85.
[12]G. A. Thomas and Ya. E. Pokrovskiĭ, Phys. Rev. 18 B, 864 (1978).
[13]A. Manoliu and C. Kittel, Sol. State Comm. 21, 635 (1977).
[14]V. F. Gantmakher and I. B. Levinson, Zh. Eksp. Teor. Fiz. 74, 261 (1978) [Sov. Phys. JETP 47, 133 (1978)].

Translated by J. G. Adashko

Instability of plane evaporation front in interaction of laser radiation with a medium

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(Submitted 18 October 1979)
Zh. Eksp. Teor. Fiz. 78, 1597–1605 (April 1980)

The stability of the boundary between condensed matter and its vapor is investigated in the case when the evaporation is produced by a laser beam. It is shown that at sufficiently high radiation intensities—more than 10^6 W/cm^2 for most metals—the evaporation front is unstable. The maximum instability growth rate is possessed by perturbations with wavelengths on the order of the depth of penetration of the light in the evaporated matter. The growth time of these perturbations is much shorter than the time of establishment of the quasistationary evaporation regime.

PACS numbers: 52.50.Jm, 52.35.Py, 52.40.Db

1. INTRODUCTION

The results of numerous experiments on the evaporation of condensed substances by laser radiation can be satisfactorily explained by using the surface-evaporation model.[1-3] According to this model evaporation comes from a thin (of the order of the interatomic distances) surface layer of condensed phase, to which the energy is transferred from the light-absorption region by heat conduction. Since the depth of penetration of the radiation into the condensed medium is always much larger than the thickness of the layer from which the evaporation takes place, the temperature near the phase boundary increases with increasing distance from the boundary, and reaches a maximum at a certain depth. It is easy to show that in this case the phase separation boundary is unstable. Indeed, when a certain section of the boundary shifts towards the more heated condensed phase, the heat flux to this section increases, and this accelerates the boundary and increases further the initial perturbation. Understandably, the amplitude of the boundary displacement can not exceed the thickness of the surface layer in which the heat flux is directed from the condensed phase toward the evaporation front. This surface layer, as will be shown below, can be destroyed by short-wave perturbations within a time much shorter than the time of establishment of the stationary evaporation regime.

The described evolution of the process presupposes absence of the volume evaporation that could develop in principle in a liquid layer or on the grain boundaries in a polycrystalline solid phase. It is important, however, that in those cases when the vapor is formed in the volume, the result is the same as in the considered instability of the surface evaporation, namely destruction of the superheated surface layer and dispersion of the liquid phase. The formation of metastable states of the condensed phase is therefore not very likely in experiments on laser evaporation. There is no point in discussing this question.

The considered instability-producing mechanism is, of course, not a feature of the laser-induced (or any other) evaporation process. The necessary condition for the instability is that when the heat is released in the volume (for example, under the influence of a shock wave or an electron beam), the phase transition with energy absorption take place not in the volume but on some surface. This situation is typical of melting and evaporation of solids, where the nucleation of the new phase calls for noticeable additional expenditure of energy.[4,5] An increase in the area of the surface on which the phase transition takes place leads to a decrease of the free energy of the system as a whole. For this reason, a plane phase boundary is unstable. We note that the boundary perturbations with the shortest wavelengths should be damped because of the increase in the surface energy.

Thus, the discussed phenomenon is of quite general character. Manifestations of the instability in question can be expected in a large group of experiments. We deal in the present article with experiments on laser evaporation of solids, since this topic has been well investigated experimentally and theoretically, so that the stability problem can, in particular, be correctly formulated. It seems to us, in addition, that the most suitable conditions for the observation of the evaporation instability are realized in experiments and technological processes in which lasers are used.

We point out that the mechanism of the onset of the thermal instability has been previously considered[6] in connection with the study of phase transitions accompanying electric explosion of conductors. The an-

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alysis in Ref. 6, however, dealt mainly with a model of the Stefan type with a given transition temperature. This model is not adequate for the description of laser evaporation,[2] since it does not take into account the real kinetics of the phase transition. Although the instability in question is of thermal origin, the perturbations of the evaporation front are determined in final analysis by the dependence of the evaporation rate on the temperature of the evaporating surface. This dependence should therefore be taken explicitly into account when the stability problem is formulated. It will be shown that within the framework of the Stefan problem it is impossible to determine the qualitatively correct behavior of the instability growth rate in the short-wave region. The short-wave perturbations of the temperature and of the shape of the front have in the Stefan problem an infinitely large growth rate, i.e., an initial quasistationary solution cannot exist at all during any finite arbitrary time interval. This situation is, of course, unsatisfactory, and a more adequate formulation becomes necessary.

