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Estimate of pressure produced during explosive boiling of a liquid film on a substrate heated by laser pulses

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***Abstract*. Experimental data of Lang and Leiderer [*New J. Phys. 8, 14 (2006)*] on the movement of a thin liquid film due to its explosive boiling on a substrate heated by a nanosecond laser pulse are analysed. It is shown that the maximum value of the pressure pulse produced in this case without any singularities may be much lower than the estimate obtained by these authors and is in good agreement with the assumption that the explosion is initiated in the vicinity of the spinodal.**

Keywords: *explosive boiling, thin films, evaporation kinetics.*

1. Introduction

Apart from its various practical applications, the explosive boiling of a liquid initiated by laser pulses is also a source of information about the behaviour of matter under extreme thermodynamic conditions and the kinetics of highly nonequilibrium phase transitions (see, for example, [1 – 4] and references therein). The ab initio simulation of such processes is a quite complicated problem that has not been completely solved so far. Therefore, it is reasonable to use simpler phenomenological approaches like the one employed recently by the authors of [1] for analysing the behaviour of pressure created in a cavity following the onset of explosive boiling between a transparent liquid film and the surface of a substrate absorbing laser radiation. However, the value of the maximum pressure obtained by these authors, which almost coincides with the critical pressure of the liquid (isopropanol) used in their investigations seems overstated due to an unsubstantiated choice of the pressure dependence on the cavity volume.

The pressure in the cavity was not measured in [1]. The second derivative of the experimental curve describing the time dependence of the cavity width was also not used for calculating the pressure perhaps due to the large errors involved in such calculations. It will be shown below that for the maximum value of pressure considered in [1], the characteristic time of pressure variation turns out to be considerably (by two orders of magnitude) shorter that the duration of the positive acceleration of the film, when the pressure in the cavity exceeds the external atmospheric pressure.

The aim of this paper is to estimate the maximum pressure in a cavity using the experimental data on the ejection of a film from the substrate after explosive boiling and more general assumptions about pressure variation than in [1].

2. Formulation of the problem

In the one-dimensional case corresponding to the experimental conditions [1], the velocity $V(t) = \mathrm{d}l/\mathrm{d}t$ of an escaping film of thickness h and density ρ is determined by the difference between the pressure $P(t)$ in a cavity of width l and the external atmospheric pressure P_{at}: $\Delta P(t) = P(t) - P_{\mathrm{at}}$ (Fig. 1). The equation for l can be written in several equivalent forms:

$$\rho h V = \int_0^t \Delta P(t)\mathrm{d}t, \tag{1}$$

or

$$\rho h \frac{\mathrm{d}^2 l}{\mathrm{d}t^2} = \Delta P[l(t)]. \tag{2}$$

The pressure $P(t)$ is the result of explosive boiling of a very thin film layer (of the order of ten nanometres) formed in the immediate vicinity of the pulse-heated substrate and superheated to a temperature exceeding the normal boiling point of the liquid. The thickness of this superheated layer is

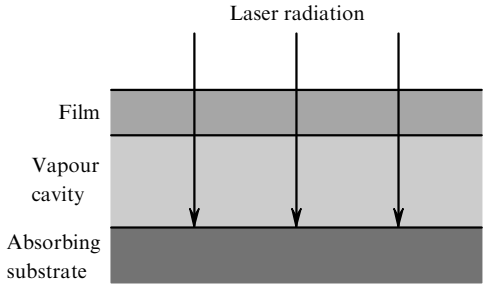

Figure 1. Scheme showing one-dimensional ejection of a transparent liquid film after explosive boiling of its thin lower layer initially adjoining the absorbing substrate heated by laser radiation, and the formation of an expanding vapour cavity.

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assumed to be small in comparison to h and the pressure build-up time is assumed to be much shorter than the characteristic time of its further evolution. In other words, the highest pressure P_0 is attained at the beginning of the pressure pulse at the instant $t = 0$. In the approach used by us, the last constraint is not significant and can be easily discarded.

