DYNAMICS OF LASER-INDUCED 3D MICROBUBBLES

IN AN ABSORBING LIQUID

Thesis

Submitted to

The School of Engineering of the

UNIVERSITY OF DAYTON

In Partial Fulfillment of the Requirements for

The Degree of

Master of Science in Electro-Optics

By

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Dayton, Ohio

August 2019

DYNAMICS OF LASER-INDUCED 3D MICROBUBBLES

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2019

ABSTRACT

DYNAMICS OF LASER-INDUCED 3D MICROBUBBLES IN AN ABSORBING LIQUID

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Optical trapping and manipulation of such microbubbles in a liquid can be used in precise drug delivery and other biological applications.

In this work, the generation and subsequent dynamics of a microbubble in a liquid are investigated, both experimentally and theoretically. When a laser beam is focused into an absorbing liquid comprising colloidal red dye particles in isopropanol alcohol inside a thick quartz cuvette, microbubbles can be generated at around the focus due to nucleation and thermal cavitation. It is experimentally shown that in some cases, the generated microbubble initially moves away from the focus due to the longitudinal optical gradient force, and is later attracted towards the focus due to the longitudinal thermo-capillary force. The thermo-capillary force on the microbubble is determined by solving the heat equation using Fourier transform methods. When developing the complete force model in microbubble dynamics, the thermo-capillary force, optical force, buoyancy force, gravity, and the viscous force have been considered. Dedicated to my families,

For all the support that they gave me to reach this level

ACKNOWLEDGMENTS

First of all, I would like to especially thank my advisor, Dr. Partha Banerjee, for his wonderful guidance, vast knowledge, advice, and continuous encouragement and support throughout this work. Without him, this would have been impossible. I would also like to thank my committee members Dr. Imad Agha and Dr. Chenglong Zhao for their support and helpful suggestions in both my studies and applications.

I also would like to especially thank Dr. Ujitha Abeywickrema for spending his valuable time in the lab with me helping on my experiments and helping in editing my thesis. I would like to thank my friends and my group members for their support in this journey. Also, I would specially thank the Department of Electro-Optics and Photonics staff and SPIE/OSA student chapter officers for giving me wonderful support and creating a pleasant environment for me and all others at the University of Dayton.

Finally, I would like to especially thank my family and my boyfriend for supporting me all the time unconditionally.

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LIST OF ABBREVIATIONS

2D	Two dimensions/dimensional
3D	Three dimensions/dimensional
CW	Continuous wave
NA	Numerical apertures
QEO	Quadratic electro-optic
SPM	Self-phase modulation
ТВ	Thermal blooming
М.О.	Microscope objective

LIST OF SYMBOLS

λ	Wavelength
f	Effective focal length
u(x, y, z; t)	Temperature
<i>x</i> , <i>y</i> , <i>z</i>	Spatial variables in cartesian coordinate
t	Time variable
Γ	The heat generated per unit volume per unit time
α	Thermal diffusivity
k	Thermal conductivity
<i>c</i> _p	Specific heat capacity
ρ	Mass density
ΔT	Change of temperature
α_l	The absorption coefficient of liquid
Ι	Laser beam intensity
P_i'	Power of the laser beam entering the liquid
w(z)	The $1/e$ Beam size at position z
<i>w</i> ₀	The laser beam waist in the liquid
Z _R	Laser beam Rayleigh range in the liquid
n_l	Refractive index for the liquid
<i>z</i> ₀	Distance where the laser beam focuses after entry in the liquid
$M_1 \sim M_6$	ABCD matrix

P _i	Power of the incident laser beam entering the container
Po	Power of the incident laser beam exiting the container
<i>P</i> _o ′	Power of the laser beam exiting the liquid
l_1	The length that laser travels in the air after the microscope
	objective
l_2	Thickness of the wall of the cuvette
l_3	The distance that laser travels in the liquid
F_{TC}	Thermo-capillary force
R ₀	The radius of the microbubble
F _{lTC}	Longitudinal component thermo-capillary force
F _{tTC}	Transverse component thermo-capillary force
σ	Surface tension coefficient
T ₀	The ambient temperature
$lpha_q$	The absorption coefficient of the quartz
L	Inner length of the cuvette
α_l	The absorption coefficient for the liquid
$\mathfrak{I}_{x,y,z}$	Fourier transform operator in x , y and z
$k_{x,y,z}$	Spatial frequency variable corresponding to x, y, z
Ra	Rayleigh number
β	Thermal expansion coefficient
g	Acceleration due to gravity
η	Viscosity of liquid

V	The volume for the microbubble
т	The mass of the microbubble
F_B	The buoyancy force on the microbubble
G	Gravity
$ ho_l$	Density of liquid
$ ho_a$	Density of gas
v	Velocity
F_v	Viscosity force on the microbubble
F _{SN}	Surface tension force
R _s	Contact radius of the microbubble with the container wall
σ	Surface tension coefficient
θ	The contact angle of the bubble
F _F	Friction force
μ	Coefficient of friction (kinetic) between the inner face of the
	cuvette and the microbubble.
α_V	The total polarizability
ϵ_0	The free space permittivity
ϵ_{r1}	The dielectric constant of the medium
ϵ_{r2}	The dielectric constant of the particle
ϵ_{rl}	The dielectric constant of the liquid
ϵ_{rg}	The dielectric constant of the mixture gas in the bubble
F _o	Optical force

С	Velocity of light
Q	Trapping efficiency
Q_g	Gradient trapping efficiency
W _{ini}	Initial waist of the optical field
W _{air}	The laser beam waist at the beam focus in the air after passing
	through the microscope objective
n _a	Refractive index of air
n _q	The refractive index for quartz
q	The q parameter in laser propagation
q_0	The q parament of the plane wavefronts at the entrance surface
	of the microscope objective
Z _{Rini}	The Rayleigh range of the laser in the air

CHAPTER I

INTRODUCTION

1.1 Introduction and Objectives

In 1970, the detection of optical scattering and gradient forces on micron-sized particles was first reported by Ashkin¹. In the late 1980s, the first application of optical tweezers to the biological sciences was demonstrated by Ashkin and Dziedzic², where an optical beam was used to trap an individual tobacco mosaic virus and Escherichia coli bacterium. Nowadays, optical tweezers have proven useful in lots of areas of biology. Due to the attraction between nanoparticles and microbubbles, microbubbles can be controlled to deliver drug particles to a specific location³ or remove waste nanoparticles out of the bloodstream⁴. Thus, the stable and controllable trapping and manipulation of a microbubble are exciting research areas.

Microbubbles can be generated thermally, optically using pulse-laser-induced breakdown, mechanically using porous plates or by using ultrasound, to name a few^{5,6}. After microbubble generation, the stable growth of the existing bubbles, the stationary existence of the bubbles, and the shrinkage and collapse of the microbubbles have been extensively studied^{7,8}. Where needed, such as in targeted drug delivery systems, microbubbles can be collapsed by using ultrasound⁹.

The main mechanisms used for the manipulation of microbubbles are based on thermal, optical, acoustic phenomena, etc¹⁰. Thermal manipulation, which is based on the change of surface tension, produces tangential stress at the gas-liquid interface called the

thermo-capillary force or the Marangoni force^{10,11}. In the Marangoni effect, the spatial gradient of temperature around the bubble causes a surface tension differential along its surface. The surface tension induced flows, and flows due to other factors like free convection can be distinguished by calculating the Rayleigh number of the fluid¹².