Laser-induced evaporation has a number of other characteristic features that have been investigated in detail in the study of quasistationary evaporation waves[1-3,7]; owing to these features, the problem of the stability of laser evaporation calls for a special analysis. This is in fact the topic of the present article.

The exposition proceeds as follows. In Sec. 2 is described the formulation of the problem of laser-induced evaporation. In Sec. 3 we obtain a stationary solution of this problem and obtain its asymptotic form in the case when the temperature on the phase boundary is much less than the heat of evaporation. Section 4 is devoted to the linearized problem for small perturbations of the temperature field and of the evaporation front. It contains also a brief description of the instability problem in the Stefan formulation. The results are discussed in Sec. 5.

2. FUNDAMENTAL EQUATIONS

It is known[8,7] that in a rather wide range of laser intensities the processes that occur in the gas phase do not exert a noticeable influence on the motion of the evaporation front. In this case the front velocity and the condensed-phase temperature are determined by solving the heat-conduction problem with appropriate boundary conditions.[2,3,8] Assuming that the vapor does not absorb the radiation, we write the heat conduction equation in the form

$$c\rho\partial T/\partial t=\varkappa\Delta T+Q,$$
$$Q=(1-R)q_0\mu\exp\{-\mu[x-X(y,t)]\},\quad x>X(y,t). \tag{1}$$

It is assumed in (1) that the thermal-conductivity coefficient $\varkappa$ and the light-absorption coefficient μ do not depend on the temperature. This assumption is well satisfied for metals. In the general case, allowance for the temperature dependences of $\varkappa$ and μ does not lead as a rule to new qualitative results. We shall assume that the laser intensity changes little within a time on the order of χ/v_0^2, where $\chi=\varkappa/c\rho$ is the thermal diffusivity and v_0 is the velocity of the evaporation front in the quasistationary regime. The shape of the phase-separation boundary $X(y,t)$ is not known beforehand and should be determined from a solution of the problem (1) with appropriate boundary conditions. Following Refs. 7–9, we write them in the form[1)]

$$\varkappa|\nabla T|=\dot{X}(y,t)\rho\Delta w \quad \text{at} \quad x=X(y,t),$$
$$\Delta w=L-(c-0.67c_{ps})T(X,y,t)+v_g^2/2;$$
$$T\to T_\infty \quad \text{as} \quad x\to\infty. \tag{2}$$

Here $\dot{X}(y,t)$ is the rate of displacement of the phase boundary as a result of the evaporation, Δw is the jump of the enthalpy on the phase boundary and includes the heat of transition L and the kinetic energy $v_g^2/2$ of the expanding vapor. In the calculation of Δw we took into account the temperature jump in the Knudsen layer as determined in Ref. 8. We assume henceforth for simplicity $T_\infty=0$.

The evaporation rate $\dot{X}(y,t)$ is connected with the surface temperature of the condensed phase by the evaporation-kinetics equation, which is usually written in the form

$$\dot{X}(y,t)=c_0\exp[-U/T(X,y,t)]. \tag{3}$$

The activation energy in (3) depends generally speaking on the local curvature of the surface. The constants U and c_0 can be obtained from the experimental data on the saturated-vapor pressure. In estimates we can assume c_0 to be equal to the speed of sound in the condensed phase, and U equal to the heat of evaporation per atom.

Ready[1] used in place of conditions (2) and (3) the Stefan condition with a specified transition temperature equal to the boiling temperature at normal pressure. The parameters calculated in this formulation for the quasistationary state in a wide range of conditions do not differ greatly from those obtained by solving the problem (1)–(3). However, the character of the development of the evaporation instability are qualitatively different in the two cases, especially in the region of the short-wave perturbations.

