Contents lists available at ScienceDirect

## Applied Mathematical Modelling

journal homepage: www.elsevier.com/locate/apm

# Tuning the characteristics of photoacoustic pressure in a laser-induced photoacoustic generator: A numerical study

### Sangmo Kang<sup>a,\*</sup>, Jeeseong Hwang<sup>b</sup>

<sup>a</sup> Department of Mechanical Engineering, Dong-A University, Busan 49315, Republic of Korea <sup>b</sup> Applied Physics Division, National Institute of Standards and Technology, Boulder, CO 80305, USA

#### ARTICLE INFO

Article history: Received 7 September 2020 Revised 16 December 2020 Accepted 20 December 2020 Available online 19 January 2021

Keywords:

Coupled acoustic and elastic wave equations Hyperbolic heat conduction equation Integrated thermal-elastic-acoustic multiphysics simulations Laser-induced photoacoustic generator Perfectly matched layers Staggered-grid finite-difference time-domain method

#### ABSTRACT

We present a new numerical approach to model a laser-induced photoacoustic generator (LIPAG) by integrating thermal, elastic, and acoustic multiphysics simulations in a linear regime. Our unique approach implements a fully-explicit staggered-grid finite-difference time-domain method with perfectly matched layers to remove common artifacts in numerical simulations due to finite-sized computational domains. The approach simulates the dynamics of photoacoustic (PA) pressure signals and local temperature fields using realistic LIPAG models to elucidate details of the underlying PA pressure generation mechanism. We also report on how the pressure wave characteristics are affected by the variation of the key parameters of the pulsed light and material properties. Our simulation technique, capable of adjusting a wide range of key parameters in the LIPAG model, may provide a guidance in the design of LIPAGs to achieve desired PA pressure characteristics for testing the performance of acoustic transducers and their calibrations.

© 2021 Elsevier Inc. All rights reserved.

#### 1. Introduction

Ultrasound represents sound waves with frequencies above the human hearing limit, from 20 kHz up to several GHz [1]. Due to low scattering properties of the ultrasound signal in light scattering media that scatters optical wavelengths, ultrasound technologies have been used in a broad range of applications in biomedical science especially for medical imaging and diagnosis of deep tissues [2,3]. For improved spatio-temporal resolution, much effort has been made in developing transducers capable of detecting broadband and high frequency acoustic or pressure signals [4]. However, developing accurate performance tests of these new transducers has been challenging due to lack of proper sources that generate broadband and high frequency ultrasound involve piezoelectric materials driven by a time-variant voltage. However, the frequency spectral bandwidth is limited by the bandwidth of the piezoelectric response of the transducers, making it hard to generate a broadband ultrasound signal with a single transducer. To this end, exploration of new types of ultrasound generators with improved signal bandwidth and adjustable pressure characteristics has been of paramount interest.

In the past decades, laser-induced photoacoustic generators (LIPAGs) have been introduced to generate acoustic signals by modulated light sources to achieve broadband and high intensity ultrasound sources [5–9]. The pioneering works of experiments and models of LIPAG techniques are described elsewhere [10–13]. In LIPAGs, a sound or acoustic wave is generated by a photoacoustic (PA) mechanism [14,15] by a pulsed optical source with a pulse duration shorter than those of thermal

\* Corresponding author.

E-mail address: kangsm@dau.ac.kr (S. Kang).







relaxation time and stress relaxation time, producing an acoustic pressure in the form of a local elastic wave by photothermal absorption and thermoelastic expansion processes. Then the elastic wave is converted to a pressure wave propagating through a surrounding medium, water or gas. In the recent decades, a variety of PA technologies employing LIPAGs have been demonstrated in various practical applications such as characterization of turbid media, trace gas and chemical reaction analysis, medical diagnosis and interventions, and biomedical imaging in a variety of state-of-the-art platforms such as photoacoustic spectroscopy, microscopy, and tomography [3,10,12,14,16–21].

Despite the rigor of the physics-based models mentioned above, development of a flexible model universally applicable to different types of LIPAGs is still in its infancy. Most theoretical studies still rely on simplified analytical models for qualitative interpretation of experimental results. Such analytical models often require oversimplified assumptions about the characteristics of the materials involved or are solved within a limited parameter space to keep the mathematics simple enough to easily model and interpret. For example, an analytical solution to a time-resolved LIPAG involving optically turbid media is solved assuming that the material's absorption coefficient is limited to a few hundred cm<sup>-1</sup> [16]. As an alternative approach, a numerical simulation technique has been introduced [22]. The technique does not need mathematics-limited simplifications and enables solutions even when analytical solutions are impossible due to the complexity of systems and material properties.

Despite their advantages, numerical techniques have seen limited application to the modeling of LIPAGs with their complex geometry due to the requirement of large computational resources. The numerical technique has been partially demonstrated by evaluating an analytical solution to the PA signal in a low absorption coefficient breast-mimicking phantom with the aid of a Monte Carlo simulation of the photothermal field [23]. Since Zhou et al. [7] performed a finite-element analysis on laser-based thermoelastic ultrasound generation, Tian et al. [22] has remained as the only reported whole numerical study of a 3D optical fiber-based LIPAG but reduced to a 2D axisymmetric geometry, using the commercial COMSOL Multiphysics software based on a finite-element method (FEM) and a fully-implicit scheme. Open sources or commercial finitedifference time-domain (FDTD) have recently been developed to simulate more complex electromagnetic (EM) problems but have not been implemented for the EM problems coupled with pulsed acoustics yet [24–26].

Highly resolved multiscale simulations such as those using COMSOL or other software packages can be time-consuming. Further reduction of the computation time is possible with a geometric symmetry in a LIPAG model to reduce the 3D simulation to 2D. Our new numerical approach enables fast and flexible simulations with adjustable parameters for the properties and geometries of the materials and an optical source in a model LIPAG. In essence, it is based on the integration of thermal, elastic, and acoustic multiphysics simulations. In brief, a heat equation is solved for a time-variant temperature field induced by the absorption of pulsed optical energy in a light absorbing layer (typically a solid material). Next, two wave equations, one for an acousto-elastic wave localized in the solid and the other for a pressure wave in a surrounding fluid, are linked with a heat equation and are solved for a space and time-varying pressure wave propagating into the surrounding medium. We derive three sets of the first-order hyperbolic differential equations involving the above multiphysics and apply a fully-explicit staggered-grid finite-difference time-domain (FDTD) method with perfectly matched layers (PMLs) [27,28] allowing for a pressure wave to escape through the finite-size computational domain. This approach not only increases computational speed and accuracy but also avoids artifacts due to a finite-sized domain, which are common problem in any numerical simulation. Details on implementing the FDTD and PML are described later.

Our simulation also enables delineation of the underlying dynamics of PA pressure generation, such as the spatiotemporal evolution of the photothermally induced temperature field and propagating pressure field. Our approach can also easily adjust the thermal, mechanical, or optical properties of the light absorbing and elastic layers, including their thermal expansion coefficient, thermal conductivity, and heat capacity, or mass density, Young's modulus, and Poisson's ratio. Characteristic parameters such as pulse width, beam size, repetition rate, and peak pulse energy are also adjustable. Our method with a wide range of adjustable parameters would be instrumental to guide the design of PA pressure generators with desired properties.

#### 2. Numerical method

Our numerical approach, consisting of an integrated thermoelastic and acoustic multiphysics simulation, is capable of simulating LIPAGs in 3D. Due to the axisymmetric geometry of the pulsed light and the sample, our model LIPAG is reduced to 2D geometry problem. Our multilayer model sample consists of two solid layers and a water layer: a sufficiently thick elastic backing layer; a thin planar light absorbing elastic wave generating layer above; and a top-most semi-infinite extent of water as a pressure wave propagating layer (Fig. 1). Only a limited number of realistic assumptions are used. All materials in the LIPAG model are assumed to be isotropic and homogeneous. For simplicity, no acoustic absorption in the water medium is accounted for in the governing equations, but the acoustic absorption or attenuation in the solid layers is taken into account. A pulsed Gaussian laser pulse irradiates the top of a light absorbing layer from above through the optically transparent water layer, and its absorbed photonic energy is converted into thermal energy in the layer. A cylindrical coordinate, respectively.

Once the pulsed laser irradiates a light absorbing layer initially in a thermally and mechanically equilibrium state, the local temperature increases in the irradiated region and its vicinity, resulting in a local stress field to induce local material strains or displacements of thermally excited molecules. The displacements are expressed as elastic wave equations in space



**Fig. 1.** Structure of a LIPAG for the numerical simulation based on an integrated thermoelastic and acoustic multiphysics model. A focused pulsed laser irradiates the origin point (O). PML is a perfectly matched layer to avoid artifacts due to a finite-sized computational domain. See text for details of the other parameters.

and time-varying displacement velocity fields. Simulations in this study focus on a linear regime where the pulsed laser energy is limited within a certain level to avoid nonlinear effects such as nonlinear temperature dependencies of stress, heat flux, and local pressure. Thus, all governing equations involving heat conduction and coupled acoustic and elastic waves can be linearized from the equilibrium state in our model. Unless specified otherwise, changes of all the primary variables in this study reflect their deviations from the equilibrium steady states. In the steady-state water medium, convection flow is negligible so that the flow velocity becomes zero, namely  $D/Dt \approx \partial/\partial t$ .