It has been shown both experimentally and theoretically that microbubbles confined in a very thin container can be trapped and steered by a focused laser beam^{13–15}. The forces acting on a thermally generated microbubble due to a focused CW laser have been studied, both without and in the presence of agglomerated silver nanoparticles¹⁶. Since the size and position of the microbubble are limited by the thin container, it can be viewed as a twodimensional (2D) microbubble trapping and manipulation experiment.

The objectives of this work are to study the dynamics of a three-dimentional (3D) microbubble in a 3D cuvette and to perform a detailed force analysis to understand the motion of the microbubble, especially the analysis on the thermo-capillary force, which is one of the dominant forces acting on the microbubble. This work reports the experimental observations of the laser-assisted motion of microbubbles in a liquid generated through nucleation and optical cavitation caused by the laser in the absorbing liquid. The liquid is a mixture of red dye and 91% isopropanol alcohol contained in a 3D transparent quartz cuvette, which in total make the liquid the colloid. Incidentally, the refractive index of the gas in the bubble is less than that of the surrounding liquid, which has implications on the optically generated net force on the microbubble.

Specifically, in Chapter II, the experimental setup for the generation of the microbubble and for observing its subsequent dynamics is discussed.

Chapter III introduces the method for generating microbubbles in the experiment. In this work, the microbubbles are thermally generated directly due to evaporation in the liquid: an explosive phase transition to vapor takes place producing a bubble¹⁷. This process requires that the local temperature exceeds the evaporation temperature and the heated area in the liquid is superheated (metastable). One way of achieving this is to focus a laser using a microscope objective into a transparent cuvette filled with liquid with an absorbing dye. The colloidal dye particles assist with the local temperature increase in the medium to initiate nucleation^{18,19}.

In Chapter IV, the relevant theory for the thermo-capillary force is presented, starting with the solution of the steady state heat equation in the presence of conduction where the source term is from the heating by the focused laser beam. The heat equation has firstly been solved in the transverse and longitudinal dimensions separately and the solutions matched at the focal point. Then the 3D steady state heat equation is solved directly by using a 3D Fourier transform approach to get the 3D temperature profile in the liquid, thereby yielding the corresponding 3D temperature gradient and 3D thermocapillary force.

Other forces (viz., the buoyancy force, gravity, viscous force, friction, optical force and surface tension force) acting on the microbubble are shown in Chapter V to give a detailed force analysis and determine the equilibrium condition for microbubble trapping. Chapter VI shows experimental results of microbubble generation and its subsequent dynamics: three types of dynamics are identified and discussed, depending on the location of the focus of the laser beam inside the liquid. Chapter VII concludes the thesis with a short description of possible future work.

1.2 Microbubbles and Their Applications

Microbubbles indicate bubbles which are smaller than one millimeter in diameter, but larger than one micrometer. The generation and controllable manipulation of gas or vapor microbubbles in aqueous liquids are interesting areas for many fundamental and research problems.

Microbubbles are used for thermodynamic studies of liquid superheating and phase transitions²⁰. They can also be used in medical diagnostics as a contrast agent for ultrasound imaging²¹. With the exception of contrast enhancement and molecular imaging, the utilization of microbubbles as targeted delivery vehicles is one of the most intensely researched applications of ultrasound contrast agents²². Nanoparticles can be attached to a laser-induced thermal microbubble, and the microbubble can be steered using a laser beam to a target location. The microbubble can be made to disappear using ultrasound or just simply turning off the laser, and nanoparticles are left behind at the desired position. Microbubbles can also be used for biofilm removal²³ and degradation of toxic compounds, water disinfection, and cleaning/de-fouling of solid surfaces including membranes²⁴.

1.3 Nonlinear Phenomena Induced by Laser in Absorbing Liquid

When an intense beam of light impinges into an absorbing medium, different nonlinear optical phenomena like self-phase modulation (SPM)²⁵, thermal blooming (TB)¹⁸, optical cavitation²⁶, dielectric breakdown²⁷, and photo-capillary effect²⁸ can be induced.

1.3.1 Self-phase modulation and self-lensing

The Kerr effect, which also called the quadratic electro-optic (QEO) effect, is an instantaneous change in the refractive index of a material in response to an applied optical or optical/electrical field. The optical Kerr effect is the case in which the electric field is due to the light itself and causes a change in the polarizability of the optical medium. This, in turn, causes a variation in refractive index which is proportional to the local irradiance of the light ²⁹. This effect only becomes significant with very intense laser beams.

Alternatively, a highly absorbing medium such as a liquid can absorb heat which is generated by a focused laser beam. The spatial gradient of temperature distribution is generated, which in turn generates a change in the refractive index, giving an effect which is similar to the Kerr effect. As shown in Figure 1-1, this can be also responsible for SPM of the laser beam as it propagates through the medium³⁰.

SPM is a self-induced phase change of light as it travels through a nonlinear medium. The change in the refractive index of the medium caused by heat absorption and/or Kerr effect manifests itself in the change of the transverse intensity pattern of the beam. For example, a Gaussian beam traveling through the nonlinear medium results in an almost Gaussian-type refractive index profile, similar to that of a gradient-index lens. This may cause the beam to focus by itself, a phenomenon is known as self-focusing³¹ if the induced nonlinearity is positive. In contrast, self-defocusing occurs if the induced nonlinearity, as in the case of heating, is negative³². The rings in the far field, observed as a result of SPM, have been interpreted as the result of the self-diffraction of light from the nonlinearly induced refractive index profile¹⁸.

SPM due to the Kerr effect has stimulated many applications in the field of an ultrashort pulse including spectral broadening³³ and supercontinuum, temporal pulse compression³⁴ and spectral pulse compression³⁵. The nonlinear properties of a Kerr medium are also beneficial for various optical pulse processing techniques such as optical regeneration³⁶ or wavelength conversion³⁷.



Figure 1-1: Nonlinear effect induced by laser in the absorbing liquid

1.3.2 Thermal blooming

Since the medium becomes lens-like due to the refractive index change, the propagation of the laser beam is substantially affected. The laser beam is a fundamental Gaussian profile, so the generated nonlinear lens due to temperature change resulting from heat absorption is defocusing. Thermal defocusing lenses have been reported for different types of medium like liquid solvents, dye solutions, glasses, and gases ^{38–42}.

The self-induced "thermal distortion" phenomenon of the medium is called TB (see Figure 1-1). This, in turn, causes a spontaneous liquid-gas phase transition, which can be registered as micro-explosions.

The bubble created in the liquid during the experiment affects light propagation. A new type of far field pattern due to TB is observed¹⁸, as the sequence of pictures in Figure 1-2 shows. The usual thermal lens effect can be regarded as SPM, while with the further increase of laser power, the microbubble forms due to TB.



Figure 1-2: Evolution of far-field pattern of the pump from SPM to TB in 2D optical trapping configuration⁴³. (a) initial rings due to SPM; (b) transition from SPM to TB; (c) far-field pattern due to TB.

1.3.3 Optical cavitation

The applications of microbubble described in the last section require a stable and controllable single microbubble generator, which is capable of producing a single cavitation bubble of known size and other given parameters at the given time and location. Optical cavitation is one of the common methods to generate cavitation bubbles.

The most common method to generate optic cavitation involves the focusing of short pulsed lasers⁴⁴ in a transparent liquid media, where nonlinear light absorption and/or cascading ionization produces a hot and supersonic expanding plasma bubble which, upon collapse, generates shockwaves of several *GPa* of pressure⁴⁵. The method of optical

cavitation which uses CW lasers incident in highly absorbing liquids is also called thermal cavitation¹⁷, which will be explained in detail in the next chapter.