The pressure dependence $P(l)$ in the cavity was simulated in [1] by the polytropic dependence $Pl^{\gamma} = \text{const}$ with a constant polytropic exponent $\gamma = \text{const}$. However, it is hard to substantiate such an assumption since the number of particles in the cavity does not remain constant on account of evaporation. A more appropriate phenomenological approach to this process involves the investigation of various versions of the time dependence $\Delta P(t)$ leading to simple analytic expressions connecting experimentally measured quantities with the highest pressure (which was not measured directly in [1]). These formulas can then be used for obtaining the dependence of the highest pressure on the general behaviour of the pressure pulse normalised to unity at the peak.

3. Discussion of results

We consider the following time dependence $\Delta P(t)$ for the pressure pulse:

$$\Delta P(t) = P_0 f(t) = P_0\left[1 - \left(\frac{t}{t_1}\right)^{\alpha}\right], \tag{3}$$

where t_1 is the duration of the pressure pulse at the base, and the exponent $\alpha > 0$. We will not consider the situation in which the time $t > t_1$ and the quantity $\Delta P(t)$ becomes negative while the film escape velocity begins to decrease. It follows from Eqns (1) and (3) that

$$V(t) = \frac{P_0 t}{\rho h}\left[1 - \frac{1}{1+\alpha}\left(\frac{t}{t_1}\right)^{\alpha}\right], \tag{4}$$

$$l(t) = \frac{P_0 t^2}{\rho h}\left[\frac{1}{2} - \frac{1}{(1+\alpha)(2+\alpha)}\left(\frac{t}{t_1}\right)^{\alpha}\right]. \tag{5}$$

For $t = t_1$, we obtain from Eqns (4) and (5)

$$\frac{P_0 t_1}{\rho h V_1} = \beta, \quad \frac{l_1}{V_1 t_1} = \delta, \tag{6}$$

where

$$\alpha = \frac{1}{\beta - 1} = \frac{3 - 4\delta}{2\delta - 1}. \tag{7}$$

Expressions (6) define in fact the parameters β and δ, while Eqns (7) indicate that the following inequalities are valid:

$$\beta > 1, \quad \frac{1}{2} < \delta < \frac{3}{4}, \tag{8}$$

since $\alpha > 0$. The limiting value $\delta = 1/2$ corresponds to the case of a uniformly accelerated film when $\alpha \gg 1$ and the pressure $\Delta P(t)$ is almost constant during the interval t_1. In the opposite limiting case ($\alpha \ll 1$), the pressure at the very beginning of the pulse decreases quite rapidly from its highest value P_0 so that the pulse half-width at half-maximum $t_{1/2}$ becomes much smaller than t_1. This property of pulse (3) will be used in the following.

For $\alpha = 2$, we can derive a simple expression for the dependence of pressure on the cavity width:

$$\Delta P(l) = P_0\left[3\left(1 - \frac{5l}{9l_1}\right)^{1/2} - 2\right]. \tag{9}$$

In contrast to the polytropic dependence $Pl^{\gamma} = \text{const}$ with $\gamma = \text{const}$, formula (9) leads to a finite value of $\Delta P(l)$ for $l = 0$.

The model form of pulse (3) contains three parameters P_0, α and t_1, and the values of β and δ are not fixed when only P_0 and t_1 are assigned fixed values; instead, these quantities are only connected through formula (7). Using the experimental values $V = 50.9$ m s^{-1}, $t_1 = 8.3$ ns, $h = 100$ nm and $\rho = 0.78$ g cm^{-3} from [1] and assuming that ΔP must not exceed significantly the critical pressure $P_c = 4.7$ MPa of isopropanol, we obtain the following values for β and δ from Eqns (6) and (7) respectively at nearly critical value $P_c = 4.5$ MPa of the highest pressure:

$$\beta = 9.4, \quad \delta = \frac{3\beta - 2}{2(2\beta - 1)} = 0.74. \tag{10}$$

