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Tutorial

Optothermal generation, steady-state trapping, and 3D manipulation of bubbles: an experimental and theoretical analysis of the Marangoni effect

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Abstract

Since Nobel Laureate Arthur Ashkin first introduced the trapping and manipulation of microparticles using light, numerous studies have explored this technique not only for dielectric/metallic particles but also for organic matter. This advancement has significantly expanded the landscape of non-contact and non-invasive micromanipulation at the nanometric scale. However, micromanipulation of particles with a refractive index smaller than the host medium, $n_p < n_m$, proves challenging with Gaussian beams. To overcome this obstacle, a force known as thermocapillary, or the Marangoni force, has emerged as a straightforward trapping mechanism for bubbles in liquids. The Marangoni force results from the surface tension of bubbles, induced either thermally or chemically—by creating a temperature gradient or adding surfactants, respectively. The surface tension gradient on the liquid host induces tangential stress on the bubble wall, causing the bubble to move toward the region of lower surface tension, where it faces less opposing force. When the Marangoni force is generated by a laser beam's temperature gradient, it becomes an exceptionally effective mechanism for the steady-state trapping and three-dimensional manipulation of bubbles, even with low optical power lasers. This force produces both longitudinal and transversal forces, resembling optical forces, creating a three-dimensional potential well capable of handling bubbles with radii of tens to hundreds of microns. This work provides guidance and demonstrates, both experimentally and theoretically, the step-by-step process of quasi-steady-state trapping and three-dimensional manipulation of bubbles through optothermal effects. The bubbles in question are tens of microns in size, significantly larger than those that optical tweezers can trap/manipulate.

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Furthermore, the study emphasizes the crucial role of the Marangoni force in this process, outlining its various advantages.

Keywords: Marangoni effect, optothermal effects, thermal gradient, micromanipulation

1. Introduction

Bubbles are ubiquitous in our daily lives whether we are bathing, washing dishes, drinking our cup of coffee, or in boiling water. It is hard to imagine that such a seemingly mundane and minuscule object as a bubble could involve a multitude of intricate processes governing its formation and dynamics. Regardless of whether they are composed of air or vapor, bubbles play a crucial role in various fields, including medical diagnostics [1–4], drug delivery [5–7], lithography [8], microfluidics [9–11], hydraulic systems [12–14], and sorting applications [15–17]. However, achieving success in these applications necessitates precise spatial control over the bubbles. While the terms ‘trapping and manipulation’ may appear commonplace, as we often manipulate objects with our hands or tools, our ability to trap and manipulate diminishes as we delve into the micro and nano world. This microscopic realm has provided us with valuable insights into our environment and has paved the way for new avenues of understanding life [18–20]. Discussing trapping and manipulation inevitably brings to mind the Nobel Laureate Arthur Ashkin [21, 22]. Indeed, our current comprehension of life would be incomplete without acknowledging his revolutionary invention: the optical tweezers. This tool provides a non-contact and non-invasive mechanism [23] not only for trapping and manipulating dielectric [24] and metallic particles [25] but also for biological matter [20, 26].

While optical tweezers have significantly broadened the scope of contactless trapping and manipulation techniques, the challenge persists in trapping and sorting particles with a refractive index n_p smaller than that of the surrounding medium n_m , i.e., $n_p/n_m < 1$, as seen in the case of microbubbles. This difficulty arises from the repulsive force generated by typical Gaussian beams. In response to this, various trapping mechanisms based on non-Gaussian profile beams have been proposed, including Bessel beams Sokolovskii [27], Laguerre–Gaussian beams [28, 29], also referred to as bottle beams, and fast-scanning Gaussian-based optical tweezers [30, 31], among others. Nevertheless, trapping and manipulating bubbles with radii exceeding 20 μm [31–33] continue to present a formidable challenge.

Beyond optical methods, various authors have suggested alternative techniques for bubble manipulation grounded in thermal and acoustic [34–36] phenomena. Nevertheless, these approaches frequently demand complex, bulky, and costly setups. Furthermore, the forces generated typically fall within the nano-Newton range.

The thermocapillary effect, also known as the Marangoni force, has proven to be a valuable and effective mechanism for trapping and manipulating bubbles, including those in the hundreds of micrometers in size [37–40]. The initial exploration

of the thermocapillary effect dates back to James Thompson’s experimental study in 1855 [41]. In that study, he observed the displacement of a spherical microbubble immersed in a solution, attributing the phenomenon to the difference in surface tension between the liquid and the bubble. A decade later, in 1865, the Italian physicist Carlo Marangoni, in his doctoral thesis, established that the surface tension between two fluids could induce a force for the displacement of the fluids, giving rise to the term ‘the Marangoni effect’ [42].

Moving forward, in 1911, Hadamard [43] and Rybczynski [44] derived an axial symmetry solution for the flow around a spherical bubble, known as the Hadamard–Rybczynski equation. This solution is based on the Navier–Stokes equation coupled with the buoyancy effect and drag force [37, 38]. In 1939, Hershey [45] determined the temperature dependence of the surface tension of a pure liquid, indicating that, in most cases, the surface tension decreases with increasing temperature. In 1948, a mathematical description of the thermocapillary effect was introduced by Fedosov in his doctoral thesis [46]. However, it was not until 1959 that the first experimental evidence of microbubble displacement within a measured temperature gradient was demonstrated by Young *et al* [38]. Over the years, numerous experimental and theoretical works have been conducted to demonstrate the thermocapillary capabilities for 2D trapping and manipulation of bubbles, including studies by Thompson *et al* [47] in 1980, Wozniak *et al* [48] in 1988, Merritt and Subramanian [49] in 1989, Takahashi *et al* [50] in 1999, among others.

Recently, the Marangoni effect, driven by the optothermal effect [51, 52], where laser radiation is absorbed by a thin film, colloidal suspensions, or bulk light absorption in liquids, has emerged as an affordable, straightforward, and effective non-contact technique for trapping and manipulating bubbles [53–58]. The optothermal effect offers several noteworthy advantages at the microscale. It provides non-contact and non-invasive approaches, operating effectively across a broad spectrum of laser beam wavelengths in various absorbing media. Additionally, thermal phenomena exert forces that are orders of magnitude greater than optical forces [59–61]. Consequently, optothermal effects combine the strengths of both domains, offering a comprehensive solution.

For instance, similar to optical forces, the Marangoni force generates a 3D potential well that allows precise spatial control over significantly large bubbles with radii exceeding 100 μm . Specifically, the Marangoni force generates both longitudinal and transverse forces, enhancing the capability for spatial manipulation and control [57, 62–64].

Here, we present a theoretical and experimental study on the generation and 3D manipulation of bubbles with radii in the tens of microns range, employing the Marangoni force driven by optothermal effects using continuous wave (CW)

and nanosecond pulsed lasers. In section 2, we elucidate the working principles of hydrodynamic forces and the Marangoni effect involved in bubble manipulation. Subsequently, in section 3, we provide a brief overview of the theory behind bubble nucleation induced by light absorption from both continuous and pulsed lasers. Moving on to section 4, we offer detailed information about the numerical calculation of optothermal effects, accomplished by solving the heat and Navier–Stokes equations in the finite element software (COMSOL Multiphysics). Section 5 outlines the experimental setup and presents results regarding the generation, steady-state trapping, and 3D manipulation of bubbles by optothermal effects. In section 6, our discussion expands to cover quasi-steady-state trapping and three-dimensional manipulation of bubbles, incorporating analytical expressions for temperature profiles and presenting analytical expressions for the Marangoni force regardless of the laser source employed. Finally, in section 7 the conclusions of this work are presented. Our work provides both compressible and valuable insight into the field of optothermal manipulation, confirming quasi-stationary trapping and 3D manipulation of bubbles up to hundreds of microns in size through the Marangoni effect activated with low power levels. Moreover, our novel insights into optothermal trapping hold promise for a wide range of applications, including medical, microfluidics, and lithography, among others.

2. Hydrodynamics forces and the Marangoni effect

2.1. Hydrodynamics forces

To start with, it is crucial to note that the dimensions of the bubbles are assumed to be significantly smaller than the overall dimensions of the incompressible and Newtonian liquid host (<1 mm). The flow around a bubble (or spherical particle) is characterized by low Reynolds numbers, and u is the magnitude of the relative velocity of the bubble ($u = \mathbf{u}_{x,y} - \mathbf{V}_{x,y}$). With these considerations, solving for the fluid velocity $\mathbf{V}$ and pressure p of the liquid host is typically sufficient to address the bubble motion equation. This can be accomplished by utilizing the Navier–Stokes equations to estimate all the forces acting on the bubble and equating the total fluid force [65], $\mathbf{F} = m_B d\mathbf{u}/dt$, where m_B represents the mass of the bubble (assumed constant) and $\mathbf{F}$ represents fluid forces that may include buoyancy, inertia, and drag.

The fluid force is governed by the Navier–Stokes equation given by [65]:

$$\frac{\partial \mathbf{V}}{\partial t} + (\mathbf{V} \cdot \nabla) \mathbf{V} = -\frac{\nabla p}{\rho} + \nu \nabla^2 \mathbf{V}, \quad (1)$$

where $\mathbf{V}$ is the fluid velocity vector, ρ , and ν are the density and kinematic viscosity of the liquid host, respectively, and the continuity equation given is by:

$$\rho \nabla \cdot \mathbf{V} = 0. \quad (2)$$

To simplify the equations (from 3D to 2D) and account for the circular shape of the bubble, polar coordinates (r, θ) are

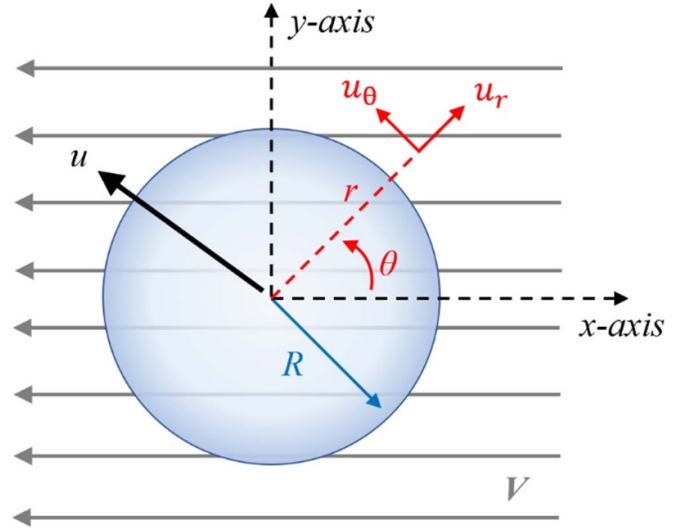


Figure 1. Schematic representation of a bubble with radius R moving in a liquid host. $\mathbf{V}$ and $\mathbf{u}$ are the fluid and bubble velocity vectors, respectively.

employed with the center of the bubble fixed as the origin system. The velocity of the bubble $\mathbf{u}$ with radius R can be defined as $\mathbf{u}_r$ and $\mathbf{u}_\theta$ as depicted in figure 1.

Therefore, the equations (1) and (2) turn into:

$$\begin{aligned} \rho \left[\frac{\partial u_r}{\partial t} + u_r \frac{\partial u_r}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta^2}{r} \right] \\ = -\frac{\partial p}{\partial r} + \rho \nu \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_r}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_r}{\partial \theta} \right) - \frac{2u_\theta}{r^2} - \frac{2}{r^2} \frac{\partial u_\theta}{\partial \theta} \right], \end{aligned} \quad (3)$$

$$\begin{aligned} \rho \left[\frac{\partial u_\theta}{\partial t} + u_r \frac{\partial u_\theta}{\partial r} + \frac{u_\theta}{r} \frac{\partial u_\theta}{\partial \theta} - \frac{u_r u_\theta}{r} \right] \\ = -\frac{1}{r} \frac{\partial p}{\partial \theta} + \rho \nu \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_\theta}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_\theta}{\partial \theta} \right) + \frac{2}{r^2} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta}{r^2 \sin^2 \theta} \right], \end{aligned} \quad (4)$$

$$\frac{1}{r^2} \frac{\partial}{\partial r} (r^2 u_r) + \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (u_\theta \sin \theta), \quad (5)$$

this equation is a form of the continuity equation in spherical coordinates, specifically written for a compressible fluid.

In the regime of low Reynolds number $Re < 1$ ($Re = 2VR/\nu$) where the viscous force dominates over the inertia force on the fluid velocity, the density can be neglected and only the viscous term is maintained. So, equations (3) and (4) become:

$$0 = -\frac{\partial p}{\partial r} + \rho \nu \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_r}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_r}{\partial \theta} \right) - \frac{2u_\theta}{r^2} - \frac{2}{r^2} \frac{\partial u_\theta}{\partial \theta} \right], \quad (6)$$

$$0 = -\frac{1}{r} \frac{\partial p}{\partial \theta} + \rho \nu \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u_\theta}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial u_\theta}{\partial \theta} \right) + \frac{2}{r^2} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta}{r^2 \sin^2 \theta} \right]. \quad (7)$$

The Stokes stream function, ψ , is defined to satisfy the continuity equation automatically [65]:

$$u_r = \frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta}, \quad (8)$$

$$u_\theta = -\frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r}, \quad (9)$$

and the solution for the inviscid potential flow is:

$$\psi = \sin^2 \theta \left(-\frac{ur^2}{2} + \frac{A}{r} + Br \right), \quad (10)$$

$$u_r = \cos \theta \left(-u + \frac{2A}{r^3} + \frac{2B}{r} \right), \quad (11)$$

$$u_\theta = -\sin \theta \left(-u - \frac{A}{r^3} + \frac{B}{r} \right), \quad (12)$$

where the constants A and B are determined from the boundary conditions on the surface of the bubble. Thus, the net force exerted by the isotropic liquid host over the bubble does not depend on the θ -direction and is given by [65]:

$$\mathbf{F} = \frac{4}{3} \pi R^2 \rho \nu \left(-\frac{4u}{R} + \frac{8A}{R^4} + \frac{2B}{R^2} \right) \vec{r}. \quad (13)$$

Surface tension σ is the tension of the surface film s of a liquid that results from the attraction of the particles in the surface by the liquid. This attraction tends to minimize the surface area [65]. On the other hand, the Marangoni force originates from the dependence on surface tension gradients along the surface, denoted as $\frac{\partial \sigma}{\partial s}$. These gradients in surface tension can be induced through various methods, including surfactant concentration, electric potential, and temperature gradients. Notably, raising the temperature of the liquid leads to a reduction in surface tension force, following Eötvös' law [66]. The adjustment of the surface tension gradient through temperature manipulation is referred to as the thermocapillary force or Marangoni force. Surface tension gradients affect the flow of free surfaces because the tangential surface gradient requires a shear stress in the negative direction for surface equilibrium. This shear stress modifies the boundary conditions and alters the flow dynamics and forces acting on the bubble [37, 65].