1.4 Optical Tweezers

Optical tweezers, which are also called single-beam gradient force traps, are scientific instruments that use a highly focused laser beam to provide an extremely small attractive or repulsive force to hold and manipulate nanometer and micron-sized dielectric particles². Due to the transfer of momentum from the scattering of incident photons, a dielectric particle will experience an optical force which is decomposed into a scattering force and a gradient force. The scattering force is always in the direction of light propagation and the gradient force is in the direction of the spatial light gradient.

The beam is typically focused by sending it through a microscope objective. The narrowest point of the focused beam, known as the beam waist, contains a very strong electric field gradient. The conventional configuration traps dielectric particles which have a larger refractive index than the surrounding medium with a high numerical aperture (NA), viz., NA>1.2, microscope objective⁴⁶. This is because the trapping requires a tightly focused laser at the sample to achieve a peak intensity gradient in order to obtain a high trapping force.

1.5 Significance of This Work

In the conventional configuration of optical trapping, a high NA microscope objective is used to create sharp focusing, and low NA is considered less useful. Nevertheless, a low NA microscope objective lens does have important advantages, such as a long "working" distance (i.e., larger Rayleigh range around focus), reduced aberrations and a wider field of view in comparison with high NA lenses⁴⁷. Therefore, the realization of optical trapping with low NA microscope objective could still be of significant value, viz., in our case of manipulating microbubbles.

Furthermore, in contrast with the conventional setup for dielectric particles, vapor/gas microbubbles have a lower refractive index than the surrounding medium, so that the conventionally known optical trapping force is repulsive, the detailed calculation will be discussed in Chapter V. Instead, the temperature gradient which gives rise to thermo-capillary force is used to trap and manipulate the microbubbles, which can be viewed as an extension of optical trapping.

The configuration in this experiment uses a quartz cuvette which is $5 mm \times 12.5 mm \times 45 mm$, therefore, the size and position of microbubble are not limited by the container, and the whole generation process of the microbubble and its motion under the thermo-capillary force and other forces in 3D can be observed clearly. Once the microbubble detaches from the inner wall, there is no surface tension force on the microbubble due to the wall effect, and the microbubble can be viewed as a sphere to simplify the force analysis. The laser beam in our experiment travels horizontally through the cuvette, instead of vertically as in our previous work in a thin container¹³, so that the buoyancy force and gravity-are perpendicular to the longitudinal part of the thermo-capillary force, and the longitudinal gradient optical force.

When determining the total thermo-capillary force, both transverse and longitudinal temperature change profiles are calculated by separately solving transverse and

longitudinal heat equations and matching the peak temperature change first. In addition, in this work, the more exact thermo-capillary force on a microbubble is determined by directly solving the 3D heat equation by 3D Fourier transform methods.

CHAPTER II

EXPERIMENTAL SETUP

2.1 Introduction

In this Chapter, the experimental setup is introduced. The experimental setup is used for generating the microbubble and for monitoring microbubble's subsequent dynamics.

2.2 Experimental Setup

The experimental setup used for microbubble formation and for monitoring its evolution and motion is shown in Figure 2-1.



Figure 2-1: Schematic of the experimental setup (top view) showing microscope objective (M.O.) in front of quartz cuvette containing the liquid (a mixture of isopropanol alcohol with red dye).

Microbubbles are thermally generated in a liquid contained in a transparent 3D quartz cuvette (inner dimensions length \times width \times height of 5 $mm \times 12.5 mm \times 45 mm$, wall thickness 1 mm) using a focused fundamental mode Gaussian beam from a CW Arion laser with a typical power of 100 mW at 514.5 nm.

The liquid in which bubbles form is a mixture of red dye and 91% isopropanol alcohol. The laser is focused on the liquid using a microscope objective lens (magnification 20X, numerical aperture NA = 0.4, effective focal length f = 9 mm). The microbubble is generated at the focus of the Gaussian beam, where has the maximum average temperature increase, and can thereafter be manipulated by the same beam. By moving the microscope objective, the location of the beam focus in the liquid can be changed to study microbubble dynamics according to the different beam focus locations. To achieve a specific beam focus location, the position of the microscope objective can be determined using the *ABCD* matrix approach, to be described later in Chapter VI. The *ABCD* matrix can also be used to determine the focal spot size of the beam in the liquid. An observation screen is placed 50 cm behind the cuvette, where the far field pattern can be monitored.

CHAPTER III

MICROBUBBLE GENERATION

3.1 Introduction

In this Chapter, the method which is used in this experiment to generate microbubble is explained. The red dye increases the net absorption of 91% isopropanol alcohol, and the nanoscale red dye particles serve as nucleation sites. Gas bubbles can be generated in the liquid when the power reaches a threshold and the temperature around the red dye particle exceeds the evaporation temperature through thermal cavitation.

3.2 Nucleation

In this work, the microbubble is generated thermally by focusing a 514.5 *nm* CW laser into the liquid, which is a mixture of red dye (water, propylene glycol, FD&C reds 40&3, and 0.1% propylparaben, McCormick & Co., Inc.) and 91% isopropanol alcohol. The dye is widely used in previous studies^{18,19} in solutions when studying the laser-induced nonlinear effects in the absorbing liquid in order to have sufficient absorption. Besides increasing the net liquid absorption since the red dye particles in the alcohol can absorb the heat at created by the focused green laser beam, the small particles in the red dye can also serve as nucleation sites⁷ when forming microbubbles.

The phenomenon of nucleation is a thermodynamic process that governs phase separation in both natural and technological processes. Nucleation is affected by both temperature and supersaturation level of the liquid. When nucleation is influenced by temperature, like in case of boiling, the change in temperature can cause changes in local pressure that favors the formation of vapor nuclei⁴⁸. In supersaturated systems, however, the nucleation happens because of the system's attempt to recover equilibrium by phase separation at a constant temperature.⁴⁹

3.3 Thermal Cavitation Process

The other phenomenon related to the microbubble generation when a focused CW laser beam is introduced to the liquid is thermal cavitation. The thermal cavitation can be obtained in any solution like ethanol¹⁸ as long as the absorption coefficient at the illumination wavelength is large¹⁷.

In order to enable evaporation of isopropanol alcohol (temperature for vaporization at standard atmospheric pressure is about 80 °C), the minimum power of the CW 514.5 *nm* Ar-ion laser should be about 100 *mW*, based on our observations. As the laser power is increased above this critical value, the laser-heated region becomes metastable (superheated), and the liquid becomes unstable to random density fluctuations⁵⁰. When the local temperature around the red dye particle exceeds the evaporation temperature, an explosive phase transition to vapor takes place, thereby producing a bubble¹⁷. The microbubble contains a nonhomogeneous mixture of gas (possibly air) and vapor⁵¹. The growth or decay of bubbles may occur through the evaporation or condensation of molecules at the gas-liquid interface⁵⁰.

3.4 Experimental Observations

Whether the microbubbles are formed directly by the local pressure change caused by the laser or are caused due to nucleation and subsequent thermal cavitation from the red dye particles can be readily verified based on a simple control experiment.

As shown in Figure 3-1(a), it has been observed that no microbubbles are generated for an incident 100 mW focused 514.5 nm laser beam after 5 minutes when there is only pure isopropanol in the cuvette. However, it has been observed that when one drop of red dye is introduced into the 91% isopropanol alcohol and diffuses to the vicinity of the laser beam, nucleation occurs. The radius of the red dye particle is estimated as 100 nm ^{7,52} (which is less than the laser beam wavelength of 514.5 nm). For this particle size, the red dye is a colloid in the alcohol, giving rise to the Tyndall effect or scattering of the light, as shown in Figure. 3-1 (b).



