

Dynamics of the heating and evaporation of a plane target by a laser pulse of arbitrary shape

V. S. Vorob'ev

Institute of High Temperature Physics, Russian Academy of Sciences, 127412 Moscow, Russia

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A general solution is derived for the 1D single-phase Stefan problem with a surface heat source for constant values of the thermal properties and the absorption coefficient. This solution is used to describe the dynamics of the heating and evaporation of a plane target by a laser pulse of arbitrary shape. A family of isotherms is plotted in the plane of dimensionless coordinates which do not depend on the thermal properties of the target material. Also plotted in this plane is a line which separates the plane into two parts: one dominated by the thermal-conductivity losses and one dominated by evaporation losses. Laser pulses of arbitrary shape are constructed in the same coordinates. Their intersections with isotherms and characteristic lines give a clear picture of the changes in the surface temperature during the heating, of the time of the transition to rapid evaporation, and of the nature of the vapor outflow. Pulses of this sort of microsecond and millisecond length are constructed for a copper target.

The problem of the heating and evaporation of metals by laser light dates back a long way. Many aspects of this phenomenon are discussed in monographs and reviews.^{1–6} The evaporation of metals by the lasers available at the time, with pulses of definite shape and differing greatly in length has been studied. The depth of the evaporation has been measured as a function of the incident-energy surface density for various metals. Ready¹ points out that pulses with lengths in the nanosecond range are relatively ineffective in causing evaporation. The reason is that the evaporation begins not immediately but only after a certain time interval, once the target has warmed up to a certain temperature. This particular temperature is determined by the dynamics of the heating and is not known at the outset. Anisimov *et al.*² have found simple formulas for the “delay times” after which rapid evaporation begins for pulses of constant intensity. The meaning here is that the energy of the laser beam is expended primarily on evaporating particles from the condensed phase and partly on transferring kinetic energy to these particles. The energy losses due to thermal conductivity are small in this regime. For pulses of arbitrary shape, in contrast, there is no simple method of this sort for determining the time at which rapid evaporation begins. As a result, it is difficult to obtain information on how effective one laser or another will be on a target, information on the losses due to thermal conductivity, etc. The situation becomes particularly serious when the lasers in question are relatively of low power and designed for technical operations involving the evaporation of target material.

In the present paper we use the standard formulation of the 1D problem of heating a plane target by laser light under the assumption that the thermal coefficients and the absorption coefficient remain constant. This formulation of the problem dates back to Ref. 2. We write a time-dependent heat-conduction equation with a boundary condition which incorporates evaporation (the Stefan prob-

lem). We derive a formal solution of this problem, which reduces to a nonlinear integral equation for the surface temperature. This equation does not contain spatial derivatives. In this approximation the term associated with evaporation is an exponential function of the temperature. A modified method for expanding an exponential function was developed in the course of the present study in order to find an approximate solution of equations of this sort. This method is a refinement of the exponential expansion method which is widely used in problems of the theory of combustion and explosions.⁷ The solution yields an algebraic expression in dimensionless variables which relates the intensity q of the laser light, the quantity

$$L = \frac{1}{\sqrt{\pi}} \int_0^t d\tau q(\tau) / (t - \tau)^{1/2},$$

which depends on this intensity, and the surface temperature at an arbitrary time t . Using this expression, we construct a universal family (i.e., one independent of the particular metal) of isotherms in the plane of the dimensionless variables L and q . The intersection of the curve describing (in the same coordinates) the actual laser pulse, on the one hand, with the isotherms, on the other, gives us definite information about the target-heating dynamics. Since the evaporation rate is exceedingly sensitive to the temperature, one can draw in the L, q plane a line which divides this plane into two parts. In the first part, the energy of the laser beam is carried off primarily by the heat flux (this is the thermal-conductivity regime). In the second part, the energy is expended on evaporation (this is the rapid-evaporation regime).^{1–6} The point at which the laser pulse intersects this line corresponds to the beginning of the evaporation regime. The boundary corresponding to the onset of rapid evaporation in the L, q coordinates is a universal boundary. To determine the time at which rapid evaporation begins during the application of a laser pulse

of arbitrary shape, it is sufficient to plot the pulse in the same coordinates. This has been done for some characteristic laser pulses with lengths in the microsecond and millisecond ranges. As an example we calculate the boundary of the region of rapid evaporation, in dimensional variables, for the case in which a millisecond pulse is applied to a copper target. It is important to note that the threshold intensity is a decreasing function of the time, and the values of this intensity early in the process can be fairly high. This circumstance sheds some light on the low efficiency of evaporation by short-pulse lasers.