A spherical bubble immersed in an incompressible and Newtonian liquid host under the presence of a temperature gradient $\frac{\partial T}{\partial x}$ in the direction of the bubble motion and considering that thermal conduction governs the heat transfer ($\nabla^2 T = 0$) is assumed, i.e. the heat transfer takes place through the bubble and not along the bubble surface. From the solution to Laplace's equation for the conductive heat transfer problem [65], it follows that:

$$\left(\frac{1}{R} \frac{ds}{d\theta} \right)_{r=R} = -\frac{3}{2} \sin \theta \left(\frac{d\sigma}{dT} \right) \left(\frac{dT}{dx} \right). \quad (14)$$

Since the bubble shape is considered to remain spherical, the surface tension differences are small compared with the absolute value of the surface tension σ , so the stress normal to the bubble surface s is mainly governed by the surface tension.

Therefore, the boundary condition of the tangential stress for the bubble is given by:

$$\rho \nu \left(\frac{\partial u_\theta}{\partial r} - \frac{u_\theta}{r} \right)_{r=R} + \frac{1}{R} \left(\frac{d\sigma}{d\theta} \right) = 0. \quad (15)$$

By applying equation (14) and the tangential stress presented in equation (15), and the kinematic condition $(u_r)_{r=R} = 0$ to the solution for the inviscid potential flow given by equations (10)–(12), now the constants A and B can be rewritten as follows:

$$A = -\frac{R^4}{4\rho\nu} \left(\frac{d\sigma}{dT} \right) \left(\frac{dT}{dx} \right), \quad (16)$$

$$B = \frac{uR}{2} + \frac{R^2}{4\rho\nu} \left(\frac{d\sigma}{dT} \right) \left(\frac{dT}{dx} \right). \quad (17)$$

Substitution of equations (16) and (17) into equation (13), the net force exerted on a bubble under the influence of temperature gradients turns into:

$$\mathbf{F} = -4\pi R \rho \nu u - 2\pi R^2 \left(\frac{d}{dT} \right) \left(\frac{dT}{dx} \right) \vec{r}, \quad (18)$$

where the drag force is accounted for by the first term (following the Hadamard–Rybczynski solution where the condition of zero shear stress on the sphere surface is considered [38, 43, 44]) while the second term illustrates the surface tension gradient induced by a temperature gradient, called the Marangoni force [65]. Thus, $\mathbf{F}_M$ is given by:

$$\mathbf{F}_M = -2\pi R^2 \nabla T \frac{d}{dT} \vec{r}, \quad (19)$$

the negative sign in the first term on the right-hand side of equation (18) indicates that drag force opposes the liquid flow direction while in the second term, the force is in the direction of the heat source (hotter liquid) while the liquid moves towards colder regions [37, 65], as depicted in figure 2.

In addition to the Stokes force, a bubble immersed in a liquid will experience an upward force from the surrounding liquid host, following Archimedes' principle. This force, known as the buoyancy force $\mathbf{F}_b$ [37, 57], always acts in the upward direction and is given by:

$$\mathbf{F}_b = \frac{4}{3} \pi (\rho_l - \rho_v) \mathbf{g} R^3, \quad (20)$$

where ρ_l and ρ_v represent the density of the liquid host and the vapor, respectively, $\mathbf{g}$ is the gravitational acceleration. Besides, the gravity force $\mathbf{F}_g$ given by [37, 57]:

$$\mathbf{F}_g = \frac{4}{3} \pi \rho_v \mathbf{g} R^3. \quad (21)$$

Typically, the density of the vapor/gas bubble is several orders of magnitude less than the density of the liquid host. Furthermore, if the bubble or particle collides with another one, the inertia force $\mathbf{F}_i$ will come into play. Inertia force, which is also known as added mass, is governed by Newton's

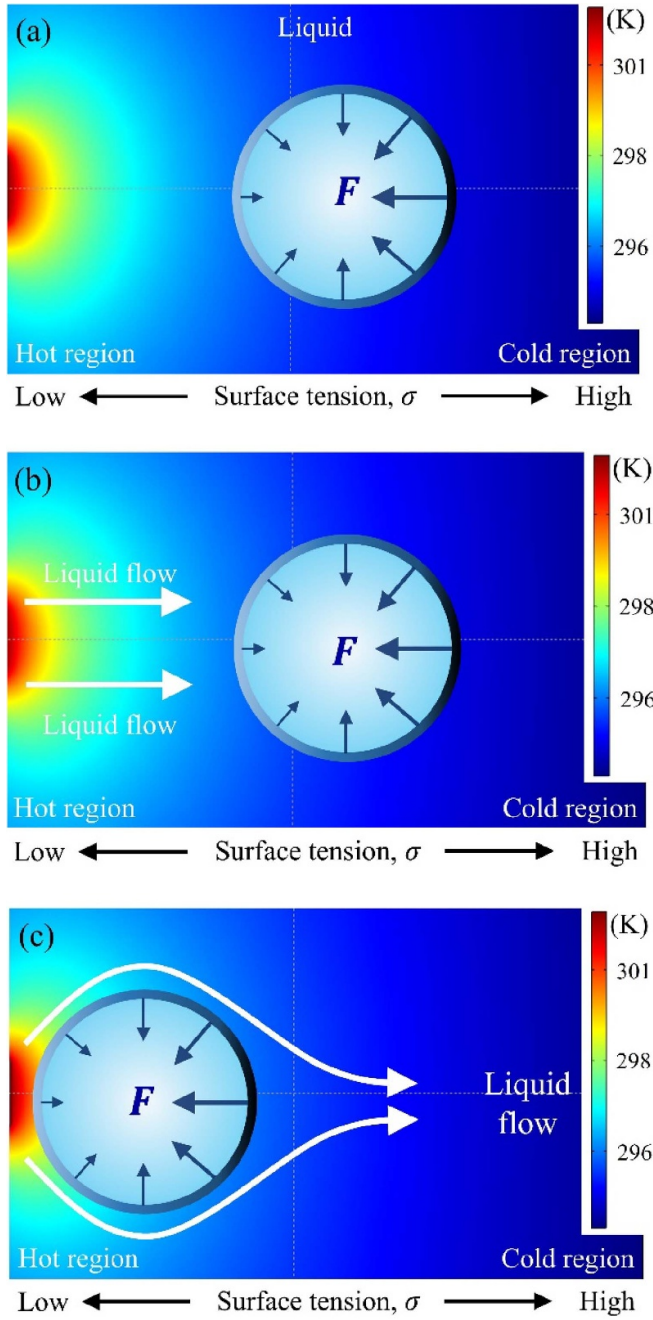


Figure 2. The Marangoni force acts on a bubble. (a) A bubble is immersed in an infinite liquid and exposed to a temperature gradient created by a heat source. (b) The bubble surrounded by the temperature gradient experiences tangential stress on its wall. (c) The bubble moves towards the hot spot while the liquid moves towards the cold regions.

second law. This law states that the acceleration of an object/particle is dependent on the net force acting upon it and its mass. Bubbles immersed in liquids have a specific added mass that characterizes the inertia force of the surrounding fluid when the bubble accelerates. F_i is described by [67, 68]:

$$F_i = \frac{4}{3} \pi R^3 \rho_l C_b \frac{du}{dt}, \quad (22)$$

where $C_b = \frac{1}{2}$ is the bubble-shape coefficient.

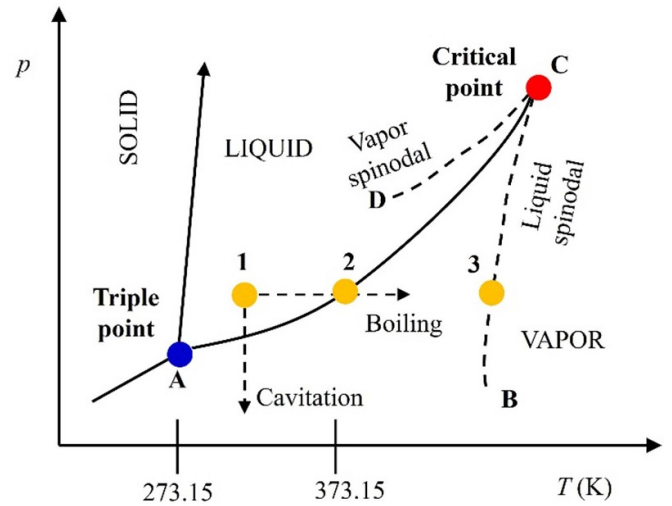


Figure 3. Phase diagram of water (pressure p versus Temperature T). The yellow dot represents the ambient pressure at room temperature, whereas the arrows indicate the paths to induce either boiling or cavitation. Reproduced with permission from [65]. Credit: Brennen, Christopher Earls. *Cavitation and Bubble Dynamics*. Oxford University Press, 1995. CC-BY-NC 4.0.

3. Phase changes and nucleation

This section discusses the thermodynamic phase changes responsible for bubble generation and the temperatures at which they occur under CW and pulsed radiation. It also highlights the physical mechanism of bubble formation and the distinctions between the two types of radiation. Lastly, the dynamics of a spherical bubble collapse near a solid wall are briefly explored.

Figure 3 shows the phase diagram of water pressure p versus temperature T . The triple point, indicated by the blue dot and marked by the letter A, represents the point at which the solid, liquid, and vapor phases coexist ($T = 273.15$ K and $p = 611.65$ Pa). At this point one can obtain ice, liquid, and vapor of water by making small and random changes in pressure and temperature. The line AC, called binodal, represents the separation limit between liquid and vapor phases, i.e. the saturated liquid/vapor line. The line CD connecting the highest points on the theoretical isotherms is known as the vapor spinodal line. It indicates the maximum subcooling of metastable vapor. The line BC connecting the lowest points is called the liquid spinodal line, indicating the maximum superheating of metastable liquids. Both spinodal lines terminate at the critical point C, beyond which there is no thermodynamic difference between the liquid and vapor phases. Point 1 represents ambient conditions ($T_1 = 298.15$ K and $p_1 = 101$ kPa), point 2 denotes the boiling temperature, i.e. saturated liquid at atmospheric pressure ($T_2 = 373.15$ K and $p_2 = 101$ kPa), and point 3 is the location the spinodal at ambient pressure ($T_3 = 578.15$ K and $p_3 = 101$ kPa) [65, 69, 70].

Two processes in the phase diagram (figure 3) are indicated, starting from room temperature and ambient pressure conditions: boiling and cavitation (represented by horizontal and vertical arrows, respectively). Both processes entail a phase

change from liquid to vapor, but they differ in their thermodynamic paths leading to the vapor phase. A straightforward way to distinguish between cavitation and boiling is to define cavitation as the process of nucleation in a liquid when the pressure drops below the vapor pressure while boiling is the nucleation process occurring when the temperature rises above the saturated vapor/liquid temperature. Cavitation is typically a faster process than boiling.

3.1. Nucleation theory

Nucleation, as defined by Oxtoby [71], represents a first-order phase transition in liquids. During this transition, the molecules within the liquid attain sufficient kinetic energy to overcome intermolecular forces that typically bind them together, allowing the formation of small voids. This marks the initial stage in the spontaneous creation of a new thermodynamic phase. Nucleation can be triggered through two mechanisms: homogeneous and heterogeneous [71]. Homogeneous nucleation arises from thermal fluctuations within a pure liquid, temporarily generating microscopic voids that act as nucleation sites [69, 72]. In contrast, heterogeneous nucleation relies on existing weak points or inhomogeneities to initiate bubble formation such as micro-bubbles trapped on solid surfaces or impurities or roughness/defects on submerged solid surfaces. These sites serve as the medium for the rupture and growth into macroscopic bubbles. Therefore, heterogeneous nucleation is more prevalent than homogeneous nucleation due to the difficulty in avoiding impurities in the liquid or voids in the solid wall [69, 72]. However, it is possible to superheat a liquid above its boiling point under specific conditions, such as using a perfectly smooth container and avoiding any stirring. In instances where the superheated liquid undergoes an explosive phase transition, a fast-expanding bubble can be produced. This phenomenon generally occurs near the spinodal temperature of the liquid, approximately 578.15 K for water [37, 69, 72].

In a pure liquid, intermolecular forces are macroscopically represented by the surface tension σ (see figure 4). The interior pressure of the bubble p_{bubble} can be related to the pressure of the surrounding liquid p_{∞} by the surface tension. The pressure p_{∞} exerted over the bubble of radius R , will be related to the interior pressure in the bubble p_{bubble} by:

$$p_{\text{bubble}} - p_{\infty} = \frac{2\sigma}{R}. \quad (23)$$

If the temperature T is uniform and the bubble contains only vapor, the bubble interior pressure will be the saturated vapor pressure $p_{\text{sat}}(T)$. Nevertheless, to produce equilibrium conditions, the exterior liquid pressure $p_{\infty} = p_{\text{sat}} - \frac{2\sigma}{R}$ will have to be less than p_{sat} . Subsequently, if the exterior liquid pressure is maintained at a constant value just slightly less than $p_{\text{sat}} - \frac{2\sigma}{R}$, the bubble will grow, thus R will increase, the excess pressure causing growth will increase, and eventually rupture will take place. It follows that if the maximum size of the vacancy

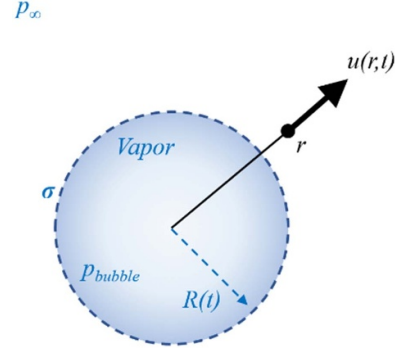


Figure 4. Spherical vapor bubble of radius $R(t)$ in an equilibrium within a liquid. p_{∞} represents the pressure of the surrounding liquid, p_{bubble} is the vapor pressure within the bubble, σ is surface tension, and $u(r,t)$ corresponds to the liquid velocity induced by the evolution of the bubble.

present is R_c (termed the critical radius or cluster radius), then the tensile strength of the liquid, Δp_c , will be given by:

$$\Delta p_c = \frac{2\sigma}{R_c}. \quad (24)$$

Now, considering a superheated liquid at a constant pressure p and temperature T above the boiling temperature, T_b or, equivalently, at a constant temperature T at a pressure below the saturation pressure $p_{\text{sat}}(T)$, the minimum work required to generate a vapor bubble of radius R_c within the liquid is given by:

$$W_c = 4\pi R_c^2 \sigma - \frac{4}{3}\pi R_c^3 \Delta p_c. \quad (25)$$

Equation (25) comprises two terms: the first term represents the energy that must be deposited and stored in the bubble surface, while the second term represents the volume of liquid displaced outward to create the bubble. The critical deposited energy required is determined by the difference between the inner and outer pressure of the bubble. The work done by the system is calculated as the product of the volume of the bubbles and the pressure difference. Therefore, equation (25) can be expressed as:

$$W_c = 4\pi R_c^2 \sigma - \frac{16\pi}{3} \frac{\sigma^3}{(\Delta p_c)^2}. \quad (26)$$

The phase change from liquid to vapor, also known as boiling, occurs when the temperature of the liquid exceeds its saturated liquid/vapor temperature [73].

