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Optoacoustical Transducer Based on Plasmonic Nanoparticles

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The problem of increasing the efficiency of optoacoustic conversion of energy is discussed. For this task, an array of metal nanoparticles is proposed to apply as a photoacoustic transducer. The absorption coefficient of optical energy can be significantly increased due to the presence of resonance lines in the absorption spectrum of the nanoparticle structure. The task of design of a spatially non-uniform absorbing optoacoustic transducer has also been studied. The application of non-uniformly absorbing transducers provides generation of acoustic pulses with a non-uniform phase front, including acoustic vortices. The optical properties of the nanoparticle array, the initial temperature profile in a single particle and the dynamics of the generated acoustic wave were calculated.

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1. Introduction

Nowadays, optoacoustic (photoacoustic) effect is widely used in science and technology. Commercially available systems for photoacoustic spectroscopy there exist, and the systems of optoacoustic tomography are being created. Such a systems can be used for example, in biological tissues investigations [1, 2] or for non-destructive control of characteristics of materials and structures [3, 4]. Recently, studies of the processes of excitation of acoustic pulses in absorbing micro- and nanostructures are of particular interest [5–11]. The frequency of sound oscillations generated in such structures upon absorption of the energy of short (nano- and picosecond) and ultrashort (femtosecond) laser pulses lies in the range from GHz to THz [5, 6]. In work [7], using the optoacoustic excitation of short acoustic pulses, the magneto-photonic nanostructure Al_2O_3−Co–Au was studied. A number of works on the modulation of light by subterahertz pulses excited by the optoacoustic method have appeared. For example, in [8, 9] a layered structured media based on a photonic crystal resonator and on a plasmon lattice were investigated using the pump-probe method.

Optoacoustic excitation with a subterahertz pulse occurs as follows. A thermal pulse is excited by irradiating by a femtosecond laser pulse of a transducer, which is a thin aluminum film. As a result of heating the film expands and creates an initial strain or a pressure nonuniformity. An acoustic pulse passes through the substrate in the medium and disturbs the nanostructure

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layers, that leads to the modulation of its optical parameters. The change of these parameters is visualized by the probe laser pulse. However, the main disadvantage of the schemes described in [8, 9] is the low modulation efficiency of the probe pulse.

To increase the efficiency of modulation of optical parameters of the medium in [10] it was proposed to apply a layered nanostructure which is a combination of a plasmonic lattice and a phononic crystal. The phononic crystal was a periodic structure of GaAs–AlAs. It provided formation of an acoustic resonator. As a result, amplification of acoustic oscillations occurred at a frequency of about 250 THz. Due to the presence of a phononic crystal, the optoacoustic excitation of oscillations near the plasmonic lattice was not possible from the substrate side (such a geometry was used in works [8, 9]). Therefore, there was investigated the configuration in which the pump beam and the probe beam propagate from the plasmonic lattice side. In this case, it was assumed that the grating has a sufficiently small period and heats uniformly when a pump pulse illuminates it. It means that from the point of view of excitation of an acoustic wave the plasmonic lattice can be considered as a homogeneous metallic optoacoustic transducer. Acoustic oscillations changed the parameters of the plasmonic lattice, which led to the modulation of the probe laser beam. Also, in [10] the problem of finding the optimal mode of the plasmonic lattice which provides the highest modulation of the probe beam, was solved.

Another possibility of increasing the amplitude of the generated acoustic signal, and hence the efficiency of acoustic modulation of the optical parameters of the medium, is associated with the use of plasmon resonances in the absorption of the energy of the exciting laser radiation. This possibility is analyzed in this work. In addition, the use of an array of absorbing elements that are non-uniformly distributed over the surface or have different absorption coefficients is promising for creating optoacoustic transducers that would generate an acoustic beam with a given wave front.