3. THE STATIONARY PROBLEM

The thermal-conductivity problem (1)–(3) formulated above has a quasistationary solution in the form of a plane evaporation wave propagating with constant velocity v_0. The time of establishment of the stationary regime is of the order of[2,3] χ/v_0^2.

To obtain the solution, we change over in (1)–(3) to a moving coordinate system with constant velocity v_0 directed along the x axis. We introduce the dimensionless variables defined by the relations

$$\xi=\mu(x-v_0t),\quad \eta=\mu y,\quad \tau=\mu v_0t,\quad \lambda=aL/c,$$
$$u(\xi,\eta,\tau)=aT(x,y,t),\quad \delta(\eta,\tau)=-\tau+\mu X(y,t),$$
$$a=\mu\varkappa/(1-R)q_0,\quad b=v_0/\mu\chi.$$

After the transformation we obtain the equation

$$b\left(\frac{\partial u}{\partial\tau}-\frac{\partial u}{\partial\xi}\right)=\frac{\partial^2u}{\partial\xi^2}+\frac{\partial^2u}{\partial\eta^2}+\exp(\delta-\xi) \tag{4}$$

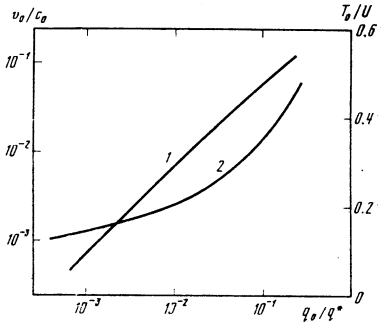

FIG. 1. Velocity of the front (1) and temperature on the phase boundary (2) in stationary evaporation.

with boundary conditions at $\xi = \delta$

$$b\left(1+\frac{\partial\delta}{\partial\tau}\right)(\lambda-0.26u)=|\nabla u|. \tag{5}$$

The problem (4)–(5) has a stationary solution

$$u_s(\xi)=Ae^{-\xi}+Be^{-b\xi}, \quad \delta(\eta,\tau)=0;$$
$$A=(b-1)^{-1}, \quad B=1.35A(0.26+\lambda(1-b)-b^{-1}). \tag{6}$$

From (6) and (3) we obtain an equation for $z \equiv \lambda b$:

$$z=\nu\exp\left(-\frac{mz}{1-z}\right), \tag{7}$$

where $\nu=\rho c_0 L/(1-R)q_0$ and $m=0.74cU/L$. Solving (7), we can obtain the stationary velocity of the evaporation front and the temperature of the condensed phase on the boundary. The calculated value of v_0/c_0 and T_0/U are shown in Fig. 1. Given m, the temperature and velocity depend on a single dimensionless parameter . The curves in Fig. 1 were calculated for the value $m = 2.22$, which corresponds to a heat capacity $3k_B$ per atom of the condensed phase (k_B is Boltzmann's constant).

We note that in all cases of practical interest we have $m \sim 1$, and ν is large compared with unity. This leads to a simple approximate solution of (7):

$$z=1+l-(m+l^2)^{1/2}, \quad l=0.5(m+\ln\nu),$$

from which we can obtain the front velocity and the temperature at all values of m.

4. THE STABILITY PROBLEM

Introducing the temperature perturbation

$$u(\xi,\eta,\tau)=u_s(\xi)+f(\xi)\exp(ik\eta+\gamma\tau)$$

($|f(\xi)| \ll |u_s(\xi)|$) and the perturbation of the phase boundary

$$\delta(\eta,\tau)=\beta\exp(ik\eta+\gamma\tau),$$

we obtain from (4) and (5), in the approximation linear in small f and β, an equation for $f(\xi)$:

$$f''+bf'-(b\gamma+k^2)f=-\beta e^{-\xi} \tag{8}$$

with the boundary condition at $\xi = 0 (u_0 \equiv u_s(0))$

$$f'(0)+0.26bf(0)=\beta[\gamma b(\lambda-0.26u_0)-0.26bu_0'-u_0'']. \tag{9}$$

We note that the initial condition (5) is imposed on the perturbed boundary $\xi = \delta(\eta, \tau)$. To change over to condition (9) at $\xi = 0$ we must expand the functions in (5) in powers of the small δ.