3.2. Superheated liquid

A superheated liquid is a state of matter where a substance remains in a liquid state despite being at a temperature higher than its boiling point at a given pressure. This occurs due to the absorption of additional energy, typically in the form of heat, which causes the temperature of the liquid to rise well above its boiling point without undergoing a phase transition into a vapor, i.e., reaching the spinodal temperature (~ 573.15 K

for water). At this point, a slight perturbation breaks the quasi-equilibrium state, and nucleation can suddenly induce an explosive phase transition [74, 75]. Superheated liquids can be induced by different methods depending on the source of energy to produce heating such as: microwave [76], electric [77], and laser radiation [75], among others.

3.2.1. Laser heating. Shortly after the invention of the laser, the demonstration of light-induced cavitation emerged as a powerful tool, gaining increased relevance in several technological areas [78, 79]. Light-induced cavitation can be produced by either short-pulsed or CW lasers and their respective physical mechanisms differ drastically. The cavitation produced by short-pulsed lasers involves the formation of plasma, typically accompanied by a visible flash [80]. An ultrashort-pulsed laser ($100 \text{ fs} < \tau < 1 \text{ ns}$) induces what is known as optical breakdown. The mechanism that triggers liquid breakdown depends on various factors, including laser pulse duration, pulse intensity, wavelength, and polarization, among others. The use of CW lasers or longer-pulsed lasers ($\tau > 1 \mu\text{s}$) generates a process called laser-induced thermal breakdown [81].

The optical breakdown is a nonlinear absorption process that ionizes the media, usually nominally transparent liquids, leading to plasma formation. This process demands high energy densities to reach the liquid breakdown threshold I_{th} [80]. Plasma formation takes place in distilled water at irradiances $I_{\text{th}} > 10^{10} \frac{\text{W}}{\text{cm}^2}$ [80]. There are two mechanisms of optical breakdown: avalanche ionization, also known as cascade ionization, and multiphoton ionization [82]. Both ionization mechanisms must exceed the critical density of ionized electrons ρ_{cr} during the laser pulse [83]. On one hand, avalanche ionization requires a few seed electrons, also known as free electrons or lucky electrons. These seed electrons, which are already present in the focal volume before the arrival of the laser pulse, are accelerated by the laser electric field. The increased kinetic energy of the electrons is transferred to the atoms/molecules through collision and generating more free electrons. These seed electrons can be produced either by the multiphoton ionization process itself or by the heating of linearly absorbing impurities present in the liquid. Besides, the impurities heating can decrease the optical breakdown threshold. On the other hand, multiphoton ionization does not require these seed electrons, however, requires a pure liquid and demands higher energies. The multiphoton ionization rate is proportional to I^k , where k is the number of photons needed to achieve the ionization potential. The avalanche ionization mechanism rate is proportional to the incident irradiance [84]. Therefore, the interplay between avalanche ionization and multiphoton ionization has been elucidated as the cornerstone for optical breakdown.

On the other hand, laser-induced thermal breakdown is a linear absorption process produced by either CW or long-pulsed laser, also known as thermocavitation [85, 86]. This process was introduced by Rastopov and Sukhodolsky in the early 1990s [85, 87]. When laser radiation is focused on absorbing liquids, it rapidly elevates the temperature beyond the boiling point without causing boiling, resulting in a superheated liquid reaching a critical temperature known as the

spinodal limit. For water, this critical temperature is approximately 647 K. Upon reaching the spinodal limit, the liquid enters a metastable state. Consequently, the superheated liquid becomes unstable to random density fluctuations, leading to an explosive liquid-to-vapor phase transition. This results in the generation of a rapidly expanding vapor bubble with a radius ranging from micrometers to millimeters [88]. However, as reported by Padilla-Martinez *et al* [75] and Ramirez-San-Juan *et al* [88], the shockwave amplitude of thermocavitationally-induced bubbles is at least three orders of magnitude smaller than the shockwave produced by optical breakdown. The thermocavitation process converts approximately 20% of input energy into mechanical energy [75], in contrast to optical breakdown, which loses around 10%–30% of input energy [89]. Nevertheless, thermocavitation can be induced at a threshold irradiance of approximately $I_{\text{th}} \approx 10^3 \frac{\text{W}}{\text{cm}^2}$, which is at least seven orders of magnitude lower than that of optical breakdown [75]. Furthermore, the threshold irradiance can be reduced by matching or tuning the wavelength used with the liquid absorption coefficient, maximizing the absorption of input energy.

One notable characteristic of thermocavitation is that the bubble induced with the lowest optical power generates the largest radius and, consequently, the highest pressure shock waves [88]. This phenomenon arises because the thermal diffusion time τ_{dif} for absorbing liquids is typically shorter than the cavitation time τ_{cav} . This condition allows for more liquid to be available for evaporation due to the efficient transfer of heat to a larger volume, resulting in the formation of larger bubbles. Additionally, thermocavitation presents the advantage of making use of inexpensive light sources.

3.2.2. Boiling crisis. The boiling crisis is characterized by a high rate of heat transfer from the heating interface to the bulk fluid. With the increase in heating energy, the temperature of the heating interface also rises. If the heat flux from the heating source reaches a threshold value, the suddenly generated vapor bubbles will transform into vapor films. These films cover the heating surfaces, acting as an insulating layer that isolates the bulk fluid from the heating source [90, 91].

3.3. Dynamics of spherical bubbles

Following the introduction of the nucleation theory for bubble generation, the focus shifts to bubble dynamics, specifically addressing bubble growth and collapse. The discussion also delves into the distinctions between symmetric and asymmetric bubble collapse, occurring when the bubble is far from an interface/wall and when it is near or attached to a wall/solid boundary.

3.3.1. Rayleigh–Plesset equation. The symmetric spherical bubble is a simple case that proves useful for deriving the Rayleigh–Plesset equation, governing the dynamics of a spherical bubble in an infinite liquid body. This equation is derived under the simplest conditions such as the bubble is immersed in an incompressible Newtonian liquid with an

infinite volume; the density of the liquid surrounding the bubble is greater than the gas density of the bubble; the gravitational force is not taken into account, and the gas content remains constant. It is important to note that these assumptions are a first approximation only, and in reality, both the exchange of energy and the transfer of mass between the bubble and its surrounding liquid may occur. The function to be determined in the liquid domain, induced by the evolution of the bubble, are the velocity $u(r,t)$ and the pressure $p(r,t)$. Mass transfer across the interface is neglected, so the liquid velocity at the interface $u(R,t)$ is equal to the interface velocity dR/dt . The continuity (or mass conservation) equation states that in an incompressible fluid, $\nabla \cdot u = 0$. By utilizing spherical symmetry (spherical coordinates), the velocity of the bubble interface bubble can be determined as [92]:

$$u(r,t) = \frac{\partial R}{\partial t} \frac{R^2}{r^2}. \quad (27)$$

In this particular case, the viscous term in the Navier-Stokes equation is zero. Therefore, for a non-viscous, the momentum equations are given by:

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial r} = -\frac{1}{\rho} \frac{\partial p}{\partial r}. \quad (28)$$

By substituting equation (27) into equation (28):

$$\frac{\partial^2 R}{\partial t^2} \frac{R^2}{r^2} + 2 \left(\frac{\partial R}{\partial t} \right)^2 \left[\frac{R}{r^2} - \frac{R^4}{r^5} \right] = -\frac{1}{\rho} \frac{\partial p}{\partial r}, \quad (29)$$

and integrating with respect to r employing the conditions at infinity $p_\infty(t)$ on the interface $r = R$, it is obtained [69]:

$$\frac{p(r,t) - p_\infty(t)}{\rho} = R \frac{\partial^2 R}{\partial t^2} + \frac{3}{2} \left(\frac{\partial R}{\partial t} \right)^2. \quad (30)$$

The pressure on the cavity interface is given by [69]:

$$p(r,t) = p_{\text{bubble}} + p_i \left(\frac{R_0}{R} \right)^{3\Psi} - \frac{2\sigma}{2R} + 2 \frac{\mu}{R} \frac{\partial u}{\partial t} \Big|_{r=R}, \quad (31)$$

where p_i is the initial pressure, R_0 is the initial bubble radius at $t = 0$, Ψ is the ratio of heat gas capacities C_{p_g} and C_{p_v} , and μ is the kinematic viscosity of the liquid, and considering that [92]:

$$\frac{\partial u}{\partial t} \Big|_{r=R} = -\frac{2}{R} \frac{\partial R}{\partial t}, \quad (32)$$

thus equation (30) turns into:

$$\rho \left[R \frac{\partial^2 R}{\partial t^2} + \frac{3}{2} \left(\frac{\partial R}{\partial t} \right)^2 \right] = p_{\text{bubble}} + p_\infty(t) + p_i \left(\frac{R_0}{R} \right)^{3\Psi} - \frac{2\sigma}{R} - 4 \frac{\mu}{R} \frac{\partial R}{\partial t}. \quad (33)$$

Equation (33), known as the Rayleigh-Plesset equation, which determines the temporal evolution of the bubble radius R [65, 92, 93]. On the right-hand side, the pressure term is

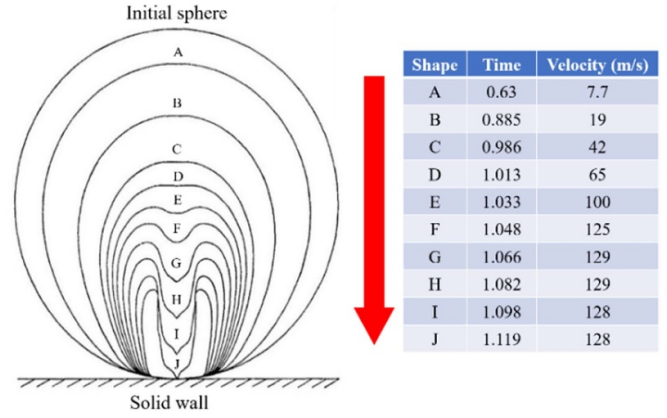


Figure 5. Non-spherical bubble collapse at $\gamma = 1$. Reproduced from [95] Credit: Chapman, Richard Bruce (1970) Nonspherical Vapor Bubble Collapse. Dissertation (Ph.D.), California Institute of Technology. doi:<https://doi.org/10.7907/TDCB-D645>. <https://resolver.caltech.edu/CaltechTHESIS:04142011-083038797>. Used with permission from the California Institute of Technology. CC BY 4.0.

the driving force, while the other three terms account for the effects of non-condensable gas, surface tension, and liquid viscosity while the term on the left-hand side denotes the interface dynamics.

In the ideal scenario, the bubble contains only vapor without any condensable gases, and the liquid is non-viscous ($\mu = 0$) and the effect of the surface tension is neglected ($\sigma = 0$), equation (33) simplifies to [65, 69, 92]:

$$\frac{\partial R}{\partial t} = -\sqrt{\frac{2}{3} \left[\frac{p_\infty - p_{\text{bubble}}}{\rho} \right] \left[\frac{R_0^3}{R^3} - 1 \right]}. \quad (34)$$

This expression determines the velocity of the bubble interface $\frac{\partial R}{\partial t}$ at the point of bubble collapse. Bubble collapse occurs when the external pressure exceeds the pressure inside the bubble, i.e. $p_\infty > p_{\text{bubble}}$. The interface velocity is negative $\frac{\partial R}{\partial t} < 0$ and the initial radius is greater than the bubble radius at any moment, i.e. $R_0 > R$. Equation (34) shows that the radius tends to 0 and the radial inwards motion accelerates without limit [92].

3.3.2. Bubble collapse near a solid wall. The collapse of a spherical bubble can be perturbed when its close occurs in the proximity of a solid or elastic wall leading to an asymmetric bubble collapse. Several researchers have delved into the dynamics of bubbles near or attached to solid surfaces or interfaces using techniques such as high-speed photography, short pulse pump-probe techniques, and acoustic methods. It is widely acknowledged that asymmetric bubble collapse gives rise to a counter jet, as elucidated by Rattray's non-spherical bubble collapse theory in his Ph.D. thesis, as shown in figure 5 [94–96].

Before collapse, the semi-spherical bubble takes on a toroidal shape due to the formation of a counter jet directed towards the wall/interface. This counter jet is capable of penetrating the bubble and striking the wall/interface. Erosion of solid substrates or the penetration into elastic walls has been

attributed to this jet [97, 98]. In cases where the walls or substrates are heated, the counter jet is immediately vaporized upon hitting the substrate, resulting in the formation of a secondary bubble [99–101]. The collapse of a cavitation bubble near a solid boundary is greatly influenced by the non-dimensional stand-off parameter γ . This parameter is defined as the ratio of the distance from the center of the bubble to the solid boundary to the maximum radius of the bubble.