(b)

(c)

Figure 3-1: (a) A focused 514.5 nm CW laser beam (100 mW) incident in the pure 91% isopropanol alcohol showing no formation of microbubbles. (b) One drop of red dye introduced from the top with a dropper diffusing in the solution showing nucleation and cavitation (marked by light scattering and light flashes) in the vicinity of the focused CW laser beam. (c) Nucleation and cavitation in the colloidal solution of red dye in alcohol.

(a)

The red dye is able to remain as a colloidal solution after shaking the cuvette and mixing in the liquid completely, as shown in Figure. 3-1(c). Thermal cavitation is accompanied by micro explosions, marked by observable light flashes, implying microbubble formation.

3.5 Conclusion

Due to nucleation and thermal cavitation, microbubble can be formed thermally when a CW laser is introduced into absorbing the liquid. Nanoscale dye particles increase the net absorption coefficient of the liquid. After the microbubble generation, its motion is determined by the competition of forces acting on it. Since the presence of spatial gradient of temperature generated in the liquid with the laser incidence, a microbubble tends to move to the maximum temperature area. In the next Chapter, the estimation of thermo-capillary force which is induced by the spatial gradient of temperature is derived based on the steady state heat equation.

CHAPTER IV

EXACT SOLUTION OF THERMAL CAPILLARY FORCE

4.1 Introduction

As mentioned in Chapter I, the thermo-capillary force due to the temperature spatial gradient of temperature is responsible for attracting the bubble towards the focus of the laser beam¹². In this Chapter, the absorption coefficient of the liquid is calculated with the measured powers, and the estimation of thermo-capillary force is presented.

The attractive thermo-capillary force can be calculated by the spatial gradient of temperature, and this gradient is related to the laser intensity and absorption coefficient of liquid. Since the transverse thermo-capillary force has been calculated in 2D condition by Abeywickrema *et al.*¹³, the longitudinal thermo-capillary force is estimated by solving the longitudinal part of the steady state heat equation in 3D condition first. In this calculation condition, the thermo-capillary force is considered as the overall effect of transverse thermo-capillary force and longitudinal thermo-capillary force. Thus, the transverse and longitudinal parts are adjusted to be matched at the beam focus point. In addition, the more exact thermo-capillary force is estimated by directly solving the 3D steady state heat equation with the 3D Fourier transform method.

The justification of using the heat equation based on predominantly conduction of the heat is given later in this Chapter.

4.2 Heat Equation

In Cartesian coordinates, the heat equation is expressed as

$$\frac{\partial u}{\partial t} = \alpha \left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \right) + \frac{1}{c_p \rho} \Gamma, \qquad (4-1)$$

where u(x, y, z; t) is the temperature and dependent on three spatial variables (x, y, z) and the time variable t; Γ is the heat energy generated by the heat source per unit volume per unit time. The thermal diffusivity which is given by⁵³

$$\alpha = \frac{k}{c_p \rho},\tag{4-2}$$

is a material-specific quantity depending on the thermal conductivity k, the mass density ρ , and the specific heat capacity c_p .

As $t \to \infty$, and if the temperature *u* is not dependent on time, it is assumed conditions exist such that

$$\frac{\partial u}{\partial t} = 0. \tag{4-3}$$

In this work, even though the local temperature around the red dye particles can reach the evaporation temperature, an average temperature profile is established, and this temperature distribution can be calculated via the heat equation. For simplicity, only the steady state temperature distribution after the microbubble generation is considered. In this steady state case, any spatial gradient of temperature does not change in time. The steady state heat equation, therefore, describes the final steady state result in all thermal problems in which a source is switched on (in this experiment, it means turning on the laser), and enough time has passed for all permanent spatial gradients of temperature to establish themselves in space, after which these spatial gradients no longer change in time.

The steady state heat equation for a volume that contains a heat source (the inhomogeneous case), is the Poisson's equation. The change in temperature u over the ambient temperature can be represented by ΔT . It follows that the change in temperature in the steady state can be found as²⁵

$$-k\nabla^2(\Delta T) = -k\left(\frac{\partial^2 \Delta T}{\partial x^2} + \frac{\partial^2 \Delta T}{\partial y^2} + \frac{\partial^2 \Delta T}{\partial z^2}\right) = \Gamma.$$
(4-4)

4.3 The Heat Source: Fundamental Gaussian Beam

The heat source is from the laser radiation that is absorbed by the liquid, in this case,

$$\Gamma = \alpha_l I, \tag{4-5}$$

where α_l is the absorption coefficient of the liquid medium and I(x, y, z) is the laser beam intensity profile with respect to three position variables *x*, *y* and *z*.

The intensity can be written as:

$$I(x, y, z) = \frac{2P_i'}{\pi w^2(z)} \exp(-\frac{2(x^2 + y^2)}{w^2(z)} - \alpha_l(z + z_0)),$$
(4-6)

where P_i' is the power of the laser beam entering the liquid (see Figure 4-1). w(z) is the 1/e beam size at position z and given by $w^2(z) = w_0^2 (1 + (\frac{z}{z_R})^2)$, where z = 0 is the location of the beam waist and w_0 is the beam waist (focal spot size) in the liquid. The Rayleigh range is $z_R = \pi n_l w_0^2 / \lambda$, where n_l is its refractive index of surrounding material,
in this work, n_l is the refractive index of the liquid, and z_0 denotes the distance where the laser beam focuses after entry in the liquid. In writing Eq. (4-6), an exponential decay term has been incorporated in accordance with Beer's law⁵⁴.



Figure. 4-1: Sketch of beam propagation from the effective microscope objective lens through the cuvette. The laser beam is incident from the right to left. The *ABCD* matrices $M_1 \sim M_6$ are used to calculate the beam width and Rayleigh range (see Chapter VI). P_i and P_o are powers of the laser beam entering and exiting the cuvette, and P_i' and P_o' are the powers of the laser beam entering and exiting the liquid, respectively. The distance l_1 is the length that laser travels in the air after the objective lens, l_2 is the thickness of the wall of the cuvette, and l_3 is an arbitrary distance of propagation of the laser beam through the liquid measured from the inner wall of cuvette.

