lat}}(z, t) = T_m$ , and as long as at a lattice point in question no energy, which is equal to the specific heat of fusion ( $\Delta H_m = 10.8$  kJ mol<sup>-1</sup>), is transmitted [21]. In addition, the frequency of collisions between the electrons in the melt is increased by about 50% [22] (cf. [13]), which is taken into account by the temperature dependence of the form

$$v_{\text{ep}}[T_{\text{lat}}(z, t)] = v_{\text{ep}}(T_0) \frac{T_{\text{lat}}(z, t)}{T_0} [1 + 0.5\eta(T_{\text{lat}}(z, t) - T_m)], \quad (15)$$

where  $\eta(\dots)$  is the unit Heaviside step function. After melting functions  $G(z, t)$  and  $C_e(z, t)$  are taken the same, as in the solid phase. Note that in the case of rapid heating of the metal, the lattice may be overheated when the temperature  $T_{\text{lat}}$  exceeds  $T_m$ , but the disorder does not occur within a few picoseconds [23]. Since the time scale, on which the ablation takes place, amounts to tens of picoseconds, this effect is neglected.

Typical time dependences of the temperatures of the electrons and the lattice on the surface of the film obtained by solving equations (6), (7) are shown in Fig. 1 for  $I_L = 3 \times 10^{12}$  W cm<sup>-2</sup> and  $L \rightarrow \infty$ ,  $R \rightarrow \infty$ . It can be seen that in the absorption of light, the electrons are rapidly heated while the lattice remains relatively cold. Then, during the time  $t_* \approx C_{\text{lat}} T_{\text{lat}}^{\text{max}} / (G_{\text{Al}} T_e^{\text{max}})$ , which is a few picoseconds, the temperatures of the electrons and the lattice become equalised. Thus there is a transfer of heat into the metal by the conduction electrons. Note that in the conditions discussed the fraction of the energy spent on the melting of aluminium is small compared to the total absorbed energy and the melting process is quick enough, as shown by a short horizontal section of the dependence  $T_{\text{lat}}(z = 0, t)$  in Fig. 1.



**Figure 1.** Typical time dependences of (1) electron and (2) lattice temperatures at the irradiated sample surface ( $z = 0$ ). Dependences are plotted for  $I_L = 3 \times 10^{12}$  W cm<sup>-2</sup> and sufficiently large  $L$  and  $R$ .

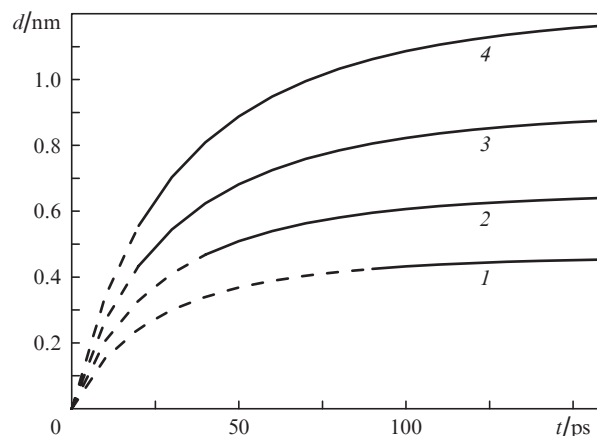
### 3. Ablation of aluminium

The heating of aluminium is accompanied by ablation. To calculate the depth of ablation we will use the relation

$$d(t) = \int_0^t v[T_{\text{lat}}(z = 0, t')] \exp\left[-\frac{U_0}{k_B T_{\text{lat}}(z = 0, t')}\right] dt', \quad (16)$$

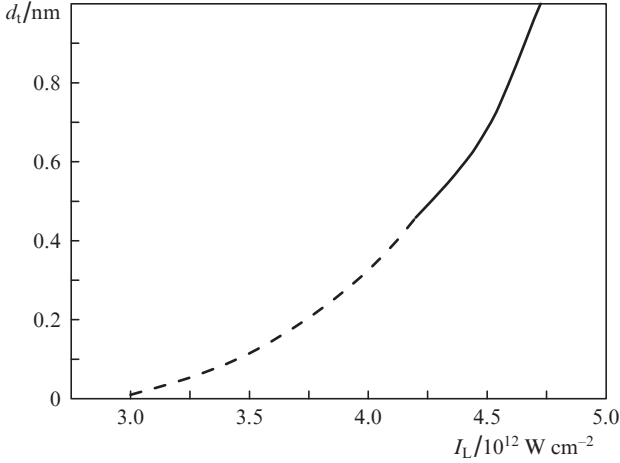
where  $U_0 = \Delta H_b / N_A = 3.05$  eV is the energy corresponding to the specific heat of vaporisation  $\Delta H_b = 293$  kJ mol<sup>-1</sup> for aluminium [21];  $N_A$  is Avogadro's number; and the ratio for the velocity  $v(T_{\text{lat}})$  is given in the Appendix.

