Laser-induced formation of holograms for generation of plasmons

A I Ignatov\(^{1,2}\), V V Zhakhovsky\(^{1,3}\), A M Merzlikin\(^{1,2}\) and N A Inogamov\(^{3,1}\)

\(^{1}\) Dukhov Research Institute of Automatics (VNIIA), Sushchevskaya 22, Moscow 127055, Russia
\(^{2}\) Institute for Theoretical and Applied Electromagnetics of the Russian Academy of Sciences, Izhorskaya 13, Moscow 125412, Russia
\(^{3}\) Landau Institute for Theoretical Physics of the Russian Academy of Sciences, Akademika Semenova 1a, Chernogolovka, Moscow Region 142432, Russia

E-mail: nailinogamov@gmail.com

Abstract. Methods of fabrication of precisely adjusted surface structures are indispensable to development of new technologies. Different surface structuring techniques resulting in solitary nanobumps, random surface structures and laser induced periodic surface structures were in focus in the past. Here we consider a physical model and numerical simulation allowing understanding a high field plasmonics formation process of a smooth periodic perturbation of surface on a metal film with a period equal to the surface plasmon-polariton wavelength at a frequency of laser wave. Such surface structure is a hologram produced by thermomechanical response to interference between an incident laser wave, a reflected laser wave, and an electromagnetic field in the running plasmon-polariton wave. The following laser irradiation of the manufactured hologram generates a new plasmon-polariton wave identical to that used for formation of the hologram.

1. Introduction

Today physics of laser–matter interaction is in the focus of applications based on manipulations with different photon beams including the Hermite-Gaussian and Laguerre-Gaussian beams. Those beams are generated by phase plates or focal spots at diffraction limit on target surface. Another method of generation of beams with complicated electromagnetic (EM) fields utilizes a combination of surface plasmon-polariton (SPP) waves and incident laser beams. This method is considered here.

We examine the generation of complicated EM fields together with how they are coupled with material, and how they leave the solidified surface structures on solid targets.

Laser–matter interactions were considered in works done by many authors and in our previous works \[1–16\]. But therein a simple Gaussian beam or even surface homogeneous irradiation were studied. Here for the first time we consider namely action of geometrically complicated EM field. We present results concerning the case when electrodynamic problem is solved together with coupled thermal and hydrodynamic problems involving hot plasmonics. In the thermal part we analyze two-temperature phenomena (duration of pulse is shorter than electron–ion thermalization time), melting, evaporation, and re-crystallization. Hydrodynamic motion of
material is solved with inclusion of first order phase transitions (melting, evaporation, re-crystallization), capillary forces, and vapor pressure.

We calculate dissipation in a skin-layer of electromagnetic field which is a sum of a plasmon mode and external (relative to this mode) laser field. Such dissipation is non-homogeneous along the target surface resulting in interference fringes. Our thermomechanical simulation shows how the process of imprinting of the hot fringes into a film proceeds. This is not darkening of a photographic plate thanks to cold photochemistry. The imprint appears as a result of thermomechanical effects produced by fast melting followed by deformation of surface shape and its solidification later time.

2. Hologram reconstruction of plasmon waves

Optics of surface plasmon-polariton (SPP) wave is essential for modern applications. Promising approaches have been proposed for using of the SPP waves for optical interconnection and as modulators [17–19]. Here we consider how to make the rippled surface structures for generation of SPP waves using hologram reconstruction. For this purpose it is necessary first to fabricate a periodic surface structure (hereafter is referred as a hologram) with a period exactly equal to the wavelength of a SPP wave that is intended for generation later. The fabrication procedure is presented in figure 1(a). Then the running SPP wave with \( k_{\text{SPP}} \) will be excited by irradiating such structure by an incident plane electromagnetic wave with \( k_\parallel, \alpha \) as shown in figure 1(b). This electromagnetic wave is termed a reconstructing wave while the excited SPP wave with \( k_{\text{SPP}} \) is a reconstructed image.

