http://dx.doi.org/10.35596/1729-7648-2021-19-8-58-62

Original paper

UDC 535.21:534

PHOTOACOUSTIC EFFECT IN MICRO- AND NANOSTRUCTURES: NUMERICAL SIMULATIONS OF LAGRANGE EQUATIONS

OLEG G. ROMANOV, YAKOV K. SHTYKOV, IGOR A. TIMOSHCHENKO

Belarusian State University (Minsk, Republic of Belarus)

Submitted 22 November 2021

Abstract. The work provides the description of theoretical and numerical modeling techniques of thermomechanical effects that take place in absorbing micro- and nanostructures of different materials under the action of pulsed laser radiation. A proposed technique of the numerical simulation is based on the solution of equations of motion of continuous media in the form of Lagrange for spatially inhomogeneous media. This model allows calculating fields of temperature, pressure, density, and velocity of the medium depending on the parameters of laser pulses and the characteristics of micro- and nanostructures.

Keywords: short laser pulses, nanoacoustics, Lagrange equations, numerical simulations.

Conflict of interests. The authors declare no conflict of interests.

For citation. Romanov O.G, Shtykov Y.K, Timoshchenko I.A. Photoacoustic effect in micro- and nanostructures: numerical simulations of Lagrange equations. Doklady BGUIR. 2021; 19(8): 58-62.

Introduction

The photoacoustic effect is the formation of sound waves due to pulsed heating of the absorbing medium upon the absorption of electromagnetic radiation. Currently, the photoacoustic effect is widely used in biomedical research [1], photoacoustic spectroscopy [2], and other applications. The studies of the interaction of pulsed laser radiation with absorbing micro- and nanostructures are of particular interest. The absorption of laser pulse energy in micro- and nanostructures makes it possible to excite acoustic vibrations in the frequency range from gigahertz to terahertz. Such oscillations are of particular interest for fundamental research and have many potential applications (acoustic visualization of nanoobjects, acoustic nanocavities, phonon crystals) [3–7].

This work is devoted to the development of a technique for modeling the problems of thermooptical excitation of acoustic vibrations in absorbing micro- and nanostructures, based on the numerical solution of the equations of motion of continuous media in the Lagrange form. Using the developed technique, the problems of excitation of ultra high-frequency oscillations in spherical micro- and nanoparticles of various materials are considered.

Theoretical model

The Lagrange equations for one-dimensional spherically symmetric motion of a continuous medium have the following forms [8]:

– equation of continuity:

$$V = V_0 \left(\frac{R}{r} \right)^2 \frac{\partial R}{\partial r}; \tag{1}$$

– equation of motion:

$$\frac{\partial u}{\partial t} = -V_0 \left(\frac{R}{r} \right)^2 \frac{\partial P}{\partial r}; \tag{2}$$

– equation of the Euler coordinate R change:

$$\frac{\partial R}{\partial t} = u. \tag{3}$$

Here V_0, V are the initial and current specific volumes $V_0 = 1/\rho_0$, $V = 1/\rho$, where ρ_0, ρ are the corresponding densities, r is the Lagrangian coordinate, t is the time.

As an equation of state in the problems under consideration, it is advisable to use the Mie – Grüneisen equation in its two-term form [9]:

$$P = \rho_0 u_0^2 \left(1 - \frac{V}{V_0} \right) + \Gamma \frac{C_V (T - T_0)}{V}. \tag{4}$$

Here $\Gamma = \frac{u_0^2 \beta}{C_V}$ is the Grünheisen coefficient, β is the volume expansion coefficient, C_V is the heat capacity, and u_0 is the sound speed in the medium.

To find the change in the temperature of the medium, we will use the following heat conduction equation:

$$\rho C_V \frac{\partial T}{\partial t} = k_h \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right) + Q_S. \tag{5}$$

Here k_h is the coefficient of thermal conductivity of the medium. The quantity Q_S is determined by the source of energy release: $Q_S = k_{abs} I(r,t)$, where k_{abs} is the absorption coefficient of the medium, $I(t,r) = I_0 f_t(t) f_r(r)$ is the intensity of the light beam. A power-exponential function $f_t(t) = t/t_p e^{-t/t_p}$ was chosen as the temporal shape of the laser pulse, where t_p is the duration of the laser pulse; the spatial function has the form $f_r(r) = \begin{cases} 1, r \le R_0 \\ 0, r > R_0 \end{cases}$, which, in the case of the spherical geometry of the absorbing particle, corresponds to a sphere of radius R_0.