1. SOLUTION OF THE 1D STEFAN PROBLEM

A heat flux which is a given function of the time, $q(t)$, is supplied from the left to the surface of the material, $x=0$; the material fills the half-space $x>0$. As a result, the surface warms up, begins to evaporate, and recedes at a velocity v_s . The heat-conduction problem in the coordinate system moving with this interface can be written as follows under the assumption that the thermal properties and the absorption coefficient remain constant:

$$\frac{\partial T}{\partial t} = \chi \frac{\partial^2 T}{\partial x^2} + v_s \frac{\partial T}{\partial x}, \quad (1)$$

where $T(x,t)$ is the temperature and χ is the thermal diffusivity. We supplement Eq. (1) with the boundary condition

$$-\kappa \frac{\partial T}{\partial x} \Big|_{x=0} = \zeta q(t) - \rho v_s \left(\Delta H + \frac{v_1^2}{2} \right), \quad (2)$$

where $\kappa = \chi \rho C$ is the thermal conductivity, ρ and C are the density and specific heat of the condensed phase, ζ is the dimensionless fraction of the heat which is absorbed by the surface, and ΔH is the difference between the specific enthalpies of the solid and gaseous phases, given by

$$\Delta H = \Lambda - CT + C'T. \quad (3)$$

We impose the initial condition $T(x,0) = 0$. The quantity Λ in (3) is the specific heat of vaporization at $T=0$ K, and C' is the specific heat of the gas phase. The quantity v_1 in (2) is the velocity at which the gas escapes from the surface directly behind the Knudsen layer.¹ The magnitude of this velocity depends on the ambient pressure, and it should in general be found from the solution of the problem of the motion of vapor in a surrounding gas.

In limiting cases, however, there is a simpler way to find the values of v_1 . If the vapor pressure at the surface is high in comparison with the ambient gas pressure, we have the Chapman–Jouguet condition:

$$v_1 - v \approx (\gamma RT_1/A)^{1/2}, \quad (4)$$

where (4) is the Chapman–Jouguet condition, γ is the ratio of specific heats, R is the universal gas constant, A is the atomic weight of the medium, and T_1 is the gas temperature behind the Knudsen layer ($T_1 = 0.67 T_s$, where $T_s = T|_{x=0}$ according to Ref. 1). Setting $C_v = 3R/A$, and assuming a monatomic gas, we find

$$\Delta H + \frac{v_1^2}{2} = \Lambda - 0.77 \frac{RT_s}{A}. \quad (5)$$

In the other limiting case, $v_1 \ll (\gamma RT_1/A)^{1/2}$, there are no discontinuities in the thermodynamic properties in the Knudsen layer; in this case we have

$$\Delta H + \frac{v_1^2}{2} = \Lambda - 0.5 \frac{RT_s}{A}. \quad (6)$$

The latter value was used in Ref. 1. We see that the difference between (5) and (6) is small. We can thus treat the problem of the surface evaporation and the problem of the vapor-expansion dynamics separately. The system of equations (1) and (2) is supplemented by an expression for the surface-evaporation velocity. This velocity can be found from the equation of state of the metal; the result is

$$v_s = v_0 \frac{T_b}{T_s} \exp\left(-\frac{\Lambda A}{RT_s}\right), \quad (7)$$

where v_0 is a quantity on the order of the sound velocity in the cold metal, and T_b is the boiling point for the given pressure of the surrounding gas.

Equation (1) with boundary condition (2) can be reduced to a single equation for the temperature T_s . We integrate Eq. (1) from over x from 0 to ∞ , and we make use of boundary condition (2). We find

$$c\rho \partial \left[\int_0^\infty T(x,t) dx \right] / \partial t = \zeta q(t) - \rho v_s (\Lambda + bRT_s/A), \quad (8)$$

where $b = 3 - a$, and a is the numerical coefficient in Eq. (5) or (6).