Scheme describing the manufacturing method of a hologram is presented in figure 2 and discussed in details below in the text. Below it is outlined briefly:

- A laser beam 1 inclined at a resonance angle excites a SPP wave propagating to the right side from the illuminated spot 1 in figure 2.
- There is a beam 2 consisting of incident wave and a wave reflected from a film.
- The excited SPP interferes with beam 2 forming a standing wave in the beam spot. The standing wave is schematized in figure 2 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 nodes.
- Later the motionless chain of the hot and cold rectangles imprints into film forming the required nanostructure. Below we performed the quantitative description of the processes according to the scheme shown in figure 2.

3. Splitting the time scales

The problem stated above is very complicated for direct solution. By the term “direct solution”, we imply development of such computer code, which will solve the problem from beginning to the end. To follow formation of nanostructures at a surface of metal film exposed to the electromagnetic field, the electrodynamic and thermomechanical processes, both, have to be taken into account in one straight-through code. At the input of the code we give a target and a laser pulse. At the output we obtain spatially deformed film which has been subjected to heating, melting, and shifting before freezing and forming the final nanostructure. Such code may be developed in future.

Presently the problem can be solved by splitting into three time scales. There are three codes and three stages corresponding to these three time scales. The output of the first code is the input for the second one, while the second output is the input for the third code.

The deformed surface is created from initially smooth plane film. We use a gold film. Its thickness \( d_f \) is of the order of few tens nanometers. The film is deposited on a dielectric prism. The shortest or the first time scale is \( \sim 1 \) ps. It is defined by duration of a laser pulse \( \tau_L \) and
Figure 1. (a) Writing a hologram—a periodic ripple with length $L$ on a surface of metal film. Interference between the incident electromagnetic wave $k_{\parallel}, \alpha$ from one side and the running surface plasmon-polariton (SPP) wave $k_{SPP}$ from another side is used to imprint the hologram. The wave $k_{\parallel}, \alpha$ comes from air or vacuum under angle $\alpha$. Due to interference a standing wave is formed within the length $L$ determined by the diameter of laser beam in the plane of target. The standing wave produces an alternation of hot and cold zones in surface layer. This spatial temperature modulation is imprinted on the surface after cooling and solidification of metal. (b) Reconstruction of a SPP wave. The manufactured hologram is illuminated by the exciting wave $k_{\parallel}, \alpha$. An image, which is a SPP wave, is reconstructed by scattering of the exciting wave $k_{\parallel}, \alpha$ at the hologram. The wave $k_{\parallel}, \alpha$ should equal to the electromagnetic wave used for writing the hologram.

duration needed to homogeneously heat a film through its thickness $d_f$; spread of dissipated heat along a spatial period of plasmon wave within $\approx 0.7 \, \mu m$ takes much longer time (nanoseconds) and relates to the third stage. We use an electrodynamic code to describe excitation of plasmon wave by the first laser beam, its interference with the second laser beam, formation of a standing wave, and dissipation in the standing wave.

The first stage is short relative to the acoustic time scale $t_a$ introduced above. Mechanical interaction of a rapidly heated film with its supporting substrate occurs during the second stage because it lasts over the acoustic time. Material motion is modeled using the second computational program: the in-house one-dimensional two-temperature (2T) hydrodynamic (HD) code.

Thickness of our film must be of the order of few skin depths $\delta_{\text{skin}} \approx 10–20 \, \text{nm}$ or less to be thin enough, since an amplitude of an excited plasmon wave decreases with increase of the ratio $d_f/\delta_{\text{skin}}$. Because the sound speed in gold is $c_s = 3.1 \, \text{nm/ps}$, then the acoustic time scale $t_a$ is in the range of 7–20 ps.