It can be found from Fig. 3 in [1] that at the instant $t = t_1$, the quantity $l(t) = l_1$ is about 380 nm. Hence, using (6), we obtain $\delta_{\text{exp}} \simeq 0.9$, which is slightly higher than the value of δ in formula (10). Using Eqn (3) in [1] and taking into consideration the assumption made there about the constancy $Pl^{\gamma} = \text{const}$ for $\gamma = 1$, we arrive at the relation $P_{\text{at}} l_1 = (3.5 \pm 1.5)$ Pa cm, which leads to the minimum value of $l_1 = 335$ nm. For such a value of l_1, the value of $\delta_{\text{exp}} = 0.79$ obtained from (6) is found to be closer to the limiting value $\delta = 0.75$.

However, the most important conclusion that can be drawn from Eqns (10) is the fact that for such large values of β ($\beta = 9.4$), the pulse halfwidth $t_{1/2} = 2^{1-\beta} t_1 = 3 \times 10^{-3} t_1 = 25$ ps turns out to be quite small, i.e., the time dependence of pressure acquires a very narrow peak. Assuming that the pressure pulse must not have such singularities, the highest value of pressure P_0 must be much lower than 4.5 MPa.

By setting $P_0 = 1.3$ MPa, we obtain

$$\beta = 2.72, \quad \delta = 0.69. \tag{11}$$

For such a decrease in the value of P_0, the value of δ does not change significantly in comparison with that given in (10), while the pulse halfwidth $t_{1/2} = 0.3 t_1$ increases by two orders of magnitude. The choice of the maximum pressure $P_0 = 1.3$ MPa also seems to be more appropriate than the nearly critical value $P_c = 4.5$ MPa since in this case the explosive boiling is likely to begin near the spinodal or at even lower temperatures $T < 0.9T$ and not near the critical temperature T_c itself. Recall that a 10 % – 15 % decrease in the liquid temperature relative to its critical value decreases the saturated vapour pressure many times, while the pressure in the cavity does not exceed the saturated vapour pressure corresponding to the temperature at the inner surface of the film.

It is interesting to note that the same values of β and δ as those presented in (11) can also be obtained with the help of the following simple form of the pressure pulse $\Delta P(t)$:

$$\Delta P(t) = P_0 f(t), \quad f(t) = 1 - \sin\frac{\pi t}{2t_1}. \tag{12}$$

Unlike formula (3) in which three parameters are involved, this expression contains just two parameters P_0 and t_1. It follows from Eqns (1) and (12) that

$$\beta = \frac{\pi}{\pi - 2} = 2.75, \quad \delta = \frac{\pi^2 - 4\pi + 8}{2(\pi - 2)} = 0.74, \tag{13}$$

$$t_{1/2} = \frac{t_1}{3}, \quad P_0 = 1.3\text{MPa}.$$

Using other simple forms $\Delta P(t) = P_0 f(t)$ for the pressure pulse, we arrive at slightly lower, but close values of β, δ and P_0:

$$\beta = \frac{1}{\delta} = \frac{\pi}{2}, \quad P_0 = 0.75 \text{ MPa for } f(t) = \cos\frac{\pi t}{2t_1}, \tag{14}$$

$$\beta = \frac{1}{\delta} = 2, \quad P_0 = 0.95 \text{ MPa for } 2f_-(t) = 1 - \cos\frac{2\pi t}{t_1}. \tag{15}$$

In contrast to all other pulses considered by us (which have a sharp leading front and a peak at $t = 0$), the function $f_-(t)$ corresponds to the pressure pulse having a symmetric shape with respect to its peak at the instant $t = t_1/2$.

Since all the functions $f(t)$ – from (3) for $\beta = 2.72$ (11) and from (12), (14) and (15) – give close values for the highest pressure P_0, P_0 does not depend on the shape of the pressure pulse without any singularities of the type emerging in function (3) with small values of α ($\alpha \ll 1$), when $t_{1/2} \ll t_1$. The methodical advantage of using function (3) lies in that the pressure pulse distortion and its dependence on the highest value are revealed quite clearly when this function is used.