4.4 Thermo-Capillary Force

Due to the tangential stress from surface tension on the microbubble, the bubble should "flow" from a higher surface tension (lower temperature) region to lower surface tension (higher temperature) region. The thermo-capillary force can be expressed as¹⁰

$$\boldsymbol{F}_{TC} = -2\pi R_0^{\ 2} \left(\frac{\partial \sigma}{\partial T}\right) \boldsymbol{\nabla}(T), \tag{4-7}$$

where R_0 is the radius of the microbubble, and $\frac{\partial \sigma}{\partial T}$ is the temperature coefficient of the surface tension, and this coefficient can be considered to be practically constant from room temperature up to 50 °C⁵⁵. Since

$$\nabla T = \nabla (T_0 + \Delta T) = \nabla (\Delta T), \tag{4-8}$$

where T_0 is the ambient temperature. Eq. (4-7) reduces to

$$\boldsymbol{F}_{TC} = -2\pi R_0^{\ 2} \left(\frac{\partial\sigma}{\partial T}\right) \boldsymbol{\nabla}(\Delta T). \tag{4-9}$$

4.5 Estimation of Absorption Coefficient

The absorption coefficient of the liquid α_l can be determined by measuring the input and output powers P_i , P_o , respectively, and using the relation

$$P_o \approx P_i e^{-L\alpha_l} e^{-2l_2\alpha_q} \left(1 - R_{qa}\right)^2 \left(1 - R_{ql}\right)^2, \tag{4-10}$$

where

$$R_{qa} = R_{aq} = \left(\frac{n_q - n_a}{n_q + n_a}\right)^2, R_{ql} = R_{lq} = \left(\frac{n_q - n_l}{n_q + n_l}\right)^2$$
(4-11)

are the reflectivities at the quartz-air/air-quartz and quartz-liquid/liquid-quartz interfaces, respectively (see Figure 4-1), α_q is the absorption coefficient of quartz, and $n_{a,q,l}$ denote the refractive indices of air, quartz and liquid, respectively. The refractive index for quartz at 514.5 nm is $n_q = 1.4616^{56}$ and the refractive index of air is $n_a = 1$. For simplicity, the (real part of) refractive index of the colloidal solution of red dye in isopropanol alcohol is taken to be $n_l = 1.3772$, which is the refractive index of isopropanol alcohol. Also, in Eq. (4-10), l_2 is the thickness of the quartz wall of the cuvette and *L* is the inner length of the cuvette between the walls in the direction of propagation of the laser beam, as shown in Figure 4-1.

However, as seen from our previous work ¹³, reflectivity at the quartz-liquid interface can be neglected compared to the reflectivity at the air-quartz interface since $n_a =$ $1, n_q = 1.4616, n_l = 1.3772$; also, the absorption coefficient α_q of quartz can be neglected compared to the absorption coefficient α_l of the liquid. Under these approximations, Eq. (4-10) is simplified to $P_o \approx P_i e^{-L\alpha_l} (1 - R_{qa})^2$. The laser power incident on the liquid filled cuvette is assumed as $P_i = 100 \ mW$, and P_i' is calculated using $P_i' \approx P_i (1 - R_{aq})$. Setting $L = 5 \ mm$, the absorption coefficient for the liquid is deduced as $\alpha_l = 280/m$.

4.6 2D Solution of Thermo-Capillary Force

Since the general 3D solution for the heat equation is complicated, the total thermocapillary force will be first calculated as the sum of transverse (tangential) thermo-capillary force F_{tTC} and longitudinal (axial) thermo-capillary force F_{lTC} .

To solve for the transverse thermo-capillary force, the intensity is assumed to be only a function of x and y, with z as a constant parameter, thus, the Eq. (4-6) is:

$$I(x, y; z) = \frac{2P_i'}{\pi w^2(z)} \exp(-\alpha_l(z+z_0)) \exp(-\frac{2(x^2+y^2)}{w^2(z)}).$$
(4-12)

so that combining Eq. (4-12) with Eq. (4-4) and (4-5),

$$\frac{\partial^2 \Delta T}{\partial x^2} + \frac{\partial^2 \Delta T}{\partial y^2} = -\frac{2P_l' \alpha_l}{k\pi w^2(z)} \exp(-\alpha_l(z+z_0)) \times \exp(-\frac{2(x^2+y^2)}{w^2(z)}).$$
(4-13)

Numerical methods based on Fourier transform and MATLAB[®] can be used to directly solve the differential equation (4-13). The solution can be expressed symbolically as

$$\Delta T(x, y; z) = \Im_{x, y}^{-1} \left\{ \frac{1}{k_x^2 + k_y^2} \Im_{x, y} \left[-\frac{2P_i' \alpha_l}{k \pi w^2(z)} \exp(-\alpha_l (z + z_0)) \times \exp\left(-\frac{2(x^2 + y^2)}{w^2(z)}\right) \right] \right\},$$
(4-14)

where $\mathfrak{I}_{x,y}$ is the Fourier transform operator in x, y, and $k_{x,y}$ are corresponding spatial frequencies. Once ΔT is known, its transverse gradient $\nabla_t(\Delta T)$ can be calculated to find the transverse thermo-capillary force F_{tTC} .

To solve for the longitudinal thermo-capillary force F_{lTC} , the intensity is considered to be only a function of z, with x = y = 0 (on-axis) for simplicity:

$$I(z) = \frac{2P_{i}'}{\pi w^{2}(z)} \exp(-\alpha_{l}(z+z_{0})) = \frac{2P_{i}' z_{R}^{2}}{\pi w_{0}^{2}} \cdot \frac{1}{z_{R}^{2}+z^{2}} \cdot \exp(-\alpha_{l}(z+z_{0})).$$
(4-15)

The solution can be expressed symbolically as

$$\Delta T(z) = \Im_z^{-1} \{ \frac{1}{k_z^2} \Im_z \left[-\frac{2P_l' z_R^2 \alpha_l}{k \pi w_0^2} \times \frac{1}{z_R^2 + z^2} \times \exp(-\alpha_l (z + z_0)) \right] \},$$
(4-16)

where \Im_z is the Fourier transform operator in *z*, and k_z is a spatial frequency variable corresponding to *z*. Once $\Delta T(z)$ is found, its longitudinal gradient can be numerically determined to find the longitudinal thermo-capillary force F_{lTC} .

The transverse and longitudinal thermo-capillary forces F_{tTC} , F_{lTC} at various locations in the liquid can be calculated by using Eqs. (4-9), (4-14) and (4-16). It is

assumed that the position of laser beam focus is about $z_0 = 2 mm$ away from the entry wall of the cuvette. The laser power is assumed as $P_i = 100 mW$. The temperature coefficient of surface tension is taken as $\frac{d\sigma}{dT} = -0.0789mN/(m \cdot K)^{57}$ and the thermal conductivity of the liquid is taken as $k = 0.136Wm^{-2}\circ C^{-158}$, which is the thermal conductivity of isopropanol alcohol. The microbubble radius is taken as $R_0 = 100\mu m$.

When solving Eq.(4-14), to avoid singularity, $(k_x^2 + k_y^2)$ is replaced with $(k_x^2 + k_y^2 + C_1)$, where C_1 is a value much less than the maximum values of k_x^2 , k_y^2 . The value of C_1 is changed until the change of temperature goes to zero at the numerical boundaries and until the difference between the computed profiles of ΔT for two values of C_1 is within 1%¹³. Typical plots for the transverse thermo-capillary force for microbubbles at different positions are shown in Figure 4-2.



Figure 4-2: For $P_i = 100 \ mW$ and $R_0 = 100 \ \mu m$, plots of transverse thermo-capillary force profiles for $z = -1 \ mm$, $z = 0 \ mm$, $z = 1 \ mm$ separately. The values of z are with respect to the location of the focus in the liquid; $z = -2 \ mm$ is the location of the inner wall of the cuvette where the laser beam is incident.

Similarly, a typical plot for the longitudinal thermo-capillary force (on-axis) are shown in Figure 4-3. As mentioned earlier when solving Eq. (4-16), a similar method is followed as in the numerical solution of the transverse case: viz., replacing k_z^2 with k_z^2 + C_2 . In this case C_2 is changed until the maximum value of the longitudinal temperature profile is equal to the maximum value of the transverse temperature profile at the focus of the laser beam, i.e., z = 0 mm.



Figure 4-3: For $P_i = 100 \ mW$ and $R_0 = 100 \ \mu m$, the variation of the longitudinal thermo-capillary force vs. propagation distance around the focus.