2. Optical properties of a nanoparticle array

In this work the problem of the interaction of a pumping laser pulse with a 2D array of plasmonic nanoparticles is considered. Fig. 1 shows a scheme of a numerical experiment. An optical pulse propagating in the air falls orthogonally onto a glass substrate. A periodic 2D array of gold nanoparticles was located on the substrate surface. A study of nanoparticles optical spectrum was conducted for the rectangular nanoparticles with smoothed edges, which represent the simplest option for fabrication by the method of electron-beam lithography. Hemispherical nanoparticles were also considered. The particles in the array were equidistant and the array of nanoparticles had an axis of symmetry of the 4th order. The simulation was carried out by the finite element method in the frequency domain. For the calculation the periodic boundary conditions were applied, so modeling could be carried out for only one period of the array. The dispersion of the refractive index of air and the substrate was not taken into account. The dielectric constant of gold was described according to the Drude–Lorentz model by the formula

$$\varepsilon_{DL}(\omega) = \varepsilon_{\text{inf}} - \frac{\omega_{p,D}^2}{\omega^2 + i\gamma_D\omega} - \frac{s_1\omega_{p1,L}}{(\omega^2 - \omega_{p1,L}^2) + i\gamma_{1,L}\omega} - \frac{s_2\omega_{p2,L}^2}{(\omega^2 - \omega_{p2,L}^2) + i\gamma_{2,L}\omega}. \quad (1)$$

Here ε_{inf} is the permittivity limit at high frequencies, $\omega_{p,D}$ and γ_D denote the plasma frequency and damping coefficient from the Drude model, respectively. The third and the fourth term in Eq. (1) are the two additional Lorentzians with resonance frequencies $\omega_{p1,L}$ and $\omega_{p2,L}$, where $\gamma_{1,L}$ and $\gamma_{2,L}$ represent the spectral widths of the two resonances with s_1 and s_2 being their weighting

factors. The coefficients in Eq.(1) are taken from [12].

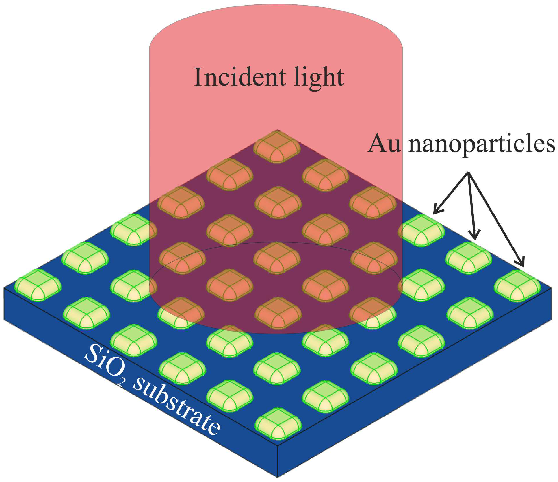

FIG. 1: Scheme of nanoparticles array. (In color)

Fig. 2(a) shows the absorption, transmission, and reflection spectra of an array of rectangular nanoparticles. The following parameters were used in the calculations: nanoparticle width 500 nm, array period 750 nm, nanoparticle height 100 nm. The dashed line shows the absorption of a homogeneous metal film with a thickness of 100 nm. It can be seen that the absorption spectrum contains lines that substantially exceed the absorption level of a homogeneous film. Thus, it proves that the use of a structured optoacoustic transducer leads to an increase in the efficiency of optical energy conversion. However as can be seen from Fig. 2(b) the distribution of the energy absorption coefficient inside a single nanoparticle is non-uniform regardless of the chosen mode. The strongest heating occurs at the edges of the nanoparticles. It should be noted that in the case of heating by a continuous laser beam this absorption distribution would lead to the formation of a uniform close to rectangular temperature distribution inside the nanoparticle. As a rule, it is this distribution that is used as the initial one when calculating the dynamics of acoustic oscillations [8–10]. However as shown in [13], in the case of a femtosecond laser pump pulse the initial temperature distribution must coincide with the absorption distribution. Thus, when considering the acoustic problem, it is necessary to use the initial pressure distribution similar to the absorption distribution in Fig. 2(b).