Figure 2 shows the time dependences of the ablation depth of a rather thick film at various energy flux densities of the incident unfocused pulse ( $R \rightarrow \infty$ ), i.e. under conditions when the influence of the transverse heat transfer is insignificant. During a few tens of picoseconds aluminium evaporates, and in about a hundred picoseconds the ablation depth reaches a quasi-stationary value  $d_t$ , which is weakly dependent on time. From Fig. 3, which shows the dependence of the ablation depth per pulse on  $I_L$ , one can see a sharp increase in  $d_t$  with increasing  $I_L$ . In this case, one atomic layer of aluminium (i.e.  $d_t = 0.4$  nm) is removed at  $I_L \approx 4.2 \times 10^{12}$  W cm<sup>-2</sup>, corresponding to an energy density of 420 mJ cm<sup>-2</sup>. This value is close to those previously found for aluminium: 400 [24, 25] and 540 mJ cm<sup>-2</sup> [26]. In Figs 2–5 the parts of the curves, where  $d_t$  is less than 0.4 nm, are shown by dashed lines.

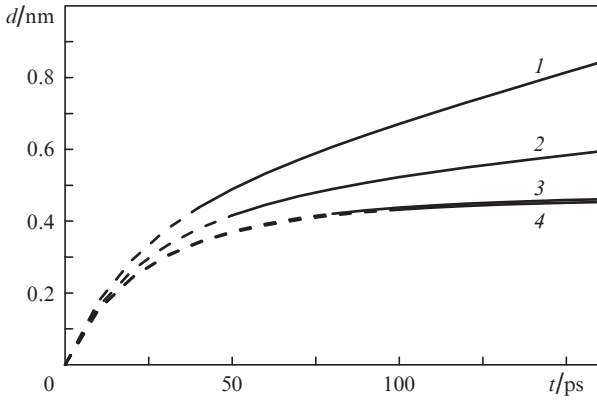


**Figure 2.** Time dependences of the ablation depth for a thick film at maximum energy flux densities  $I_L = (1) 4.2 \times 10^{12}$ , (2)  $4.4 \times 10^{12}$ , (3)  $4.6 \times 10^{12}$  and (4)  $4.8 \times 10^{12}$  W cm<sup>-2</sup>.

Figure 4 shows the time dependence of the ablation depth for films of different thickness. In experiments the film is usually located on the substrate. However, the boundary condition at  $z = L$  does not take into account the transfer of heat to the substrate. This approximation is justified for relatively thick films, or at a relatively low thermal conductivity of the substrate. It is seen that for 120-nm-thick films the transfer of heat inside the film provides a sufficiently rapid cooling, which leads to the fact that  $d(t)$  reaches a quasi-stationary value at times of a few tens of picoseconds. By reducing the film thickness down to 100 nm or less, for a time of about 100 ps a slightly nonuniform temperature distribution is attained in the entire film thickness, and its magnitude is relatively large. As a result, the depth of ablation monotonically increases with time. For a quasi-stationary value of the abla-



**Figure 3.** Dependence of the ablation depth per pulse on the maximum energy flux density in the incident pulse.



**Figure 4.** Time dependences of the ablation depth for films having a thickness of (1) 100, (2) 120, (3) 200 and (4) 300 nm. The maximum energy flux density is  $I_L = 4.2 \times 10^{12} \text{ W cm}^{-2}$ .

tion depths of thin films to be reached it is necessary, in particular, to consider the heat transfer in the transverse direction, which is important for a small focal spot size.