To evaluate the integral in (8), we find a solution of Eq. (1) for an arbitrarily varying temperature $T_s(t)$. Generalizing the calculations in Ref. 8 to the case of a heat-conduction equation with a moving interface, (1), we find

$$T(x,t) = \frac{1}{\sqrt{\pi\chi}} \int_0^t \frac{d\tau T_s(\tau)}{(t-\tau)^{3/2}} [x + \Delta x(t-\tau)] \times \exp\left[-\frac{[x + \Delta x(t-\tau)]^2}{4\chi(t-\tau)}\right], \quad (9)$$

where $\Delta x(t) = \int_0^t v_s(\tau) d\tau$ is the displacement of the evaporation front over a time t . In the absence of such a displacement, expression (9) becomes the solution given in Ref. 8. Substituting (9) into (8), integrating over x , and then differentiating with respect to t , we find

$$c\rho \sqrt{\frac{\chi}{\pi}} \int_0^t \frac{d\tau}{(t-\tau)^{1/2}} \frac{dT_s}{d\tau} \exp[-\Delta x(t-\tau)^2/4\chi(t-\tau)] = \zeta q(t) - \rho v_s \left(\Lambda + \frac{bRT_s}{A} \right). \quad (10)$$

Closed equation (10) gives the $T_s(t)$ dependence of the 1D Stefan problem.

The nature of the solution of Eq. (10) is clear at a qualitative level. In the early stages, the evaporation is negligible, and we have the purely thermal-conductivity

solution. As the heating proceeds, the term associated with evaporation grows exponentially, and at some time we find

$$\xi q \approx \rho v_s \left(\Lambda + \frac{bRT_s}{A} \right). \quad (11)$$

This is the well-known condition for rapid evaporation.^{1,3,4} Under these conditions, the terms on the right side in (8) are large in comparison with the integral term. The latter is determined from the difference between two large quantities. Since the evaporation rate is an exceedingly strong function of the temperature, the transition from one regime to the other occurs in a narrow interval of parameter values.

2. HEATING AND EVAPORATION OF A PLANE SURFACE BY LASER LIGHT

Let us examine the effect of laser light with an intensity $q(t)$ on a plane metal surface. We assume that the vapor is transparent to the light. We also assume $v_s t \ll \sqrt{\chi t}$, i.e., that the "thermal wave overtakes the evaporation wave" (if the opposite inequality held, the regime of rapid evaporation would set in immediately, and the thermal conductivity would be unimportant). In this case we can omit the exponential function from (10); this simplification is equivalent to ignoring the term with v_s in (1) (but not in the boundary condition).

We use some dimensionless variables: $\Lambda A/R$ for the temperature, $\Lambda \rho v_0/\xi$ for the intensity, and $9\chi/v_0^2$ for the time. For copper with $\xi=0.05$ we have $\Lambda A/R=40\ 600$ K, $\Lambda \rho v_0/\xi=2.4 \cdot 10^{10}$ W/cm², and $9\chi/v_0^2=10^{11}$ s. In terms of these dimensionless variables, Eq. (10) takes the following form, after the simplifications we just mentioned:

$$\frac{1}{\sqrt{\pi}} \int_0^t \frac{d\tau}{(t-\tau)^{1/2}} \frac{dT}{d\tau} = q(t) - (1+bT) \exp\left(-\frac{1}{T}\right), \quad (12)$$

where $T(t)=T_s(t)$. We will suppress the index s below, since only the surface temperature is involved in the discussion. We see that in terms of these dimensionless variables the dependence on the thermal properties of the material has disappeared. It is easy to show that we can rewrite (12) in the equivalent form

$$T(t) = \frac{1}{\sqrt{\pi}} \int_0^t \frac{d\tau}{(t-\tau)^{1/2}} \left[q(\tau) - (1+bT) \right] \times \exp\left(-\frac{1}{T}\right). \quad (13)$$

In order to determine the time evolution of the surface temperature we thus need to solve one of these nonlinear integral equations, either (12) or (13). However, there is yet another approach. We introduce the auxiliary function $\theta(t,\tau)$, which satisfies the integral equation

$$\theta(t,\tau) = \frac{1}{\sqrt{\pi}} \int_0^\tau \frac{d\tau'}{(t-\tau')^{1/2}} \left\{ q(\tau') - [1+b\theta(t,\tau')] \exp\left(-\frac{1}{\theta(t,\tau')}\right) \right\}. \quad (14)$$