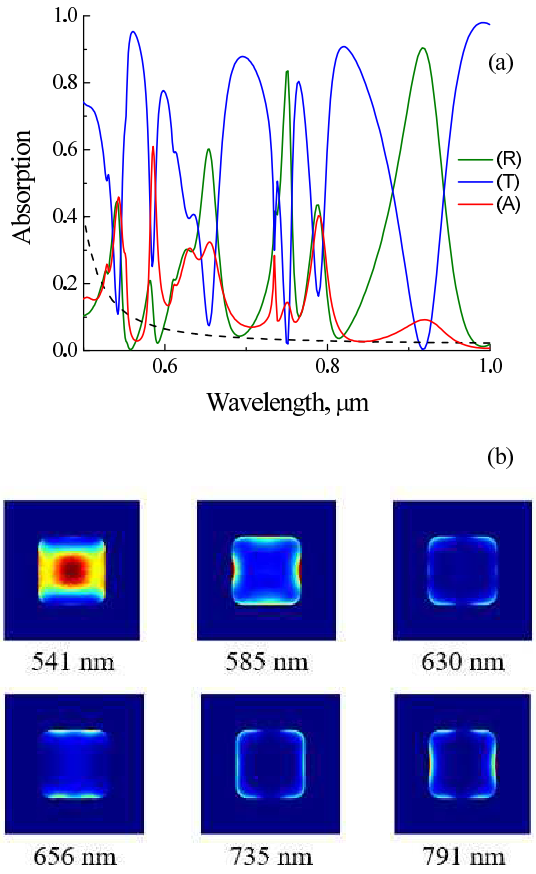

FIG. 2. Optical characteristics of the rectangular nanoparticle array: (a) - spectrum of reflection (R), absorption (A) and transmission (T); (b) - distribution of optical absorption in a nanoparticle on resonance wavelengths. (In color)

An optoacoustic transducer with an inhomogeneous absorption distribution may be designed via varying the distance between nanoparticles, as proposed in [14]. However, the spectral absorption lines for an array of square nanoparticles are strongly shifted by varying the distance between the particles (see Fig. 3). Therefore, the creation of a given distribution of the absorption coefficient by changing the distance between the particles is a less preferred method.

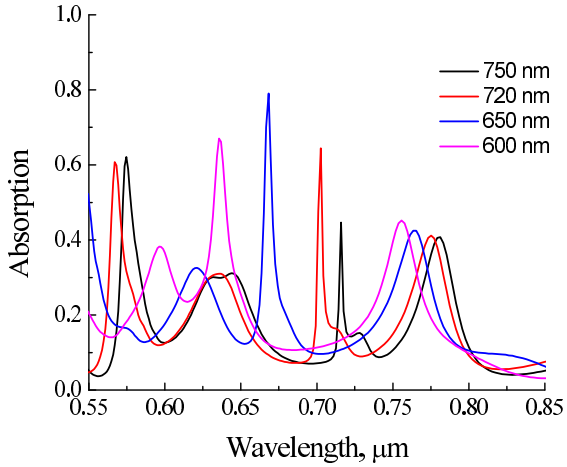

FIG. 3. Absorption spectra for different period of the rectangular nanoparticles array. (In color)

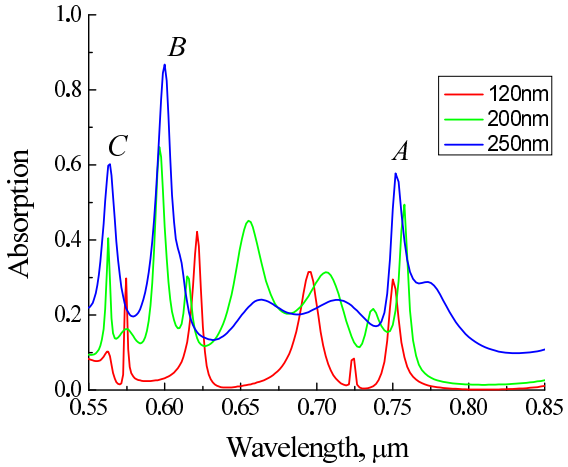

FIG. 4. Absorption spectrum of the hemispherical nanoparticles array for different radius of the particles. (In color)

Another method is to change the size of nanoparticles while the array period is constant. This method was considered on the example of an array of hemispherical nanoparticles. The period of this array was 750 nm. The hemisphere radius ranged from 120 nm to 250 nm. Fig. 4 shows the absorption spectra of the array for different radius values. In the figure, one can see three main absorption peaks. Despite the fact that peak B has a high absorption level compared to A and C, the C mode is most interesting. We calculated the optical absorption distributions inside the nanoparticle for B and C modes, see Fig. 5. The absorption distribution for mode C has hexagonal symmetry in plane of substrate and, therefore, provides a more uniform temperature distribution over the nanoparticle. Also from Fig. 4 it can be seen that the spectral position of mode C is not shifted when the size of the hemisphere changes. In this case, the absorption coefficient substantially depends on the radius of the nanoparticle. Thus, by changing the size of the nanoparticles, with a constant array period, a given absorption distribution on the surface of the transducer can be created.

