Action of a femtosecond laser pulse on thin metal film supported by glass substrate

V V Shepelev\textsuperscript{1,2}, N A Inogamov\textsuperscript{2,3}, S V Fortova\textsuperscript{1}, P A Danilov\textsuperscript{4}, S I Kudryashov\textsuperscript{4}, A A Kuchmizhak\textsuperscript{5,6} and O B Vitrik\textsuperscript{5,6}

\textsuperscript{1}Institute for Computer-Aided Design of the Russian Academy of Sciences, 19/18 Vtoraya Brestskaya St., Moscow 123056, Russia
\textsuperscript{2}Landau Institute for Theoretical Physics of the Russian Academy of Sciences, 1a Akademika Semenova Avenue, Chernogolovka, Moscow Region 142432, Russia
\textsuperscript{3}Dukhov Research Institute of Automatics (VNIIA), 22 Sushchevskaya, Moscow 127055, Russia
\textsuperscript{4}Lebedev Physical Institute of the Russian Academy of Sciences, 53 Leninsky Avenue, Moscow 119991, Russia
\textsuperscript{5}Institute of Automation and Control Processes of the Far Eastern Branch of the Russian Academy of Sciences, 5 Radio St., Vladivostok, Primorsky Krai 690041, Russia
\textsuperscript{6}School of Natural Sciences, Far Eastern Federal University, 8 Sukhanova St., Vladivostok, Primorsky Krai 690041, Russia

E-mail: vadim.aries@gmail.com

Abstract. Numerous papers are devoted to the problem of irradiating thin films on the substrate for surface nano-modification. But all of them concern the dynamics of the dynamics, paying almost no attention to substrate just as object of the first shockwave absorption. A different point of view with an emphasis on the dynamics of a substrate is presented. Under powerful laser action upon a thin metal film a hole arises. Its radius depends on the absorbed laser energy. Experimental results, quantitative theoretical model and numerical research are presented and show that the hole formation is influenced by propagation of the shock wave in the substrate, but not in the film itself. Four stages are considered. (i) Shockwave generation in a support because of an impact of a contact. (ii) Transition from one-dimensional to two-dimensional propagation of the shockwave. (iii) Lateral propagation of the shockwave along a film-support contact. And (iv) calculating pressure in the compressed layer behind the decaying shockwave. This positive pressure acting from substrate on the film accelerates the film in direction to vacuum. Above some threshold, velocity of accelerated film is enough to separate the film from support. In these cases the circle of separation is significantly wider than the focal laser spot on film surface.

1. Introduction
Laser surface nanostructuring is modern and intensively growing filed of studies. It has several subdirections, which differ in the ways of laser action and in resulting structures. At relatively weak intensities, wide spots, optical frequencies, bulk targets, and multiple repetitions the plasmon-polariton
mediated structures (called LIPSS) appeared; see recent review [1] and numerous references cited in this review.

At larger, than for LIPSS, absorbed energies and ultrashort laser pulse durations the random or chaotic surface structures are formed [2,3] in the chain of consecutive processes including melting, thermomechanical kick-off, nucleation of cavities, inertial inflation of foam, capillary deceleration of inflation, breaking of foam, and late stage freezing of remnants of cells and membranes of the broken foam.

![Diagram](image)

**Figure 1.** (a) shows initial bulging 4 of the film inside the illuminated spot and propagation of the shock 5 into glass. (b) presents situation later in time when expansion of the film into the substrate side stops and the region of negative pressures appears. (c) shows the instant pressure distribution in the shock compressed layer between the red and blue curves.

There are intermediate structures linked to the transition range of energies near the thermomechanical ablation threshold. Porous surface is formed in this range of energies [4]. If central fluence $F_c$ of a Gaussian beam is above the ablation threshold then the porous structures form a circle around the main crater. The solitary figures called also bumps or domed formations appeared after laser heating of a small spot. The paper is devoted to the analysis of solitary figures in the case of a thin film supported by substrate. We investigate the regime of rather large absorbed fluences when the interior of the film inflates after thermomechanical kick-off and flies away leaving a hole in the film. We managed to show that above some threshold a diameter of the hole is defined by the amplitude of a shock propagating in the substrate under the film.

Shock initiation in substrate by the impact of a laser driven plate was considered in works [5–7] for the case of an approximately planar shock in substrate with the reflection of the shock from a rear-side of a relatively thin substrate layer. On the contrary, here substrate is thicker in comparison to the spot diameter. Transition from planar to semi-spherical shock shape is described.

2. General view

There are many experimental and theoretical papers devoted to the problem of the thin films ultrafast laser irradiation, mounted on supporting substrate. Laser action transfers an initially smooth film into nanostructured one.
Thin films in consideration are thinner in comparison to a heat affected zone $d_f$. For gold $d_f \approx 150\text{nm}$. Their dynamics is qualitatively different from bulk targets cases; one-dimensional (1D) dynamics of bulk targets was studied in papers [33–41]. Experimental papers [9–15, 18, 20, 27] devoted to the film-substrate problem, do not consider processes which take place in substrate, so molecular dynamics-based numerical research or relatively faster hydrodynamic one is required (e.g. [23–25]).