#### 2.1. Relevant governing equations

A numerical simulation of the LIPAG model described above solves the following three sets of differential equations: (i) the equations of a heat conduction in the whole computational domain, including two solid layers and a water medium; (ii) the elastic wave equations in the two solid layers (light absorption and elastic backing layers below); and (iii) the propagating pressure wave equation in the water medium. The first set, involving a temperature field change, consists of the following first-order hyperbolic heat conduction equations written in the temperature and heat-flux formulation [29,30]:

$$\rho C_p \frac{\partial T}{\partial t} = -\left[\frac{\partial Q_x}{\partial x} + \frac{\partial (rQ_r)}{r\partial r}\right] + \dot{q}_s,\tag{1}$$

$$Q_x + \zeta \frac{\partial Q_x}{\partial t} = -k \frac{\partial T}{\partial x},\tag{2}$$

$$Q_r + \zeta \frac{\partial Q_r}{\partial t} = -k \frac{\partial T}{\partial r},\tag{3}$$

where *T* [K] denotes the temperature field change,  $(Q_x, Q_r)$  [W/m<sup>2</sup>] the heat flux field expressed in a component form  $(Q_\theta = 0$  in the axisymmetric configuration),  $\rho$  [kg/m<sup>3</sup>] the mass density,  $\dot{q}_s$  [W/m<sup>3</sup>] the power density from a single laser pulse,  $C_p$  [J/(kg·K)] the isobaric specific heat (capacity), k [W/(m·K)] the thermal conductivity, and  $\zeta$  [s] the thermal relaxation time of heat conduction (the time lag between the heat flux introduction and consequent temperature gradient) as [30]

$$\zeta = \frac{\alpha}{c_h^2} = \frac{\kappa}{\rho C_p c_h^2},\tag{4}$$

where  $c_h$  [m/s] is the speed of heat propagation and  $\alpha$  [m<sup>2</sup>/s] the thermal diffusivity. Note that, with zero relaxation time ( $\zeta = 0$ ), Eqs. (2) and (3) reduce to differential forms of Fourier's law of thermal conduction. To solve the heat conduction Eqs. (1)–(3), the following continuity conditions are applied to the interface between the light absorbing layer ( $s_a$ ) and the water medium (w). Assuming no loss across the interface, the temperature and normal component of the heat flux are continuous:

$$T|_{s_a} = T|_{w}, \tag{5}$$

$$Q_x|_{s_a} = Q_x|_w, \tag{6}$$

where the subscript *x* denotes a normal direction to the interface which coincides with the axial direction  $(Q_n = Q_x)$ . Note that the same conditions are also applied to the interface between the light absorbing layer and the elastic backing layer  $(s_b)$ .

In the heat conduction Eq. (1), the volumetric power density,  $\dot{q}_s$  [W/m<sup>3</sup>], originates from the conversion of the absorbed optical energy from a pulsed light irradiation to the thermal energy in the light absorbing layer. This power density from a single laser pulse into heat can be written as [22,31]

$$\dot{q}_{s}(x,r,t) = I_{o}(r,t)\frac{1}{\delta_{p}}\exp\left(\frac{x}{\delta_{p}}\right),\tag{7}$$

where *x* denotes the *negative* axial coordinate away from the top surface,  $\delta_p$  [m] the penetration depth of the light or a reciprocal of the optical absorption coefficient (i.e.  $\delta_p = 1/\mu_a$ ) and  $I_o$  [W/m<sup>2</sup>] the incident areal density of the optical power on the top surface of the light absorbing layer, expressed as

$$I_{o}(r,t) = A \, \exp\left(\frac{-2r^{2}}{w_{p}^{2}}\right) \exp\left[\frac{-4 \, \ln 2 \, \left(t - t_{c}\right)^{2}}{\tau_{p}^{2}}\right],\tag{8}$$

$$A = \frac{4 (\ln 2)^{1/2} E_p}{\pi^{3/2} w_p^2 \tau_p} .$$
(9)

Here, the spatio-temporal density distribution,  $I_o(r, t)$ , of the irradiated pulsed light is assumed to be Gaussian in time as well as in the radial direction,  $w_p$  [m] is the full radius of the  $1/e^2$  maximum of the Gaussian pulsed laser at the focus, and  $\tau_p$  [s] the full width of the half maximum (FWHM) of the Gaussian pulse in time, and  $t_c$  the peak time of the pulsed light. The coefficient, A, is derived by equating the total energy of the light irradiation energy,  $E_p$  [J], to that of the heat release as follows:

$$E_p = \int_{-\infty}^{\infty} \int_{0}^{\infty} \int_{-\infty}^{0} 2\pi r \dot{q}_s dx dr dt .$$
<sup>(10)</sup>

To simulate an elastic wave in the solid layers and a propagating pressure wave in a water medium, we simultaneously solve the six elastic wave equations in the solid layers and a pressure wave equation in water (Fig. 1). The sound generation and propagation in the solid layers are governed by the following six first-order elastic wave equations written in a velocity and a stress formulation [7,27]:

$$\rho \frac{\partial v_x}{\partial t} = \frac{\partial \tau_{xx}}{\partial x} + \frac{\partial (r\tau_{xr})}{r\partial r},\tag{11}$$

$$\rho \frac{\partial v_r}{\partial t} = \frac{\partial \tau_{xr}}{\partial x} + \frac{\partial (r\tau_{rr})}{r\partial r} - \frac{\tau_{\theta\theta}}{r}, \qquad (12)$$

$$\frac{\partial \tau_{xx}}{\partial t} = (\lambda + 2\mu) \frac{\partial \nu_x}{\partial x} + \lambda \frac{\partial (r\nu_r)}{r\partial r} - \left(\lambda + \frac{2}{3}\mu\right)\beta \frac{\partial T}{\partial t},\tag{13}$$

$$\frac{\partial \tau_{rr}}{\partial t} = \lambda \frac{\partial \nu_x}{\partial x} + \lambda \frac{\partial (r\nu_r)}{r\partial r} + 2\mu \frac{\partial \nu_r}{\partial r} - \left(\lambda + \frac{2}{3}\mu\right)\beta \frac{\partial T}{\partial t},\tag{14}$$

$$\frac{\partial \tau_{\theta\theta}}{\partial t} = \lambda \frac{\partial v_x}{\partial x} + \lambda \frac{\partial (rv_r)}{r\partial r} + 2\mu \frac{v_r}{r} - \left(\lambda + \frac{2}{3}\mu\right)\beta \frac{\partial T}{\partial t},\tag{15}$$

$$\frac{\partial \tau_{xr}}{\partial t} = \mu \left( \frac{\partial \nu_x}{\partial r} + \frac{\partial \nu_r}{\partial x} \right). \tag{16}$$

In the first two equations above, the velocity field vector,  $(v_x, v_r) = (du_x/dt, du_r/dt)$  [m/s] is defined by the time derivative of the material's local displacement vector,  $(u_x, u_r)$  [m], with  $v_{\theta} = 0$ , considering axi-symmetry. In rest of the equations, the mechanical stress field components ( $\tau_{xx}$ ,  $\tau_{rr}$ ,  $\tau_{\theta\theta}$ ,  $\tau_{xr}$ ;  $\tau_{x\theta} = \tau_{r\theta} = 0$ ) [Pa] are equated to the derivatives of the velocity field

components, taking into account thermal expansion of the material with a volumetric thermal expansion coefficient of  $\beta$  [1/K]. Here,  $\lambda$  and  $\mu$  [Pa] are the first and the second Lamé constants defined by

$$\lambda = \frac{E\nu}{(1+\nu)(1-2\nu)},$$
(17)

$$\mu = \frac{L}{2(1+\nu)},$$
(18)

where *E* [Pa] is the Young's modulus and  $\nu$  the Poisson's ratio ( $-1 < \nu < 0.5$ ).

In a water medium, the sound propagation is governed by the following first-order pressure wave equations written in a velocity and pressure formulation [32]:

$$\rho \frac{\partial v_x}{\partial t} = -\frac{\partial p}{\partial x},\tag{19}$$

$$\rho \frac{\partial v_r}{\partial t} = -\frac{\partial p}{\partial r},\tag{20}$$

$$\frac{\partial p}{\partial t} = -\frac{1}{\kappa} \frac{\partial v_x}{\partial x} - \frac{1}{\kappa} \frac{\partial (rv_r)}{r\partial r} + \frac{1}{\kappa} \beta \frac{\partial T}{\partial t} .$$
(21)

Here, *p* [Pa] is the pressure field and  $\kappa = 1/(\rho c_s^2)$  [1/Pa] the isothermal compressibility, where  $c_s$  [m/s] is the speed of sound. Note that Eqs. (19)–(21) are derived by plugging  $\tau_{xx} = \tau_{rr} = \tau_{\theta\theta} = -p$ ,  $\tau_{xr} = 0$ ,  $\lambda = 1/\kappa$  and  $\mu = 0$  into Eqs. (11)–(16).