Unfortunately, the inhomogeneous absorption distribution inside a single nanoparticle, in particular, an increase in absorption near the edges of the nanoparticle can disrupt the homogeneity of the phase front of an acoustic wave formed as a result of the interference of acoustic waves from single nanoparticles. From Fig. 5 it can be seen that the absorption is maximal at the nanoparticle-substrate interface and has an annular distribution. In this case, mode C provides the most homogeneous distribution of absorption. Therefore, the initial pressure distribution in the acoustic wave will be the most homogeneous both in the transducer plane and in the orthogonal direction. So the absorption distribution for mode C (see Fig. 5(b)) was chosen as the initial one when solving the acoustic problem.

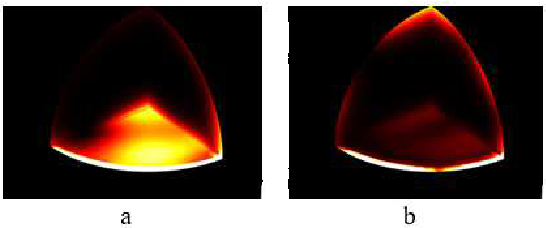

FIG. 5. 3D cross-section of absorption in the hemispherical nanoparticle for B and C modes. (In color)

3. Dynamics of acoustic waves generated by optoacoustic transducer

The principle of operation of the optoacoustic transducer on plasmon nanoparticles is as follows. When the energy of an ultrashort laser pulse is absorbed by a metal nanoparticle, its local heating, expansion and generation of an acoustic pulse occur. These processes can be effectively modeled by numerically solving the equations of motion of the medium in the Lagrange form [15]. For many practically important cases of interaction of radiation with an absorbing structure of a specific geometry (flat, cylindrical, or spherical), numerical solutions of a one-dimensional problem have been obtained that adequately describe the generation and propagation of an acoustic pulse [16, 17]. However, the effect of pulsed laser radiation on the absorbing structure of an arbitrary geometry, which can initiate the excitation of an acoustic pulse of a spatially inhomogeneous structure, requires the development, in general, of a three-dimensional model.

In this paper, a model of the thermo-optical effect of ultrashort laser pulses on metal absorbing targets (structures) of arbitrary geometry is developed on the basis of solving three-dimensional equations of the motion of continuous media in the Lagrange form.

The Lagrange equations for the three-dimensional motion of a continuous medium have the form [18]:

- a continuity equation:

$$V = V_0 \left[\frac{\partial x_\mathrm{e}}{\partial x_\mathrm{l}} \left(\frac{\partial y_\mathrm{e}}{\partial y_\mathrm{l}} \frac{\partial z_\mathrm{e}}{\partial z_\mathrm{l}} - \frac{\partial y_\mathrm{e}}{\partial z_\mathrm{l}} \frac{\partial z_\mathrm{e}}{\partial y_\mathrm{l}} \right) - \frac{\partial y_\mathrm{e}}{\partial x_\mathrm{l}} \left(\frac{\partial x_\mathrm{e}}{\partial y_\mathrm{l}} \frac{\partial z_\mathrm{e}}{\partial z_\mathrm{l}} - \frac{\partial x_\mathrm{e}}{\partial z_\mathrm{l}} \frac{\partial z_\mathrm{e}}{\partial y_\mathrm{l}} \right) + \frac{\partial z_\mathrm{e}}{\partial x_\mathrm{l}} \left(\frac{\partial x_\mathrm{e}}{\partial y_\mathrm{l}} \frac{\partial y_\mathrm{e}}{\partial z_\mathrm{l}} - \frac{\partial x_\mathrm{e}}{\partial z_\mathrm{l}} \frac{\partial y_\mathrm{e}}{\partial y_\mathrm{l}} \right) \right]; \quad (2)$$