The temperature $T(t)$ in which we are interested is obviously related to $\theta(t,\tau)$ by

$$T(t) = \lim_{\tau \rightarrow t} \theta(t,\tau). \quad (15)$$

The meaning of this auxiliary function becomes clear when we differentiate (14) with respect to τ . Doing this, we find the differential equation

$$\sqrt{\pi}(t-\tau)^{1/2} \frac{d\theta(t,\tau)}{d\tau} = q(\tau) - [1+b\theta(t,\tau)] \times \exp\left[-\frac{1}{\theta(t,\tau)}\right], \quad (16)$$

which holds for $0 < \tau \leq t$. The problem then reduces from one of solving integral equation (13) to one of solving differential equation (16) and taking the limit in (15). The latter approach is simpler.

3. SOLUTION OF EQUATION (16)

Equation (16) belongs to a class of equations analyzed in the Appendix. The method described there can be used to solve it. We set

$$A = -\frac{\theta(\tau)}{T_b} \frac{1+b\theta(\tau)}{[\pi(t-\tau)]^{1/2}},$$

$$B = \frac{q(\tau)}{[\pi(t-\tau)]^{1/2}}.$$

Since Eq. (14) is compatible with homogeneous boundary conditions, the solution is given by Eq. (A8). Substituting the corresponding A and B into it, we find

$$\theta = L + \frac{\theta^2}{f} \ln \left\{ 1 - f(1+b\theta) \exp\left(-\frac{1}{\theta} - \frac{fL}{\theta^2}\right) \frac{1}{\sqrt{\pi\theta^2}} \int_0^\tau \frac{d\tau'}{(t-\tau')^{1/2}} \exp\left[\frac{fL(\tau')}{\theta^2}\right] \right\}, \quad (17)$$

where

$$\theta = \theta(t,\tau), \quad f = 1 - 2\theta + b\theta^2/(1+b\theta),$$

$$\frac{1}{\sqrt{\pi}} \int_0^\tau \frac{q(\tau') d\tau'}{(t-\tau')^{1/2}} = L(t,\tau). \quad (18)$$

The function $L(t,\tau)$ depends on only the shape of the laser pulse. The differential equation

$$\sqrt{\pi} \frac{dL(t,\tau)}{d\tau} = \frac{q(\tau)}{(t-\tau)^{1/2}} \quad (19)$$

also holds for this function. Using (19), we can eliminate $d\tau'/(t-\tau')^{1/2}$ from the integral in (17). The function $\exp[L(\tau')/T^2]$ has an extremely strong time dependence. Therefore, the function $q(\tau')$ in the integral which results can be taken outside the integral sign at the upper limit, $\tau'=t$, without any great error, since this integral is dominated by values of τ' close to this limit. Evaluating the integral in this manner, and taking the limit $\tau \rightarrow t$, we find

$$T = L + \frac{T^2}{f} \ln \left[1 - \frac{(1 + bT) \exp(-1/T)}{q} \right], \quad (20)$$

where $L(t) = \lim_{\tau \rightarrow t} L(t, \tau)$. Here are some values of f and f/T for certain temperatures:

T	0.07	0.08	0.0866	0.12	0.15
f	0.86	0.85	0.84	0.79	0.70
f/T	12.3	10.7	9.7	6.6	4.7

Expression (20) gives T as a function of q and L and does not have an explicit time dependence. Using (20), we can easily write the dimensionless evaporation velocity in the form

$$\exp\left(-\frac{1}{T}\right) = \frac{q}{1 + bT} \left[1 - \exp\left[-\frac{f(L - T)}{T^2}\right] \right]. \quad (21)$$

If we take the limit $L \rightarrow T$, which corresponds to the temperature variation in the purely thermal-conductivity regime, in (19), we find $\exp(-1/T) \rightarrow 0$. If, however, the argument of the exponential function satisfies $(f/T^2)(L - T) > 1$, then we have

$$\exp\left(-\frac{1}{T}\right) = \frac{q}{1 + bT}. \quad (22)$$

This is the case of rapid evaporation. The transition from one regime to the other occurs very sharply, because of the large quantity f/T^2 in the argument of the exponential function. Setting $(f/T^2)(L - T) \approx 1$, we thus find