The scheme of the problem in consideration is presented in figure 1, the substrate is situated under the film.

The shock propagating in a substrate is generated by expansion of a fast heated film into the substrate: a film is heated faster than a sound wave crosses the film: $t_{\text{heating}} \sim t_s = d_f / c_s$. The film is heated homogeneously across its thickness but not across the lateral extension $\sim R_L$ of a laser heated spot, because $d_f \ll d_r$. Thus the film-substrate contact in figure 1 acts as a piston driving a shock in substrate. This acceleration occurs during the time interval which duration has the similar order as acoustic time scale $t_s$, see [23–25].

After this time interval, the rarefaction from the vacuum side of a film arrives to the contact and the piston stops its pressing on substrate. Therefore the shock in the substrate transfers into the so called triangular shock. Shock compression of matter decreases behind the shock front. Shockwave decays during its propagation. If the absorbed fluence $F_{abs}$ is high enough, the shock is strong enough to cause the process of film separation even outside the spot $R_L$.

![Figure 2](image-url)

**Figure 2.** (a) shows the simulation box with equal horizontal and vertical spatial scales. Film zone is denoted as 1; silica substrate is 2, vacuum is 3. (b) shows velocities calculated with temporal pressure dependence $p(x_i, y = -40 \text{ nm}, t)$ taken from the simulation at $x_i = 0\text{nm}$, $x_i = 600\text{nm}$, $x_i = 1080\text{nm}$. We consider the two-dimensional (2D) plane motion, coordinate is $x, y$, where the horizontal axis $x$ is going along the initial contact line between the film and the substrate below the film, see figure 1. The vertical line $x = 0$ is the axis $y$ and the axis of a laser beam irradiating a film.

For modeling radiation absorption we start with temperature distribution, proportional to Gaussian absorbed energy distribution $F = F_r \exp(-r^2 / R_L^2)$, taking into account that for thin films the temperature distribution along normal direction is almost constant, but lateral temperature variation is significant.

Figure 2 presents the simulation box. We impose transmissive boundary conditions upon all four walls of the rectangular box to avoid reflection of SW. We use one-temperature two-dimensional
The hydrodynamic code (1T-2D-HD) thus neglecting some minor peculiarities linked to the two-temperature (2T) state. These peculiarities were studied in papers [23–25, 44, 45]. For bulk gold a 2T stage is analyzed in [46].

The initial stage of the process in its heated region is shown in figure 1(a). Shortly after subpicosecond pulse (at \( t \approx 10\,\text{ps} \)) approximately one-dimensional motion takes place. This means that pressure behind the shock wave (SW) (red curve in figure 1(a)) is proportional to the local absorbed fluence; indeed \( R_s \) is significantly larger than thickness \( d_f \) of a film. Pressure between the SW and

![Figure 3](image-url)

Figure 3. SEM visualization of the holes [27] created after the action of single laser pulses. Scale bars have 1 micron length. Energies of pulses are equal to 48, 64, 100, 240, 320, 400, 800, and 960 nJ. the film-glass contact is positive. Later in time the rarefaction wave propagating from the film-vacuum boundary crosses Au-glass (film is made from Au—gold) contact, stops motion of the Au-glass contact in the downward direction, and reduces pressures above the blue curve in figure 1(b) to negative values. Pressure field in the moving region inside the silica substrate is shown in figure 1(c).

Figure 1(b) shows expansion of the SW in glass substrate under the gold film. SW front shown as red curve in figures 1(a) and 1(b) moves down and along the film. The point A in figure 1(a) is the point where SW begins to interact with the film. Pressure is positive in the compressed layer of glass between the blue and red curves in figure 1(b). Compressed layer forms a half-spherical shell. Pressure in the shell accelerates film in the upper direction, towards vacuum at the interval \( A-B \) in figure 1(b). Acceleration has been occurring during a finite time interval. Let us consider the observation point \( A \). The SW achieves it at the instant shown in figure 1(b). After that during the time interval needed for the compressed layer to pass by the observation point. This point accumulates vertical component of momentum

\[
\rho_{Au}(t)d_f(t)v_{cm}(t) = \int_{-\infty}^{t} (p(\tau) - p_{init})d\tau
\]

(1)
because of the pressure difference between vacuum semi space and compressed layer. Resulting force directed into vacuum side. \( \rho_{Au}(t) \) and \( d_f(t) \) are current average density and thickness of the gold film; the product \( \rho_{Au}(t)d_f(t) \approx \text{const} \) due to conservation of mass. The local (along x) centre of mass of the film is denoted ‘cm’.