Under the conditions discussed, at  $t \approx 2\text{--}4$  ps (see Fig. 1) the temperatures of the electrons and the lattice for aluminium are equalised. Starting from this moment of time ( $t = t_*$ ), we can use a single equation for the overall temperature:

$$\frac{\partial T(r, z, t)}{\partial t} = \frac{\lambda_0}{C_{\text{lat}}} \left[ \frac{\partial^2 T(r, z, t)}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T(r, z, t)}{\partial r} \right) \right]. \quad (17)$$

The effect of the electron–electron collisions on the thermal conductivity of aluminium at temperatures that do not exceed  $4 \times 10^3$  K is insignificant in comparison with the influence of collisions with phonons. By neglecting  $v_{ee}$  and taking into account the fact that at similar temperatures of the electrons and the lattice and at  $T_{\text{lat}} > T_m$  the dependence of  $\lambda_0$  on the temperature disappears [see (10) and (15)], for the thermal conductivity coefficient in (17) we have  $\lambda_0 = C_{\text{Al}} T_0 v_F^2 / 3v_{\text{ep}}(T_0) = \text{const}$ .

The boundary conditions for equation (17)

$$\lambda_0 \frac{\partial T(r, z, t)}{\partial z} \Big|_{z=0} \simeq 0, \quad -\lambda_0 \frac{\partial T(r, z, t)}{\partial z} \Big|_{z=L} = 0, \quad (18)$$

$$T(z, r \rightarrow \infty, t) = T_0$$

are similar to those used previously [see (9)]. In this case, the heat loss due to evaporative cooling is considered insignificant. The corresponding boundary condition must take into account the removal of the heat by evaporating particles, each of which carries an energy  $U_0$  (see [27]). However, because we consider the conditions under which only a few atomic layers of the material are evaporated, the heat flux outgoing from the metal surface during its evaporation is small compared with the flux inside the metal.

The temperature at time  $t = t_*$  is obtained from the numerical solutions of equations (6), (7) and is given as a function that is independent of  $r$  in the focal spot region of radius  $R$ :

$$T(z, r, t = t_*) = T_0 + [T(z, t_*) - T_0] \eta (R - r). \quad (19)$$

Typically, the focal spot radius  $R$  is greater than the wavelength of incident radiation, i.e. more than 1000 nm. It should be noted that during the time  $t_*$  heat propagates by about  $v_F \sqrt{3t_*/v_{\text{ep}}} \approx 300$  nm, which is smaller than the typical values of  $R$ .

The solution of equation (17), corresponding to conditions (18) and (19), has the form

$$\begin{aligned} T(r, z, t) = & T_0 + \frac{C_{\text{lat}}}{L \lambda_0 (t - t_*)} \sum_{n=0}^{\infty} \exp \left[ -\frac{\lambda_0}{C_{\text{lat}}} (t - t_*) \frac{\pi^2 n^2}{L^2} \right] \\ & \times \cos \left( \frac{\pi n z}{L} \right) \int_0^L dz' [T(z', t_*) - T_0] \cos \left( \frac{\pi n z'}{L} \right) \\ & \times \int_0^R dr' r' \exp \left[ -\frac{r^2 + r'^2}{4\lambda_0 (t - t_*)} C_{\text{lat}} \right] I_0 \left[ \frac{r r' C_{\text{lat}}}{2\lambda_0 (t - t_*)} \right]. \quad (20) \end{aligned}$$

The prime at the sum indicates that the term with  $n = 0$  is taken with a weight of  $1/2$ , and  $I_0$  is the modified Bessel function of the first kind. For  $r = 0$  at the centre of the focal spot we derive from (20) a simpler expression

$$\begin{aligned} T(0, z, t) = & T_0 + \frac{2}{L} \left\{ 1 - \exp \left[ -\frac{C_{\text{lat}} R^2}{4\lambda_0 (t - t_*)} \right] \right\} \\ & \times \sum_{n=0}^{\infty} \exp \left[ -\frac{\lambda_0}{C_{\text{lat}}} (t - t_*) \frac{\pi^2 n^2}{L^2} \right] \cos \left( \frac{\pi n z}{L} \right) \\ & \times \int_0^L dz' [T(z', t_*) - T_0] \cos \left( \frac{\pi n z'}{L} \right). \quad (21) \end{aligned}$$

Equation (17) and formula (21) show that there are two characteristic time scales of the relaxation temperature. In the plane of the film the relaxation temperature is determined by the time

$$\tau_R = \frac{C_{\text{lat}} R^2}{4\lambda_0}, \quad (22)$$

which, for example, for  $R \approx 3 \mu\text{m}$  is about ten nanoseconds. In turn, the temperature equalisation time over the film thickness is given by

