Thermomechanical ablation under plasmonic field excited by ultrashort laser pulse. Part I: Plasmonics

A.I. Ignatov\textsuperscript{1,2,3}, V.V. Zhakhovsky\textsuperscript{1,4}, A.M. Merzlikin\textsuperscript{1,2,3}, and N.A. Inogamov\textsuperscript{4,1}

\textsuperscript{1} Dukhov All-Russia Research Institute of Automatics, 22 ul. Sushchevskaya, Moscow 127055, Russia
\textsuperscript{2} Moscow Institute of Physics and Technology, 9 Institutskiy per., Dolgoprudny, Moscow Region, 141700, Russia
\textsuperscript{3} Institute for Theoretical and Applied Electromagnetics of the Russian Academy of Sciences, 13 Izhorskaya ul., Moscow 125412, Russia
\textsuperscript{4} Landau Institute for Theoretical Physics of the Russian Academy of Sciences, 1a prospekt Akademika Semenova, Chernogolovka, Moscow Region 142432, Russia

E-mail: nailinogamov@gmail.com

Abstract. Ultrashort laser pulse with duration \( \tau_L \sim 1 \) ps, e.g. acting in Kretschmann geometry, supports a standing plasmonic wave in a thin metal film deposited onto a surface of transparent dielectric prism during the pulse. It is very important that the standing plasmonic wave spatially redistributes energy of the 1st laser beam which excites a surface plasmon polariton (SPP), where SPP traps this energy, and laser energy incident on SPP (laser 2, region where standing wave locates). Thus a spatially periodic heating pattern is formed with dissipative hot spots around the antinodes and cold spots around the nodes of the standing wave. Distance between two subsequent nodes equals to plasmonic wavelength \( \sim 0.7 \ \mu \text{m} \).

Another important circumstance is the ultrashort duration of the non-homogeneous heating. It is shorter than the acoustic time scale \( t_s = d_f/c_s \) defining mechanical response of the film to heating; here \( d_f \) is film thickness and \( c_s \) is a sound speed in metal. \( t_s \) is about a few tens of picoseconds for our range of thicknesses. This means that mechanical response of a film to the heating is retarded in time relative to the heating stage, which leads to the subsonic motion of material and large generated stresses because they are directly proportional to volume density of absorbed energy. Fast freezing of moving molten film fixes its deformed configuration. This opens a new way for fabrication of desirable structures at nanoscales. In the first part of our work we present a picture of standing wave. The second part given separately is devoted to the thermal and hydrodynamic response to the fast heating by the standing wave.

1. Introduction. Modification of surface properties by structuring can be done by different techniques listed in the paper II which follows this paper I. An important weakly studied approach using a joint action of ultrashort laser pulse and surface plasmon-polariton (SPP) wave is studied in our work divided on two parts. Here in the Part I we consider an electromagnetic (EM) configuration which creates a sequence of hot spots along a thin gold film mounted above the dielectric substrate. These hot spots are the antinodes of an EM standing wave. Below a scheme of such standing wave generated by interferences between a propagating SPP wave and a laser beam is considered. In the next part II we describe thermomechanical consequences of this joint fast thermal action onto a film. It is shown that this action results in appearance of ripples looking like periodic bending of a film.
2. Scheme of splitting of a laser spot 2 into a sequence of small hot spots. General scheme describing the new method of fine manufacturing is presented in figure 1. Briefly, (i) the laser beam 1 inclined at a resonance angle excites a SPP (surface plasmon polariton) according to the Kretschmann scheme [1]. The SPP propagates to the right side from the illuminated spot 1 in figure 1; (ii) there is the laser beam 2 consisting of incident and reflected waves (reflection from a film); (iii) the excited SPP interferes with the beam 2 forming a standing wave in the beam spot. The standing wave is schematized in figure 1 by the red and blue alternating motionless rectangles. The red ones relate to antinodes and places of high light dissipation power, while the blue ones correspond to the nodes. (iv) Later in time the motionless chain of the hot and cold rectangles imprints into geometry of a film forming designed nanostructure, see details in the Part II.

3. Hierarchy of three time scales. The problem concerning SPP-assisted structuring is difficult because formally the computational code has to include electrodynamic (ED), thermodynamic, and hydrodynamic subcodes combined consistently. We resolve these difficulties by separating physical processes according to their rates: fast, moderate, and slow, as described in Part II. The fastest one taking $\sim 1 \text{ ps}$ is EDs and the heating of film through its depth. Separation of a film from substrate, inflation of ripples, and freezing of them take longer time. Thus we can neglect deformations in description of the fastest process discussed here in Part I.

4. Electrodynamic simulation. Scheme of electrodynamic problem is shown in figure 1, and computation technique is partially presented in [2]. A gold film $d_f = 80 \text{ nm}$ thick is deposited onto substrate made from fused silica. Two Gaussian $\lambda_L = 785 \text{ nm}$ 2D laser pulses 1 and 2 are used. 2D means that in electrodynamic simulation the laser beams are infinitely long in the direction perpendicular to the plane of figure 1. Polarization vector $E$ of electromagnetic field is in the plane of figure 1. Refractive indexes for this wavelength are $n = 0.149 + 4.78i$ [3] and $n = 1.454$ for gold and fused silica, respectively. The axis $x$ of the reference frame in figure 1 is directed along a film in the plane of figure; the axis $y$ is directed up; while the axis $z$ is directed to us from the plane of figure 1 (right-hand triple). The level $y = 0$ corresponds to the contact boundary between gold and vacuum (or air). The level $y = -80 \text{ nm}$ is the film-silica contact.