The bubble lifetime τ_r close to a substrate is given by [94, 96, 102]:

$$\tau_r \approx \tau_c (1 + 0.205\gamma), \quad (35)$$

where $\gamma = \frac{H}{R_M}$, is the standoff parameter, H is the distance between the wall and the center of the bubble, R_M is the maximal semi-spherical radius, and τ_c is the collapse time for a spherical bubble, also called the Rayleigh collapse time given by [94]:

$$\tau_c \approx 0.915 \sqrt{\frac{\rho}{p_\infty - p_v}} R_M, \quad (36)$$

where p_v is the vapor pressure (bubble pressure) [65, 92, 93].

4. Numerical simulation of the light absorption

This section presents numerical simulations related to the manipulation of bubbles through optothermal effects. To understand the mechanisms responsible for steady-state trapping and 3D manipulation of bubbles, we conducted simulations on COMSOL Multiphysics where the heat equation is coupled to the Navier–Stokes equations. This ensures that thermal diffusion from the liquid to the optical fiber and silver nanoparticles is taken into account. These simulations helped analyze the resulting heat transfer and fluid dynamics caused by light absorption, whether from bulk absorption or metal absorption.

In our simulation, we utilized a 2D configuration given the radial symmetry of the problem. The origin of the coordinate system is placed at the center of the optical fiber. This approach helped us to efficiently capture the requisite dynamics and simultaneously minimize computational complexity. To define the boundary conditions of the simulation area, we established the initial temperature as equivalent to the room temperature (293.15 K). Moreover, we implemented a non-slip boundary condition to model the absence of fluid motion at the limits of the simulation domain [59, 60]. The glass cell has a rectangular shape, measuring 2 mm wide and 5 mm high, as shown in figure 6. In our numerical model, the laser beam's optical intensity follows a Gaussian profile, whose core radius corresponds to the waist of the core of the optical fiber (4.5 μm and 52.5 μm for single-mode and multi-mode optical fiber, respectively).

When the light is incident upon an absorbing medium, a portion is absorbed, producing an increase of the temperature at the surrounding medium. This is in accordance with the heat transfer equation (where a steady-state condition is assumed), given by [59, 61]:

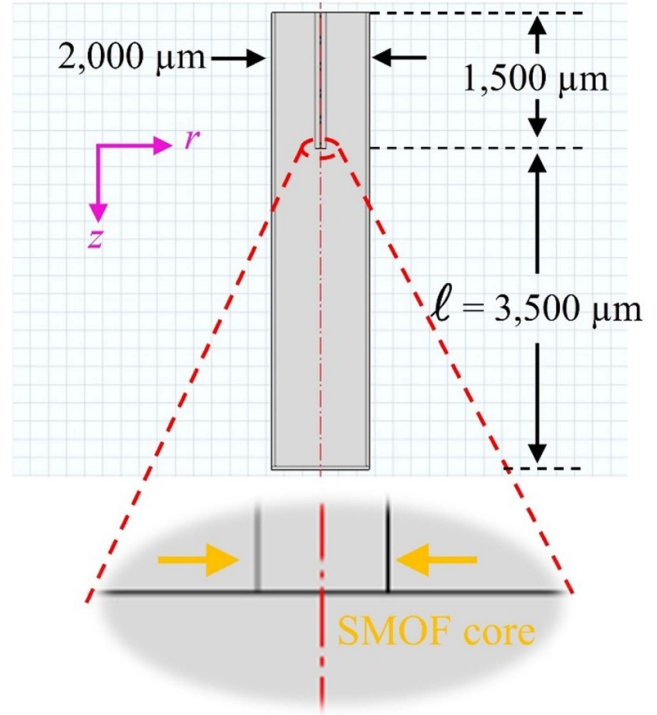


Figure 6. Geometry model of the glass cell and a single-mode optical fiber (SMOF) immersed in ethanol by solving the heat transfer equation coupled to Navier–Stokes equations on COMSOL Multiphysics.

$$\rho c_p \mathbf{u} \cdot \nabla T = \nabla \cdot (K \nabla T) + Q, \quad (37)$$

where $\rho = 789 \text{ Kg m}^{-3}$ the liquid ethanol density at room temperature [57], $c_p = 2.42 \text{ J g}^{-1} \text{ }^\circ\text{C}^{-1}$ is the ethanol heat capacity [103], ∇T is the temperature gradient, $K = 0.17 \text{ W m}^{-1} \text{ K}^{-1}$ is the thermal conductivity [103], and $Q = \alpha T_0$ is the heat source per volume unit where α is the absorption coefficient and I_0 is the optical intensity of the Gaussian laser beam. The fluid field velocity $\mathbf{u}$, i.e. convective currents, is modeled by the Navier–Stokes equation for an incompressible fluid given by [59, 61]:

$$\rho (\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla \cdot \left\{ -p \mathbf{I} + \mu \left[\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right] \right\} + \mathbf{F}_B, \quad (38)$$

$$\rho \nabla \cdot \mathbf{u} = 0, \quad (39)$$

where $\mathbf{I}$ is the identity matrix, $\mu = 1.17 \times 10^{-3} \text{ Pas}^{-1}$ is the dynamic viscosity of ethanol [63], and $\mathbf{F}_B = -\mathbf{g} [\rho(T) - \rho]$ is the volumetric force per volume unit where $\rho(T)$ corresponds to the density of ethanol as a function of the temperature.

To continue with, we describe two scenarios for light-matter interactions that can be used to trap and manipulate bubbles by optothermal means: heating by light bulk absorption at ethanol and heating by light absorption at thin films deposited on a substrate. In our case, a thin film of silver nanoparticles deposited on the core of an optical fiber.

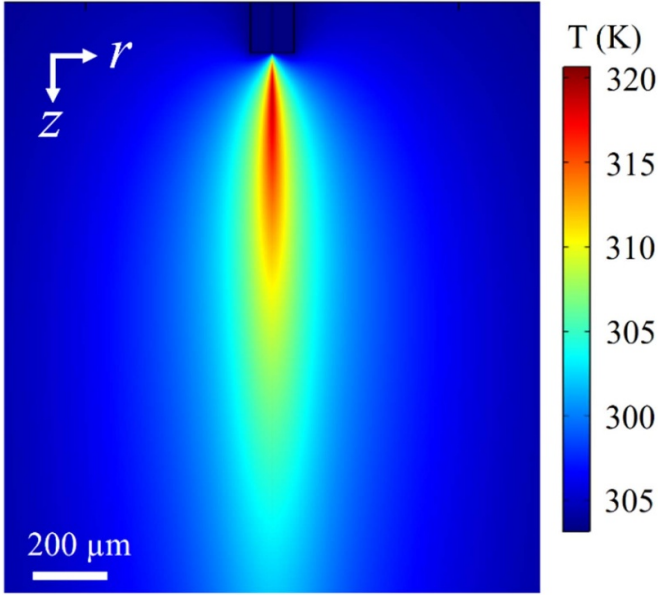


Figure 7. The temperature distribution in ethanol caused by light bulk absorption is extracted from COMSOL simulations using $\lambda = 1550$ nm and 15 mW of optical power. The scale bar represents 200 μm . Reprinted with permission from [62]. © 2021 Optical Society of America.

4.1. Liquid heating by light bulk absorption using CW lasers

In the first case, we model the light bulk absorption from ethanol employing a CW laser emitting at 1550 nm and 15 mW of power. The absorption coefficient of ethanol is $\alpha_{\text{eth}} = 5.63 \text{ cm}^{-1}$ [104].

The light from the 1550 nm laser is delivered into ethanol through a single-mode optical fiber. The light absorption heats the ethanol and generates a spatially varying temperature distribution. Figure 7 shows the spatial temperature distribution resulting from a 15 mW of laser power. The temperature rises as it penetrates the liquid, reaches a peak (~ 320 K at $z \sim 80 \mu\text{m}$), and then decreases to room temperature far from the optical fiber. The thermal diffusion of ethanol is $\sim 85.19 \times 10^{-9} \frac{\text{m}^2}{\text{s}}$ whereas for silica is $\sim 8.66 \times 10^{-7} \frac{\text{m}^2}{\text{s}}$, so the silica will radiate heat more efficiently but ethanol is a bad radiator. Therefore, the temperature of ethanol will increase from the initial temperature (room temperature, 293.15 K) [63].

Figure 8(a) displays the temperature distribution profile along the propagation axis (z -axis), i.e. the longitudinal temperature profile. It is noteworthy that the temperature abruptly increases from room temperature to the peak temperature at a distance of $z \approx 80 \mu\text{m}$ then decreases exponentially in accordance with the heat transfer equation until it reaches room temperature for $z \geq 2000 \mu\text{m}$. Figure 8(b) shows the temperature distribution profile along with radial axis, i.e. transversal temperature profile, at several transversal planes: $z = 20 \mu\text{m}$, 80 μm (highest longitudinal temperature z_{HT}), 360 μm , and 1440 μm . The temperature profile perpendicular to the beam shows a wider spatial distribution compared to the Gaussian beam profile due to heat diffusion.

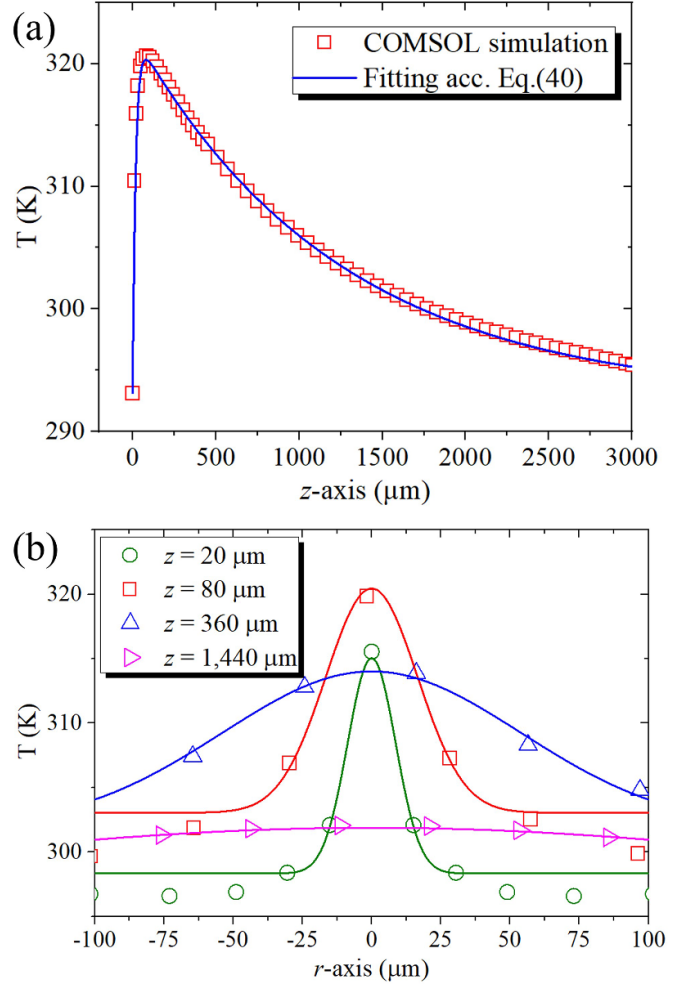


Figure 8. (a) Longitudinal temperature profile along the propagation axis z ($\lambda = 1550$ nm & 15 mW of optical power). The blue line is fitted to equation (40). (b) Transversal temperature distribution obtained at different positions along the z -axis ($\lambda = 1550$ nm & 15 mW of optical power). Continuous lines are fit to Gaussian functions in equation (34). (a), (b) Reprinted with permission from [62]. © 2021 Optical Society of America.

According to the COMSOL simulation results shown in figures 8(a) and (b), a growth followed by an exponential decay function and a Gaussian function were identified as suitable descriptions for the transversal and longitudinal temperature distribution profiles, respectively.

Thus, the longitudinal temperature profile along the z -axis has the form [62]:

$$T_z(z, P) = T_0 + \Delta T_{z1}(P) \left(1 - \exp\left(-\frac{z}{z_1}\right) \right) - \Delta T_{z2} \left(1 - \exp\left(-\frac{z}{z_2}\right) \right), \quad (40)$$

where $\Delta T_{z1}(P)$ represents the temperature difference between the highest temperature and the offset temperature T_0 , while z_1 and z_2 are the characteristic distances representing the increase and decrease of the temperature profile, respectively.

Likewise, the transversal temperature profile has the form [62]:

$$T_r(r, z, P) = T_0 + \Delta T_r(z, P) \exp \left[-\frac{1}{2} \left(\frac{r}{r_0(z, P)} \right)^2 \right], \quad (41)$$

where r is the transversal radial coordinate, z is the longitudinal axial coordinate, ΔT_r represents the temperature difference between the maximum temperature and the offset temperature T_0 (which corresponds approximately to the room temperature for power levels of mW) finally, r_0 is the characteristic distance at which the temperature decays to e^{-1} from its peak value [62].

On the other hand, figure 9(a) shows the convective currents $\mathbf{u}$, leading to the coupling of equations (37)–(39), induced by light bulk absorption. The longitudinal convective currents velocity profile (on the z -axis) is shown in figure 9(b). Figure 9(c) shows the transversal convective currents profile (on the r -axis) taken at $z = 40, 80$, and $120 \mu\text{m}$.

4.2. Liquid heating by light absorption from a CW laser at silver nanoparticles

To continue with our numerical study, absorption from silver nanoparticles deposited on the distal end of the optical fiber is considered. It is central to determine the temperature increase at the AgNPs-ethanol interface. Due to both the timescales and the small volume heated, it is quite complex to experimentally determine the temperature reached, i.e. to our best knowledge, there is no sensor capable of handling nanosecond time scales. However, since the AgNPs are densely packed and uniformly distributed on the core of the optical fiber, forming a film (see figure 13(b)), let us consider them as a thin film of silver [63, 105, 106].