- equations of motion:

$$\frac{\partial^2 x_\mathrm{e}}{\partial t^2} \frac{\partial x_\mathrm{e}}{\partial x_\mathrm{l}} + \frac{\partial^2 y_\mathrm{e}}{\partial t^2} \frac{\partial y_\mathrm{e}}{\partial x_\mathrm{l}} + \frac{\partial^2 z_\mathrm{e}}{\partial t^2} \frac{\partial z_\mathrm{e}}{\partial x_\mathrm{l}} = -\frac{1}{\rho} \frac{\partial P}{\partial x_\mathrm{l}}, \quad (3)$$

$$\frac{\partial^2 x_\mathrm{e}}{\partial t^2} \frac{\partial x_\mathrm{e}}{\partial y_\mathrm{l}} + \frac{\partial^2 y_\mathrm{e}}{\partial t^2} \frac{\partial y_\mathrm{e}}{\partial y_\mathrm{l}} + \frac{\partial^2 z_\mathrm{e}}{\partial t^2} \frac{\partial z_\mathrm{e}}{\partial y_\mathrm{l}} = -\frac{1}{\rho} \frac{\partial P}{\partial y_\mathrm{l}}, \quad (4)$$

$$\frac{\partial^2 x_\mathrm{e}}{\partial t^2} \frac{\partial x_\mathrm{e}}{\partial z_\mathrm{l}} + \frac{\partial^2 y_\mathrm{e}}{\partial t^2} \frac{\partial y_\mathrm{e}}{\partial z_\mathrm{l}} + \frac{\partial^2 z_\mathrm{e}}{\partial t^2} \frac{\partial z_\mathrm{e}}{\partial z_\mathrm{l}} = -\frac{1}{\rho} \frac{\partial P}{\partial z_\mathrm{l}}; \quad (5)$$

- equations of the changing of Euler coordinates:

$$u_{x_\mathrm{e}} = \frac{\partial x_\mathrm{e}}{\partial t}, u_{y_\mathrm{e}} = \frac{\partial y_\mathrm{e}}{\partial t}, u_{z_\mathrm{e}} = \frac{\partial z_\mathrm{e}}{\partial t}; \quad (6)$$

- an equation of state:

$$P = P\left(V,\ E\right). \quad (7)$$

Here $x_\mathrm{e}, y_\mathrm{e}, z_\mathrm{e}$ are the Euler coordinates; P is the pressure; $V_0 = 1/\rho_0$, $V = 1/\rho$ are the initial and current specific volumes; ρ_0, ρ are the corresponding densities; E is the specific internal energy. The system of equations (2–7) are used to describe thermomechanical processes both in a metal structures and in a crystal substrate with the appropriate choice of material constants.

Theoretical consideration of the interaction

of ultrashort laser pulses with metals requires the development of models that take into account the essentially nonequilibrium nature of the process of laser heating of metals. To solve the problem of heating metallic nanostructures upon absorption of the energy of an ultrashort laser pulse, it is customary to use the so-called two-temperature model for the electron gas and the ion lattice [19]. In the framework of this approach, the following equations are written for the electron and ion subsystems:

$$\rho_e C_e \frac{\partial T_e}{\partial t} = k_T^e \left(\frac{\partial^2 T_e}{\partial x_1{}^2} + \frac{\partial^2 T_e}{\partial y_1{}^2} + \frac{\partial^2 T_e}{\partial z_1{}^2} \right) + Q_S - \gamma \left(T_e - T_i \right), \quad (8)$$

$$\rho_i C_i \frac{\partial T_i}{\partial t} = \gamma \left(T_e - T_i \right). \quad (9)$$

Here the quantities ρ (density), C (heat capacity), T (temperature) with the index "e" refer to the electronic subsystem, with the index "i" – to the ion one. The parameter γ determines the rate of the energy relaxation from the electron gas to ions of the crystal lattice. The value of Q_S in equation (8) is determined by the energy release source: $Q_S = I\left(x_1, y_1, z_1, t\right) k_{abs}$, where $I\left(t, x_1, y_1, z_1\right) = I_0 f_t\left(t\right) f_{xyz}\left(x_1, y_1, z_1\right)$ is the intensity of the light beam, k_{abs} is the absorption coefficient of the medium, x_1, y_1, z_1 are Lagrangian coordinates. To describe the time shape of a laser pulse, a power-exponential function was used $f_t\left(t\right) = \left(t/\tau_{\text{p}}\right) \exp\left[-t/\tau_{\text{p}}\right]$, where τ_{p} is a pulse duration.