Comparing the first and the second time scales we conclude that the electrodynamic code neglecting appearance of a gap between the film and substrate, and neglecting deformations of film at the spatial region of the order of plasmon wavelength $\approx 0.7 \, \mu m$ can be employed. The process of separation of the film and formation of the gap begins after finishing of the acoustic stage, because the pressure at the film–substrate contact remains positive during the acoustic stage: the film and substrate are clasped to each other. Thus the electrodynamic
Figure 2. The scheme of manufacturing a nanostructured ripple from the flat thin film using plasmon assisted management of dissipative spots location. We see the upper part of the Kretschmann prism with a gold film above. Laser 1 excites a SPP wave. The SPP achieves the illuminated spot 2 where SPP together with laser beam 2 produces the standing wave. The standing wave creates the sequence of hot and cold zones in the film. This sequence imprints into the final surface nanostructure.

code gives a heat distribution along the film; this distribution serves as a starting condition for thermomechanical simulations within the second and third stages. Operating the electrodynamic code we neglect motion of material. During the second (i.e. acoustic) stage the distribution of internal energy changes slightly—mainly due to work performed for material expansion. The conductive cross-flow of energy along the film is negligible.

Only later at the third stage a slow thermal cross-flow between the places, where the hot antinodes and the cold nodes of the standing wave were localized, becomes significant in the energy balance. Final cooling of the nanostructure down to freezing proceeds due to thermal outflow shown in figure 3 outward the region occupied by spot 2 of laser beam 2 in figure 2. Freezing time should be less than tens nanoseconds to keep structures formed when the film moves in a liquid state following the electrodynamic imprint. For too long times of cooling the structure will be smoothed out by the capillary effects before freezing. Cooling rates impose a strong limitation on the lateral size of the structured spot at the surface which should be less than the few tens microns, otherwise we have to design alternative scheme of cooling through a conductive substrate; the structured spot is shown in figure 3.

Laser pulses 1 and 2 in figure 2 should be synchronized and have the identical frequencies. It can be achieved by splitting one ultrashort pulse to two pulses traveling the optical paths with different lengths.

4. Electrodynamic simulation

Scheme of the problem is shown in figure 2. Computation technique is partly presented in [20]. A gold film $d_f = 80$ nm thick is deposited onto substrate made from fused silica. Two Gaussian $\lambda_L = 785$ 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 2. Vector of polarization of electromagnetic field (vector $E$) is in the plane of figure 2. Refractive indexes
Figure 3. The top view of spot 2 which is an intersection of laser beam 2 in figure 2 with film surface. Here the 2D geometry is considered. Thus the spots 1 and 2 in figure 2 are the long strips perpendicular to the plane of figure 2. Such strip is created by a cylindrical lens. In figure shown here we changed the strip to the elongated ellipsoid (spot 2). The red stretched ellipsoids are the ripples of film imprinted by the antinodes of standing wave where the maximal rate of energy dissipation is localized. The red arrows mark an outward thermal flux to cold regions. This flux leads to freezing of ripples.

for this wavelength are $n = 0.149 + 4.78i$ (gold) [21] and $n = 1.454$ (fused silica). The axis $x$ of the reference frame in figure 2 is directed along a film in the plane of the figure; the axis $y$ is directed up; while the axis $z$ is directed to us from the plane of figure 2 (right-hand triple). The level $y = 0$ corresponds to the contact boundary between gold and vacuum (or air). The level $y = -80$ nm is the film–silica contact.

Laser beam 2 in figure 2 is a Gaussian beam limited in its width in plane ($x, y$) and unlimited in the direction $z$ (because it is the 2D beam). Beam 2 comes from vacuum at normal angle of incidence. The e-fold decrease of beam intensity occurs at the radius $\omega_{0}^{inc} = 6\lambda_{L}$. Intensity distribution of this beam is shown in figure 4(b). For the considered wavelength of 785 nm the reflection coefficient of gold is large. Thus the interference of incident and reflected waves produces a chain of bright horizontal intervals.