Let \( t_B \) is the instant when the whole compression layer shown in figure 1(b) rolled past the observation point. If velocity

\[
v_{cm}(t_B) = \frac{\int_{-\infty}^{t_B} (p(\tau) - p_{init})d\tau}{\rho_{Au}d_f}
\]

(2)
accumulated during the shock acceleration stage is high enough then the film in the observation point separates from substrate. The same arguments are used in [23–25] to explain the entire film separation process due to pressure in substrate accelerating local (along a film) centre of mass in the thermomechanical mechanism of separation.

![Figure 4](image)

**Figure 4.** Dependence of a surface structure radius against natural logarithm of laser pulse energy, creating a particular structure.

3. Experimental results

Our experimental results are presented in figures 3 and 4. Figure 3 shows the scanning electron microscope (SEM) images of the holes created by the solitary laser impact. In experiments, the second harmonics with 515 nm wavelength was used. Duration of a pulse was 200 fs (FWHM—full width half maximum). Numerical aperture NA=0.25. We use a single laser impact onto the thin 50 nm silver film. Silver was deposited onto 1 mm thick silica plate. The images are placed from left to right and from the upper series to the lower series according to laser pulse energies. Radius of a hole increases with energy. The plot which shows dependence of a hole radius square against energy is given in figure 4.

![Figure 5](image)

**Figure 5.** $p - p_{in}$ diagram at time moments t=40 ps (a) and t=60 ps(b) show the lateral propagation of the shockwave in the substrate.
Sharp kink is apparent in figure 4. Position of the kink relates to energies \( \sim 100 \text{nJ} \). The kink in figure 4 is inside the interval of energies corresponding to the holes.

4. Shock driven separation of the film from substrate

We consider the process of shock driving of a film. Temporal behavior of velocities \( \nu_{\text{cm}}(t) \), see (2), for the three observation points are presented in the inset in figure 2 (b). In the growing plot of the graph, velocities \( \nu_{\text{cm}}(t) \) are gradually accumulated during SW acceleration of the observation point, when the shock compressed half-spherical shell passes the point and pressure under the point in figure 1(b) is positive. The first dependence taken in the point \( x = 0 \) should be omitted because it belongs to the inner part where the thermomechanical process occurs. The two other observation points at \( x=600 \text{ nm} \) and \( x=1080 \text{ nm} \) are far apart from the heating radius \( R_k = 250 \text{ nm} \) of the Gaussian beam in figure 2. Nevertheless they acquire significant velocities \( \nu_{\text{cm}}(t) \) (2) equal to 360 and 240 m/s; these are the maximum values of the functions \( \nu_{\text{cm}}(t) \) in figure 2(b). These velocities are high enough to separate the film from the substrate [27]. Thus we come to the main conclusion of the paper. The regime with shock driven separation of a film exists.

To calculate velocities shown in figure 2(b) we substitute the pressure dependence \( p(x, y = -40, t) \) into integral (2). \( x = 0, 600 \) and \( 1080 \text{ nm} \), see the inset in figure 2 (b); this inset corresponds to the right part of region near and slightly below the film shown in the computational box in figure 2(a). We use the level \( y = -40 \text{ nm} \) in glass (below the moving film) for calculation of pressure acting on a film, because golden film is dense and shifts slowly, thus this level is always inside glass.

5. Advantages and shortcomings of gaseous approximation

It is interesting that there are many computational works devoted to laser action onto films, but all of them are about MD [18, 19, 21–27] simulations, or for 1D hydrodynamic modelling [23,24,27,49]. No 2D hydrodynamic works considering ultrashort laser pulses are known, but [16, 17]. This means that 2D hydrodynamic finite difference approximation of ablation problem is difficult.

Results of our simulation for the time period from 0 to 100 ps are presented in movies posted at YouTube [50–52]. Some pressure diagrams are presented in figure 4.

We take finite initial pressure \( p_{\text{ini}} \) instead of \( p_{\text{ini}} = 0 \). By putting the level of initial pressure in our gas \( p_{\text{ini}} = 100 \text{GPa} \) at normal densities of gold and silica we obtain semi quantitative correspondence of the processes within ideal media to ones in the condensed matter. Speed of sound in our effective silica is equal to 8.5 km/s which being approximately twice higher than sound speed in real silica. Another semi quantitative disadvantage is too low negative \( (p - p_{\text{ini}}) \) pressure \( p \) in gaseous approximation.

Only positive part of pressure \( (p - p_{\text{ini}}) \) and the edge of the half-spherical shell travelling at the speed of the order of the speed of sound where \( p - p_{\text{ini}} = 0 \) are important for calculation of accumulated velocity (1). This is the main advantage of our scheme of solution. This gives us the opportunity to describe the stage of loading of the Au-film from its bottom boundary by the half-spherical shocked shell sliding along the film. In movies, full pressure \( p \) is presented, while figure 4 shows just deviation from background \( p - p_{\text{ini}} \).

6. Conclusion

Above we compare the thermomechanical kick-off a film inside its laser heated spot versus the shock-driven kick-off connected with a shock propagating inside substrate. In previous literature only the first case was investigated detail.

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