To approximate the equation of state of a metallic film, we use the Mie-Grüneisen equation, which, taking into account the separation of two subsystems (electronic and ionic ones), takes the form [20, 21]:

$$P = \rho_{i0} u_0^2 \left(1 - \frac{V_i}{V_{i0}}\right) + \Gamma_i \frac{C_i \left(T_i - T_0\right)}{V_i} + \Gamma_e \frac{C_e \left(T_e - T_0\right)}{V_e}, \quad (10)$$

where $\Gamma_{i,e}$ are the Grüneisen coefficients, u_0 is the speed of sound in the metal. As the equation of state of the substrate, the Mie-Grüneisen equation in its binomial form has been also used:

$$P = \rho_0 u_l^2 \left(1 - \frac{V}{V_0}\right) + \Gamma \frac{C\left(T - T_0\right)}{V}, \quad (11)$$

where Γ is the Grüneisen coefficient of the substrate, u_l is the speed of sound in the substrate.

The simultaneous numerical solution of the system of equations (2–11) makes it possible to calculate the space-time dependencies of pressure, temperature, density and velocity of motion, both in the metal nanostructures and in the crystal substrate. Simulation has been carried out using the finite-difference approximation of the equations of motion in the form of Lagrange. When implementing the numerical method, the calculation region has been divided into cells of the size of the order $\Delta x_1, \Delta y_1, \Delta z_1 = 0.05$ nm. The time step has been determined by the Courant criterion $\Delta t = k \Delta x_1 / u$, where u is the sound velocity in the material, $k = 0.1 \div 0.01$.

A numerical experiment was carried out for the case of the excitation of acoustic pulses when the energy of an ultrashort laser pulse with the intensity $I_0 = 10^8$ W/cm^2 and duration $\tau_{\text{p}} = 10^{-13}$ s is absorbed by a gold nanoparticle in the form of a rectangular parallelepiped located on the surface of the GaAs crystal. Geometrical parameters of the particle: height 70 nm, width 30 nm. Calculations were carried out for typical thermophysical parameters of Au and GaAs. When solving the thermal problem, all material parameters were considered constant, without regard for their temperature dependence.

Figures 6 and 7 show the spatial distributions of the pressure field in the structure at different time moments under chosen conditions of excitation. Fig. 6 shows the change in the pressure field in the crystal substrate in the plane perpendicular to the propagation of the laser beam. It can be seen that after the transition process that lasts 1-2 picoseconds, and equalizes the initial temperature and pressure inside the nanoparticle, the nanoparticle is a quasispherical source of acoustic oscillations

with a period determined by the longitudinal particle size and sound velocity in the material $T = 2L/u_0$.

For the case of an array of metal nanoparticles, Fig. 7 shows the cross section of the pressure distribution in the structure in the plane containing the interface between the metal nanoparticles — the surface of the crystalline substrate and the direction of propagation of the acoustic pulse deep into the material. As can be seen, at a distance of 3–4 wavelengths of the acoustic train from the substrate surface, the wave fronts of individual sources of acoustic vibrations largely overlap, forming a common wave front. This result allows us to consider an array of metal nanoparticles as a prospective optoacoustic transducer for efficient generation of a macroscopic acoustic beam, whose structure will be determined by the spatial distribution of the absorption coefficient of the array of nanoparticles.