The solution of the system of equations (1) – (5) makes it possible to calculate the space-time dependences of pressure, temperature, density, and velocity, to estimate the contribution of thermal and acoustic mechanisms to the change in the physical parameters of a continuous medium. Note that if it is necessary to describe thermomechanical phenomena in metallic nanostructures under the action of ultrashort laser pulses, this approach should be supplemented with the usage of a two-temperature model of metal heating, which makes it possible to study processes at times shorter than the electron-phonon relaxation time. In this case, the heat equation takes the following form [10]:

$$\rho_e C_e \frac{\partial T_e}{\partial t} = k_h^e \frac{1}{r^2} \frac{\partial}{\partial r}\left(r^2 \frac{\partial T_e}{\partial r} \right) + Q_S - \gamma\left(T_e - T_i\right); \quad (6)$$

$$\rho_i C_i \frac{\partial T_i}{\partial t} = \gamma\left(T_e - T_i\right). \quad (7)$$

Here the values ρ (density), C (heat capacity), T (temperature), k_h (thermal conductivity) with the index "*e*" refer to the electronic subsystem, with the index "*i*" – to the ionic one. The parameter γ determines the rate of energy relaxation from the electron gas to the ions of the crystal lattice.

To approximate the equation of state of a metal nanoparticle, we use the Mie – Grüneisen equation, which, taking into account the separation of two subsystems (electronic and ionic), takes the form [6]:

$$P = \rho_{i0} u_{0m}^2 \left(1 - \frac{V_i}{V_{i0}}\right) + \Gamma_i \frac{C_i\left(T_i - T_0\right)}{V_i} + \Gamma_e \frac{C_e\left(T_e - T_0\right)}{V_e}. \quad (8)$$

Here V_{i0}, $V_{i,e}$ are the initial and current specific volumes, $V_{i0} = 1/\rho_{i0}$, $V_{i,e} = 1/\rho_{i,e}$, where ρ_{i0}, $\rho_{i,e}$ are the corresponding densities, $\Gamma_{i,e}$ are the Grünheisen coefficients, u_{0m} is the sound speed in a metal particle.

The numerical solution of this system of equations was carried out by constructing a finite-difference approximation using artificial viscosity to stabilize the solution in the presence of pressure jumps [11].

Results of numerical simulations and discussion

Before proceeding to the discussion of the results of numerical simulations, let us establish approximate requirements for the duration of a laser pulse for efficient excitation of acoustic signals in micro- and nanoparticles. To implement isochoric heating, it is necessary that the duration of the laser pulse be less than the acoustic relaxation time t_a of the heated region (less than the time of passage of a sound wave through it). For a spherical particle $t_a = 2R_0 / u_0$. For typical values of the sound speed, this value can be estimated for micron-sized particles as $t_a \approx 10^{-9}$ s, and $t_a \approx 10^{-11}$ s for particles with a radius of 10 nm. Consequently, in the first case, it is advisable to use laser radiation with a pulse duration of the picosecond range, and to excite acoustic vibrations of nanometer particles, it is necessary to use radiation of a subpicosecond (femtosecond) duration.

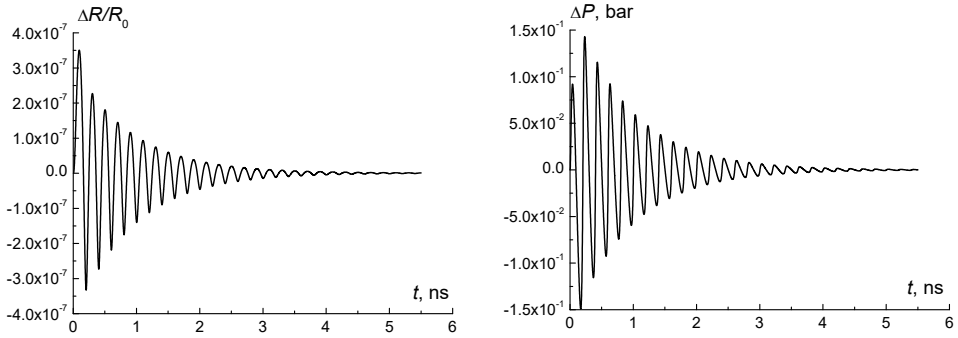
As the first example, consider the problem of the excitation of acoustic pulses in a spherical microparticle simulating an agglomerate of close-packed carbon nanotubes [12]. We assume the radius of the absorbing particle $R_0 = 1$ μm, the absorption coefficient $k_{abs} = 2.4 \cdot 10^5$ cm^{-1}, the duration of the laser pulse $t_p = 10^{-11}$ s, and the intensity $I_0 = 10^6$ W/cm^2. The rest parameters used in the calculations are the typical values for single-walled carbon nanotubes and water as environment.