The AgNPs are irradiated by a CW laser emitting at 445 nm and 10 mW of power. The absorption coefficient of silver $\alpha_{\text{Ag}} = 638.2 \times 10^3 \text{ cm}^{-1}$ [107]. Heat is transferred to ethanol from the irradiated zone due to diffusion. The diffusion time is given by [63, 108]:

$$\tau_{\text{diff}} = \frac{d^2}{4D_t}, \quad (42)$$

where d is the thickness of the absorbing medium and D_t is the thermal diffusivity, i.e. $171.92 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ and $85.19 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ for silver [109] and ethanol [110], respectively. Based on the SEM image shown in figure 13(b), the thickness of the silver nanoparticles photodeposited on the core of the optical fiber form a single-layer of nanoparticles with a thickness of $\sim 100 \text{ nm}$ (like a thin film), so $\tau_{\text{diff}} \sim 15 \text{ ps}$ while in ethanol is much larger. This indicates that the thin film of silver nanoparticles efficiently radiates, while ethanol is a poor radiator, leading to an increase in its temperature [63]. Figure 10 shows the spatial temperature distribution field due to light absorption at AgNPs photodeposited in the core of a multi-mode optical fiber (105/125 μm) extracted from the COMSOL Multiphysics.

Similar to section 4.1 (where no NPs are considered), temperature profiles along with the propagation axis and the radial

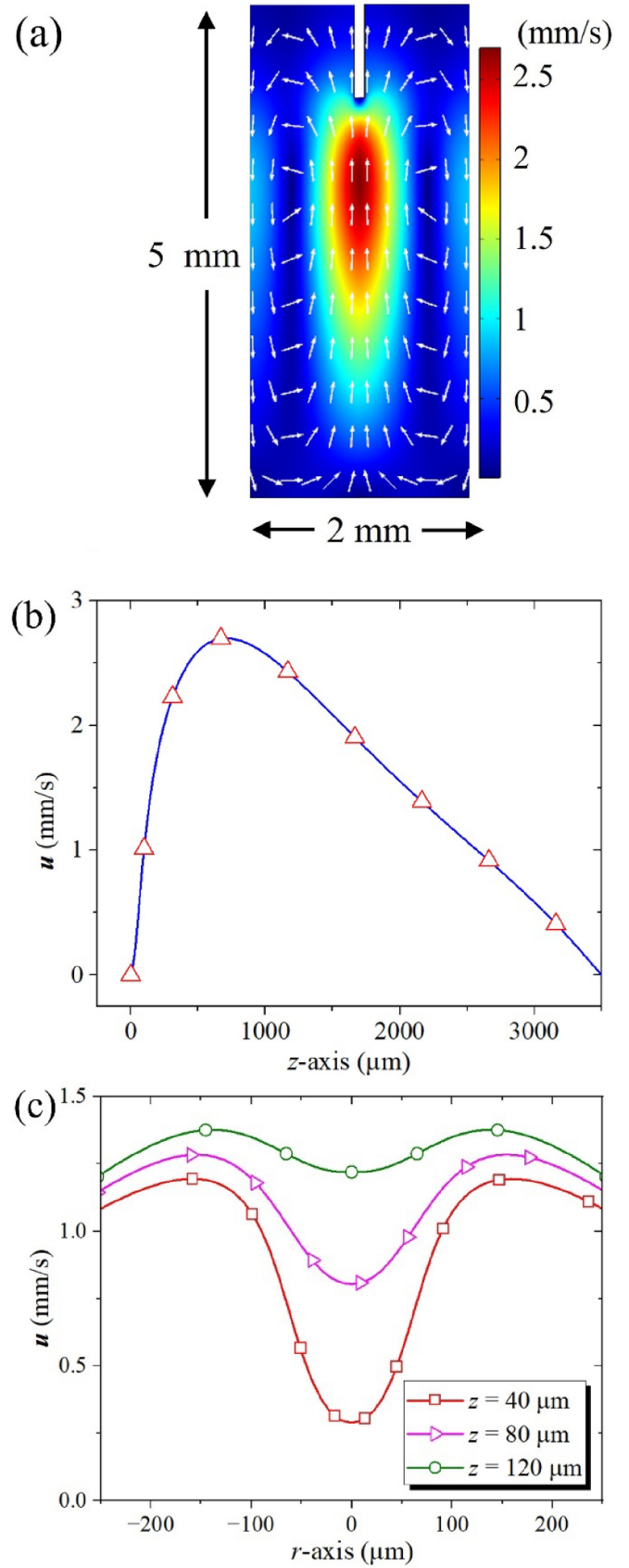


Figure 9. Convective currents field (a), longitudinal (b), and (c) transversal convective currents velocity profile induced by light bulk absorption at $\lambda = 1550$ with 15 mW of optical power. (a), (b) Reprinted with permission from [62]. © 2021 Optical Society of America.

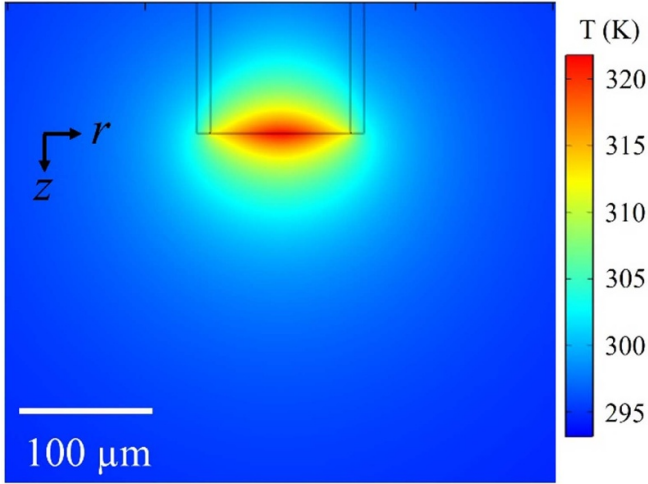


Figure 10. The temperature distribution in ethanol caused by light ($\lambda = 445$ nm) absorption at the AgNPs photodeposited at the core of a multi-mode optical fiber with a power of 10 mW power, extracted from COMSOL simulations. The scale bar represents 100 μm .

one are extracted from the COMSOL simulations, i.e. longitudinal and transversal temperature profiles. Unlike the case of light bulk absorption, the longitudinal temperature distribution exhibits an exponential decay as shown in figure 11(a). The maximum temperature is reached just on the AgNPs/ethanol interface.

Figure 11(b) shows the transversal temperature distribution profile at several transversal planes: $z = 20 \mu\text{m}$, $40 \mu\text{m}$, $60 \mu\text{m}$, $80 \mu\text{m}$, and $100 \mu\text{m}$. The temperature profile perpendicular to the beam shows a wider Gaussian spatial distribution compared to the Gaussian beam profile due to heat diffusion, likewise to the case of light bulk absorption.

The distribution of temperature along the longitudinal axis is given by [57, 62]:

$$T_z(z, P) = T_0 + \Delta T_z(P) \exp\left(-\frac{z}{z_D}\right), \quad (43)$$

where z_D is the characteristic distance representing the decrease of the temperature profile, while the transversal temperature distribution has a similar form to the case with no AgNPs (equation (43)). Convective currents field exhibits a similar trend of section 4.1 (see figure 9).

4.3. Liquid heating by light absorption at silver nanoparticles using a pulsed laser

Let us consider a nanosecond pulsed laser (described in section 5.1.2) impinging on the AgNPs photodeposited on the core of an optical fiber. Determining the temperature increase at the AgNPs-ethanol interface is crucial. However, direct measurement is challenging due to the lack of a thermal sensor capable of operating at the timescales involved ($\tau_p < 10$ ns) in a small heated volume. To address this, reasonable assumptions were made to numerically estimate both the temperature increase and its spatial distribution using COMSOL

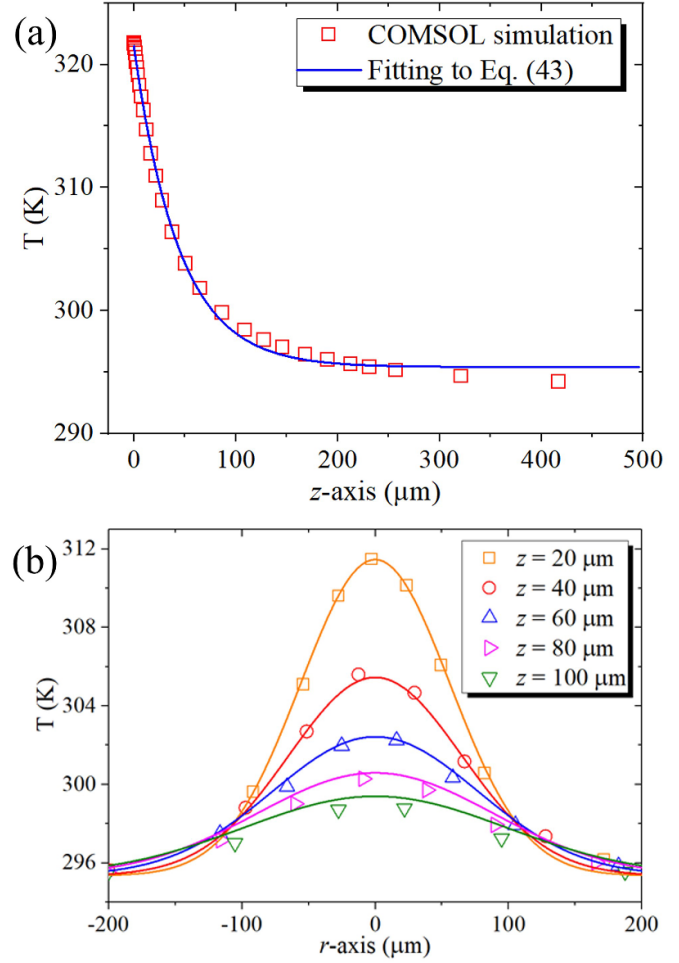


Figure 11. Longitudinal (a) and transversal (b) temperature distribution in ethanol caused by the light absorption at AgNPs film ($\lambda = 445$ nm at 10 mW of optical power) extracted from COMSOL simulations.

Multiphysics. The absorption coefficient of ethanol ($\alpha_{\text{eth}} = 6.8 \times 10^{-2} \text{ m}^{-1}$) is much smaller than that of silver one ($\alpha_{\text{Ag}} = 7.2086 \times 10^{-6} \text{ m}^{-1}$). Therefore, the silver film is the only source of heat.

Figure 12 displays the temperature profile resulting from a single laser pulse (5 ns) operating at a repetition rate of 10 kHz. At the AgNPs/ethanol interface for varying laser pulse energies. In all cases, the temperature quickly rises beyond the boiling point of ethanol ($T_b = 351.4$ K) without boiling and even surpassing the thermodynamic critical temperature of ethanol ($T_c = 516.15$ K). After reaching a metastable state, ethanol undergoes a rapid phase transition to vapor, resulting in the formation of a rapidly expanding bubble. It is important to note that our simulation does not include the liquid-to vapor phase transition. Once the vapor phase is reached, further heating of the ethanol is not possible due to the well-known boiling crisis [72]. Once the laser pulse has passed through, the temperature slowly decreases. When the next pulse arrives 100 μs later, the film almost reaches the steady-state temperature, and the process repeats. Therefore, a bubble is produced with each laser pulse.

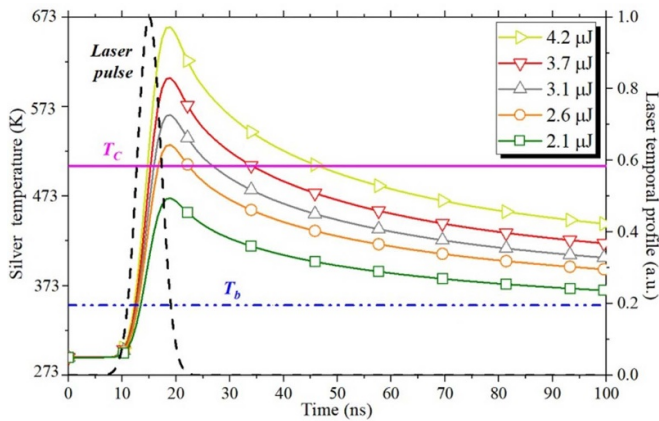


Figure 12. The temperature profile at the AgNPs-ethanol interface was obtained by solving equations (37)–(39) using COMSOL Multiphysics. The phase explosion is more likely to occur around $T_c \sim 516.15$ K (continuous magenta line). The temperature increase at the interface is a linear function of the laser energy. Solid lines represent the temporal profile of the temperature at the AgNPs/ethanol interface due to light absorption. The black broken line represents the temporal profile of one laser pulse. The blue double-dot line represents the pure ethanol boiling temperature $T_b \sim 351.4$ K. Reprinted with permission from [63]. © 2020 Optical Society of America.

5. Experimental generation, trapping, and 3D manipulation of bubbles

This section presents the experimental results related to the manipulation of bubbles through optothermal effects in pure ethanol, including their generation, quasi-steady-state trapping, and 3D manipulation.

5.1. Bubble generation

As we pointed out in section 3, bubble nucleation can be generated by optical means, either by CW or pulsed laser. The selection of the optical source will rely on the available resources.

5.1.1. Bubble generation using a CW laser. Depending on the optical power used, bubble nucleation can occur through thermal breakdown or boiling processes via CW laser (see figure 3). It is important to note that the CW lasers used have an optical power in the milliwatt range, which makes the experimental setup affordable. Additionally, the laser source has a fiber optic mounted on a 3D translation stage that enables spatial control of the laser spot within the liquid.

Figure 13(a) shows the experimental setup used to produce bubbles optothermally by a CW laser in pure ethanol. At the distal end of the optical fiber, silver nanoparticles AgNPs were immobilized following the photodeposition technique reported in [111]. Silver nanoparticles act as both an absorbing medium and a thermal source. In other words, AgNPs absorb light and transfer it to the surrounding liquid. In most cases, the absorption coefficients of metal nanoparticles are several orders of magnitude greater than those of the pure liquid.