Figure 4(a) presents an intensity field in the gold film illuminated from the substrate side solely by beam 1 shown in figure 2. The p-polarized beam 1 illuminates the film from the substrate of (optically dense fused silica) side. The e-fold decrease of Gaussian beam intensity on the illuminated surface occurs at the radius of $\omega_{0}^{inc} = 20\lambda$ in figure 4(a) and in our electrodynamic simulation. As was said above, beam 1 comes at the resonant angle $\alpha = 44.8^\circ$ relative to normal direction to a film. At these conditions (propagation from optically dense medium to metal film) according to Kretschmann scheme [22] the SPP is excited at the metal–vacuum contact. The angle $\alpha = 44.8^\circ$ corresponds to a resonance for excitation. The SPP wavelength for the considered pair of metal–substrate and the chosen wave $\lambda_{L} = 768$ nm. It is slightly less than $\lambda_{L} = 785$ nm.

In the right side of figure 4(a) at the upper boundary of a film we see the excited SPP wave going from spot 1 in the right direction. The interference pattern of the SPP and beam 2 is clearly seen in figure 4(c) which presents an intensity distribution summed up from the SPP and
Figure 4. Side views of electric field intensity in a top vacuum layer and a thin metal film lying on a thick layer of substrate. The color bar shows intensity in arbitrary units identical for all panels. (a) Intensity distribution for the Gaussian beam 1 shown in figure 2. The angle of incidence $\alpha = 44.8^\circ$ corresponds to excitation of surface plasmon in Kretschmann configuration. A SPP wave is excited at the upper boundary of a film in figure 2. Beam 1 from figure 2 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 propagating to the right side SPP. 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 for beam 2 shown in figure 2. The beam comes normally from vacuum side. It is polarized in the plane of this figure. Horizontal width of the intervals is defined by width of Gaussian beam, see text. Vertical distance between two successive intervals is $\lambda_L/2$. (c) Time-averaged intensity of field for both beams 1 and 2. The SPP propagating from spot 1 in figure 2 interferes with incident and reflected waves of beam 2, which results in appearance of the standing wave. The last produces hot zones in its antinodes. Those hot zones are seen on the bottom of film as a sequence of the short red intervals, with length equal to the SPP wavelength. Energy deposition happens within these zones in the skin layer producing the hot rectangles shown in figure 2.

the incident and reflected beams 1 and 2. In the particular run of electrodynamic simulation presented here distance between axes of the two beams is 20 $\mu$m. To achieve the maximum visibility of the interference picture in spot 2 (see figures 2 and 4) the power of beam 1 should be four times larger than power of beam 2.

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 figures 5 and 6. In the hydrodynamic and molecular dynamic simulations we use an absorbed fluence $F_{abs}(x)$, which is $F_{abs}(x) = \int Q(x, y) \, dy$, where the integral is taken along the film thickness. As was said above in section 3 the heating with picosecond time scale becomes homogeneous through the film thickness. While the heat spreading along the SPP spatial period lasts much longer than the acoustic time, at which a partial film separation from substrate takes place. This is why we use the fluence $F_{abs}(x)$ as the input data for the simulation of the second acoustic stage.
5. Thermomechanical response to plasmon heating

As it was mentioned in section 3) the thermomechanical evolution consists of the fast acoustic stage 2 and slow stage 3 with movement and separation of film. For stage 2 we use our in-house developed one-dimensional two-temperature $T_c > T_i$ hydrodynamics (2T-HD) code [9, 23, 24];
the code continuously describes both the 2T and one-temperature (1T) $T_e \approx T_i$ states. For stage 3 we employ the molecular dynamics code combined with the Monte-Carlo subcode [7]. Monte-Carlo subcode provides the required electron heat conductivity in metals. The combined MC-MD code simulates material motion, melting, cooling, and recrystallization. It should be emphasized that at the given spatiotemporal scales the non-equilibrium recrystallization takes place in supercooled liquid at temperature much below the melting point.

1D 2T-HD code is applicable in stage 2 which duration is of the order of acoustic time scale $t_s$. Indeed the SPP wavelength $\lambda_{spp} = 768$ nm is ten times longer than the film thickness of $d_f = 80$ nm. Acoustic waves cross a film with speed of sound, but material motion proceeds with significantly smaller velocities in a subsonic regime at the considered range of absorbed fluences. Thus the film displacement from its initial position at stage 2 is significantly less than the film thickness $d_f$ which validates the 1D approach.