In order to simultaneously solve the governing wave Eqs. (11)–(16) and (19)–(21), the following interface conditions are applied. Assuming no energy loss across the interface between the light absorbing layer ( $s_a$ ) and the backing layer ( $s_b$ ), the element's velocity and stress components are continuous at their interface:

$$\nu_i|_{s_a} = \nu_i|_{s_b} \ (i = x, r), \tag{22}$$

$$\tau_{ij}|_{s_a} = \tau_{ij}|_{s_b} \ (ij = xx, \ rr, \ \theta\theta, \ xr).$$
(23)

Across the interface (at x = 0) between  $s_a$  and water (w),

$$\nu_X|_{S_a} = \nu_X|_W, \tag{24}$$

$$\tau_{xx}|_{s_a} = -p|_W . \tag{25}$$

#### 2.2. Numerical solution schemes

To numerically solve the aforementioned equations, we apply a leap-frog scheme of the fully-explicit FDTD method with staggered grid systems separately constructed for the time domain or for the space domain as well. Fig. 2 shows such grid systems adopted for the integrated thermal, elastic, and acoustic multiphysics simulations. In the time domain shown in panel (a), all components of the heat flux and velocity ( $Q_i$  and  $v_i$ ) are located at the center of the temporal grid cell, while the temperature, pressure and all components of the stress (T, p and  $\tau_{ij}$ ) are placed at the face of the grid cell. In the space domain in panel (b), the temperature, pressure, and normal components of the stress [T, p and  $\tau_{ij}$  (i=j)] are located at the center, the components of heat flux and velocity ( $Q_i$  and  $v_i$ ) at the corresponding faces, and the shear stress [ $\tau_{ij}$  ( $i \neq j$ )] on the edge. Such staggered-grid systems have a great advantage that the first order temporal and spatial derivatives can be readily discretized using the central finite-difference scheme of a second-order accuracy in numerical analysis without the need for additional interpolation [33].

Reducing computation time is achieved by adopting a finite-sized computational domain. However, bounding the domain often results in artifacts due to reflections of the waves at the domain ends, but the artifacts are eliminated by implementing a PML [32]. The PMLs in Fig. 1 are thin additional layers ( $\delta_i$ ) surrounding the entire computational domain. To implement the PMLs into the wave equations, we introduce the complex coordinates,  $\tilde{\eta}$  as follows:

$$\tilde{\eta} = \int_{0}^{\eta} \left[ 1 + i \frac{\omega_{\eta}(\eta')}{\omega} \right] d\eta', \quad \eta = x, \quad r; \quad \omega_{\eta} \ge 0.$$
(26)

Here, *i* is the imaginary unit,  $\omega_{\eta} / \omega$  the attenuation factor in the complex coordinates. Note that  $\omega_{\eta} = 0$  outside the PMLs. Lastly, the differential Eqs. (1)–(3), (11)–(16) and (19)–(21) are rewritten by a complex coordinate transformation in (26) and subsequently by splitting the field components (not shown here). For the final equations involving the PMLs, refer to Eqs. (43)–(59) with details described elsewhere [27].



**Fig. 2.** Grid systems for the leap-frog scheme of the fully-explicit staggered-grid finite-difference (SGFD) method constructed for the time domain (a) or for the space domain (b) to implement the integrated thermal, elastic, and acoustic multiphysics simulations.

#### Table 1

Thermal and mechanical properties of the materials selected for the integrated thermal-elastic-acoustic multiphysics simulations on the photoacoustic ultrasound generator. Note that, for the light absorption layer, the same physical properties for PDMS employed in the backing layer are used under the assumption that the content of the light absorbing additives embedded into the nanocomposite is low enough.

Properties	Light Absorbing layer (nanocomposite)	Backing layer (PDMS)	Wave propagation layer (Water)
$\rho  [kg/m^3]$	970	970	1000
C <sub>p</sub> [J/kgK]	1460	1460	4180
$k [W/m^2K]$	0.16	0.16	0.61
$\beta$ [10 <sup>-4</sup> /K]	9.1	9.1	2.1
c <sub>s</sub> [m/sec]	-	-	1490
E [kPa]	1650	1650	-
ν	0.499	0.499	-

#### 2.3. Structural details and initial parameters of a simulated LIPAG

The model LIPAG consists of a highly absorbing thin planar layer sandwiched between an elastic backing layer and a water medium which are sufficiently extended laterally. The backing layer is optically transparent ( $\mu_a = 0$ ) polydimethylsiloxane (PDMS) in a solid form, and the light absorption layer is a nanocomposite of PDMS with a small amount of light absorbing additives (e.g. gold nanoparticles or carbon nanotubes) dispersed within it. Typical mass ratio of the light absorbing additives to PDMS in experiment is less than 2% [22], therefore our simulation uses the same thermal and mechanical properties as PDMS for the initial set of parameters. These thermal and mechanical parameters for the light absorbing layer are readily replaceable with experimentally verified values, but we believe that this is a good approximation for the objectives of this study without changing the underlying physics. Table 1 lists the thermal and mechanical properties of PDMS and water from various literatures except the Poisson's ratio of PDMS,  $\nu = 0.499$ , suggested to avoid mathematical singularity of Eq. (17) if a literature value of  $\nu \approx 0.5$  were used [34,35].

The layers in Fig. 1 are circular discs consisting of a 10 µm thick light absorbing layer ( $\mu_a = 1000 \text{ cm}^{-1}$ ), a backing layer and a water medium with thicknesses of  $h_a = 10 \text{ µm}$ ,  $L_+= 250 \text{ µm}$  and  $L_-= 125 \text{ µm}$ , respectively, and with the radius of R=250 µm for sufficiently larger computational bounds than  $h_a$ . The dimensions of PMLs are:  $\delta_{x,+} = 0.2L_+$ ,  $\delta_{x,-} = 0.2L_-$  and  $\delta_r = 0.2R$ . Characteristics of the pulsed laser,  $E_p = 20 \text{ nJ}$ ,  $w_p = 2.5 \text{ µm}$ ,  $\tau_p = 10 \text{ ns}$ ,  $\delta_p = 10 \text{ µm}$  (i.e.  $\mu_a = 1000 \text{ cm}^{-1}$ ), and  $t_c = \tau_p$ were used in Eqs. (7)-(9). The coordinate origin is at the center on the top surface of the top layer, coinciding with the center



**Fig. 3.** Validation of the numerical results. (a) Simulated time-traced pressure amplitudes at  $l_m = 50 \,\mu\text{m}$  to compare the results obtained at two different spatio-temporal resolutions, 'present resolution' ( $0.1 \,\mu\text{m} < \Delta x, \Delta r < 0.8 \,\mu\text{m}; \Delta t = 0.04 \,\text{ns}$ ) and 'higher resolution' ( $0.05 \,\mu\text{m} < \Delta x, \Delta r < 0.4 \,\mu\text{m}; \Delta t = 0.02 \,\text{ns}$ ). (b) Simulated time-traced pressure amplitudes at  $l_m = 50 \,\mu\text{m}$  to compare between the results obtained by our numerical technique and the ones by the commercial COMSOL Multiphysics package.

of the irradiating Gaussian pulsed light. The time-variant pressure wave intensity in water was calculated at a distance  $(l_m)$  away from the origin along the *x*-axis into the water medium.

For numerical simulations, all the relevant differential Eqs. (1)–(3), (11)–(16) and (19)–(21) are fully explicitly resolved on a staggered mesh with a finite-difference approach, and all the spatial derivatives are discretized with the second-order central difference scheme. In the radial direction, the grids are defined by a tangential-hyperbolic function with the minimum grid size of  $\Delta r = 0.1 \,\mu\text{m}$  at the central axis, gradually increasing to  $\Delta r = 0.8 \,\mu\text{m}$  at the farthest boundary. In the axial (*x* coordinate) direction, the grids are also defined by a tangential-hyperbolic function in each of the three layers so that the grids are of minimum, intermediate and maximum sizes,  $\Delta x = 0.1 \,\mu\text{m}$ , 0.25  $\mu\text{m}$  and 0.8  $\mu\text{m}$ , set respectively on the top and bottom of the light absorbing layer and at both ends. A time step of  $\Delta t = 0.04 \,\text{ns}$  is used in all simulations. Unless otherwise mentioned, all the numerical simulations start at  $t = t_0 = -1.5 \,\tau_p$  to ensure light energy is completely absorbed. To summarize, for the initial set of parameters, a total of 848×570 and 1376 nodes are distributed in the spatial and temporal directions, respectively, corresponding to CFL<sub>max</sub>  $\approx 3.7 \times 10^{-3}$ . In addition, the laser pulse width ( $\tau_p = 10 \,\text{ns}$ ) can be resolved with 250 temporal nodes, which is enough for our study. Note that, at the beginning of the numerical simulations, all the primary variables are assumed to have a value of zero.

In order to confirm the adopted grid and temporal resolutions are sufficiently high and the numerical results are independent of the grid and time resolutions, results with the resolution defined above  $(0.1 \,\mu\text{m} < \Delta x, \Delta r < 0.8 \,\mu\text{m}; \Delta t = 0.04 \,\text{ns})$  are compared with those with increased resolution  $(0.05 \,\mu\text{m} < \Delta x, \Delta r < 0.4 \,\mu\text{m}; \Delta t = 0.02 \,\text{ns})$ . Fig. 3(a) compares the resulting time-traced pressure signals at  $l_m = 50 \,\mu\text{m}$ , exhibiting that both results are in good agreement. The lower resolution result in Fig. 3(a) is also compared with one obtained by the commercial COMSOL Multiphysics package (Version 5.3, COM-SOL, Inc. Burlington, MA) in Fig. 3(b), confirming our numerical result also agrees well with that obtained by a commercial COMSOL simulation package regardless of the differences in the solution technique and associated equations. For instance, our numerical approach uses the first-order form of hyperbolic differential equations, fully-explicit FDTD method and PMLs, while the COMSOL employs the second-order form of hyperbolic differential equations, fully-implicit finite-element method and wave radiative boundary condition.