4. Conclusions

Thus, using the available experimental data [1] on the kinematics of an ejected film after its explosive boiling on a substrate heated by a laser pulse, as well as the equation of motion and the simplest assumption about the general form of the time dependence of the pressure pulse normalised to unity at the maximum, we can obtain an analytic estimate of the highest pressure which was not measured directly in [1].

This estimate is found to be about three or four times smaller than the value obtained in [1] from numerical computations and by assuming a polytropic dependence of the pressure in the cavity on its volume. Under such an assumption, almost all the evaporated particles are actually found to be in the cavity at the initial instant of time, which leads to a considerable increase in the initial (highest) pressure P_0 and to a distortion of the entire pattern of evaporation during acceleration of the escaping film. Our estimate of the quantity P_0 is also in accord with the assumption that explosive boiling begins at a temperature close to, or slightly lower than, the spinodal temperature [$T \sim (0.8 - 0.9)T_c$].

Under the experimental conditions in [1], the instability and inhomogeneity of the temperature profile may play a significant role in the superheated liquid layer whose characteristic thickness is comparable with the size of the critical nucleus formed as a result of fluctuations in the superheated metastable liquid. The radius of the critical nucleus decreases with temperature, and this may influence the initial stage of formation of the vapour cavity even before the ejection of the film from the substrate (see Fig. 4 in [1]). Subsequent increase in the substrate temperature following the formation of the vapour cavity may also affect the dependence $\Delta P(t)$ due to heating of the vapour as a result of its interaction with the substrate surface.

The versions of the phenomenological approach used in this research and in [1] do not shed any light on the microscopic processes determining the evolution of pressure in the expanding cavity whose size is found to be comparable with the mean free path of the particles in it. The behaviour of the pressure in the cavity may also be unstable in the sense that the form of the function $f(t)$ depends on the initial pressure P_0 and the film thickness h since an increase in the maximum velocity of the escaping film due to a change in these parameters makes the effect of finite evaporation rate even more significant; the maximum value of this quantity is determined by a relation of the type of Hertz–Knudsen formula [5]. For a low coefficient R of reflection of particles from the evaporation surface, the characteristic velocity is of the order of 100 m s^{-1}, which is comparable with the velocity of ejection of the film. For $R \sim 1$, this characteristic velocity may be much lower.

It should also be borne in mind that the steady-state recoil pressure acting on the evaporating surface depends on the rate of evaporation and decreases to about half the saturated vapour pressure as the evaporation flux increases from zero to its highest value (see, for example, [6, 7] and the literature cited therein). Such effects may lower the maximum momentum of the ejected film upon an increase in its velocity due to a decrease in its thickness, as observed in [1]. More detailed theoretical and experimental studies will provide qualitatively new information about the peculiarities of explosive boiling and kinetics of highly nonequilibrium evaporation.

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References

1. Lang F., Leiderer P. *New J. Phys.*, **8**, 14 (2006).
2. Kudryashov S.I., Allen S.D. *J. Appl. Phys.*, **95**, 5820 (2004).
3. Kim D., Park H.K., Grigoropoulos C.P. *Int. J. Heat Mass Transfer*, **44**, 3843 (2001).
4. Andreev S.N., Samokhin A.A. *Kr. Soobshch. Fiz. FIAN*, (8), 26 (2004).
5. Landau L.D., Lifshitz E.M. *Statistical Physics* (New York: Pergamon Press, 1980; Moscow: Nauka, 1976) Vol. 1.
6. Kartashov I.N., Samokhin A.A. *Kvantovaya Elektron.*, **33**, 435 (2003) [*Quantum Electron.*, **33**, 435 (2003)].
7. Kartashov I.N., Samokhin A.A., Smurov I.Yu. *J. Phys. D: Appl. Phys.*, **38**, 3703 (2005).