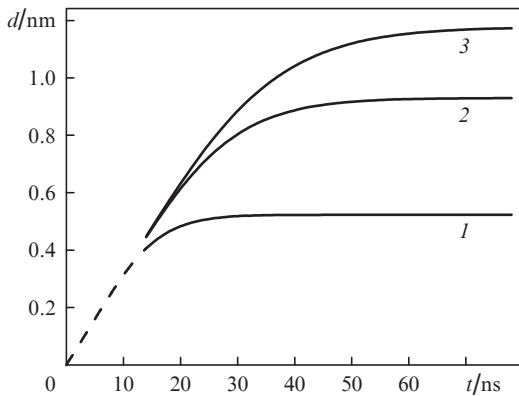
$$\tau_L = \frac{C_{\text{lat}} L^2}{\pi^2 \lambda_0} \quad (23)$$

and at  $L \approx 100$  nm is about ten picoseconds (see Fig. 1), i.e. significantly less than  $\tau_R$ .

One can see from (20), (21) that the temperature  $T(r, z, t)$  has a maximum at  $z = 0$  and  $r = 0$  and determines the maximum ablation depth. In a thin film during a time  $t_*$  a temperature distribution that is almost uniform over the thickness is established [28]. Therefore, at times greater than  $t_*$ , the sum over  $n$  in formula (21) is determined mainly by the term with  $n = 0$  and for  $T(0, 0, t)$  we have approximately

$$T(0, 0, t) \simeq T(0, t_*) \left[ 1 - \exp\left(-\frac{\tau_R}{t - t_*}\right) \right]. \quad (24)$$

The transverse heat transfer reduces the temperature at point  $z = 0$ ,  $r = 0$  and allows integral (16) to reach a quasi-stationary value at times of tens of nanoseconds. Typical dependences of the ablation depth of the thin film are shown in Fig. 5 for different focal spot sizes. It is seen that the quasi-stationary value of  $d_t$  increases with  $R$  due to an increase in time, during which a sufficiently high temperature is maintained in the centre of the focal spot. For thin films, the energy flux density at which ablation is efficient, as compared with the corresponding values for bulk samples, decreases. The reason for this reduction is that in the case of thin films, the absorbed pulse energy is redistributed in a smaller volume of the metal, which further cools during the time  $\tau_R$  that is longer than  $\tau_L$ . Therefore, the characteristic values of the energy flux density required to remove several atomic layers of aluminium are much lower in the case of thin films (see Fig. 5) than in the case of bulk samples (see Fig. 2).



**Figure 5.** Time dependence of the ablation depth of a 100-nm-thick film for the focal spot radii  $R = (1)$  3,  $(2)$  4 and  $(3)$  4.5  $\mu\text{m}$  at  $I_L = 2.8 \times 10^{12} \text{ W cm}^{-2}$ .

Given that the characteristic times of the ablation are much longer than  $t_*$ , and using (24) at  $T(0, t_*)$  that is higher than the initial temperature  $T_0$ , from (16) for the quasi-stationary value of the ablation depth we find

$$d_t \simeq v_* \tau_R \left\{ \ln \left[ \frac{U_0}{k_B T(0, t_*)} \right] \right\}^{-1} \exp \left[ -\frac{U_0}{k_B T(0, t_*)} \right], \quad (25)$$

where  $v_* \equiv v[T(0, t_*)]$ . In the calculation we take into account that a significant contribution to the integral comes from small times at which the temperature remains close to  $T(0, t_*)$ . The temperature  $T(0, t_*)$  can be estimated by assuming that for a thin film all the absorbed pulse energy in the focal spot region during the time  $t_*$  is uniformly distributed over the film thickness  $L$ , i.e.  $C_{\text{lat}} T(0, t_*) L = A I_L \tau$ , where  $A$  is the absorp-

tion coefficient averaged over the time of the pulse action. Taking into account the estimate for  $T(0, t_*)$ , from (25) we obtain for the energy flux density at which  $d_t = d_0 = 0.4 \text{ nm}$

$$I_{\text{th}} = \frac{U_0 C_{\text{lat}} L}{k_B A \tau} \left( \ln \left\{ v_* \tau_R \left[ d_0 \ln \left( \frac{U_0}{k_B T(0, t_*)} \right) \right]^{-1} \right\} \right)^{-1}. \quad (26)$$

According to (26) the value of  $I_{\text{th}}$  is logarithmically weakly dependent on the explicit form of the imprecisely known pre-exponential factor in (25). Note that a virtually linear dependence of the ablation threshold on film thickness from (26) corresponds to that found experimentally [7, 9, 10].