#### 3. Results and discussion

In the first section, results on the dynamics of PA pressure generation process with initial parameters are presented to delineate the underlying photoacoustic mechanism. In the next two sections, we report on how the pressure wave charac-



**Fig. 4.** Simulated temporal traces of the rise of the temperature (a) and the rise of the acoustic pressure (b) during the first 80 ns at the origin point, (x, r)=(0, 0) on the top surface of the light absorbing layer. For comparison, the temporal intensity change of a single laser light pulse during its irradiation,  $f(t) = \exp[-4\ln 2(t - t_c)^2/\tau_p^2]$  in Eq. (8), is also shown with dotted lines (scale on the right axis).

teristics respond to changes in the pulsed light properties and the thermal and mechanical properties of the light absorbing material. The light properties include irradiation energy ( $E_p$ ), beam size ( $w_p$ ), and pulse width ( $\tau_p$ ). The thermal properties include thermal conductivity (k), thermal expansion coefficient ( $\beta$ ), and the mechanical properties include mass density ( $\rho$ ), Young's modulus (E), and Poisson's ratio ( $\nu$ ). To this end, each parameter is varied while keeping the others unchanged.

#### 3.1. Elucidation of the underlying PA pressure generation mechanism in LIPAGs

The commonly accepted PA pressure generation mechanism in LIPAGs is described below. Upon irradiation, energy from a single light pulse is converted to heat in a solid light absorbing layer, resulting in a rapid local temperature rise. This temperature rise induces rapid thermal expansion of the irradiated region from which an elastic wave is produced and propagates outward. Upon reaching the interface with a surrounding water medium, the elastic wave is converted into a pressure wave and propagates through the water. This section demonstrates the details of such phenomena with our simulation results. To this end, the temperature and pressure characteristics at the early stage, within 100 ns after the light irradiation, were studied in detail.

Fig. 4 shows simulated temporal traces of the changes in temperature and acoustic pressure in the light absorbing layer during the first 80 ns (solid line curves). The results shown are at the origin, (x, r) = (0, 0), the top surface of the 10 µm thick light absorption layer under the illumination with a single spatio-temporal Gaussian pulse. The temporally Gaussian profile of the pulsed light (see Eq. (8)) is also displayed in the plots to compare their relative time traces against the temporal profile of the light pulse (dotted line curves). Both temperature and acoustic pressure intensity rise as soon as the pulsed light irradiation begins. Upon reaching the maximum, the temperature stays nearly constant even after the pulsed irradiation is finished (panel (a)), indicative of faster heat deposition than the thermal diffusion rate. Consequently, the temperature rise is attributed to the integral of the optical energy as shown in panel (a). Meanwhile, the time-varying acoustic pressure forms a sinusoid-like bipolar wave packet (panel (b)), where the bipolar waveform is proportional to the first time derivative of the heating pulse and originates from the expansion of water at the interface followed by rarefaction [36,37]. In a general photoacoustic wave equation, the propagating pressure wave is driven by the second time derivative of the temperature field [2]. Consistently, we note that the sinusoid-like pressure waveform in panel (b) resembles the second time derivative form of the sigmoid-like temperature curve in panel (a) but with an asymmetric amplitude and with retarded time probably due to highly localized thermal diffusion [36]. After one acoustic wave packet is generated, no additional acoustic pressure wave appears at the later time, indicating that the longer time cooling process does not contribute to the sound wave generation, and the wave packet is purely from the longitudinal pressure change with no noticeable transverse pressure component which would contribute pressure fluctuations at the later time [38].



**Fig. 5.** (a) Temporally traced amplitudes of the propagating pressure in water simulated at distances,  $l_m = 50 \,\mu\text{m}$ , 100  $\mu\text{m}$ , and 150  $\mu\text{m}$  from the pressure origin. (b) A plot of the full width of the half maximum ( $\tau_p^*$ ) values obtained from the Gaussian fittings of the positive portions in the pressure wave packets. The laser pulse width (10 ns, shown with a dotted line) was fixed for the pressure simulations.

The acoustic wave originated from the solid layer continues to propagate through the water medium as a pressure wave. Figs. 4(b) and 5(a) are the time-varying acoustic pressure at the origin,  $l_m = 0 \mu m$  [Fig. 4(b)] and the time-traced amplitude of the propagating pressure wave in water recorded at different distances,  $l_m = 50 \mu m$ , 100  $\mu m$  and 150  $\mu m$  from the pressure origin in sequence [Fig. 5(a)]. The time-lapse results across the solid-water interface show that the acoustic wave is converted to a pressure wave, propagating away from the origin with a similar waveform shape and at a constant speed, but the amplitude decreases due to the spread of the wave. The positive-to-negative waveform of the time-varying pressure signal indicates a propagating longitudinal pressure wave packet involving compression and rarefaction of the irradiated region.

The dispersion of the pressure waveform is also noted in the propagating wave packet in water as shown Fig. 5(b), a plot of the FWHM ( $\tau_p^*$ ) values obtained from the Gaussian fittings of the positive portions of the wave packets simulated at different  $l_m$  values. The FWHM exponentially increases away from the origin and then converges to a constant value at the far field ( $l_m > 20 \,\mu\text{m}$ ). This short-range FWHM increase may be due to the contribution by multiple waves originated from multiple grid points on the interfacial surface as the irradiated spot size ( $w_p = 2.5 \,\mu\text{m}$ ) is larger than the grid size ( $\Delta x, \Delta r = 0.1 \,\mu\text{m}$ ). In the long range, the converged FWHM value is still smaller than the pulse width ( $\tau_p = 10 \,\text{ns}$ ) of the light, supporting that, in experiments, the pressure signal can be measured at a far-field distance without much temporal dispersion.

Simulation of the spatio-temporally resolved temperature fields at the early stage of PA pressure generation is useful in understanding the dynamics involving thermoelastic energy conversion. In particular, an understanding on the effect of the heat conduction or diffusion on the PA pressure generation would provide useful insight in designing LIPAGs with desired characteristics. In optical resolution PA devices, like our model LIPAG involving a focused light beam with a submicrometer or micrometer size and a PA signal generating material with comparable dimensions, one would think the heat conduction effect is important. However, the heat diffusion time across the light penetration depth (10µm) in PDMS is  $\delta_p^2/\alpha = (10 \mu m)^2/(110 \mu m^2/ms) \approx 0.9 \text{ ms}$  and is much longer than the transit time of the sound wave,  $\approx 9.5 \text{ ns}$ , across the light penetration depth. Also, under a thermal confinement condition with a fast pulsed light with a pulse width of 10 ns, no heat diffusion is expected across the PA generating layer (10µm). Our results confirm this scenario of a rapid heat deposition. Fig. 6(a) and (b) are vertical cross section views of the isothermal contour lines from spatial temperature maps at two different times, (a) at  $\tau_p$  (= 10 ns) and (b) at 2  $\tau_p$  after the laser irradiation/absorption begins. The spatial distributions of the optical intensity by an irradiated laser pulse at  $\tau_p$  (= 10 ns) and at 2  $\tau_p$  are displayed in panel (c) and (d), respectively. The plots (a) and (b) compare distinct spatial distributions of the isothermal lines, one with a slow temperature gradient (loosely packed lines) into the light absorbing layer and the other with a steeper gradient (densely packed lines) near the origin. Referring to Fig. 4(a), the former is the intermediate temperature field while the sample is still being heated, and the



**Fig. 6.** Isothermal contour lines and the optical intensity map of an irradiated laser pulse. (a, b) Vertical cross section views of the isothermal contour lines from simulated spatial temperature maps at two different times, (a) at  $\tau_p$  (= 10 ns) and (b) at 2  $\tau_p$  after the laser irradiation/absorption begins. The outermost isothermal line is for  $\Delta T = 0$  K and the temperature stepwise increases by  $\Delta T = 2$  K from one line to the next toward the origin. (c, d) Spatial distribution of the optical intensity by an irradiated laser pulse at (c)  $\tau_p$  (= 10 ns) and (d) at 2  $\tau_p$ .



**Fig. 7.** Vertical cross section views of the isobaric contour lines from numerically simulated spatial pressure maps. The pressure difference between two adjacent isobaric lines is  $\triangle P = 2.5$  kPa. The numerical results were obtained at two different times, (a) at t = 43.46 ns or (b) at 61.20 ns, when the first maximum or minimum peak, respectively, in the propagating pressure wave packet reaches the vertical distance  $l_m = 50 \,\mu\text{m}$  away from the origin.

latter is when the heating is near-completed. We note that the isothermal lines are accumulated at the origin and at the interface (x = 0 surface), indicative of a rapid heat deposition at the ( $s_a$ -w) interface more heavily at the origin. On the other hand, isothermal lines are far less dense away from the interface, across the sample depth. Furthermore, away from the interface, the inter-line spacing from the time  $\tau_p$  to 2  $\tau_p$  is not significantly changed, indicative of the further heat diffusion across the light absorbing layer after the pulse irradiation time ( $t > 2\tau_p$ ) would be negligible once the sample heating is completed within the time period of  $2\tau_p$ . Negligible heat diffusion and thus rapid heat deposition and temperature rise on the light absorbing layer agrees with the predictions by the Karabutov group [13,16,23,39]. Note that the thermal boundary condition does not affect the temperature field because the computational domain is set very large compared to the thermal diffusion range, which is demonstrated in Fig. 6.