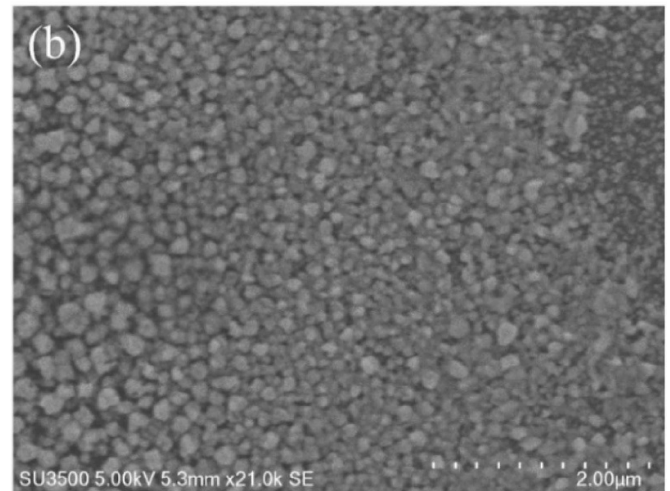
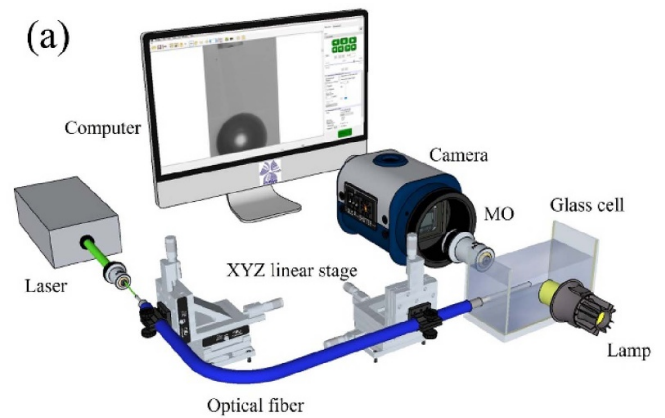


Figure 13. (a) Experimental setup for the generation of bubbles by optothermal effects. A continuous-wave laser beam emitting at $\lambda = 1064$ nm with 16 mW of optical power is coupled to a multi-mode optical fiber (MMOF, 105/125 μm) through a 10x microscope objective (MO). (b) SEM image of the distal end of the multi-mode optical with AgNPs. (a), (b) Reprinted with permission from [63]. © 2020 Optical Society of America.

Figure 13(b) provides a close-up view of the AgNPs photodeposited on the distal end of the optical fiber, captured with a scanning electron microscope (SEM, Hitachi model SU3500). The experimental setup is complemented with a visualization system composed of a microscope objective coupled to a high-speed camera. The illumination is provided by a white lamp.

After the photodeposition of the AgNPs, the distal end of the optical fiber is immersed in the glass cuvette filled with pure ethanol. Figure 14 shows snapshots of the bubble generation and its evolution. Due to the continuous vaporization of ethanol, the radius of the bubble exponentially increases over time and eventually reaches 68 μm after 60 ms [57]. The temperature reached at the interface is a function of both the laser power P and the silver absorption coefficient at the wavelength employed α_λ . Therefore, the interface temperature can be raised beyond the boiling point of ethanol (~ 351.15 K). It is necessary to clarify that bubbles cannot be generated in the absence of nanoparticles at the reported power levels.

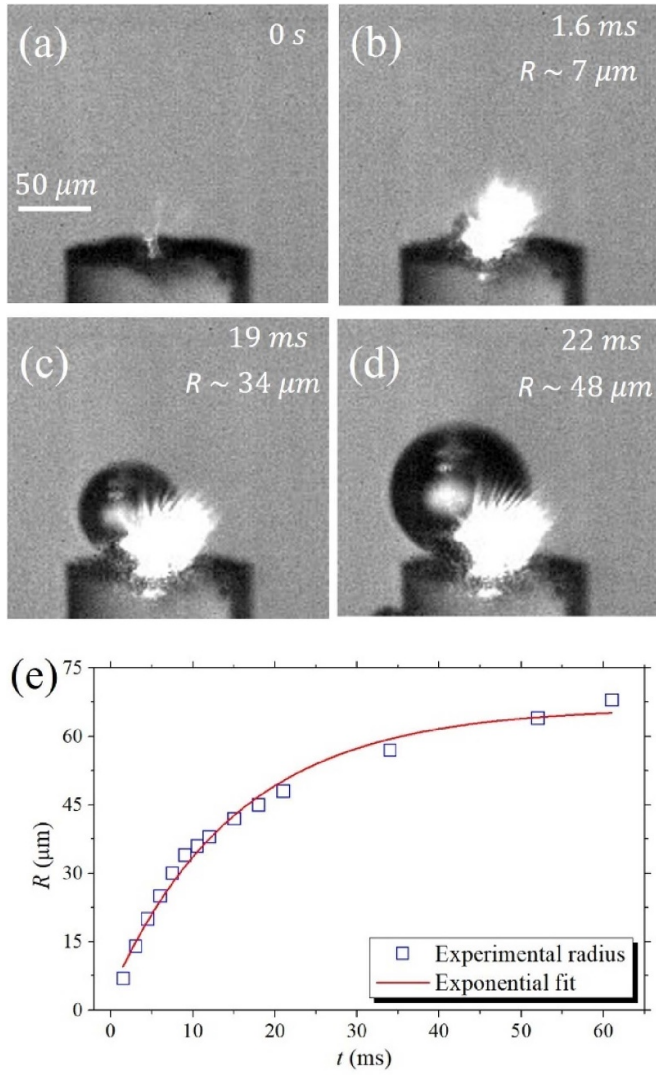


Figure 14. (a)–(d) Snapshots of the optothermal generation of a microbubble using a CW laser emitting at $\lambda = 1064 \text{ nm}$ with 16 mW of optical power. The microbubble grew attached to the core of the distal end of the MMOP with AgNPs photodeposited. (e) The bubble radius is dependent on the illumination time and power. The red solid red line is a fit to an exponential function. (a)–(e) Reprinted with permission from [57]. © 2018 Optical Society of America.

5.1.2. Bubble generation using a pulsed laser. As mentioned previously, optothermal bubble nucleation can be induced by optical breakdown resulting from a pulsed laser (see section 3). In this case, a visible nanosecond pulsed laser is coupled to an optical fiber via a microscope objective. Similar to the CW laser radiation setup (see figure 13(a)), AgNPs were photodeposited on the opposite distal end of the optical fiber.

Figure 15 shows snapshots of the bubble generation by a visible pulsed laser ($\lambda = 532 \text{ nm}$, $\tau_p = 9 \text{ ns}$ of pulse duration, 10 kHz of repetition rate, 2.1 μJ of pulse energy per pulse) coupled to an MMOF with AgNPs photodeposited on its distal end. The optical fiber is positioned upside down, meaning that the laser beam propagates downwards. Each laser pulse releases a bubble from the distal end of the optical fiber against

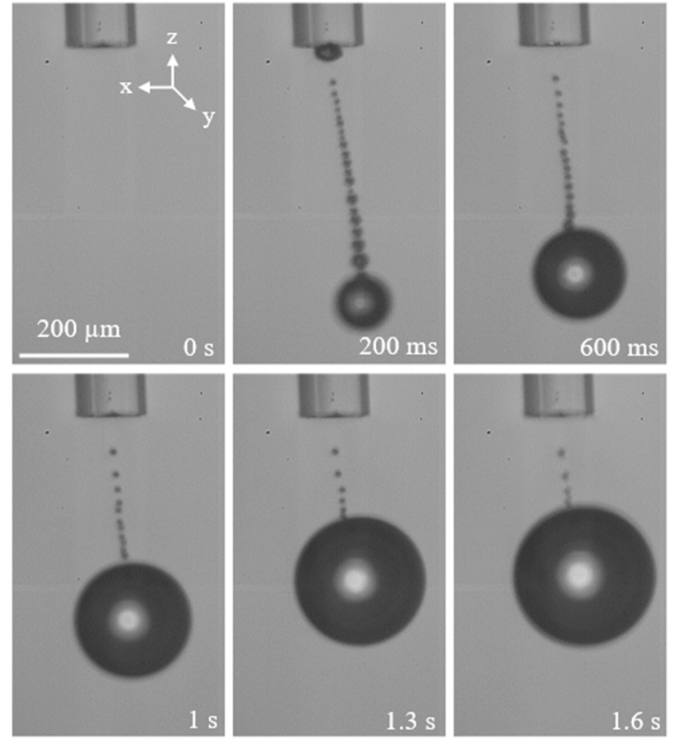


Figure 15. Snapshot of the temporal evolution of the main-bubble radius recorded at 6,600 frames per second. Reprinted with permission from [63]. © 2020 Optical Society of America.

the buoyancy force, creating a long column of bubbles. The diameter of the bubbles increases and their velocity decreases as they move away. Ohl *et al* [112, 113] reported a similar phenomenon, but they used a CW laser source or Joule heating. They did not report the formation of a larger bubble or its trapping and manipulation. As the speed of the bubbles decreases, they eventually coalesce and form a larger bubble, referred to as the main bubble (with a radius of 125 μm after 1.6 s). Our experimental results indicate that the main bubble approaches the heat source due to the Marangoni effect as it grows [63]. Recall that the Marangoni force scales as R^2 . This means that only large enough bubbles will be affected by the force. For example, a bubble with a radius of 5 μm near the optical fiber end will experience $F_M \sim 20 \text{ nN}$, while larger bubbles with a radius $R > 100 \mu\text{m}$ will experience a force more than 400 times larger.

Figure 16 shows the velocity of the ejected bubbles from the tip of the optical fiber along the light propagation (z -axis) at different laser pulse energies.

In all cases, the velocity of the ejected bubbles is faster around the fiber end and then decreases exponentially with distance. The velocity also increases as the laser energy increases. No bubbles were formed for pulse energy $< 2 \mu\text{J}$, and for pulse energy $> 4.2 \mu\text{J}$, two bubbles were expelled but with lower velocity, as shown in the inset of figure 16. To measure the velocity of convective currents, we added silica microparticles (3 μm) as tracers to the solution. By video analysis, the velocity of the tracers (velocity_{max} 0.5 ms^{-1}) (black circles) was computed. The tracers stopped around 300 μm and were then

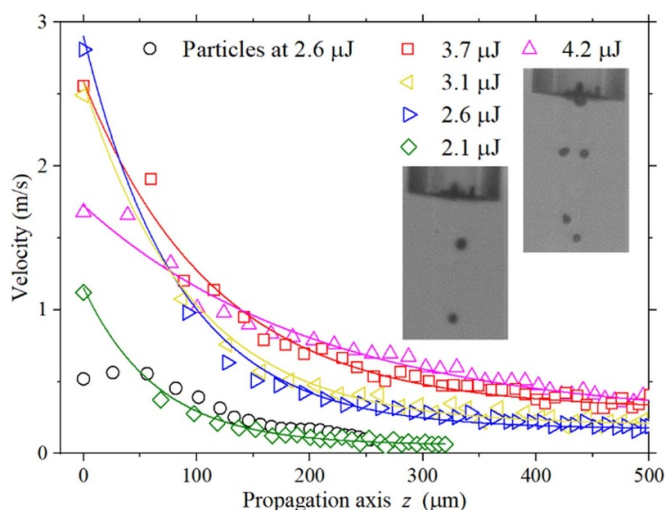


Figure 16. Velocity profile of the bubble as a function of laser energy. The data was extracted from recorded images at 43 000 fps and continuous lines were fitted to an exponential function. Reprinted with permission from [63]. © 2020 Optical Society of America.

carried out of the field of view by the convective currents. It is worth noting that the velocity of the bubbles was always faster than that of the tracers and travelled a longer distance along the propagation distance. This distance was determined by the laser pulse energy.

Figure 17 displays snapshots of the temporal evolution of bubbles generated with $3.1 \mu\text{J}$ per pulse energy, i.e. in the single bubble generation regime. A semi-spherical bubble is created with each laser pulse, which rapidly grows to its maximal radius R_{max} while remaining attached to the optical fiber. This behavior is typical of thermocavitation bubbles. After reaching its maximum radius, the bubble collapses, assuming a mushroom-shaped form produced by a jet directed towards the fiber end. Due to the high temperature of the nanoparticles, the jet is rapidly evaporated, resulting in the ejection of a small bubble.

When the pulse energy is increased beyond $4.2 \mu\text{J}$, larger bubbles are generated, however, its collapse is more complex, as two or even three bubbles are generated, with the smallest bubble always attached to the fiber end as shown in figure 18. Unfortunately, our fast camera was unable to capture the detailed time evolution of the bubble dynamics, so only snapshots of the process were taken.

5.2. Bubble trapping and 3D manipulation

After creating the bubble, whether by using a CW laser or a pulsed laser, spatial control over its displacement can be achieved by generating a temperature gradient through opto-thermal means. This can lead to induced quasi-steady-state trapping of the bubble and its 3D manipulation. To achieve this, a second or even a third laser can be coupled to the experimental setup shown in figure 13(a).

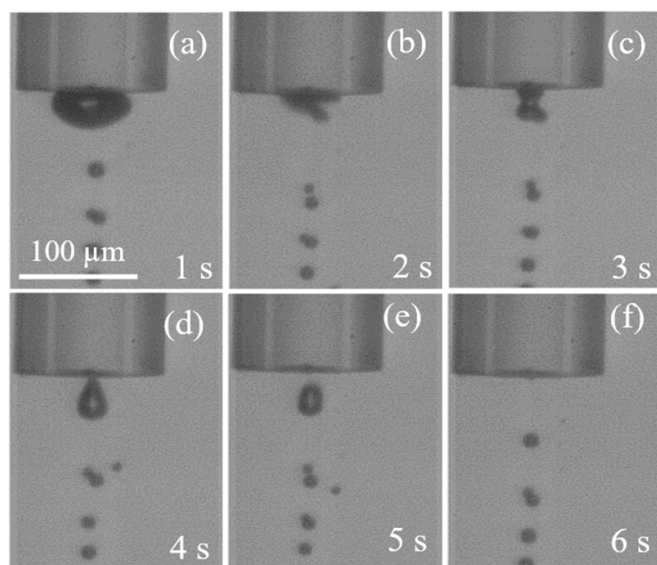


Figure 17. Snapshot of the generation of microbubbles at $3.1 \mu\text{J}$ of laser pulse energy. (a) Maximum cavitation bubble size, (b) bubble collapse, (c) rebound bubble, (d–f) bubble moves away from the optical fiber tip. The scale bar represents $100 \mu\text{m}$. The frame rate was 43 000 fps. (a)–(f) Reprinted with permission from [63]. © 2020 Optical Society of America.