$$L = T(1 + T/f) \quad (23)$$

as the value of L at which the regime of rapid evaporation is effectively established. Since we have $T/f \ll 1$, the quantity L is only negligibly higher than the values corresponding to the purely thermal-conductivity regime. A universal family of isotherms has been plotted in the plane of the dimensionless variables q and L on the basis of (20) in Fig. 1 (here $b = 2.5$; the isotherms are labeled with the corresponding values of T/Λ). We see two asymptotes: one vertical and one horizontal. The former corresponds to $T = L$, i.e., to the purely thermal-conductivity solution. The latter corresponds to condition (11), i.e., to the regime of rapid evaporation. The dashed curve was plotted through a joint solution of Eqs. (22) and (23); it forms the boundary between these two regimes. We see that the transition region is narrow. We can say that the thermal-conductivity solution is valid on the left, while the rapid-evaporation solution is predominant on the right.

We have used the same figure to draw some isotherms corresponding to the heating of the material to the boiling point T_b . For metals, the values of the ratio T_b/Λ lie in the rather narrow interval^{5,6} 0.05–0.09. The extreme values of this interval correspond to low-melting metals (Zn and Pb) and refractory metals (W and Ta). For copper we would have $T_b/\Lambda \approx 0.07$. The corresponding isotherm is singled out in Fig. 1. In the region below this line or to its left the condition $T < T_b$ holds, and there is a diffusive regime of vapor outflow. The vapor diffuses slowly into the surrounding gas. A vapor-gas mixture forms. In the region above this line, $T > T_b$, we have a hydrodynamic regime of

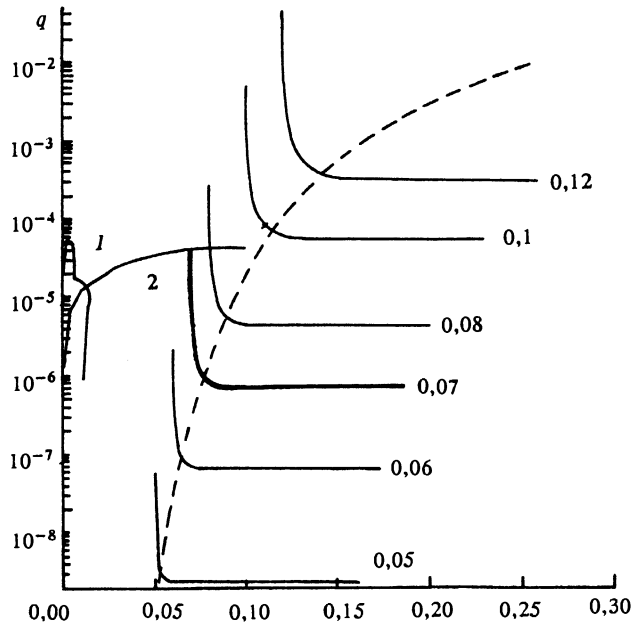


FIG. 1. Isotherms in the (L, q) plane. Dashed line—boundary between the thermal-conductivity regime (on the left) and the rapid-evaporation regime (on the right); 1—pulse from Ref. 9 characteristic of TEA lasers; 2—pulse from Ref. 10.

vapor outflow. In this case the vapor displaces the gas around the target and forms an erosion burst of vapor of the target material.

Note that a laser pulse of arbitrary shape can be plotted in the coordinates q and L . The position of the pulse in this plane depends on the surface material. From the intersection of this pulse with the isotherms we can draw conclusions about the heating dynamics, the nature of the vapor outflow, the time at which the evaporation regime sets in, and more. For pulses which vary in accordance with a power law, $q \propto t^\alpha$, it is a simple matter to calculate L :

$$L = \frac{E f(\alpha)}{\sqrt{t} \sqrt{\pi}},$$

where

$$f(\alpha) = (\alpha + 1) \int_0^1 \frac{u^\alpha du}{(1-u)^{1/2}} = \sqrt{\pi}(\alpha + 1) \frac{\Gamma(\alpha + 1)}{\Gamma(\alpha + 3/2)}$$

is a numerical factor which depends on the exponent α , and $\Gamma(x)$ is the gamma function. Here are values of $f(\alpha)$ for various values of α :

α	-0.9	-2/3	-1/2	0	1	2	3	4
f	1.91	1.41	$\sqrt{\pi}/2$	2	8/3	16/5	128/35	256/63

The quantity L has also been calculated for a very short intense pulse which is applied at the time t_1 and which can be approximated by a δ -function:

$q(t) = E\delta(t-t_1)$. In this case we have $L = E/\sqrt{(t-t_1)\pi}$. We see that we have $L \sim E(T)/\sqrt{t}$ for a wide class of functional dependences $q(t)$.