Upon immediate heat deposition, a rapid expansion of the light absorbing layer produces an acoustic wave in the layer, then it is converted to a pressure wave propagating into the water medium in contact. Simulated pressure waves are displayed in Fig. 7 of the vertical cross section views of the isobaric contour lines from spatial pressure maps obtained at two different times, t = 43.46 ns and 61.20 ns. These are the moments when the first maximum or minimum peak, respectively, in the propagating pressure wave packet reaches the vertical distance  $l_m = 50 \,\mu\text{m}$  from the origin. These plots provide rich information on the dynamics of the propagating pressure wave in water. Increasing inter-line spacing from panel (a) to (b) indicates spatial broadening in the radially propagating wave packet both in positive and in negative pressure parts, similar to the evolution in the time-resolved wave packet form in Fig. 5. The wave packet shape, a positive pressure in the leading front and negative in the trailing end, is well preserved in the far field, but the radially measured aspect ratio of the wave packet is largest along the vertical axis (+*x*-axis) and keeps decreasing toward the solid-water interface (horizontal axis). In the near-field (*x* or  $r \leq 25 \,\mu\text{m}$ ), far behind the trailing end of the negative pressure packet or along near the solid-water interface, the distribution of the isobaric lines is irregular, resulting in highly complex pressure waveforms.

#### 3.2. Tuning pressure wave characteristics by adjusting the properties of the pulsed light

One of the advantages of the LIPAG to demonstrate in this work is a flexibility to tune characteristics, such as the amplitude, spatio-temporal dispersion and frequency spectrum of the PA pressure wave. This section demonstrates simulations on the dependency of PA pressure characteristics on the changes of specific properties of the pulsed light or of the light absorbing material. To this end, the simulated time-varying amplitudes at  $l_m = 50 \,\mu\text{m}$  of the propagating pressure waves are presented in (a) panels in Figs. 8–11 under the variation of only one parameter of the pulsed light properties while keeping the others unchanged. The (b) panels of Figs. 8–11 and (a-f) panels of Fig. 12 are plots of the maximum and minimum peak values ( $p_{\text{max}}$  and  $p_{\text{min}}$ ) from the time-varying intensity versus the parameter under variation.

Fig. 8 demonstrates that the dynamic pressure intensity is readily controllable by changing the total light energy,  $E_p$ . While the temporal wave packet shape remains the same, the peak pressures both for the positive and for the negative packet linearly increase as  $E_p$  increases up to 100 nJ. This linearity is consistent with previously reported experimental results on low light absorbing metallic mirrors in the linear regime [13] and with the fact that all the relevant governing Eqs. (1)–(3), (11)–(16) and (19)–(21) can be fully normalized by  $E_p$  for small variations of the primary variables (displace-



**Fig. 8.** Pressure wave characteristics under the variation of laser energy. (a) Temporal pressure wave packets constructed from the simulated time-varying amplitudes at  $l_m = 50 \,\mu\text{m}$  of the propagating pressure waves under the variation of laser pulse energies. (b) A plot of the maximum (the positive peak) and minimum (the negative peak) peak values obtained from the time-varying wave packets under the variation of E<sub>p</sub>. Fitted lines,  $(p_{\text{max}}, p_{\text{min}}) \approx (2.2, -1.3)E_p$ , are presented along with the simulated data.

ment, temperature, and pressure) from the equilibrium state. Note that the result on metallic mirrors is from the total (peak- to-peak) PA pressure amplitude [13], but our result reports positive (compressive) and negative (rarefaction) values separately.

Since the acoustic pressure is proportional to the photon fluence *F*, as  $F \approx E_p/w_p^2$ , another way to control the acoustic pressure characteristics is by changing the beam size,  $w_p$ , while keeping  $E_p$  and  $\tau_p$  unchanged. The beam size-dependent time-traced pressure amplitude in Fig. 9(a) shows that the pressure peak values decrease as  $w_p$  increases. Intuitively, the energy disperses over a larger area at higher  $w_p$ , thereby the temperature field spatially broadens to produce less PA pressure per unit area. The peak values of both the positive and negative packet in Fig. 9(b) exhibit  $1/w_p^2$  dependency at  $w_p \gtrsim 40 \,\mu\text{m}$ . However, a gradual increase at a lower rate than  $1/w_p^2$  is observed when  $w_p$  is further decreased from 40  $\mu\text{m}$  to 10  $\mu\text{m}$  and saturates to a constant peak value below 10  $\mu\text{m}$ , demonstrating an unexpected nonlinear PA response to the photon fluence in this high fluence regime. Therefore, we recognize that controlling the PA pressure in this regime only by adjusting the beam size would be challenging.

Based on our simulated result that the time scales of both temperature-rise and pressure-increase are comparable to the laser pulse (Fig. 4), a strong influence of the pulse width on the pressure characteristics is expected. From plots of the time-varying pressure amplitude vs the laser pulse width shown in Fig. 10(a), we note a short laser pulse of 2 ns width ( $\tau_p = 2$  ns) induces a sharp rise-and-fall of the positive pressure followed by a time period with a steady pressure then a sharp fall-and-rise of the negative pressure. The pressure change rate decreases as  $\tau_p$  increases, implying that a faster deposition of thermal energy by a shorter light pulse induces a more rapidly changing PA pressure. Furthermore, the time period between the two pressure packets decreases as  $\tau_p$  increases then disappears eventually to result in a smooth transition from positive to negative pressures. The trailing negative pressure wave packet represents a rarefaction [37] indicative of a longitudinal propagation of the PA pulse through the water, consistent with Fig. 7. We also note in Fig. 10(b) that the peak pressures both for the negative and the positive packet decrease in inverse proportion as the pulse width increases. A plot of the laser pulse width vs the pressure pulse width in Fig. 10(c) displays their near-linear relationship at laser pulse widths  $\leq 10$  ns but a growing deviation from the linear relation as  $\tau_p$  increases further due to reduced contribution by high frequency PA pressures, suggesting further temporal frequency domain studies for quantitative explanation of the deviation. The above results suggest that the laser pulse width in LIPAGs determines not only the width but also the amplitude of the PA pressure wave.

#### 3.3. Tuning pressure wave characteristics by adjusting the optical properties of the light absorbing layer

In typical LIPAGs, the light absorbing layers are thin films in sub-millimeter to micrometer scale thicknesses. When the layer thickness  $(h_a)$  or  $\mu_a$  is reduced so that the penetration depth  $(\delta_p)$  becomes comparable to  $h_a$ , the ratio of  $h_a$  to  $\delta_p$ ,



**Fig. 9.** Pressure wave characteristics under the variation of laser beam size. (a) Temporal pressure wave packets constructed from the simulated timevarying amplitudes at  $l_m = 50 \,\mu\text{m}$  of the propagating pressure waves under the variation of laser beam size,  $w_p$ . (b) A plot of the maximum (the positive peak) and minimum (the negative peak) peak values obtained from the time-varying wave packets under the variation of  $w_p$ . Fitted lines,  $(p_{\text{max}}, p_{\text{min}}) \approx$  $(5.5 \times 10^4, -3.3 \times 10^4)/w_p^{-2}$ , are presented along with the simulated data.

not  $\delta_p$  by itself, strongly influences the characteristics of the generated pressure. After integrating Eq. (7) across the layer thickness, the conversion ratio of the total irradiated optical energy ( $E_p$ ) to heat energy converted from the optical energy ( $E_p^*$ ) and consequent pressure energy is:

With  $h_a$  unchanged, the larger the  $\delta_p$  (i.e. less light absorbing), the less acoustic energy is produced because the optical energy is only partially absorbed by the thin layer. Fig. 11(a) shows the effect of  $\delta_p$  change on the dynamics of PA pressure characteristics when  $h_a$  is fixed at 10 µm. Qualitatively, the peak amplitude of a propagating pressure decreases with increasing  $\delta_p$  (i.e. decreasing  $\mu_a$ ) due to decreased ratio of  $E^*_p/E_p$  as Eq. (27) describes. It is expected that the sample region from which thermoelastic energy contributes to PA pressure generation is extended into the deeper sample region as  $\delta_p$  increases, consistent with broader width of the pressure wave packet and its delayed propagation (note right-shifting wave packets) as  $\delta_p$  increases. We also note that the wave packet shape at  $\delta_p \geq h_a$  (=10 µm) becomes non-Gaussian-like, especially in the negative packet. In this regime, a thermoelastic response of the entire layer needs to be considered for quantitative explanation of the generated PA pressure, which is beyond the scope of present study. From Fig. 11(b), we also note that the pressure amplitude peaks decrease in inverse proportion as the penetration depth increases (i.e.  $p \sim 1/\delta_p$ ), which will be explained later by Eq. (28).