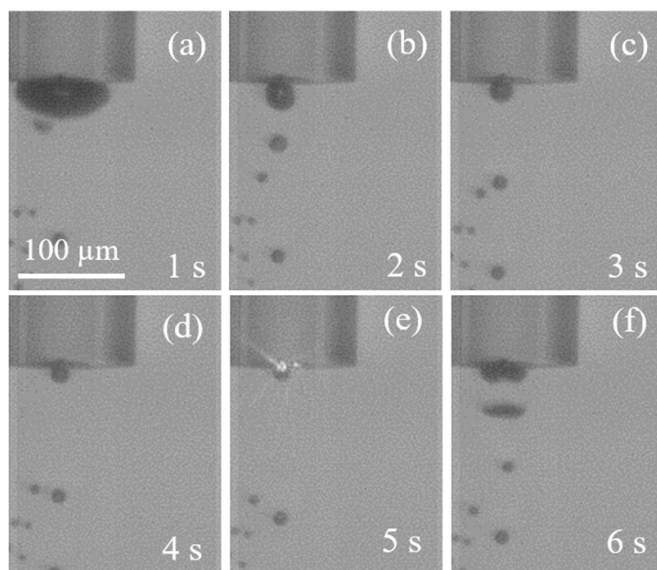


Figure 18. Snapshot of the generation of microbubbles at $4.2 \mu\text{J}$ of laser pulse energy. (a) Maximum cavitation bubble size, (b) bubble collapse, (c) rebound bubble, (d–f) bubble moves away from the optical fiber end. The scale bar represents $100 \mu\text{m}$. The frame rate was 43 000 fps. (a)–(f) Reprinted with permission from [63]. © 2020 Optical Society of America.

5.2.1. CW laser trapping—AgNPs photodeposited on the core of optical fibers. Once the bubble is created and attached to the multi-mode optical fiber (MMOF) using a CW laser (see section 5.1.1), a second MMOF with similar conditions (AgNPs photodeposited on both multi-mode optical fibers) is introduced into the glass cell using a second XYZ linear translation stage. Both MMOFs are coupled to a CW laser that emits

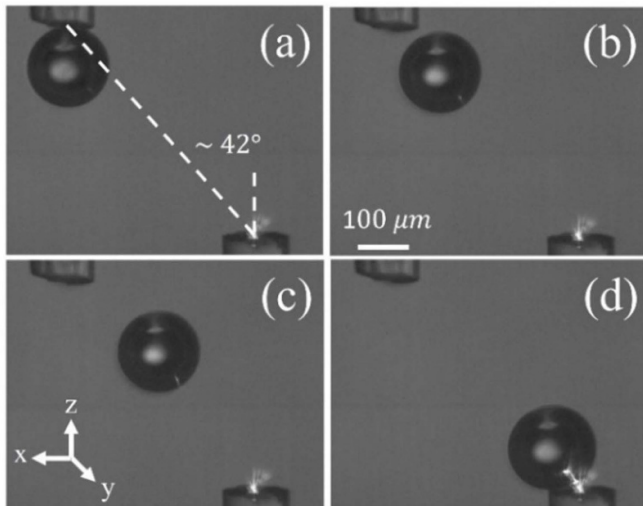


Figure 19. Trapping and 2D manipulation of a bubble between two vertically opposed and off-axis optical fibers employing 11 mW of laser power. (a) The bubble is photothermally induced at the upper MMOF at 16 mW. (b) The upper laser emission is off while the lower emission is on. (c)–(d) the bubble migrates up to the lower MMOF. (a)–(d) Reprinted with permission from [57]. © 2018 Optical Society of America.

at 1064 nm are immersed in a glass cell filled with pure ethanol. The laser light was switched on/off mechanically.

Figure 19 illustrates the trapping and three-dimensional manipulation of an 80 μm radius microbubble positioned between two vertically opposed optical fibers. These fibers are spaced 560 μm apart and are misaligned by 42° with respect to the normal axis. The microbubble is generated in the upper fiber using 16 mW of laser power (as described in section 5.1.1). Once the bubble is produced the laser is turned off and the lower laser is turned on. Thus, the temperature of the lower optical fiber modulates the surface tension of the bubble, resulting in the appearance of a Marangoni force directed towards the lower fiber, causing the bubble to move opposite to the buoyancy force. Conversely, the bubble may also be generated on the lower fiber and subsequently trapped on the upper fiber [57].

5.2.2. CW laser trapping—Bulk absorption. Now, the bubble manipulation is carried out by a second CW laser ($\lambda = 1550$ nm at 15 mW of optical power) coupled to a second optical fiber with a diameter of 9/125 μm (single-mode optical fiber, SMOF) with no AgNPs photodeposited. The laser radiation from the laser is absorbed by the liquid bulk. In particular, the absorption coefficient of the ethanol at 1550 nm is not that large ($\alpha = 2.5$ cm⁻¹) but large enough to increase the liquid temperature by a few degrees with a power of only 15 mW [64].

After the bubble is generated as we pointed out in section 5.1.1, the laser radiation from the MMOF ($\lambda = 10$ 646 nm) is turned off when the laser emitting at 1550 nm is turned on, causing the bubble to migrate towards

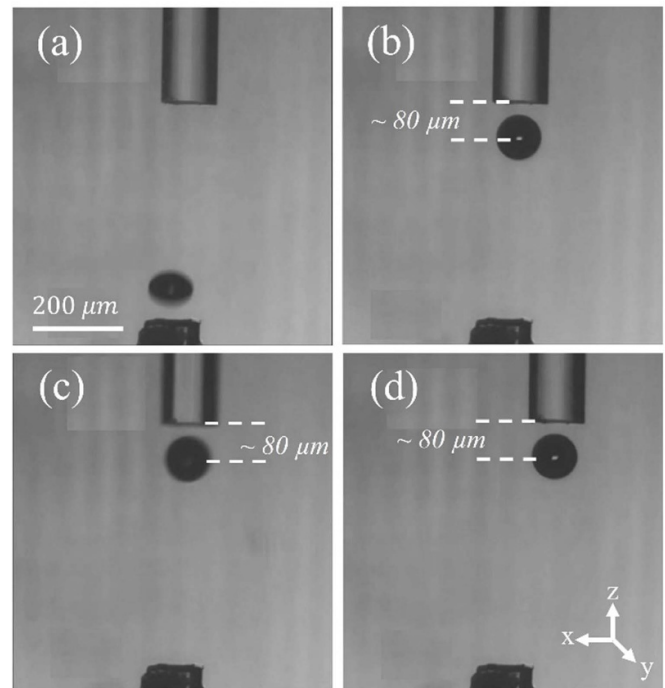


Figure 20. Snapshots of the steady-state trapping and manipulation of a bubble of 50 μm of radius ($\lambda = 1550$ nm at 15 mW of optical power). The scale bar represents 200 μm . (a)–(d) Reproduced from [64]. CC BY 4.0.

the SMOF. The bubble is trapped in a steady state approximately 80 μm away from the SMOF, as shown in figure 20. The 3D trapping of the bubble is demonstrated by moving the SMOF through an XYZ linear stage, where the defocusing of both the optical fiber and bubble corresponds to the displacement out of the focal plane (y-axis).

5.2.3. Trapping and manipulation using CW lasers in a broad spectral range. To extend the landscape of optical sources and demonstrate the ability of optothermal effects to carry out bubble manipulation, we place three different optical fibers (one MMOF for bubble generation and two SMOF for bubble trapping) in a horizontal plane. Each optical fiber, with AgNPs photodeposited onto its core, is coupled to different CW lasers whose wavelength ranges from ultraviolet to infrared light (445 nm, 980 nm, and 1550 nm) at low optical powers.

The laser emitting at 445 nm and 13 mW of laser power was used to propagate light through the optical fiber and heat the AgNPs, thereby generation a bubble (see section 5.1.1). On the other hand, the lasers emitting at 980 nm and 1550 nm with 5 mW and 4 mW power, respectively, were used to trap the bubble. The three optical fiber distal ends were immersed in a glass cell containing pure ethanol, with an ~ 800 μm separation between them. The bubble was trapped and transferred from one optical fiber to another by sequentially switching the CW lasers on and off [62], as shown in figure 21. This experiment only demonstrates the potential for generating, trapping, and manipulating bubbles in a more intricate manner.

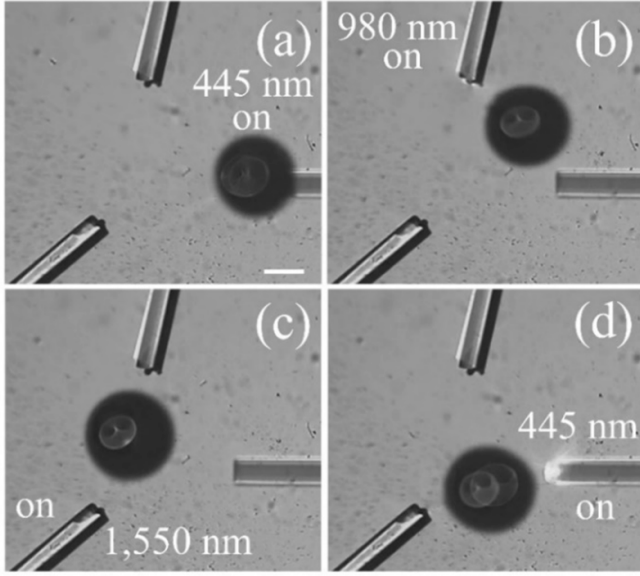


Figure 21. 2D trapping and manipulation of a bubble of $R = 100 \mu\text{m}$ within an array of three optical fibers coupled at three CW lasers driven by the light absorption of AgNPs photodeposited at the core of each optical fiber. The scale bar represents $200 \mu\text{m}$. (a)–(d) Reprinted with permission from [62]. © 2021 Optical Society of America.

5.3. Trapping and manipulation using pulsed lasers

In this case, the generation, steady-state trapping, and 3D manipulation are archived by the same pulsed laser coupled to an optical fiber with AgNPs photodeposited onto its core (refer to section 5.1.2) [63].

Figure 22 displays snapshots of a main-bubble with a radius of $R = 130 \mu\text{m}$ as it approaches the optical fiber. As discussed previously, a large bubble is generated by the coalescence of a large number of small bubbles ($\sim 5 \mu\text{m}$ radius) generated with each laser pulse of $3.7 \mu\text{J}$ of energy and 10 kHz of repetition rate. The temporal evolution of the main-bubble radius is not shown as it occurred out of the field of view. However, the displacement of the main-bubble in the $+z$ direction can be observed while a stream of bubbles moves along the $-z$ direction and eventually coalesces with the larger bubble. This means that the bubble continuously grows, so trapping is achieved for periods ranging from microseconds to milliseconds only before the buoyancy force dominates. Again, the bubble is trapped by the Marangoni effect due to the heat source located at the fiber end. The trapping position of the main-bubble occurs at $\sim 440 \mu\text{m}$ from the fiber end. Likewise, in the case of using CW laser, the trapping of the bubble in 3D can be demonstrated by moving the optical fiber through an XYZ linear stage [63].

6. Discussion

As previously mentioned, the temperature distribution induced by either continuous-wave or pulsed laser affects the surface

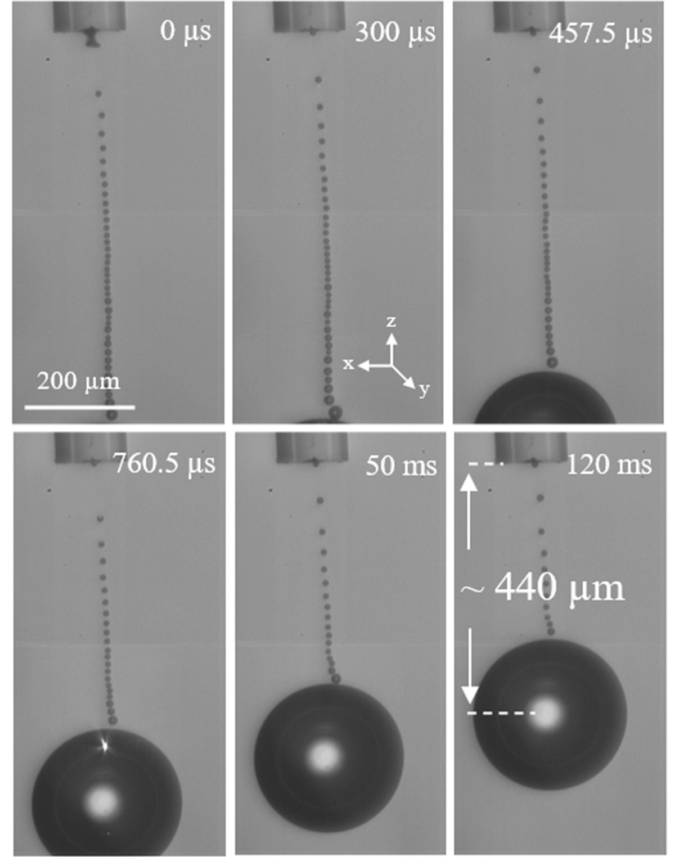


Figure 22. The main bubble is displaced spatially in the $+z$ direction, while the bubble stream moves in the $-z$ direction. Reprinted with permission from [63]. © 2020 Optical Society of America.

tension of bubbles located near the distal end of the optical fiber, resulting in a Marangoni force F_M acting on the bubble.

The Marangoni force given by equation (19) and with the temperature distributions given by equations (40) and (41), can be decomposed into two components: a radial and a longitudinal force as follows [62]:

$$F_{M_r} = 2\pi R^2 \frac{d\sigma}{dT} \left(\frac{\Delta T_t(z, P)}{r_0(z, P)^2} \right) r \exp \left[-\frac{1}{2} \left(\frac{r}{r_0(z, P)} \right)^2 \right], \quad (44)$$

$$F_{M_z} = 2\pi R^2 \frac{d\sigma}{dT} \left\{ \left(\frac{\Delta T_{z1}}{z_1} \right) \exp \left(-\frac{z}{z_1} \right) - \left(\frac{\Delta T_{z2}}{z_2} \right) \exp \left(-\frac{z}{z_2} \right) \right\}. \quad (45)$$

This is like the optical trapping force; however, the Marangoni force is much larger than the optical forces as shown below. Thus, equations (44) and (45) allows three-dimensional manipulation of bubbles.

It is important to note that the analytical expressions developed in this work are valid for both continuous and pulsed lasers. Specifically, equations (44) and (45) describe the Marangoni force regardless of the laser source used.