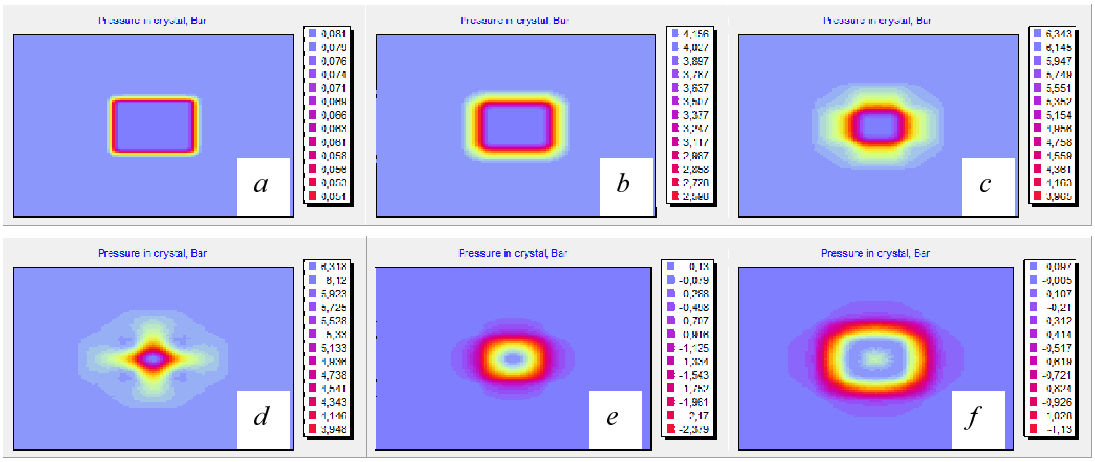

FIG. 6. Distribution of pressure in crystal at different time moments: 400 fs (a), 700 fs (b), 1000 fs (c), 1300 fs (d), 2400 fs (e), 3000 fs (f). (In color)

4. Conclusions

We performed a detailed study of the absorption of a periodic array of plasmonic nanoparticles. It has been showed that the application of plasmonic structures as an optoacoustic absorber can leads to a significant increase in the efficiency of conversion of optical energy to acoustic one due to absorption in conditions of plasmon excitation resonance. The numerical calculations show that it is necessary to consider each plasmonic nanoparticle deposited on the surface as a source of a quasi-spherical subterahertz acoustic signal, and in the far-field a superposition these partial signals leads to formation of acoustic beam with a wave front which depends on distribution of energy source absorbed by the array of plasmonic nanoparticles.

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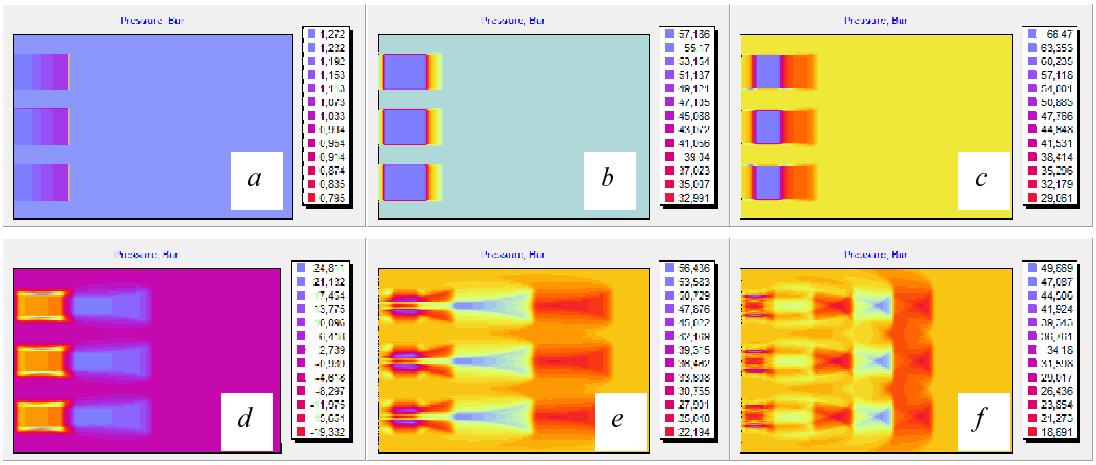

FIG. 7. Distribution of pressure in the structure at different time moments: 50 fs (a), 500 fs (b), 1 ps (c), 3.5 ps (d), 7 ps (e), 13 ps (f). (In color)

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