#### 3.4. Tuning pressure wave characteristics by adjusting the physical properties of the light absorption layer

As the PA pressure generation involves a photothermal excitation followed by an impulsive mechanical expansion of the material, the PA pressure characteristics are expected to be controllable by adjusting the thermal and mechanical properties of the light absorbing layer. In experiments, different base polymeric materials may be chosen, or the concentration of light absorbing additive nanomaterials may be adjusted to tune those properties.

For studies of the influence by changes of essential thermal properties, pressure amplitudes at different values of thermal conductivity (k), thermal expansion coefficient ( $\beta$ ), and isobaric specific heat ( $C_p$ ) are displayed in Fig. 12(a), (b), and (c), respectively. Fig. 12(a) shows insensitive dependency on the change of thermal conductivity (k), which was also expected from an analytical solution described below, where the pressure peak value is independent of k. In LIPAGs, an analytical solution of the peak value  $p_o$  of the leading packet of the acoustic wave is known as [2,15]:

$$p_o = \Gamma A_e = \frac{\beta F E}{3\rho C_p (1 - 2\nu)\delta_p},\tag{28}$$



**Fig. 10.** Pressure wave characteristics under the variation of laser pulse width. (a) Temporal pressure wave packets constructed from the time-varying amplitudes at  $l_m = 50 \,\mu\text{m}$  at various laser pulse widths,  $\tau_p$ . (b) A plot of the maximum (the positive peak) and minimum (the negative peak) peak values obtained from the time-varying wave packets under the variation of  $\tau_p$ . Fitted lines,  $(p_{\text{max}}, p_{\text{min}}) \approx (4.9 \times 10^2, -2.0 \times 10^2)/\tau_p$ , are presented along with the simulated data. (c) A plot of the width of the positive-part packet in the propagating pressure wave versus  $\tau_p$  as shown in panel (a). The laser pulse widths  $(\tau_p^* = \tau_p)$  are plotted also for comparison.

where the Grüneisen parameter  $\Gamma$  is defined as  $\Gamma = \beta K/\rho C_p$ , with the bulk modulus  $K = E/3(1 - 2\nu)$ , which is a reciprocal of the isothermal compressibility, and the volumetric optical absorption  $A_e = \mu_a F = F/\delta_p$ . The result is also consistent with what Fig. 6 demonstrates: the heat diffusion across the sample domain  $(10 \,\mu\text{m})$  is much slower than the pulse duration under a thermal confinement condition, making the *k*-dependency insignificant. Although not covered in the present study, this result raises a potential concern in an experiment involving repeated irradiation by a high repetition rate laser. In this situation, regardless of the thermal conductivity value of the material, localized accumulation of the thermal energy is expected to be significant, which may result in excessive temperature rise to induce thermal damage to the material or cavitation, preventing an effective generation of the PA pressure. Furthermore, significant non-linear response would make a quantitative analysis challenging.

While properties of the PA pressure are not readily controllable by thermal conductivity (*k*), Eq. (28) suggests that the adjustment of thermal expansion coefficient ( $\beta$ ) or specific heat ( $C_p$ ) would enable its control. Fig. 12(b) and (c) display that the simulation resulted in what the analytical Eq. (28) also predicts: both positive and negative peak values in both plots fit well to the expected  $\beta$  and  $C_p^{-1}$  dependencies, respectively. In LIPAGs, under a thermal confinement condition characterized by negligible *k*-dependency, the adjustment only of  $\beta$  and/or  $C_p$  is important in controlling the PA pressure characteristics. As seen in Eqs. (13)–(15) and (21), the backing layer and water medium are also be considered for thermal expansion effect in the wave equations. However, thermal expansions in those layers turned out not to contribute to the pressure intensity because temperature changes in those layers during the short pressure generation time scale are negligible.

Eq. (28) also implies that the adjustment of the density ( $\rho$ ), Young's modulus (*E*), and Poisson's ratio ( $\nu$ ) of the light absorbing layer would allow for the control of the PA pressure properties. Note that the change in mechanical properties of the light absorbing layer will also change the impedance of the layer, resulting in different transmission of the PA signal



**Fig. 11.** Pressure wave characteristics under the variation of laser penetration depth. (a) Temporal pressure wave packets constructed from the simulated time-varying amplitudes at  $l_m = 50 \,\mu\text{m}$  of the propagating pressure waves under the variation of laser penetration depth, ( $\delta_p = 1/\mu_a$ ) (b) A plot of the maximum (the positive peak) and minimum (the negative peak) peak values obtained from the time-varying wave packets under the variation of  $\delta_p$ . Fitted lines,  $p_{\text{max}} \approx 580/(\delta_p + 3.2)$  and  $p_{\text{min}} \approx -260/(\delta_p + 1.7)$ , are presented along with the simulated data.

into the water medium. In this study, the influence by the impedance change is not considered because the PA pressure is generated from the layer's surface in contact with the water medium. Also, the focus of this study is on elucidating the trend how the PA pressure is influenced by the change of one mechanical property at a time. The impedance factor can readily be implemented as a constant transmission factor in Eq. (28), which will not change our trend analyses discussed below. Fig. 12(d), (e), and (f), respectively display how the peak pressures depend on these parameters. Fig. 12(d) clearly exhibits  $\rho^{-1}$  dependency of both the positive and negative pressure peaks. Along with Fig. 12(c), note that, as either of the density or specific heat increases, the PA pressure amplitude is inversely proportional to the volumetric heat capacity,  $\rho C_p$ . In practice, the product  $\rho C_p$ , with the values  $\rho$  and  $C_p$  values multiplied, is adjusted as the change in one value always leads to the concatenated change of the other. For instance, as the concentration of the additive metal particles and carbon nanotubes increases in a polymer base material, mass density, thermal conductivity, and Young's modulus increase, but the specific heat, thermal expansion coefficient and Poisson's ratio decrease concatenately [40–42].

In biomedical applications of PA elastography, Young's modulus measurement is important in assessing the stiffnesses of tissues as a meaningful disease biomarker as it has been demonstrated by photoacoustic strain imaging under applied stresses [43] or by ultrasound evaluations of the speed of shear or longitudinal sound wave [38]. In the present study, PA pressure intensity is simulated with the change of the sample's Young's modulus to directly relate the dependency of pressure intensity on the Young's modulus of the light absorbing layer. The analytical expression of the PA pressure increase is expressed also by Young's modulus and Poisson's ratio as shown in Eq. (28), so the pressure increase is linearly proportional to the Young's modulus. Fig. 12(e) shows that the simulated peak-to-peak pressure value linearly increases as the Young's modulus increases up to  $\approx 3$  MPa, where the Hooke's law is valid in this linear regime with small strains and stresses. The upper limit of the Young's modulus is well below of that of biological tissues except bones, suggesting that the tissue's Young's moduli or stiffnesses might be calculated from the measured PA pressure characteristics with a full knowledge of other physical properties of the materials and the pulsed light. However, the nonlinearity at Young's modulus material. A detailed description of this nonlinear behavior and why the nonlinearity is not shown in the negative peak pressure values may be a subject for future studies.

As the three parameters, elastic moduli, Young's, shear, and bulk moduli, are related with each other, Poisson's ratio ( $\nu$ ) dictates their interdependency which is described in Eq. (28). Poisson's ratio ( $-1 \le \nu \le 0.5$ ) is the ratio of the transverse contraction to the longitudinal extension in the direction of applied stretching force. For highly elastic PDMS,  $\nu$  is close to 0.5, which is theoretically the largest possible value, making the first Lamé constant mathematically singular when  $\nu = 0.5$  is used, making numerical simulation impossible. To avoid such singularity,  $\nu = 0.499$  for PDMS was used for this study [34,35]. For different polymer materials, the Poisson's ratio varies between 0.3 and 0.5, and the Eq. (28) predicts the pressure peak



**Fig. 12.** Plots of the maximum (the positive peak) and minimum (the negative peak) peak values obtained from the time-varying wave packets under the variation of the light absorbing layer's properties, including (a) thermal conductivity (*k*), (b) thermal expansion coefficient ( $\beta$ ), (c) isobaric heat capacity ( $C_p$ ), (d) mass density ( $\rho$ ), (e) Young's modulus (*E*), and (f) Poisson's ratio ( $\nu$ ). Fitted curves using Eq. (28) are presented along with the simulated data points. The first, the second or the third number in the parenthesis is the coefficient of each fitting equation for  $p_i$  ( $p_{max}$ ,  $p_{min}$ , or  $\Delta p$ ), respectively.

values proportional to  $(1-2\nu)^{-1}$ . The simulated positive and negative peak pressure values in Fig. 12(f) for  $(0.45 \le \nu \le 0.499)$  demonstrate that the simulated results are well-fitted to the  $(1-2\nu)^{-1}$  dependency from the analytical solution. The PA pressure change is sensitive to the change in Poisson's ratio near  $\nu = 0.5$ . For instance, only 0.2% decrease in  $\nu$ , from 0.499 to 0.498, the peak-to-peak PA pressure value drops by a factor of two. This strong sensitivity near  $\nu = 0.5$  for PDMS in our LIPAG model suggests that knowing the accurate value of the Poisson's ratio of PDMS is important for accurate computation of the PA pressure. However, we note that the pressure change is not sensitive to the change in Poisson's ratio below 0.49. Other polymeric materials including poly(vinyl chloride) plastisol ( $\nu = 0.381-0.387$ ) [44] and polyvinyl alcohol ( $\nu = 0.455-0.485$ ) [45] have recently been introduced for biologically relevant PA phantoms [46–49], and these materials may be used for LIPAGs. Numerical simulations in these LIPAGs would allow for numerical simulations under no influence of the change of the Poisson's ratios.