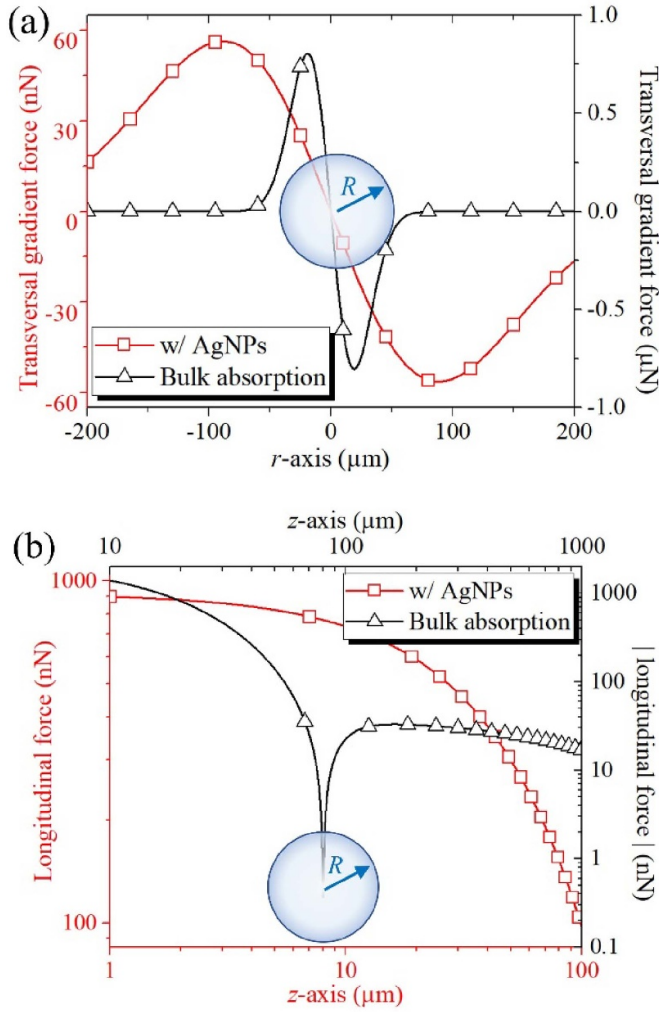


Figure 23. The Marangoni force acting on a bubble with $R = 50 \mu\text{m}$ due to light bulk absorption of ethanol (black) and from absorption of light by AgNPs that are photodeposited on the core of the optical fiber (red). (a) Transversal Marangoni force measured at $z = 80 \mu\text{m}$ away from the distal end of the optical fiber. Figure (b) shows the longitudinal Marangoni force. (a), (b) Reprinted with permission from [62]. © 2021 Optical Society of America.

Figure 23(a) shows the transversal Marangoni force from equation (44) measured at $z = 80 \mu\text{m}$ away from the distal end of the optical fiber over a vapor bubble of $R = 50 \mu\text{m}$. One can notice the profile of the F_{M_r} (with and without AgNPs) exhibits similar performance to the optical transversal gradient force, i.e. displacing the bubble along the r -axis will generate a restoring force aimed at $r = 0 \mu\text{m}$. It should be noted that the magnitude of both force components is determined by various factors, including the ratio of temperature increase and the extent of the temperature distribution.

On the other hand, figure 23(b) depicts the longitudinal Marangoni force from equation (45). It is worth noting that during bulk absorption, the Marangoni force is negative. This indicates that the bubble is repelled when it is located at $z < 80 \mu\text{m}$ and attracted when it is situated at $z > 80 \mu\text{m}$. Consequently, the bubble remains fixed at a distance of $80 \mu\text{m}$ from the optical fiber end (heat source). However, in the case

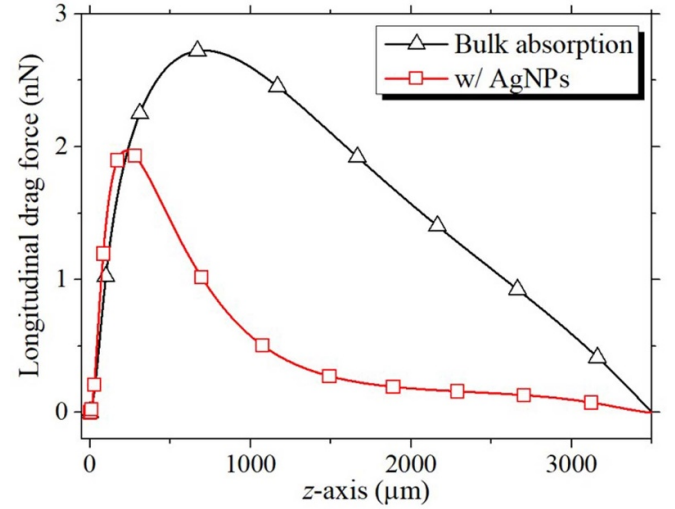


Figure 24. Longitudinal drag force gradient along the z -axis due to light bulk absorption of ethanol and silver nanoparticles for a bubble of $R = 50 \mu\text{m}$.

of AgNPs deposited on the core of the optical fiber end, the bubble is always trapped there. During bulk absorption, the distance where the bubble is trapped depends on the absorption coefficient. As the absorption coefficient increases, the distance to the optical fiber end decreases. It is worth mentioning that the magnitude of the Marangoni force is within the μN range, which is 10^6 larger than optical forces.

Recently, it has been proposed convective currents may contribute an extra force to the trapping force [59, 61, 62, 114]. The drag force F_d can be divided into two orthogonal components, transversal and longitudinal. Since the transversal force behaves similarly to the transversal Marangoni force discussed previously, the bubble is more likely to be trapped along the propagation distance. Figure 24 illustrates the longitudinal drag force (along the z -axis) resulting from both light bulk absorption and light absorption from AgNPs for a bubble with a radius of $50 \mu\text{m}$. As the longitudinal velocity progresses towards the optical fiber, the drag force propels the bubble towards it.

In addition to the Marangoni forces and drag forces, the bubble that is immersed in ethanol experiences the buoyancy force. Although the buoyancy force scales with the bubble radius to the third power and can eventually exceed the Marangoni force, causing the bubble to detach from the optical fiber, in the present case, for a bubble with $R = 50 \mu\text{m}$, $F_b \sim 4 \text{ nN}$. The gravity force, which acts in opposition to the buoyancy force, exerts $\sim 20 \text{ pN}$ over the vapor bubble.

Optical forces may indeed exist, however, only the radiation pressure force will exist given the large radius of the bubbles. Consequently, when the beam reaches the trapping site (located 80 mm from the fiber end), it will encounter an almost flat surface (beam spot size $\sim 4.5 \mu\text{m}$ and the Rayleigh distance $\sim 120 \mu\text{m}$). As a result, the beam will only push the bubble along the propagation distance. Given the power levels used in this work, the optical force falls within the pN range [62, 63], rendering it negligible when compared to the

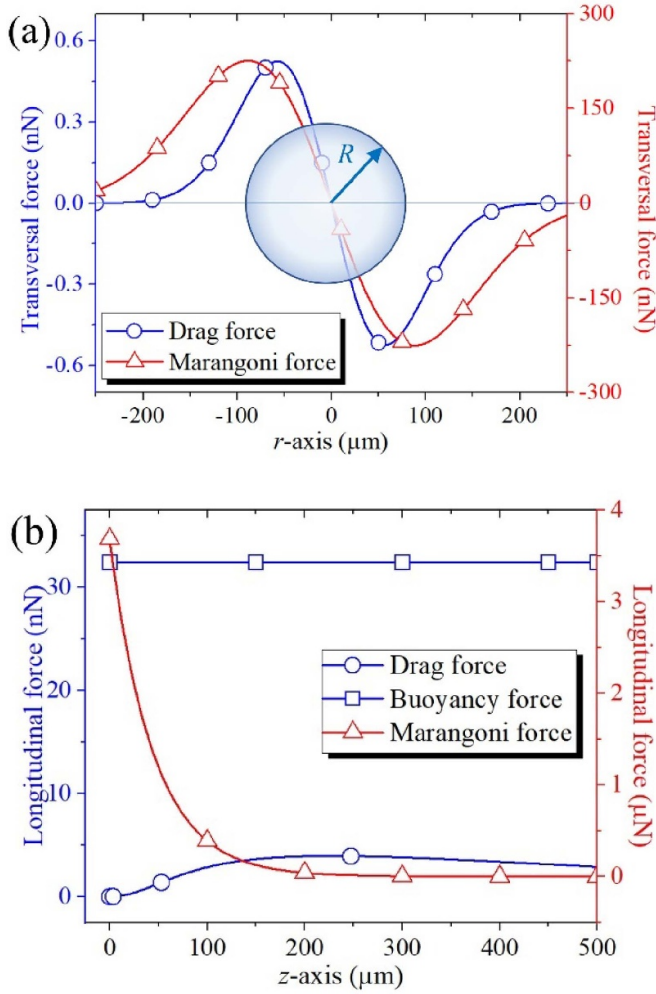


Figure 25. Optothermal forces over a vapor bubble of $R = 100 \mu\text{m}$. (a) Transversal components of the gradient forces F_d (blue circles) and F_M (red triangles). (b) The longitudinal component of F_M (red triangles), F_d (blue circles), and F_b (blue squares). As F_M is greater than F_d and F_b , the microbubble is attracted towards the optothermal source, i.e. the distal end of the optical fiber. (a), (b) Reprinted with permission from [62]. © 2021 Optical Society of America.

Marangoni, buoyancy, and drag forces. Hence, optical forces need not be accounted for in the analysis of steady-state trapping and three-dimensional manipulation of bubbles.

The transversal components of the optothermal effects (F_M , F_d) due to light absorption from AgNPs over a bubble of $R = 100 \mu\text{m}$ are shown in figure 25(a). As previously mentioned, the transverse component of both the Marangoni force and the drag force act as attractive forces, aiding in the steady-state trapping of bubbles. Furthermore, the maximal transverse Marangoni gradient force is ~ 250 times greater than the drag force associated with the convective currents. On the other hand, figure 25(b) shows the longitudinal components of Marangoni, drag, and buoyancy forces. Similarly, the longitudinal Marangoni force is dominant over the drag and buoyancy ones near the distal end of the optical fiber. However, at a distance far away from the optical fiber, the buoyancy force becomes dominant.

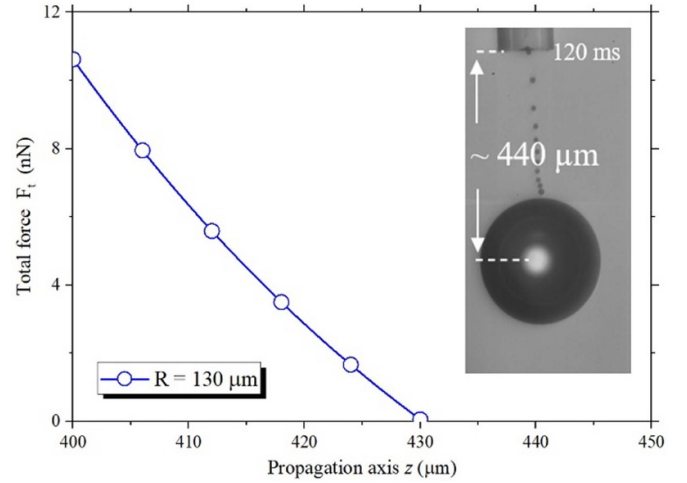


Figure 26. Total force on a bubble with $R = 130 \mu\text{m}$ that achieves quasi-steady-state trapping at $z \sim 430.2 \mu\text{m}$. The experimental trapping distance is depicted in the inset at $z \sim 440 \mu\text{m}$. Reprinted with permission from [63]. © 2020 Optical Society of America.

Based on our theoretical results, the use of a continuous-wave laser in pure ethanol without AgNPs will result in transversal confinement of the bubble of $50 \mu\text{m}$ of radius at $r = 0 \mu\text{m}$ and longitudinal bubble trapping at $z \sim 80 \mu\text{m}$, as shown in figures 23(a) and (b) (extracted from equations (44) and (45)). This is consistent with our experimental results, as shown in figure 20, where the bubble of $R = 50 \mu\text{m}$ is steady-state trapped and 3D manipulated approximately $80 \mu\text{m}$ away from the distal end of the optical fiber at $r = 0 \mu\text{m}$.

On the other hand, our theoretical analysis indicates that the use of a pulsed laser with AgNPs photodeposited on the core of an optical fiber will induce a total force acting on the bubble moving along (against) the propagation axis $\pm z$ -axis given by [63]:

$$F_T = F_b \pm F_M \mp F_d \pm F_i. \quad (46)$$

where the sign $\pm$ denotes along (against) the direction of the light propagation. Thus, F_T drives the trapping of a bubble with a radius of $130 \mu\text{m}$ at $z \sim 430 \mu\text{m}$ away from the optical fiber, as shown in figure 26. The experimental trapping distance occurs at $z \sim 440 \mu\text{m}$ for a bubble of $R = 130 \mu\text{m}$ (see figure 22), it means a mismatch $< 5\%$. However, these results support our theoretical analysis and give us confidence to trapping distance for reaching quasi-steady-state 3D trapping.

7. Conclusions

The optothermal mechanism showcased in this study not only outlines a framework for bubble generation but also for quasi-steady-state trapping and three-dimensional manipulation of bubbles. We showed that 3D trapping is due to the Marangoni effect as a result of the heat produced by light absorption on thin films deposited on the fiber distal end or by bulk absorption on the liquid. The heating can be produced using either CW or pulsed lasers. The heating of the liquid induces a surface tension gradient on the bubble wall, exerting tangential

stress on the bubble wall and propelling it towards the region of lower surface tension, or ‘hot spot’. Despite several proposed mechanisms for bubble manipulation, the optothermal effect emerges as a direct, cost-effective, and efficient non-contact technique, primarily driven by the Marangoni force.

The precise spatial control observed, particularly for large bubbles with a radius exceeding 100 μm , is attributed to the three-dimensional potential well created by the transverse and longitudinal gradient force components of the Marangoni force. The remarkable agreement between our theoretical predictions and experimental results not only provides deeper insights into bubble trapping and manipulation via optothermal effects at low optical powers but also underscores the reliability of the optothermal mechanism.

Moreover, our novel findings in optothermal trapping hold promise for a myriad of applications, spanning medical diagnostics, drug delivery, lithography, microfluidics, hydraulics, and sorting, among others.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflict of interest

The authors declare that there are no conflicts of interest related to this article.

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