After this analysis it is a simple matter to determine the particular shape a laser pulse should have if the energy required to heat the target to a given temperature T over a time t is to be minimized. To answer this question we write E as $E = \sqrt{t\pi T/f_\alpha}$ or, for a δ -function pulse, $E = \sqrt{\pi(t-t_1)T}$. We see that from this point of view the pulses should have a front which initially rises slowly and then progressively faster: $q(t) \propto t^\alpha$, $\alpha > 1$. In the limit $\alpha \rightarrow \infty$ a pulse of this sort can be modeled by a δ -function which is "turned on" at a time $t_1 < t$. The energy required for the heating is minimized in this case.

For a pulse of a given shape which is not a power law or a δ -function, we need to work from Eq. (18). There is no particular difficulty in doing so with the computer facilities available today. Figure 1 shows the microsecond-length pulse which was used in Ref. 9, applied to a copper target with $\xi=0.05$ (curve 2). This is a typical pulse for TEA lasers. Its total energy is 20 J/cm^2 at a length of 3500 ns . The peak intensity at $t=80 \text{ ns}$ is $2.8 \cdot 10^7 \text{ W/cm}^2$. This intensity falls off linearly from the peak to a value of $0.86 \cdot 10^7 \text{ W/cm}^2$ at $t=270 \text{ ns}$, and from this value it falls to zero at the end of the pulse. We see that the evaporation regime is not reached during heating of an ideally smooth plane target by a pulse of this sort.

Also shown in Fig. 1 is the millisecond-length pulse which was used in Ref. 10, again for copper. Its length is $\sim 1 \text{ ms}$, its intensity is a constant $\sim 20 \text{ MW/cm}^2$, and its rise time is $\sim 0.1 \text{ ms}$. In this case the evaporation regime is reached immediately after the hydrodynamic regime is reached. This pulse would be extremely efficient for heating and evaporating plane targets.

The nature of the vapor outflow and the evaporation of target material has been discussed in the literature for a long time. It has been suggested in many papers that under the condition $T > T_b$ the regime of rapid evaporation and the regime of a hydrodynamic vapor outflow coexist. It follows from Fig. 1 that a rapid evaporation can occur in either the case $T < T_b$ or the case $T > T_b$. To explain, we assume that a long pulse of relatively low-intensity, is incident on a target. This pulse intersects the dashed line before the target is heated to T_b . Intense evaporation occurs in the course of a diffuse outflow of vapor. When an intense short pulse is applied to the target instead, the target is heated to $T = T_b$, when the laser pulse intersects the vertical branch of the $T = T_b$ isotherm. However, rapid evaporation occurs when the pulse again intersects the dashed line. For a pulse of constant intensity, heating of the target to $T = T_b$ means a simultaneous transition to the hydrodynamic regime of vapor outflow.

4. BOUNDARY OF THE REGION OF RAPID EVAPORATION

Rapid evaporation occurs when some pulse intersects the dashed line, which determines the threshold values of L and q corresponding to this regime [Eqs. (22) and (23)].

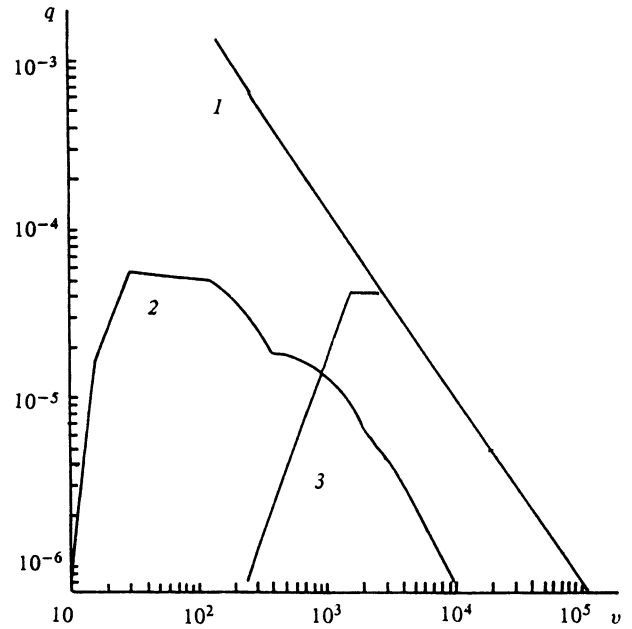


FIG. 2. 1—Dimensionless threshold values of q as a function of ϑ (curve 1); 2, 3—pulses from Refs. 9 and 10.