In 1D 2T-HD calculations we use the following features developed before:

- the heat conduction coefficient valid in 2T and 1T states [25];
- the electron–ion coupling parameter [25];
- the equation of state for gold [26], which is also valid for 2T and 1T states.

The 2T effects are significant because the duration of 2T relaxation 7–10 ps is comparable to acoustic time scale $t_s$ in gold. 2T gold is softer than 1T one at the same density and total internal energy, because the part of pressure is associated with the excited electrons which have 2–3 times lower Grüneisen parameter [26] in 2T state. This circumstance appreciably decreases the expansion velocities for the same absorbed energy in comparison with the hypothetical case when gold always stays in 1T state after absorption of plasmon energy. Let us mention that the equation of state used in the 2T-HD code describes well the melting and crystallization transitions.

In 1D 2T-HD simulations we include cohesion between gold and fused silica. Resistance to breaking of the contact below 1 GPa weakly influences dynamics of a film. There are two tensile stress thresholds for metal on substrate: the first corresponds to the contact cohesion and second one is determined by the tensile strength of metal itself. If the contact cohesion is strong enough then the first threshold disappears because it cannot be higher than the second threshold. A whole film separates if the applied stress is between them [9]. Above the second threshold the film breaks into two part [9].

We use 1D 2T-HD code to define temperature $T(x)$ and velocity $v(x)$ distributions. The code also defines which a particular piece of film will be ruptured from substrate; a piece of film remains on substrate if the absorbed fluence in this piece is below than the first threshold determined by cohesion between a film and fused silica. The functions $T(x)$ and $v(x)$ are the output functions from stage 2. They are utilized as input conditions for stage 3. To calculate these functions the dissipated fluence $F_{\text{abs}}(x)$, which is the output function from stage 1 (electrodynamics), is employed as an input condition for stage 2 (2T-HD).

6. Thin films approach to thermomechanical response

Stage 3 was analyzed by two approaches. In the simple one, which is presented in this section, we use a thin film approximation: $\lambda_{spp}/d_f \to \infty$ [6]. In the more sophisticated approach discussed in the next section we employ the molecular dynamics code combined with the Monte-Carlo subcode [6, 7]. MD-MC simulations last up to formation of solidified structures and cessation of motion [7]. In the thin film approach the only mechanical part of problem is solved: competition between inertia and capillary action is quantitatively evaluated. Instead of description of temporal propagation of crystallization process (as in MD-MC) in thin film approach we can stop simulation when the moving piece of the film has to be solidified. As it
Figure 7. Slow development of perturbation of a film described in the thin film approximation: \( \lambda_{\text{app}} \gg d_f \). Perturbation is imposed by the standing wave shown in figures 4–6. Variation of dissipated energy \( F_{\text{abs}}(x) \) is transformed into initial velocity profile for stage 3 by the 2T-HD code operating at stage 2—section 5. Initial vertical velocities are tens m/s. One period \( \lambda_{\text{app}} \) of the perturbation is considered while the less significant long-wavelength modulation connected with the e-fold radius of laser beam 2 is neglected. At the initial time \( t = 0 \) the film is plane. Simulation starts with the initial velocity profile \( v(x) \) derived from stage 2. It is supposed that the film is entirely molten. Half period of wavelength 500 nm is shown. The presented times relate to the stage of deceleration and stopping of a film under action of surface tension. Formation of jet begins at the time of 3.8 ns.

was said in section 3 recrystallization is governed by the heat flow outward into the surrounding cold film, see figure 3.

Thin film approach is necessary to prepare the initial data for the resources consuming MD-MC simulation. Because we are interested in the specially designed shapes of the formed nanostructures, for example, with frozen jet ahead, the thin film approach is used to choose a few interesting shapes from the various possible configurations to generates the starting conditions for MD-MC simulations.

Simplicity and low resource-demanding are significant advantages of the thin film approach. From the other hand we cannot include heat conductivity and freezing using this approach, while in MD-MC simulation is free from these shortcomings [7, 9, 23, 27].