The laser beam 2 in figure 1 is a 2D Gaussian beam limited in its width in plane $(x, y)$ and unlimited in the direction $z$. The beam 2 comes from vacuum at normal angle of incidence. Beam e-folding radius $\omega_0^{inc}$ on an amplitude of electromagnetic field is $6\lambda_L$. Intensity distribution of this beam is shown in figure 2(b).
Figure 2. a) Intensity distribution of electric field for the Gaussian beam 1 (see figure 1) illuminating a film from the substrate (optically dense fused silica) side. The incident angle $\alpha = 44.8^\circ$ corresponds to excitation of surface plasmon in the Kretschmann configuration. Electromagnetic field of the beam 1 is p-polarized. In this case a surface plasmon polariton (SPP) is excited at the upper boundary of a film shown in figure 1. The beam 1 is seen as the rather wide vertical light strip at the bottom left half of the substrate. The dark thin horizontal strip is the gold film dividing silica below and vacuum above. The bright thin strip above the film relates to averaged intensity of SPP propagating to the right side. Brightness and width in $y$-direction of this strip increases as it crosses the exciting beam 1, and after crossing the brightness and width remain constant at the considered distance. b) Distribution of intensity of electric field in the case of the beam 2 from figure 1. The beam comes normally from vacuum side. It is polarized in the plane of this figure. For the considered wavelength 785 nm the reflection coefficient of gold is high. Therefore the interference of incident and reflected waves produces a chain of bright horizontal intervals. Horizontal width of the intervals is defined by Gaussian beam width. Vertical distance between two successive intervals is $\lambda_L/2$. c) Time averaged intensity distribution if both laser beams 1 and 2 are present. The cases with beam 1 and beam 2 are shown in the above two panels separately. Now the SPP propagating from the spot 1 in figure 1 interferes with incident and reflected waves of the beam 2. The interference results in the standing wave with hot spots in its antinodes. These hot intervals are well seen as the bottom sequence of the short red intervals. Length of these intervals equals to the SPP wavelength. Dissipation in the skin layer of the film follows this sequence of the bottom red intervals. Thus the hot rectangles shown in figure 1 are produced. The color bar with arbitrary units is the same for all three panels (a), (b), and (c).

Figure 2(a) presents the intensity distribution for the gold film illuminated from the substrate side solely by the beam 1 shown in figure 1. The e-folding radius (at the illuminated surface) of decrease of amplitude of Gaussian beam when we go out from the beam axis is $\omega_0^K r = 20 \lambda$ in our ED simulation As was mentioned above, the beam 1 comes at the resonant angle $\alpha = 44.8^\circ$ relative to a normal direction to the film. At these conditions (propagation from optically dense medium to metal film) according to the Kretschmann scheme [1] the SPP is excited at the metal-vacuum contact, because this angle corresponds to the resonance for excitation. The SPP wavelength for the consider pair metal-substrate and the chosen wave $\lambda_L$ is equal to $\lambda_{spp} = 768$ nm, which is slightly less than $\lambda_L = 785$ nm.

In the right side of figure 2(a) at the upper film boundary we see the excited SPP wave
Figure 3. Spatial distribution of electromagnetic heating power inside the gold film. The fused silica is below the film, vacuum is above. The scheme of illumination by beams 1 and 2 is shown in figure 1. The vertical axis of the beam 2 is placed in the middle of this frame. The film thickness of 80 nm is ten times less than the SPP wavelength of $\lambda_{\text{spp}} = 768$ nm. The more intense dissipation in the skin-layers near the upper and bottom boundaries is seen. The horizontal shift of the maximums of dissipation between the upper and the bottom boundaries is observed.

propagating in the right direction from the spot 1. The pattern of interference between the SPP and the beam 2 is clearly seen in figure 2(c), which presents intensity distribution of the total sum obtained from incident and reflected beams 1 and 2, together with SPP. In the presented here particular run of electrodynamic simulation the distance between axes of the two beams is 20 $\mu$m. To achieve the maximum visibility of the interference pattern in the spot 2 (see figures 1 and 2) the ratio of powers of the beams 1 and 2 was specially adjusted. To achieve a maximum the power of the beam 1 should be four times larger than power of the beam 2.

The calculated spatial distribution of local power $Q(x,y)$ of energy dissipation in metal per unit of volume for described above conditions is presented in figure 3. In the hydrodynamic and molecular dynamic simulations in Part II we use absorbed fluence $F_{\text{abs}}(x)$. This function is $F_{\text{abs}}(x) = \int Q(x,y)\, dy$, where the integral is taken through the film thickness. Heating at a picosecond time scale becomes uniform through the vertical intervals crossing the film. While spreading of heat along the SPP spatial period lasts much longer than the duration of the acoustic stage at which the partial or total separation of a film takes place. This is why we use the fluence $F_{\text{abs}}(x)$ as the input data for the simulation of the second (or acoustic) stage described in Part II.

Conclusion
The complicated problem of how the "hot" plasmonics can be used for formation of surface nanostructures is studied. Solution is based on consecutive treatment of physical processes based on their time scales. Orders of magnitudes differ these scales from $\sim 1$ ps for EDs and heating through film thickness (subproblem #1 for the fastest processes) to 10–20 ps for sound propagation through the film (subproblem #2 for acoustic scale and separation from substrate) and to tens of ns for the slowest stage of inflation of ripples, their cooling and freezing due to lateral heat transfer (subproblem #3 for formation and freezing of ripples). Here the subproblem #1 is studied. The next two subproblems are discussed in Part II.

Authors INA and ZVV are supported by the RSF grant 14-19-01599.

References
[1] Kretschmann E and Raether H 1968 Z. Naturforsch. 23a 2135–2136
[2] Ignatov A and Merzlikin A 2018 Journal of the Optical Society of America B 35 accepted
[3] Johnson P B and Christy R W 1972 Phys. Rev. B 6(12) 4370–4379