We can find the pulses corresponding to the beginning of the regime by dividing (22) by (23). As a result we find

$$\vartheta = \frac{L}{q} = \frac{T(1+T/f)\exp(1/T)}{1+bT}. \quad (24)$$

The quantity on the left in (17) depends on the length and shape of the pulse, while that on the right depends on the temperature. By specifying values of the former, we can find the temperature corresponding to the beginning of the evaporation regime; we can then use the latter temperature to calculate threshold values of L and q . For a power-law pulse we have $L/q = \sqrt{t}\Gamma(\alpha+1)/\Gamma(\alpha+3/2)$. In this case expression (24) gives the "delay time," i.e., the time which elapses from the beginning of the pulse to the beginning of the evaporation regime:

$$\sqrt{t} = \frac{T(1+T/f)\exp(1/T)}{(1+bT)\Gamma(\alpha+1)/\Gamma(\alpha+3/2)}. \quad (25)$$

In particular, for a constant density, $\alpha=0$, expression (25) agrees within a correction T/f with the expression given in Ref. 2.

Figure 2 shows a plot of $q(\vartheta)$ corresponding to the beginning of the evaporation regime, along with pulses from Refs. 9 and 10. Figure 2 confirms the conclusions which we drew above regarding the efficiency of these pulses.

Figure 3 shows the dimensional threshold values of q as a function of ϑ^2 , which has the dimensionality of a time, for the case in which the pulse is applied to a copper target. We see that in the millisecond range we have $q \sim 10^5 \text{ W/cm}^2$. Initially, the values of q required for rapid evaporation increase sharply. We see thus the reason why short

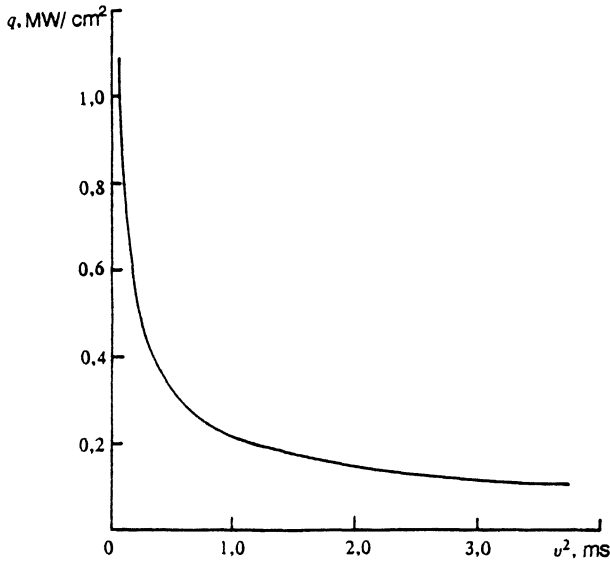


FIG. 3. Dimensional threshold values of q versus ϑ^2 for a copper target.

pulses are inefficient when applied to a cold target: Very high intensities are required to start the evaporation of such targets.

APPENDIX: MODIFIED METHOD FOR EXPANSION OF AN EXPONENTIAL FUNCTION

We consider a differential equation

$$\frac{dT}{dt} = A(T, t) \exp\left(-\frac{1}{T}\right) + B(t, T). \quad (\text{A1})$$

The variables in (A1) are dimensionless; the values of T can be small, so the exponential dependence is very strong. The functions A and B are relatively smooth. The idea of the method is to take the exponential dependence into account as accurately as possible in an approximate consideration of some functions which vary more slowly.

Let us consider a point which satisfies Eq. (A1), e.g., t_1, T_1 (point 1). We introduce the relative coordinates $T' = T/T_1$. We rewrite Eq. (A1) in terms of these coordinates, omitting the primes:

$$\frac{dT}{dt} = \frac{1}{T_1} \exp\left(-\frac{1}{T_1}\right) A(T, t) \exp\left(\frac{1-1/T}{T_1}\right) + \frac{1}{T_1} B(x, T). \quad (\text{A2})$$

At the point t_1, T_1 we have $T=1$.