Example of simulation in the thin film approach is presented in figure 7. In this example a film totally separates from a substrate. Thus there are no pieces where the film remains on fused silica. Let us mention that cohesion between gold and fused silica is weak and the situation with total separation seems plausible. In this case the standing wave shown in figure 6 may separate from silica in the central bumps remaining in contact with substrate outside the e-fold radius.

The film remains in contact with a fused silica substrate if the dissipated energy corresponds to the first threshold. For averaged absorbed fluence lying between first and second thresholds the separation takes place in the intervals where maximums of dissipated energy is located while the contact remains intact in the minima of the function \( F_{\text{abs}}(x) \). The film is rippled as a result of interference between an incident laser wave and SPP wave expanding from solitary surface
perturbation observed in [28]; the solitary perturbation was created by a preceding tightly focused laser shot. The degree of film separation (total or partial) from an underlying substrate in these experiments is unclear, but at least partial separation should take place (this follows from rippling amplitude and volume conservation; evaporation degree is small).

Conclusions that definitely follows from our electrodynamic and thermomechanical modelings are as follows:

- The film melts fast at the time scales less than the acoustic time $t_s$.
- Motion of the film in direction outward from substrate is decelerated gradually by the surface tension.
- The film separates from substrate at least partially in the considered range of fluences where temperatures in 1T state rise up to 1.5–2 kK.
- Molten metal film moves during deceleration stage in the $y$ and $x$ directions accumulating near the maximums of dissipated energy $F_{\text{abs}}(x)$; the $x$ component of velocity appears from the capillary forces.
- Freezing of a quasi-periodic structures takes place later during the final stage.

How fast will the film be recrystallized depends on a cooling rate. Cooling proceeds through heat outflow from the laser spot 2, see figure 3. Question is: how fast does crystallization proceed relative to the inertial-capillary flow? In other words—is crystallization finished before the stopping of inertial inflation of a bump or after when a film begins to return back? There are also situations with delayed solidification at which the liquid film collides with substrate, see examples with a freestanding film [23, 27].

7. Molecular dynamics of thermomechanical response
The main simplification which allows to solve the problem stated in section 1 is separation of time scales, see also section 3. Thus we divide the entire evolution to three stages. The first two stages provide the velocity $v(x)$ and temperature $T(x)$ profiles for the third stage. Simplified version of the third stage was considered above in section 6. Here we analyze the third stage including recrystallization.

Dynamics of a sample within one wavelength of the velocity profiles is studied in a series of MD-MC simulations. Typical results are presented in figure 8. It shows evolution of a film after melting and separation from substrate. Initial thickness of the gold film is $L_y = 20\text{nm}$. The horizontal length (i.e. in $x$ direction) of the rectangular box is $L_x = 200\text{ nm}$. Thickness of the simulation box in the $z$ direction (perpendicular to the plane of figure 8) is $L_z = 12\text{nm}$. The periodic boundary conditions are imposed on the $x$ and $z$-axis. Temperature of gold after fast melting is 1800 K. In this run we neglected the temperature variation along the simulation box because the main thermal process is not heat propagation along the period of a SPP wave but cooling due to heat transfer outward the hot spot 2, see figure 3. In simulation we use the EAM interatomic potential for gold developed by the stress-matching method [29]. The EAM potential provides the melting temperature of $T_m \approx 1330\text{ K}$ in close agreement with the experimental one of 1337 K.

Initially the velocity distribution $v(x) = v_0 \cos(2\pi x/L)$ with $v_0 = 50\text{ m/s}$ is applied to sample material in the run presented in figure 8. 2T-HD simulations indicate that the film separation velocities are of the order of a few tens of m/s in the considered range of dissipated energies. Capillary scale of speed is $v_{\text{cap}} = 2\sqrt{\sigma/\rho d_f}$, where $\sigma$ is a surface tension coefficient and $\rho$ is density of gold. This speed is 56 m/s for the film with $d_f = 80\text{ nm}$ and $\sigma = 1200\text{ erg/cm}^2$. In our MD-MC simulations $d_f = 20\text{ nm}$ and EAM potential has $\sigma \approx 600\text{ erg/cm}^2$, see appendix and figure 15 in paper [6]. Then $v_{\text{cap}} = 79\text{ m/s}$. MD-MC simulations show that in order to decelerate the film quickly to prevent jetting the surface tension should be sufficiently strong
against inertia. Then the dimensionless separation velocity \(2v_0/v_{\text{cap}}\) normalized by the capillary velocity should be of the order of 1 or less.