LIPAGs capable of generating PA pressure of high frequency above 20 MHz and of a broad bandwidth is instrumental to test ultrasound sensors operating at a broad frequency range. In an effort to explore the parameter space to guide the design of such LIPAGs, we further analyzed the dynamic characteristics of the PA pressure with independently varied density ( $\rho$ ), Young's modulus (*E*), or Poisson's ratio ( $\nu$ ), one at a time within the range for relevant values for potential elastic polymeric materials. Fig. 13(a), (c), and (e) demonstrate the time-traced pressure waveforms under the change of  $\rho$ , *E*, and  $\nu$ , respectively. We note that variations in *E* or  $\nu$  result in significant changes to the waveforms with noticeable temporal shifts and high frequency fluctuations as well. Except for the amplitude change, such changes were less significant under the change in  $\rho$ . These differences are characterized better from the corresponding power spectra calculated from the pressure



**Fig. 13.** Time-traced pressure waveforms under the change of  $\rho$ , *E*, and  $\nu$ , respectively and their power spectra. (a) Time-traced pressure waveforms under the change of mass density ( $\rho$ ); (b) Corresponding acoustic power spectra in magnitude calculated from the waveforms in (a); (c) and (e) are time-traced pressure waveforms under the change of Young's modulus (E) and Poisson's ratio ( $\nu$ ), respectively; (d) and (f) are corresponding power spectra in magnitude calculated from the waveforms. The inset graphs of (a), (c), and (e) are zoomed-in view of the boxed region in the main graphs. The inset graphs of (b), (d), and (f) are the main power spectra plots after normalizing them by the peak values of each graph.

waveforms, shown in panel Fig. 13(b), (d), and (f), respectively. The change of the  $\rho$  value mainly affects the magnitude of the power spectrum (Fig. 13(a)) with a subtle frequency shift toward lower frequency as the density increases, as shown in the normalized spectra in the inset of Fig. 13(b). From panel (a), although insignificant, we note that the density-dependent frequency shift is attributed to the decreasing speed of sound (see peak shifts in the upper left inset plot) as well as the broadening of the waveform (see upper right inset plot) as the density increases. On the other hand, Fig. 13(d) and (f) exhibit noticeable spectral shifts towards higher frequency as *E* or  $\nu$  increases. The causes of these frequency shifts are identified from the corresponding waveform plots in (c) and (e). These frequency shifts originate not only from the concurrent increase of the speed of sound (see peak shifts in the upper left inset plot) but also from additional high frequency fluctuations (as indicated with arrows in the upper right inset plot) with increasing *E* or  $\nu$ . These high frequency pressure fluctuations may be attributed from delayed generation of a transverse or shear wave followed by an initial longitudinal wave [38].

The current report is purely a theoretical study of a LIPAG model, from which we demonstrate an important knowledge base to enable further studies. For instance, the temporal frequency domain power spectra under the changes of light source characteristics and material properties would provide rich information on how to tune the PA properties in LIPAGs in different ways. Ultimately, our approach can be used in parallel with experimental studies to model various types of LIPAGs. A rigorous parallel study is still challenging due to lack of accurate information on the exact thermal and mechanical properties of the constituent materials and on the conversion efficiency of the optical energy into thermal energy. Upon availability of that information, our numerical simulation approach would be readily instrumental for quantitative interpretation of the experimental data as well as for guiding the design of effective PA pressure generators with desired properties.

#### 4. Conclusion

We described a new numerical approach to simulate the dynamics of photoacoustic (PA) pressure generation in a realistic laser-induced photoacoustic generator (LIPAG) model. Our technique is in essence an integration of thermal, elastic, and acoustic multiphysics simulations implemented with a fully-explicit staggered-grid finite-difference numerical method and perfectly matched layers at the boundaries of the LIPAG geometry. The simulated LIPAG consists of a PDMS-based light absorbing layer supported by a pure (non-absorbing) PDMS backing layer, and a surrounding water medium. This unique approach enables fast numerical simulations, avoiding computational artifacts which are common in any simulation approach due to a finite domain size. The simulations of the dynamic processes involving light absorption and its conversion to thermal energy reveal details of the dynamics of laser-induced local temperature field and of thermoelastic wave generation. The integration of integrated thermal-elastic-acoustic multiphysics further allows simulations on the conversion of an elastic wave through the solid-water interface to a propagating pressure wave in water beyond. These results elucidate the underlying mechanisms of the PA pressure generation in LIPAGs. In addition, simulation of the PA pressure by adjusting a wide range of parameters of optical, thermal, and mechanical properties of the laser light and materials also reports on how the changes affect the PA pressure characteristics. Our numerical simulation approach for LIPAGs with a wide range of adjustable parameters may be instrumental to guide the design of PA pressure generators with desired properties. Furthermore, our numerical approach would be a complementary tool to the analytical approach towards a full understanding of the underlying PA pressure generation mechanism in LIPAGs.

#### Acknowledgments

This research was partially supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2020R1I1A3070105). Authors thank Aaron Goldfain, Ward Johnson and Kimberly Briggman for close reading of this manuscript. Certain commercial equipment, instruments, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identification is not intended to imply recommendation or endorsement by the National Institute of Standards and Technology, nor is it intended to imply that the materials or equipment identified are necessarily the best available for the purpose.

#### References

- [1] J.P. McGahan, B.B. Goldberg, Diagnostic Ultrasound: A Logical Approach, Lippincott-Raven Publishers, New York, 1998.
- [2] LV. Wang, Tutorial on photoacoustic microscopy and computed tomography, IEEE J. Sel. Top. Quant. 14 (2008) 171–179, doi:10.1109/jstqe.2008.918134.
   [3] LV. Wang, Multiscale photoacoustic microscopy and computed tomography, Nat. Photonics 3 (2009) 503–509, doi:10.1038/nphoton.2009.157.
- [4] V.G. Andreev, A.A. Karabutov, A.A. Oraevsky, Detection of ultrawide-band ultrasound pulses in optoacoustic tomography, IEEE T. Ultrason. Ferr. 50 (2003) 1383–1390, doi:10.1109/tuffc.2003.1244756.
- [5] S.J. Davies, C. Edwards, G.S. Taylor, S.B. Palmer, Laser-generated ultrasound Its properties, mechanisms and multifarious applications, J. Phys. D Appl. Phys. 26 (1993) 329–348, doi:10.1088/0022-3727/26/3/001.
- [6] S. Kang, Y. Yoon, J. Kim, W. Kim, Thermoelastic response of thin metal films and their adjacent materials, Appl. Phys. Lett. 102 (2013) 021908, doi:10. 1063/1.4776701.
- [7] S.W. Zhou, P. Reynolds, R. Krause, T. Buma, M. O'Donnell, J.LA. Hossack, Finite-element analysis of material and parameter effects in laser-based thermoelastic ultrasound generation, IEEE T. Ultrason Ferr. 51 (2004) 1178–1186, doi:10.1109/tuffc.2004.1334851.
- [8] F. Gao, R. Kishor, X.H. Feng, S.Y. Liu, R. Ding, R.C. Zhang, Y.J. Zheng, An analytical study of photoacoustic and thermoacoustic generation efficiency towards contrast agent and film design optimization, Photoacoustics 7 (2017) 1–11, doi:10.1016/j.pacs.2017.05.001.
- [9] S. Rajagopal, T. Sainsbury, B.E. Treeby, B.T. Cox, Laser generated ultrasound sources using carbon-polymer nanocomposites for high frequency metrology, J. Acoust. Soc. Am. 144 (2018) 584–597, doi:10.1121/1.5048413.
- [10] A. Rosencwaig, Photoacoustic spectroscopy of biological materials, Science 181 (1973) 657–658, doi:10.1126/science.181.4100.657.