We introduce the new variable

$$U = f \frac{1-1/T}{T_1},$$

where f is a constant. Equation (A2) takes the form

$$\frac{dU}{dt} = \frac{f}{T_1^2} \exp\left(-\frac{1}{T_1}\right) \left(1 - \frac{T_1 U}{f}\right)^2 A(U) \exp\left(\frac{U}{f}\right)$$

$$+ B(t, U) \left(1 - \frac{T_1 U}{f}\right)^2 \frac{f}{T_1^2}.$$

The variable U ranges from $-\infty$ to f/T_1 , and in point 1 we have $U=0$ at $T=1$. We write

$$\left(1 - \frac{TU}{f}\right)^2 A(U) \exp\left(\frac{U}{f}\right) = \exp[F(U)].$$

We expand the function $F(U)$ in a power series in U around the point $U=0$:

$$F(U) \approx F(0) + F'(0)U.$$

We find then

$$F'(U) = \frac{1}{f} \left(1 - 2T_1 + \frac{d \ln A}{dT} \Big|_{T_1} \cdot T_1^2\right).$$

We set

$$f = 1 - 2T_1 + \frac{d \ln A}{dT} \Big|_{T_1} \cdot T_1^2.$$

Equation (A2) then becomes

$$\frac{dU}{dt} = a_1 e^U + b_1 B(t, T), \quad (\text{A3})$$

where

$$a_1 = \frac{A_1}{T_1^2} f \exp\left(-\frac{1}{T_1}\right) \frac{1}{T_1^2}, \quad b_1 = \frac{f_1}{T_1^2}.$$

We seek a solution of Eq. (A3) in the form

$$U = b_1 \int_{t_1}^t B(t', T_1) dt' + C(t), \quad (\text{A4})$$

where $C(t)$ is an unknown function. Substituting (A4) into (A3), we find the equation

$$\frac{dC}{dt} = a_1 \exp\left(b_1 \int_{t_1}^t B dt' + C\right). \quad (\text{A5})$$

A solution of this equation is

$$C = -\ln \left[1 - a_1 \int_{t_1}^t \exp\left(b_1 \int_{t_1}^{t'} B dt''\right) dt' \right]. \quad (\text{A6})$$

Substituting (A6) into (A4), and going back to T , we find

$$\exp\left(1 - \frac{1}{T}\right) = \exp\left(b_1 T_1 \int_{t_1}^t \frac{B dx}{f_1}\right) \left[1 - a_1 \int_{t_1}^t \exp\left(b_1 \int_{t_1}^{t'} B dt''\right) dt' \right]^g, \quad (\text{A7})$$

where $g = T_1/f_1$.

Equation (A7) is valid if T does not deviate from one too greatly. We therefore expand $1-1/T$ in powers of T near $T=1$. We find $1-1/T \approx T-1$. The final form of the solution depends on whether $\int_{t_1}^t B dt'$ exists as $t \rightarrow 0$. In this case, (A1) is compatible with homogeneous boundary con-

ditions. If this is the case, then we can set $t=0$ and $T=0$ in (A7), and we can treat point 1 as arbitrary, omitting the index 1 from (A7). We then go back to our original variable T . As a result we find

$$T = \int_0^t B dt + \frac{T^2}{f} \ln \left[1 + \left[\frac{A(T)}{T^2} f \exp\left(-\frac{1}{T}\right) \right] \right. \\ \times \exp\left(-\frac{f}{T^2} \int_0^t B dt\right) \\ \left. \times \int_0^t \exp\left(\frac{f}{T^2} \int_0^{t'} B dt\right) dt' \right]. \quad (\text{A8})$$

If, on the other hand, $\int B dx$ does not exist, then the values of T are close to one. We thus have $\exp(1-1/T) \approx T$, and from (A7) we find

$$t = \exp\left(b_1 T_1 \int_{t_1}^t \frac{B dt}{f}\right) / \left[1 - a_1 \int_{t_1}^t \exp\left(b_1 \int_{t_1}^{t'} B dt\right) dt' \right]^g. \quad (\text{A9})$$

Expression (A9) is written in terms of relative variables.

The modification of the method has formally led to the appearance of the function f in the final results.

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