In the run illustrated by figure 8, the evolution of one SPP period of a film is followed in the coordinate system of the mass center of the film. Total momentum of the film remains zero during simulation. But in the frame connected with the substrate this film moves outward. This corresponds to the few central bumps in the chain of bumps or periods of the SPP wave train covering spot 2 in figures 4(c)–6. The bumps outside this central interval remains in contact with substrate.

There are different situations depending on relation between the dimensionless separation velocity and the rate of cooling, see [9]. In the run shown in figure 8 we use the Langevin thermostat with the goal temperature of 600 K and the characteristic cooling time 400 ps. The thermostat acts only on the \(z\)-component of velocity in order to escape the thermostat-driven deceleration of \(x\) and \(z\)-components of velocity in the \(x-y\) plane. The thermostat corresponds to approximately homogeneous cooling of gold inside the SPP period due to heat transfer outward from the hot spot 2 including a train of the standing SPP wavelengths, see figure 3.

The panels in figure 8 follow evolution of phase states in the film during freezing. Molten gold goes into the deeply supercooled states until it reaches \(T/T_m \approx 0.65\) at the time of 590 ps, when the temperature drops to \(T \approx 870\) K and appreciable growth of nuclei of solid phase begins. Nucleation starts like a jump process because it is driven by explosive homogeneous nucleation of multiple seeds of solid state in whole volume of melt after such large degree of supercooling of metastable liquid is achieved. This bulk recrystallization resulting in polycrystalline solid with tiny grains takes a few hundreds picoseconds.
8. Conclusion

We considered in details the complicated problem how a “hot” plasmon can produces the specific surface nanostructures. The consequence of processes starting from excitation of the running surface plasmon-polariton wave and completing with the final solidified structure as a result of the imprint of the standing wave on the thin metal film was studied.

Acknowledgments

This work is supported by the Russian Science Foundation (grant No. 14-19-01599).