- [11] C.K.N. Patel, A.C. Tam, Pulsed optoacoustic spectroscopy of condensed matter, Rev. Mod. Phys. 53 (1981) 517–550, doi:10.1103/revmodphys.53.517.
- [12] G.A. West, J.J. Barrett, D.R. Siebert, K.V. Reddy, Photoacoustic spectroscopy, Rev. Sci. Instrum. 54 (1983) 797-817, doi:10.1063/1.1137483.
- [13] S.A. Akhmanov, V.E. Gusev, A.A. Karabutov, Pulsed laser optoacoustics: achievements and perspective, Infrared Phys. 29 (1989) 815-838, doi:10.1016/ 0020-0891(89)90130-9
- [14] A. Rosencwaig, A. Gersho, Theory of the photoacoustic effect with solids, J. Appl. Phys. 47 (1976) 64-69, doi:10.1063/1.322296.
- [15] S. Manohar, D. Razansky, Photoacoustics: a historical review, Adv. Opt. Photonics 8 (2016) 586-617, doi:10.1364/aop.8.000586.
- [16] A.A. Karabutov, N.B. Podymova, V.S. Letokhov, Time-resolved optoacoustic measurement of absorption of light by inhomogeneous media, Appl. Opt. 34 (1995) 1484–1487. doi:10.1364/ao.34.001484.
- [17] D.K. Havey, P.A. Bueno, K.A. Gillis, J.T. Hodges, G.W. Mulholland, R.D. van Zee, M.R. Zachariah, Photoacoustic spectrometer with a calculable cell constant for measurements of gases and aerosols, Anal. Chem. 82 (2010) 7935-7942, doi:10.1021/ac101366e.
- T.A. Filimonova, D.S. Volkov, M.A. Proskurnin, I.M. Pelivanov, Optoacoustic spectroscopy for real-time monitoring of strongly light-absorbing solutions [18] in applications to analytical chemistry, Photoacoustics 1 (2013) 54-61, doi:10.1016/j.pacs.2013.08.002
- [19] J. Kang, J.H. Chang, S.M. Kim, H.J. Lee, H. Kim, B.C. Wilson, T.K. Song, Real-time sentinel lymph node biopsy guidance using combined ultrasound, photoacoustic, fluorescence imaging: in vivo proof-of-principle and validation with nodal obstruction, Sci. Rep.-UK 7 (2017) 45008, doi:10.1038/srep45008. [20] C.W. Wei, T.M. Nguyen, J. Xia, B. Arnal, E.Y. Wong, I.M. Pelivanov, M. O'Donnell, Real-time integrated photoacoustic and ultrasound (PAUS) imaging
- system to guide interventional procedures: ex vivo study, IEEE T. Ultrason. Ferr. 62 (2015) 319-328, doi:10.1109/TUFFC.2014.006728. V. Kozhushko, T. Khokhlova, A. Zharinov, I. Pelivanov, V. Solomatin, A. Karabutov, Focused array transducer for two-dimensional optoacoustic tomog-
- raphy, J. Acoust. Soc. Am. 116 (2004) 1498-1506, doi:10.1121/1.1781710. Y. Tian, N. Wu, K. Sun, X.T. Zou, X.W. Wang, Numerical simulation of fiber-optic photoacoustic generator using nanocomposite material, J. Comput.
- Acoust, 21 (2013) 1350002, doi:10.1142/s0218396x13500021.
- T.D. Khokhlova, I.M. Pelivanov, V.V. Kozhushko, A.N. Zharinov, V.S. Solomatin, A.A. Karabutov, Optoacoustic imaging of absorbing objects in a turbid [23] medium: ultimate sensitivity and application to breast cancer diagnostics, Appl. Opt. 46 (2007) 262-272, doi:10.1364/ao.46.000262.
- [24] A. Fedeli, C. Montecucco, G.L. Gragnani, Open-source software for electromagnetic scattering simulation: the case of antenna design, Electronics (Basel) 8 (2019) 1506, doi:10.3390/electronics8121506.
- [25] C. Warren, A. Giannopoulos, A. Gray, I. Giannakis, A. Patterson, L. Wetter, A. Hamrah, A CUDA-based GPU engine for gprMax: open source FDTD electromagnetic simulation software, Comput. Phys. Commun. 237 (2019) 208-218, doi:10.1016/j.cpc.2018.11.007. https://en.wikipedia.org/wiki/Comparison\_of\_EM\_simulation\_software. [26]
- [27]
- Q.H. Liu, Perfectly matched layers for elastic waves in cylindrical and spherical coordinates, J. Acoust. Soc. Am. 105 (1999) 2075-2084, doi:10.1121/1. 426812
- [28] Y.Q. Zeng, Q.H. Liu, A staggered-grid finite-difference method with perfectly matched layers for poroelastic wave equations, J. Acoust. Soc. Am. 109 (2001) 2571-2580, doi:10.1121/1.1369783.
- R. Ciegis, Numerical solution of hyperbolic heat conduction equation, Math. Model. Anal. 14 (2009) 11-24, doi:10.3846/1392-6292.2009.14. [29]
- [30] J.A.L. Molina, M.J. Rivera, E. Berjano, Fourier, hyperbolic and relativistic heat transfer equations: a comparative analytical study, P. Roy. Soc. A Math. Phys. 470 (2014) 20140547, doi:10.1098/rspa.2014.0547.
- E. Biagi, F. Margheri, D. Menichelli, Efficient laser-ultrasound generation by using heavily absorbing films as targets, IEEE T. Ultrason. Ferr. 48 (2001) [31] 1669-1680, doi:10.1109/58.971720.
- Q.H. Liu, J.P. Tao, The perfectly matched layer for acoustic waves in absorptive media, J. Acoust. Soc. Am. 102 (1997) 2072–2082, doi:10.1121/1.419657.

J. Virieux, P-SV wave propagation in heterogeneous media: velocity-stress finite-difference method, Geophysics 51 (1986) 889-901, doi:10.1190/1. [33] 1442147

- [34] P. Du, I.K. Lin, H.B. Lu, X. Zhang, Extension of the beam theory for polymer bio-transducers with low aspect ratios and viscoelastic characteristics, J. Micromech. Microeng. 20 (2010) 095016, doi:10.1088/0960-1317/20/9/095016.
- I.D. Johnston, D.K. McCluskey, C.K.L. Tan, M.C. Tracey, Mechanical characterization of bulk Sylgard 184 for microfluidics and microengineering, J. Mi-[35] cromech. Microeng. 24 (2014) 035017, doi:10.1088/0960-1317/24/3/035017.
- I.G. Calasso, W. Craig, G.J. Diebold, The photoacoustic effect generated by heat diffusion, Anal. Sci. 17 (2001) s249-s250, doi:10.14891/analyscisp.17icpp. [36] 0 s249 0
- [37] I.G. Calasso, W. Craig, G.J. Diebold, Photoacoustic point source, Phys. Rev. Lett. 86 (2001) 3550-3553, doi:10.1103/physrevlett.86.3550.
- [38] J. Ketolainen, M. Oksanen, J. Rantala, J. Storpellinen, M. Luukkala, P. Paronen, Photoacoustic evaluation of elasticity and integrity of pharmaceutical tablets, Int. J. Pharm. 125 (1995) 45-53, doi:10.1016/0378-5173(95)00110-5.
- [39] V.E. Gusev, A.A. Karabutov, Laser Optoacoustics, AIP Press, New York, 1993.
- A. Goyal, A. Kumar, P.K. Patra, S. Mahendra, S. Tabatabaei, P.J.J. Alvarez, G. John, P.M. Ajayan, In situ synthesis of metal nanoparticle embedded free [40] standing multifunctional PDMS films, Macromol. Rapid Comm. 30 (2009) 1116-1122, doi:10.1002/marc.200900174.
- J. Hong, H.S. Choi, K.S. Lee, S.E. Shim, Thermal properties of poly(dimethyl siloxane) nanocomposite filled with silicon carbide and multiwall carbon [41] nanotubes, Polym. Int. 61 (2012) 639-645, doi:10.1002/pi.3224
- J. Hong, J. Lee, C.K. Hong, S.E. Shim, Effect of dispersion state of carbon nanotube on the thermal conductivity of poly(dimethyl siloxane) composites, Curr. Appl. Phys. 10 (2010) 359-363, doi:10.1016/j.cap.2009.06.028.
- [43] P.F. Hai, Y. Zhou, L. Gong, L.H.V. Wang, Quantitative photoacoustic elastography in humans, J. Biomed. Opt. 21 (2016) 066011, doi:10.1117/1.jbo.21.6. 066011.
- SHIMADZUMeasurement of Poisson's Ratio and Elongation For Polyvinyl Chloride, SHIMADZU, 2019 https://www.shimadzu.com/an/industry/ [44] petrochemicalchemical/i205.html.
- K. Urayama, T. Takigawa, T. Masuda, Poisson's ratio of poly(vinyl alcohol) gels, Macromolecules 26 (1993) 3092-3096, doi:10.1021/ma00064a016.
- S.E. Bohndiek, S. Bodapati, D. Van De Sompel, S.R. Kothapalli, S.S. Gambhir, Development and application of stable phantoms for the evaluation of [46] photoacoustic imaging instruments, PLoS ONE 8 (2013) e75533, doi:10.1371/journal.pone.0075533
- M. Fonseca, B. Zeqiri, P.C. Beard, B.T. Cox, Characterisation of a phantom for multiwavelength quantitative photoacoustic imaging, Phys. Med. Biol. 61 (2016) 4950-4973, doi:10.1088/0031-9155/61/13/4950.
- W.C. Vogt, C.X. Jia, K.A. Wear, B.S. Garra, T.J. Pfefer, Biologically relevant photoacoustic imaging phantoms with tunable optical and acoustic properties, J. Biomed. Opt. 21 (2016) 101405, doi:10.1117/1.jbo.21.10.101405.
- A. Kharine, S. Manohar, R. Seeton, R.G.M. Kolkman, R.A. Bolt, W. Steenbergen, F.F.M. de Mul, Poly(vinyl alcohol) gels for use as tissue phantoms in [49] photoacoustic mammography, Phys. Med. Biol. 48 (2003) 357-370, doi:10.1088/0031-9155/48/3/306.