As the second example, the problem of the excitation of acoustic vibrations in spherical gold nanoparticles in an aqueous environment is considered. In this case, a two-temperature model of metal heating under the action of an ultrashort laser pulse (equations (6)–(7)) and equation of state (8) were used. The radius of the gold nanoparticle was assumed equal $R_0 = 10$ nm, the absorption coefficient $k_{abs} = 5 \cdot 10^5$ cm^{-1}, the duration of the laser pulse $t_p = 10^{-13}$ s, and the intensity $I_0 = 10^8$ W/cm^2. The calculations were performed for typical thermophysical parameters of gold.

The results of numerical modeling of two problems are presented in Fig. 1 and 2. Let's carry out a comparative analysis. In the first case, the duration of the laser pulse is sufficient, according to the theoretical estimation, for effective excitation of acoustic vibrations of the microparticle (Fig. 1, *a*) and for the formation of a train of acoustic vibrations in the environment surrounding

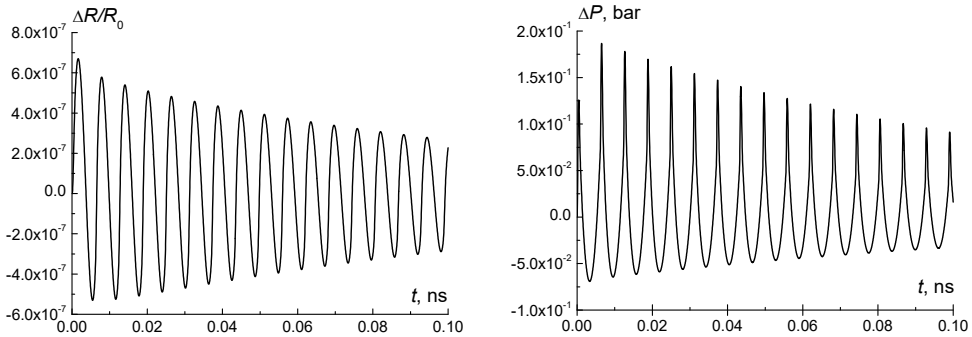
the particle (Fig. 1, *b*). A train of compression-rarefaction pulses in the environment is formed due to the reflection of a pressure wave at the interface between the absorbing particle/external environment. The period of oscillations (changes in the radius of a spherical particle) in this case is $T \approx 200$ ps.

In the case of a metal (Au) nanoparticle, a change in temperature due to the absorption of the laser pulse energy also initiates the appearance of radial pressure waves ΔP, density $\Delta\rho$, and the velocity of motion of particles of the medium u, propagating at the speed of sound inside the nanoparticle and reflected from its surface and the center of symmetry. In this case, the presence of wave-like motion of particles of the medium can be considered as the emergence of its radial oscillations. For example, Fig. 2 shows the time dependence of the relative radial displacement of the nanoparticle surface. The period of the excited acoustic oscillations is $T \approx 6.2$ ps for the case under consideration, the amplitude of the oscillations is directly proportional to the intensity of the exciting laser pulse. A pressure wave propagating inside a particle upon reflection from the interface between two media on the surface of a nanoparticle initiates pressure fluctuations in the environment. In this case, the first peak in the time dependence of ΔP (r = 10,1 nm) (Fig. 2, *b*) is associated with the heating of the near-surface layer of the liquid at the time of the laser pulse action, while the rest are the result of the partial passage of the compression-rarefaction wave through the gold–water interface.



a *b*

Fig. 1. Time dependence of changes in the radius of carbon microparticle (*a*) and pressure in the environment (*b*)



a *b*

Fig. 2. Time dependence of changes in the radius of gold nanoparticle (*a*) and pressure in the environment (*b*)

Conclusion

In conclusion, for the theoretical description of thermomechanical phenomena in absorbing micro- and nanostructures under the action of pulsed laser radiation, a technique has been developed for the numerical simulation of the equations of continuous media motion in the form of the Lagrange equation and the heat conduction equation. It is shown that, when the duration of laser pulses is shorter than the acoustic relaxation time, effective excitation of acoustic vibrations in micro- and nanostructures occurs. Examples of carbon microparticles and gold nanoparticles are considered in details.