The second boundary condition follows from the equation for the evaporation rate referred to the surface $\xi = 0$ and linearized in f. This condition calls for some additional discussion. In laser-induced evaporation experiments the vapor pressure at the irradiated surface is usually much higher than the pressure of the environment, so that an adequate model for this process is evaporation in vacuum. In this case, even when condensation is taken into account[8] the mass flux and the evaporation rate are proportional to the saturated vapor pressure, from which in fact Eq. (3) follows in the case of a plane front. When the phase boundary is not plane, the vapor pressure and the evaporation rate depend on the curvature of the boundary, with the convex sections of the surface evaporating more rapidly than the concave ones. It is clear that this should stabilize the perturbations with sufficiently large k. This effect can be taken into account[4] by introducing in (3) the effective value U_{eff}, which is a function of the local curvature radius. For the perturbations considered here we obtain, following Ref. 4,

$$U_{eff}=U-pr^{-1}, \quad p=\sigma M/\rho k_B,$$

where σ is the surface-tension coefficient, M is the mass of the atom, ρ is the density of the solid, and r is the boundary curvature radius. In the approximation linear in the amplitude we must put $r^{-1}=\partial^2 X/\partial y^2$. The kinetic boundary condition then takes the form

$$f(0)=\beta[\gamma u_0^2/aU+\Lambda u_0 k^2-u_0'], \tag{10}$$

where $\Lambda = \mu p/U$. For a rough estimate of Λ we note that both terms of U_{eff} become of the same order when r is of the order of the interatomic distances d. This leads to the estimate $\Lambda \sim \mu d \ll 1$.

Integrating (8) with the boundary conditions (9) and (10) we obtain in the usual manner the dispersion equation for the unstable modes in parametric form, with the parameter α:

$$k^2=\alpha(\alpha-b)-b\gamma,$$
$$\gamma=\frac{(\alpha-b)[h-(\alpha-b+1)^{-1}-\alpha(\alpha-0.26b)\Lambda u_0]}{h+(\alpha-0.26b)(s-\Lambda b u_0)}, \tag{11}$$

where $h = 1-bu_0 = u_0'$ and $s = u_0^2/aU$.

It is easily seen that at large and small k the growth rate $\gamma < 0$, i.e., the corresponding modes are damped. As $k \to 0$ the growth rate $\gamma(k)$ vanishes. If the intensity of the laser radiation is high enough, i.e., if the parameter ν is not too large, then the function $\gamma(k)$ has two more zeros, k_1 corresponding to $\alpha_1 \approx b-1+h^{-1}$, and $k_2 \approx \alpha_2 \approx [bh/\Lambda(1-h)]^{1/2}$. The perturbations with wave numbers lying between k_1 and k_2 are unstable. Figure 2 shows plots of the function $\gamma(k)$ for the parameter values $\nu = \lambda = 20$, $\Lambda = 10^{-3}$ (curve 1) and $\nu = \lambda = 100$, $\Lambda = 10^{-5}$ (curve 2).

Inside the instability interval, the function $\gamma(k)$ has a maximum. This maximum growth rate γ_m and the corresponding wave number k_m are shown in Fig. 3 for $\Lambda = 10^{-5}$ as functions of the dimensionless laser intensity q_0/q^* ($v = \lambda$).

When the laser intensity is low enough, $\gamma(k)$ turns out

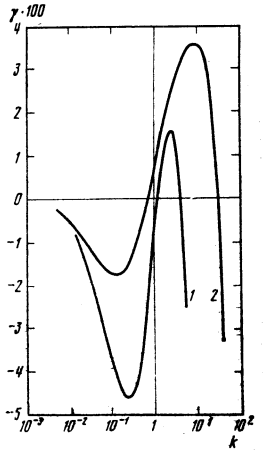

FIG. 2. Instability increment as a function of the wave number: 1—$\nu=\lambda=20$, $\Lambda=10^{-3}$; 2—$\nu=\lambda=100$, $\Lambda=10^{-5}$.

to be negative for all $k\neq 0$. This corresponds to stable propagation of the plane evaporation front. The critical intensity corresponding to the appearance of the unstable mode depends on the parameter Λ. The stability limit is shown in Fig. 4 ($\nu=\lambda$).