The final solution for the thermo-capillary force can be assumed to be the vector sum of the transverse and longitudinal thermo-capillary forces. While the transverse 2D solution was appropriate for the case studied earlier (confined bubble in thin container)^{13,16}, our case demands the more exact 3D solution of the heat equation. As will be seen, the solution for the force in the 3D case is different from that found using the method above since in the theory above, the energy distribution is considered only in either the longitudinal or transverse directions.

4.7 3D Solution of Thermo-Capillary Force

Here, the thermo-capillary force is calculated directly by solving the 3D steady state heat equation by using the 3D Fourier transform method. The solution of Eq. (4-4) can be expressed symbolically as

$$\Delta T(x, y, z) = \Im_{x, y; z}^{-1} \{ \frac{1}{k_x^2 + k_y^2 + k_z^2} \Im_{x, y; z} [-\frac{2P_i' \alpha_l}{k \pi w^2(z)} \exp(-\frac{2(x^2 + y^2)}{w^2(z)} - \alpha_l(z + z_0))] \},$$
(4-17)

where $\Im_{x,y;z}$ is the Fourier transform operator in *x*, *y* and *z*, and $k_{x,y,z}$ are corresponding spatial frequencies. Once ΔT is known, its temperature gradient $\nabla(\Delta T)$ can be calculated to find the thermo-capillary force F_{TC} .

It is assumed that the position of laser beam focus is about $z_0 = 2 mm$ away from the entry wall of the cuvette, as well. The thermo-capillary force F_{TC} at various locations in the liquid can be calculated by using Eqs. (4-9) and (4-17). The laser power incident on the liquid filled cuvette is still assumed as $P_i = 100 mW$.

When solving Eq.(4-9), to avoid singularity $(k_x^2 + k_y^2 + k_z^2)$ should be replaced with $(k_x^2 + k_y^2 + k_z^2 + C)$, where C is a value typically less than the minimum nonzero values of k_x^2 , k_y^2 and k_z^2 . The value of C is changed until the difference between the computed profiles of ΔT for two values of C is within 1%¹³.

Simulation results for the transverse component of the temperature change ΔT for different values of *z* are shown in Figure 4-4.



Figure 4-4: For $P_i = 100 \ mW$, and beam focus at $z = 0 \ mm$ with $z_0 = 2 \ mm$, (a) variation of the temperature change in the transverse direction at beam focus; (b) variation of the temperature change in the transverse direction at different longitudinal positions; (c) corresponding to (a), transverse thermo-capillary force around focus for microbubbles of different radii. When x > 0, the transverse thermo-capillary force is negative which indicates the force is in the -x direction and points towards the beam focus. Similarly, when x > 0, the transverse thermo-capillary force is positive which indicates the force is in the +x direction and again points towards the beam focus. Typical radii of microbubbles are in accordance with estimated sizes using in-line holography^{25,43}.

Figure. 4-4(a) shows the plot of $\Delta T(x, 0; 0)$. As shown in Figure 4-4(b), for other values of z, the temperature profiles have smaller peaks due to decreased on-axis intensities; furthermore, the on-axis peak for, say, z = 2 mm is less than for z = -2 mm

due to absorption of the laser beam during propagation through the liquid. The corresponding plot of the transversal thermo-capillary force at z = 0 for microbubbles of different radii has been plotted in Figure 4-3(c) with the knowledge of the temperature change ΔT . The temperature change profiles and the corresponding thermo-capillary force are the same for *y* axis due to the cylindrical symmetry of the laser intensity profile in Eq. (4-6).

Simulation results for the longitudinal component of the temperature change $\Delta T(0, 0, z)$ is shown in Figure 4-5(a). The plot is asymmetric because of light absorption in the liquid. The calculated longitudinal thermo-capillary force results for microbubbles of different radii are shown in Figure 4-5(b).



Figure 4-5: For $P_i = 100 \ mW$, and beam focus at $z = 0 \ mm$ with $z_0 = 2 \ mm$, (a) variation of the longitudinal (on-axis) temperature change *vs.* propagation distance around focus; (b) the longitudinal (on-axis) thermo-capillary force *vs.* propagation distance around focus for microbubbles of different radii. When z > 0, the thermo-capillary force is negative which indicates the force is in the -z direction and points towards the beam focus. Similarly, when z < 0, the thermo-capillary force is positive which indicates the force is in the +z direction and again points towards the beam focus.

For the remainder of this thesis, the results derived by 3D Fourier Transform are

used.

4.8 Nature of Heat Transfer

The justification for assuming conduction as the predominant heat transfer mechanism is as follows. The Rayleigh number *Ra* is calculated to verify the heat transfer form. When the Rayleigh number is below a critical value (about 2000^{12}) for that fluid, heat transfer is primarily in the form of conduction; when it exceeds the critical value, heat transfer is primarily in the form of convection⁵⁹. For a volume of fluid of size *l* in all three dimensions, the Rayleigh number can be calculated by:

$$Ra = \frac{\rho_l \beta(\Delta T) l^3 g}{\eta \alpha},\tag{4-18}$$

where $\rho_l = 785 \ kg/m^3 \ {}^{60}$; $\beta = 0.00107/{}^{\circ}C$ is the thermal expansion coefficient⁶¹, *g* is the acceleration due to gravity, $\eta = 2.859 \times 10^{-3} \ kgm^{-1}s^{-1}$ is the viscosity of liquid⁶², and α is the thermal diffusivity. The temperature change is taken as the peak value at beam focus which is $6 \times 10^{-3} \ K$, according to the calculated results in Figure 4-5(a). Since the 3D cuvette has the inner volume $5 \ mm \times 12.5 \ mm \times 45 \ mm$, and the laser radius is comparatively small, *l* is taken as $5 \ mm$ in the calculation.

According to Eq. (4-2), the thermal diffusivity is $\alpha = \frac{k}{\rho_l c_p}$, where the specific heat capacity $c_p = 2.68 \times 10^3 J/(kg \cdot K)$ at 20 °C ~ 25 °C. Using these values in Eq. (4-18), it is clear that $Ra \ll 2000$, even if the average temperature change is taken as the peak temperature at the beam focus, and hence convection can be neglected, thereby justifying the use of conduction as the predominant heat transfer process. At the inner wall of cuvette, the average temperature change is much less than that of beam focus, thus the convection can be neglected. This calculation is consistent with the experimental results from Bazhenov *et al.*, ⁶³

4.9 Conclusion

In this Chapter, the method for calculating thermo-capillary force is introduced. The generated heat by the laser is related to the laser intensity. By solving the steady state heat equation, the temperature distribution is derived. Thermo-capillary force is proportional to the temperature gradient. First, the transverse and longitudinal components of thermo-capillary forces are calculated separately, and their peak values at beam focus are adjusted to be consistent. Then the thermo-capillary force is calculated by solving the 3D steady state heat equation directly with the 3D Fourier transform method. In the next Chapter, other forces acting on the microbubble will be analyzed in detail.

CHAPTER V

ANALYSIS OF FORCES ACTING ON MICROBUBBLES

5.1 Introduction

In this Chapter, a detailed analysis of all forces acting on the microbubble is presented. After the microbubble generation by nucleation and thermal cavitation, the microbubble's motion is determined by the forces acting on microbubbles.



Figure 5-1: (a) Force model when the microbubble is attached to the inner face; (b) force model when microbubble leaves the inner surface.