References

1. Attia A.B.E., Balasundaram G., Moothanchery M., Dinish U.S., Bi R., Ntziachristos V., Olivo M. A review of clinical photoacoustic imaging: Current and future trends. *Photoacoustic*s. 2019;16:100144. https://doi.org/10.1016/j.pacs.2019.100144
2. Dumitras D.C., Petrus M., Bratu A.M., Popa C. Applications of Near Infrared Photoacoustic Spectroscopy for Analysis of Human Respiration: A Review. *Molecules.* 2020;25(7):1728. http://doi.org/10.3390/molecules25071728.
3. van Capel P.J.S., Peronne E., Dijkhuis J.I. Nonlinear ultrafast acoustics at the nano scale. *Ultrasonics*. 2015;56:36-51. https://doi.org/10.1016/j.ultras.2014.09.021.
4. Smith R.J., Perez Cota F., Marques L., Chen X., Arca A., Webb K., Aylott J., Somekh M.G., Clark M. Optically excited nanoscale ultrasonic transducers. *J. Acoust. Soc. Am.* 2015;137:219-227. https://doi.org/10.1121/1.4904487.
5. Taruttis A., Ntziachristos V. Advances in real-time multispectral optoacoustic imaging and its applications. *Nature Photonics*. 2015;9:219-227. https://doi.org/10.1038/nphoton.2015.29.
6. Romanov O.G., Romanov G.S. Thermomechanical effect of ultrashort laser pulses on single-dimension metallic nanostructures. *Bulletin of the Russian Academy of Sciences: Physics*. 2014;78(12):1299-1302. https://doi.org/10.3103/s1062873814120260.
7. Khokhlov N., Knyazev G., Glavin B., Shtykov Y., Romanov O., Belotelov V. Interaction of surface plasmon polaritons and acoustic waves inside an acoustic cavity. *Optics Letters*. 2017;42(18):3558-3561. https://doi.org/10.1364/OL.42.003558.
8. Richtmayer R.D., Morton K.W. *Difference methods for initial-value problems*. Interscience publishers a division of John Wiley & Sons; 1967.
9. Zel'dovich Ya.B., Raizer Yu.P. *Fizika udarnykh voln i vysokotemperaturnykh gidrodinamicheskikh yavlenii* (*Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena*). Moskow: Nauka; 1966.
10. Anisimov S.I., Imas Ya.A., Romanov G.S., Khodyko Y.V. Deistvie izlucheniya bol'shoi moshchnosti na metally (The Effect of High Power Radiation onto Metals). Moskow: Nauka; 1970.
11. Romanov O.G., Zheltov G.I., Romanov G.S. Numerical modeling of thermomechanical processes in absorption of laser radiation in spatially inhomogeneous media. *Journal of Engineering Physics and Thermophysics*. 2011;84(4):772-780. https://doi.org/10.1007/s10891-011-0533-5.
12. Golubewa L., Timoshchenko I., Romanov O., Karpicz R., Kulahava T., Rutkauskas D., Shuba M., Dementjev A., Svirko Yu., Kuzhir P. Single-walled carbon nanotubes as a photo-thermo-acoustic cancer theranostic agent: theory and proof of the concept experiment. *Scientific Reports*. 2020;10:22174. https://doi.org/10.1038/s41598-020-79238-6.

Authors' contribution

Romanov O.G. developed the theoretical model, prepared the manuscript of the article.

Shtykov Y.K. performed computer simulations of the photoacoustic effect in metal nanostructures.

Timoshchenko I.A. performed computer simulations of the photoacoustic effect in carbon micro- and nanostructures.

All authors took part in the discussion of the results of the work.

Information about the authors

Romanov O.G., PhD., Assosiate Professor, Head of the Department of Computer Simulations of the Faculty of Physics of Belarusian State University.

Shtykov Y.K., Postgraduate student at the Faculty of Physics of Belarusian State University.

Timoshenko I.A. Senior Lecturer at the Faculty of Physics of Belarusian State University.

Address for correspondence

220030, Republic of Belarus,
Minsk, Nezavisimosti Ave., 4,
Belarusian State University;
tel. +375-17-209-51-21;
e-mail: romanov@bsu.by
Romanov Oleg Gennadievich