At very small Λ the maximum value of the increment can be calculated in simple manner if it is noted that the maximum is reached at values $k=k_m\gg 1$, and the temperature perturbations f are at large k of the order of β/k. In other words, at large k the perturbations of the phase boundary evolve against a background of an unperturbed temperature field. Putting $f(0)=0$ in (10) and performing simple calculations, we get

$$\gamma_m\approx\frac{aU}{u_0^2}[k-0.6(u_0\Lambda)^{1/3}].$$

Changing to dimensional variables and leaving out the second term in the square brackets, which is several times smaller than the first, we obtain an approximate estimate of the maximum growth rate:

$$\tilde{\gamma}_m\approx v_0 U T_s'(0)/T_s^2(0). \qquad (12)$$

From the last expression we can see directly the connection between the unstable phase boundary and the direction of the temperature gradient in its vicinity: namely, instability takes place when the temperature gradient and the gradient and the velocity of the boundary have the same direction. Formula (12) can be interpreted also somewhat differently if it is noted that it can be written formally, with the aid of (3), in the form

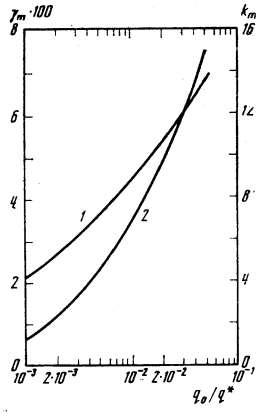

FIG. 3. Maximum instability growth rate (2) and corresponding value of the wave number (1) as a function of the radiation intensity.

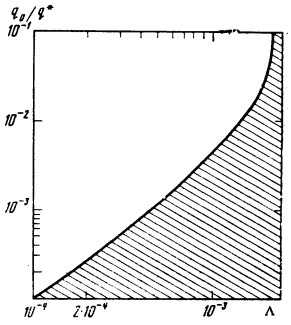

FIG. 4. Stability boundary. The shaded region corresponds to stable evaporation regimes.

$$\tilde{\gamma}_m\approx\frac{dv_0}{dT_0}T_0'=\frac{dv_0}{dx}.$$

The growth rate is positive if the forward displacement of the front causes an increase in its velocity. In this case the temperature field is regarded as unperturbed, a correct assumption in the short-wave limit.

From the derivation of (12) it is understandable that the estimate for the instability growth rate remains valid for weakly nonstationary evaporation if v_0 and T_0 are regarded as slowly varying functions of the time.

We now dwell briefly on the model of the phase transition with given temperature. This model corresponds to Stefan's classical problem and was considered, in connection with the problem of the interaction of laser radiation with matter in Ref. 1. The stability of the phase boundary in the Stefan problem was investigated by another method in Ref. 6. A calculation of the growth increment, in analogy with that described above, leads to the dispersion equation

$$\gamma=(\alpha-b)\left[1-\frac{1}{b\lambda(\alpha-b+1)}\right],$$

from which it follows that at large wave numbers the growth rate increases with increasing k linearly. In terms of dimensional quantities, the asymptotic formula $\tilde{\gamma}=v_0\tilde{k}$ is valid. In view of the unlimited increase of the growth rate, it is strictly speaking incorrect to pose the stability problem for a model of the Stefan type. It is easily seen that this circumstance is connected with a fundamental shortcoming of the Stefan model, namely with the absence of an upper bound on the phase-boundary velocity. For this reason, the model becomes incorrect at sufficiently high temperature gradients.

5. DISCUSSION OF RESULTS

We have shown that under certain conditions a plane evaporation front is unstable. The maximum instability growth rate is of the order of

$$\tilde{\gamma}_m\sim v_0 U\frac{T_s'(0)}{T_s^2(0)}\sim\frac{v_0^2}{\chi}\left(\frac{U}{T_0}\right)^2.$$

It is clear that the growth time of the corresponding perturbations is much shorter than the time of establishment χ/v^2 of the stationary evaporation regime. The wavelength of the fastest growing perturbations is

close to the characteristic spatial scale of the temperature distribution.