Referring to Figure 5-1, possible forces acting on the bubble include the buoyancy force F_B , the gravity G, the viscous force F_v , the surface tension force F_{SN} , the friction force F_F , the optical force F_o , and the thermo-capillary force F_{TC} . The last two of these forces, viz., optical force and thermo-capillary force, are due to the optical radiation. The thermos-capillary force has both longitudinal (horizontal) and transverse (e.g., vertical) components. When the microbubble is pushed to the cuvette wall against the beam

propagation direction after being generated and is attached to the cuvette inner surface, the surface tension and friction act on the microbubble. When the microbubble is traveling in the liquid, these two forces are not considered. The thermo-capillary force has been discussed earlier at length in Chapter IV. All other forces will be discussed systematically in this Chapter.

5.2 Buoyancy Force and Gravity

Let *V* denote the volume for the microbubble. For simplicity, the microbubble is assumed to be a sphere of radius R_0 with volume

$$V = \frac{4}{3}\pi R_0^{3}.$$
 (5-1)

Its size is determined by the power of the laser beam and exposure time⁴³. Once formed, the buoyancy force and gravity on the microbubble are given as

$$\boldsymbol{F}_{\boldsymbol{B}} = + \boldsymbol{V} \boldsymbol{g} \boldsymbol{\rho}_{l}, \tag{5-2}$$

$$\boldsymbol{G} = -m\boldsymbol{g} = -\rho_a \boldsymbol{V}\boldsymbol{g},\tag{5-3}$$

where $g = 9.8 m/s^2$ is the acceleration due to gravity, ρ_l and ρ_a are the densities of the liquid and the gas mixture, respectively, and *m* is the mass of the microbubble. The plus and minus sighs in Eqs. (5-2) and (5-3) indicate that the buoyancy and gravity, although both in the vertical direction, oppose each other.

Using $\rho_l = 785kg/m^3$, $\rho_a = 1.225kg/m^3$, it follows that $F_B = 32174.07R^3$, which is much larger than the force of gravity $G = 50.286R^3$. For future calculations, the force due to the force of gravity of the bubble can therefore be neglected.

5.3 Viscosity Force

The viscous force is the measure of a fluid's resistance to flow. The force is proportional to the rate at which the fluid velocity is changing in space; with the proportionality constant being the viscosity.

If the microbubble moves with a velocity v, the viscous force acting on the microbubble can be expressed using Stokes' theorem⁶⁴ as

$$\boldsymbol{F}_{\boldsymbol{\nu}} = -6\pi\eta R_0 \boldsymbol{\nu},\tag{5-4}$$

where η is the viscosity of the liquid. The negative sign indicates that the viscous force is opposite to the direction of motion of the bubble. Assuming $\eta = 2.859 \times 10^{-3} kgm^{-1}s^{-1}$, $F_{\nu} = 5.389 \times 10^{-2} R\nu$ (N).

5.4 Surface Tension Force and Friction Force

When the microbubble is attached to the inner surface, there is a surface tension force acting on the bubble which is normal to the surface. This force can be expressed as

$$F_{SN} = 2\pi R_s \sigma \sin\theta, \tag{5-5}$$

where R_s is the contact radius of the microbubble with the container wall, σ is the surface tension coefficient and the θ is the contact angle of the bubble as shown in Figure 5-1. The direction of surface tension force is in the -z direction. In previous work, albeit for a bubble contained in a thin container¹³, a typical value for the surface tension force, assuming $\sigma = 2.17 \times 10^{-2} N/m$, $\theta = 2^{\circ}$ and $R_s = R/3$, is $F_{SN} = 1.586 \times 10^{-3} R$ (N). Also in the previous work⁶⁵, the friction force was of relevance since the bubble was guided along the container wall. The friction force can be written in terms of the surface tension force as

$$F_F = \mu F_{SN},\tag{5-6}$$

where μ is the coefficient of friction (kinetic) between the inner face of the cuvette and the microbubble. A typical value for the coefficient of friction is $\mu = 0.003$.

5.5 Optical Force

Traditionally, the optical force is responsible for optical trapping and manipulation⁴⁶ of dielectric particles of relative permittivity ϵ_{r2} embedded in a medium of relative permittivity ϵ_{r1} . The optical force is proportional to the gradient of the optical field and the total polarizability α_V , which should be expressed using the Clausius-Mossotti equation⁶⁶ as:

$$\alpha_V = 3\epsilon_0 \epsilon_{r1} \left(\frac{\epsilon_{r2} - \epsilon_{r1}}{\epsilon_{r2} + 2\epsilon_{r1}} \right) V, \tag{5-7}$$

where ϵ_0 is the free space permittivity. The optical force is usually attractive since $\epsilon_{r2} > \epsilon_{r1}$. On the contrary, in our case, $\epsilon_{r1} = \epsilon_{rl} \equiv n_l^2 > 1$ is the relative permittivity of the liquid, while the relative permittivity ϵ_{rv} of the gas/vapor in the bubble, which can be assumed to approximately that of air, makes ($\epsilon_{r2} = \epsilon_{rv} \equiv n_g^2 \approx n_a^2 = 1$). It is easy to see that since $\epsilon_{r2} < \epsilon_{r1}$, the optical force is repulsive.

However, since the estimated microbubble size $(R_0 \sim 100 \ \mu m)$ from the experiment is much larger than the wavelength ($\lambda = 514.5 \ nm$), the optical force should be computed from ray optics 46,67 . The optical force on a particle in the surrounding medium of refractive index *n* can be expressed as

$$F_o = Q \,\frac{n^P}{c} \,, \tag{5-8}$$

where $\frac{nP}{c}$ is the is the incident momentum per second of a ray of power *P*, *c* is the velocity of light and *Q* is a dimensionless factor, which is defined as the trapping efficiency. The optical force has two components: a scattering force, which is always in the direction of light propagation (+*z*) and a gradient force, which is in the direction of the spatial gradient of the optical field. For spheres of diameter much larger than the wavelength λ , the gradient force dominates over the scattering force ⁶⁷.

By adopting the MATLAB[®] toolbox developed by Callegari *et al.*⁶⁸, the gradient trapping efficiency Q_g and the corresponding optical gradient force in the longitudinal (on-axis) direction can be computed, as shown in Figure 5-2 for a bubble size $R_0 \sim 100 \ \mu m$. As seen from Figure 5-2, the optical gradient force is repulsive, being negative for z < 0 and positive for z > 0. Since the scattering force is always in the direction of propagation of the laser beam (+z) ⁶⁷, it can slightly decrease the net optical force for z < 0, and slightly increase the net optical force for z > 0.



Figure 5-2: (a) the gradient trapping efficiency and (b) the longitudinal optical gradient force *vs.* propagation distance around focus for $R_0 \sim 100 \,\mu m$. When z > 0, the optical gradient force is positive which indicates the force is in the +z direction. Similarly, when z < 0, the optical gradient force is negative which indicates the force is in the -z direction (repulsive).

5.6 Conclusion

In previous work, a force model had been developed to analyze all the forces on a microbubble confined due to 2D trapping within a thin glass container¹³. All the possible forces acting on the microbubble such as thermo-capillary force, optical force, surface tension force, friction force, and buoyancy force were incorporated in the model.

In this Chapter, a detailed force analysis is again performed and the expressions for the forces on a microbubble in 3D cuvette are developed. It has been shown that gravity can be neglected because it is much less than buoyancy force. The typical values are shown for those forces. In the next Chapter, a detailed analysis of the thermo-capillary force and comparison with other forces acting on the microbubble are presented, along with the experiment results.