References

[1] Domke M, Rapp S, Schmidt M and Huber H P 2012 Opt. Express 20 10330–8
[2] Ivanov D S, Kuznetsov A I, Lipp V P, Rethfeld B, Chichkov B N, Garcia M E and Schulz W 2013 Appl. Phys. A: Mater. Sci. Process. 111 675–87
[3] Nakata Y, Miyanaga N, Momoo K and Hiromoto T 2013 Appl. Surf. Sci. 274 27–32
[4] Inogamov N and Zhakhovsky V 2014 JETP Lett. 100 4–10
[5] Zywietz U, Evlyukhin A B, Reinhardt C and Chichkov B N 2014 Nat. Commun. 5 3402
[6] Inogamov N A, Zhakhovskii V V and Khokhlov V A 2015 J. Exp. Theor. Phys. 120 15–48
[7] Inogamov N A, Zhakhovsky V V, Khokhlov V A, Petrov Yu V and Migdal K P 2016 Nanoscale Res. Lett. 11 177
[8] Ivanov D S, Blumenstein A, Ihlemann J, Simon P, Garcia M E and Rethfeld B 2017 Appl. Phys. A: Mater. Sci. Process. 123 744
[9] Wang X W, Kuchmizhak A A, Li X, Juodkazis S, Vitrik O B, Kulchin Yu N, Zhakhovsky V V, Danilov P A, Ionin A A, Kudryashov S I, Rudenko A A and Inogamov N A 2017 Phys. Rev. Appl. 8 044016
[10] Inogamov N A, Zhakhovsky V V, Ashitkov S I, Emirov Yu N, Faenov A Ya, Petrov Yu V, Khokhlov V A, Ishino M, Demaske B J, Tanaka M, Hasegawa N, Nishikino M, Tamotsu S, Pikuz T A, Skobelev I Yu, Ohba T, Kahiort T, Ochi Y, Imazono T, Fukuda Y, Kando M, Kato Y, Kawachi T, Anisimov S I, Agranat M B, Olevnik I I and Fortov V E 2015 Eng. Failure Anal. 47 328–37
[11] Mayer P N and Mayer A E 2016 J. Appl. Phys. 120 075901 (Preprint https://doi.org/10.1063/1.4959819)
[12] Wu C and Zhigilei L V 2016 J. Phys. Chem. C 120 4438–47
[13] Anisimov S I, Zhakhovsky V V, Murzov S A and Khokhlov V A 2017 Quantum Electron. 47 509–21
[14] Ashitkov S I, Inogamov N A, Zhakhovskii V V, Emirov Yu N, Agranat M B, Oleinik I I, Anisimov S I and Fortov V E 2012 JETP Lett. 95 75–81
[15] Inogamov N A, Zhakhovsky V V, Emirov S I, Emirov Yu N, Faenov A Ya, Pikuz T A, Ishino M, Kando M, Hasegawa N, Nishikino M, Kawachi T, Agranat M B, Andriash A V, Kuratov S E and Oleynik I I 2014 J. Phys.: Conf. Ser. 500 112070
[16] Karim E T, Shugaev M V, Wu C, Lin Z, Matsumoto H, Conneran M, Kleinert J, Hainsey R F and Zhigilei L V 2016 Appl. Phys. A: Mater. Sci. Process. 122 407
[17] Kretschmann E and Raether H 1968 Z. Naturforsch. 23a 2135–6
[18] Inogamov N A, Zhakhovsky V V, Emirov S I, Emirov Yu N, Faenov A Ya, Pikuz T A, Ishino M, Kando M, Hasegawa N, Nishikino M, Kawachi T, Agranat M B, Andriash A V, Kuratov S E and Oleynik I I 2014 J. Phys.: Conf. Ser. 500 112070
[19] Babicheva V E, Malureanu R and Lavrinenko A V 2013 Photonics and Nanostructures - Fundamentals and Applications 11 323–34
[20] Ignatov A and Merzikin A 2018 J. Opt. Soc. Am. B 35 308–16
[21] Johnson P B and Christy R W 1972 Phys. Rev. B 6 4370–9
[22] Kretschmann E and Raether H 1968 Z. Naturforsch. 23a 2135–6
[23] Inogamov N A, Khokhov V A, Petrov Y V, Zakhakhovsky V V, Migdal K P, lnitsky D K, Hasegawa N, Nishikino M, Yamagawa M, Ishino M, Kawachi T, Faenov S I, Pikuz T A, Baba M, Minami Y and Suemoto T 2017 AIP Conf. Proc. 1793 070012 (Preprint https://aip.scitation.org/doi/pdf/10.1063/1.4971600)
[24] Khokhlov V A, Inogamov N A, Zhakhovsky V V, lnitsky D K, Migdal K P and Shepelev V V 2017 AIP Conf. Proc. 1793 100038 (Preprint https://aip.scitation.org/doi/pdf/10.1063/1.4971663)
[25] Petrov Yu V, Inogamov N A and Migdal K P 2013 JETP Lett. 97 20–7
[26] Petrov Yu V, Migdal K P, Inogamov N A and Zhakhovsky V V 2015 Appl. Phys. B: Lasers Opt. 119 401–11
[27] Inogamov N A, Zhakhovsky V V and Khokhlov V A 2018 J. Phys.: Conf. Ser. 946 012008
[28] Kuchmizhak A A, Ionin A A, Kudryashov S I, Makarov S V, Rudenko A A, Kulchin Yu N, Vitrik O B and Efimov T V 2015 Opt. Lett. 40 1687–90
[29] Zhakhovskii V V, Inogamov N A, Petrov Yu V, Ashitkov S I and Nishihara K 2009 Appl. Surf. Sci. 255 9592–6