The foregoing analysis of the instabilities contains a number of simplifying assumptions, introduced mainly to prevent extraneous details from complicating the exposition. We have assumed, first, that the optical and thermophysical characteristics of the condensed phase are independent of temperature. A more detailed investigation shows that allowance for this dependence does not alter the qualitative results of the investigation and can be significant only in the case of weakly absorbing dielectrics whose absorption coefficient varies radically with temperature.

Second, we have confined ourselves in our analysis to the case of normal incidence of the light. In this case the reflectivity of the flat surface changes when a perturbation $\delta(\eta, \tau)$ is superimposed on a quantity of the order of δ^2. In the linear stability problem this change need not be taken into account.

Third, we did not consider nonlinear effects, particularly those leading to stabilization of the perturbations that grow in the linear approximation.

All the foregoing questions call for additional study.

Experiments on laser evaporation of solids have established a number of rules that can be connected with a manifestation of the instability in question. Many workers have noted, for example in Ref. 7, that the start of intense evaporation of a metal change is accompanied by a drastic change in its reflectivity and in the angular distribution of the reflected light. It is known furthermore that an appreciable fraction of the products of metal disintegration under the influence of laser radiation is made up of liquid drops, and the average specific energy of the distintegration is always lower than the specific heat of the evaporation.[7] Several likely mechanisms have been proposed for the appearance of liquid drops in the disintegration products. There are no grounds for excluding the considered instability and of the associated dispersal of the surface layer of the evaporated body from the list of most likely mechanisms. We note that the observed dimensions of the drops does not contradict the assumption that they were produced as a result of development of the instability in question.

Materials of interest as objects for the experimental study of the instability of evaporation induced by laser radiation are apparently metals and certain nonmetallic strongly absorbing substances with high vapor pressure at the melting temperature. A layer of molten material of this type has small thickness and the evaporation proceeds essentially from the solid phase. The development of the instability can be influenced by the presence of a polycrystalline structure in the material, since the boundaries of the single crystals can serve as centers of "volume" evaporation. In metals with single-crystal sizes on the order of a micron and with an absorption coefficient of the order of 10^5 cm^{-1} the presence of a polycrystalline structure should not affect at all the development of the instability. For substances with a smaller absorption coefficient such a structure might play the role of the initial perturbation that determines, at any rate during the initial stages, the spatial scale of the instability.

The authors consider it their pleasant duty to thank Ya. B. Zel'dovich and L. P. Pitaevskiĭ for helpful discussions.

[1)] A number of workers have proposed also other variants of boundary conditions for the solution of problems of thermal conduction with phase transitions. For the one-dimensional problem considered in Sec. 3 and for the linear stability problem in Sec. 4, all these variants are equivalent to the conditions (2).

[1] J. F. Ready, J. Appl. Phys. **36**, 462 (1965).
[2] S. I. Anisimov, A. M. Bonch-Bruevich, M. A. Él'yashevich, Ya. A. Imas, N. A. Pavlenko, and G. S. Romanov, Zh. Tekh. Fiz. **36**, 1273 (1966) [Sov. Phys. Tech. Phys. **11**, 945 (1967)].
[3] S. I. Anisimov, Teplofiz. vys. temp. **6**, 116 (1968).
[4] L. D. Landau and E. M. Lifshitz, Statisticheskaya Fizika (Statistical Physics), Moscow, Nauka, 1976 [Pergamon].
[5] I. M. Lifshitz and L. S. Gulida, Dokl. Akad. Nauk SSSR **87**, 523 (1952).
[6] A. M. Iskol'dskii, V. K. Pimus, and Ya. G. Épel'baum, Preprint No. 59, Inst. of Automation and Electrometry, Siberian Division, USSR Academy of Sciences, Novosibirsk, 1977.
[7] S. I. Anisimov, Ya. A. Imas, G. S. Romanov, and Yu. V. Khodyko, Deĭstvie izlucheniya bol'shoĭ moshchnosti na metally (Action of High-Power Radiation on Metals), Moscow, Nauka, 1970.
[8] S. I. Anisimov, Zh. Eksp. Teor. Fiz. **54**, 339 (1968) [Sov. Phys. JETP **27**, 182 (1968)].
[9] H. S. Carslaw and J. C. Jaeger, Conduction of Heat in Solids, Oxford, 1959.

Translated by J. G. Adashko