In the case when the heat transfer is significant not only in the transverse direction but also into the substrate having, as a rule, a much lower thermal conductivity than the film, the estimates by formula (26) are allowed, if the characteristic time of the transverse heat transfer,  $\tau_R$ , is replaced by the time of the heat transfer into the substrate. However, due to the logarithmic dependence of  $I_{\text{th}}$  on the characteristic time this change will not have a significant impact on the estimate of the value of  $I_{\text{th}}$  and its dependence on the film thickness.

## 4. Conclusions

We present a relatively simple description of ablation of aluminium nanofilms heated by a femtosecond laser pulse. It is shown that for sufficiently thick films ablation occurs during a time of about ten picoseconds, which is determined by the cooling of the surface as a result of the energy transfer into the metal. Calculations of the threshold energy flux densities are in agreement with the data of a number of experiments [24–26]. For films whose thickness is less than 100 nm, and in the absence of heat removal into the substrate, it is necessary to take into account the transverse heat transfer, which at a focal spot size of a few micrometres occurs during approximately ten nanoseconds. The presented study of the aluminium ablation is quite versatile and can be generalised with respect to the ablation of films of other metals.

**Acknowledgements.** This work was supported by the Russian Foundation for Basic Research (Project No. 13-02-01377) and the programme of the RAS Presidium and Physical Sciences Department.

## Appendix

Let us derive the expression for the velocity of the evaporation front. Following [29], we write the equality of chemical potentials in the different phases that are in equilibrium:

$$\frac{\varepsilon_2}{k_B T} + \ln \frac{P}{k_B T} - c_V \ln k_B T - \frac{3}{2} \ln \frac{M}{2\pi\hbar^2} = \frac{\varepsilon_1}{k_B T} + c_1 \ln \frac{\hbar\bar{\omega}}{k_B T}, \quad (\text{A.1})$$

where  $P$  is the saturated vapour pressure;  $M$  is the mass of the atom;  $c_1 = 3$  and  $c_V = 3/2$  are dimensionless specific heats of the condensed phase and gas;  $\varepsilon_1$  and  $\varepsilon_2$  are equilibrium energies of the atom in the condensed and gaseous phases; and  $\bar{\omega}$  is the average frequency of lattice vibrations. From (A.1), we find the saturated vapour pressure

$$P = k_B T \left( \frac{\hbar\bar{\omega}}{k_B T} \right)^3 \left( \frac{M k_B T}{2\pi\hbar^2} \right)^{3/2} \exp \left[ \frac{\varepsilon_1 - \varepsilon_2}{k_B T} \right]. \quad (\text{A.2})$$

According to [29] the rate of evaporation into vacuum is determined by the number of atoms leaving the surface per unit time per unit surface area of the body. Neglecting the influence of the unknown reflection coefficient of gas particles from the surface, we can estimate the rate of the evaporation front as

$$v \simeq \frac{P}{N\sqrt{2\pi M k_B T}} = \frac{\bar{\omega}^3 M}{4\pi^2 N k_B T} \exp\left[-\frac{U_0}{k_B T}\right] \equiv v(T) \exp(-U_0/k_B T), \quad (\text{A.3})$$

where  $U_0 = \varepsilon_2 - \varepsilon_1$ , and  $N$  is the number of atoms per unit volume.

Let us present an estimate for aluminium. Assuming  $M = 4.5 \times 10^{-23}$  g,  $N = 4.9 \times 10^{22}$  cm<sup>-3</sup>,  $\bar{\omega} \simeq 2.5 \times 10^{13}$  s<sup>-1</sup> and  $T \approx T_{\text{lat}}^{\text{max}} \simeq 5500$  K, we have  $v(T) \simeq 4.8 \times 10^5$  cm s<sup>-1</sup>, which is close to  $4 \times 10^5$  cm s<sup>-1</sup>, given in [6] for aluminium.

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