CHAPTER VI

EXPERIMENTAL RESULTS AND ANALYSIS

6.1 Introduction

In this Chapter, the essential theory of laser propagation is given to present the change for beam waist position and beam size. After implementing the experiment, far-field self-diffraction rings are observed and recorded to show the evolution from SPM to TB and the microbubble evolution. Finally, this Chapter illustrates the dynamics of the microbubble due to the thermo-capillary force before it starts to rise due to buoyancy.

6.2 Analysis of Gaussian Beam Propagation

The experimental setup is illustrated in Figure 2-1. By moving the microscope objective, the location of the beam focus in the liquid can be changed to study microbubble dynamics according to the different beam focus locations.

The focal spot size can be determined knowing the initial waist of the beam incident on the microscope objective. Assuming that the initial waist of the optical field is $w_{ini} = 0.3mm$. Referring to Figure 4-1, assume that the CW Gaussian beam with plane wavefronts is incident on the microscope objective, and M_1 is the *ABCD* matrix of the objective lens, which is simplified as an effective thin lens. After passing through the objective, the laser beam travels a l_1 distance in air, and M_2 is the *ABCD* matrix of this propagation. M_3 and M_5 are the *ABCD* matrices at the air-quartz and quartz-liquid boundaries, respectively. M_4 and M_6 are the *ABCD* matrices for laser propagation through the entry wall of the cuvette and in the liquid, respectively. Thus, the total *ABCD* matrix going from the entrance surface of the objective for the laser to an arbitrary distance l_3 inside the liquid is

$$\begin{split} M_{total} &= M_6 M_5 M_4 M_3 M_2 M_1 \\ &= \begin{pmatrix} 1 & l_3 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & n_q/n_l \end{pmatrix} \begin{pmatrix} 1 & l_2 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & n_a/n_q \end{pmatrix} \begin{pmatrix} 1 & l_1 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ -1/f & 1 \end{pmatrix} \\ &= \begin{pmatrix} 1 - \frac{1}{f} \Big[l_1 + \frac{n_a}{n_q} \Big(l_2 + l_3 \frac{n_q}{n_l} \Big) \Big] \quad l_1 + \frac{n_a}{n_q} \Big(l_2 + l_3 \frac{n_q}{n_l} \Big) \\ &- \frac{1}{f} \frac{n_a}{n_l} & \frac{n_a}{n_l} \end{pmatrix} \equiv \begin{pmatrix} A & B \\ C & D \end{pmatrix}. \end{split}$$
(6-1)

Therefore, the *q* parameter in the liquid at a distance l_3 from the $z = -z_0$ surface is $q = \frac{Aq_0+B}{Cq_0+D}$, where $q_0 = j \frac{\pi w_{ini}^2}{\lambda} \equiv j z_{Rini}$, which is purely imaginary, is the *q* parameter of the Gaussian beam with waist w_{ini} and with plane wavefronts at the entrance surface of the microscope objective.

When the beam focuses in the liquid after traveling a distance $l_3 = z_0$, the q is purely imaginary. Hence,

$$Im(q) = \frac{z_{Rini}n_l}{n_a\left(1 + \frac{z_{Rini}^2}{f^2}\right)} \equiv z_R = \frac{\pi n_l w_0^2}{\lambda}, \qquad (6-2)$$

where w_0 is the beam waist in the liquid. It is easily seen that w_0 does not depend on the position of beam focus. The location of the focus, $l_3 = z_0$, can be calculated by setting the real part of q to zero:

$$Re(q) = \frac{B\left(1 + \frac{z_0^2}{f^2}\right) - \frac{z_0^2}{f}}{\frac{n_a}{n_l} \left(1 + \frac{z_0^2}{f^2}\right)} = 0,$$
(6-3)

where *B* is defined in Eq. (6-1). Eq.(6-3) can be used to determine the location l_1 of the microscope objective from the cuvette to achieve the beam focus at a desired distance z_0 from the entry wall of the cuvette. For instance, for $l_3 = z_0 = 2 mm$, it follows that $l_1 = 6.87 mm$. However, since the microscope objective is simplified as a thin lens, l_1 represents the distance between the effective lens and cuvette, and the actual distance from the last surface of the microscope objective and cuvette is slightly different.

Based on the *ABCD* matrix method for Gaussian beam, the laser beam waist in the liquid is $w_0 = 4.91 \,\mu m$ and does not depend on the position of beam focus. The Rayleigh range for the focused beam in the liquid is $z_R = 202.9 \,\mu m$, which is much less than the propagation distance (5 mm) of the light through the liquid. Therefore, in the theory, propagation of the beam through the liquid incorporating diffraction is used. Also, based on the Rayleigh range, the observation plane can be considered to be in the far field.

6.3 Far-Field Patterns

As mentioned in Chapter I, when an intense beam of light impinges into an absorbing medium, a spatial temperature distribution is generated with laser incidence, which in turn induces a change in the refractive index which is proportional to the local light intensity profile. This is responsible for SPM^{25} of a (focused) laser beam as it propagates through the medium. The self-induced "thermal distortion" of the medium is TB^{18} . Since all these effects are responsible for the refractive index change, the propagation of the laser beam is substantially affected.

As shown in Figure 6-1, the self-diffraction pattern can be observed on the observation screen which is positioned about $50 \ cm$ away from the cuvette. When the

laser is just turned on, the far field pattern is like Figure 6-1(a), and once SPM forms after a few seconds, the pattern in Figure 6-1 (b) is shown. With the increase of laser power, the pattern will change into Figure 6-1 (c) which indicates TB and a more intense phase change. Due to the force of gravity and convection effects, the far-field self-diffraction rings are not concentric. The far-field patterns due to SPM and TB are similar to observations by Abeywickrema *et al.*⁴³



Figure 6-1: Far-field self-diffraction patterns of (a) initial state, (b) SPM, (c) TB.

The far-field patterns of the incident focused laser beam during SPM, TB, spatiotemporal evolution of one bubble starting from its creation, followed by its motion, till the moment it floats up due to buoyancy, is shown in Figure 6-2.



Figure 6-2: Time-lapse records of far-field patterns of focused laser beam showing spatiotemporal evolution of SPM and TB along with the effect of generated microbubbles. The clear concentric ring patterns (like the first and last pattern indicate) are from SPM, the second pattern shows TB, the others are from the effect of generated microbubble and its motion till it floats up (and out of the path of the laser beam) due to buoyancy.

However, according to the simulation results of temperature distribution caused by the laser, the peak temperature change in the liquid is not high enough to reach to the evaporation temperature for isopropanol alcohol. Thus, as argued earlier, nucleation and optical cavitation is the major reason for microbubble generation instead of thermal blooming.

6.4 Dynamics of Microbubbles

After generation, the microbubble is trapped by the focused beam and scatters the laser radiation as shown in Figure 6-3(a), pictured with the camera shown in Figure 2-1 on the side of the cuvette. A simple explanation for the downward scattering of the light from the microbubble can be understood through a simple "ray" diagram in Figure 6-3(b), which shows that the direction of scattering is due to the fact that the center of the bubble is located slightly higher than the focus spot due to buoyancy.



Figure 6-3: (a) Picture of microbubble scattering the laser radiation. (b) A simple "ray" diagram showing the downward deflection of the laser light.

Subsequent dynamics of the microbubble can be classified into three types, based on the position of laser beam focus z_0 :

Type A: This type of movement happens when the beam focus region approximately in the middle region of the cuvette ($z_0 = 1 \sim 2 mm$). The microbubble is initially pushed away from the focus to the inner surface of the cuvette wall, *against* the direction of beam propagation. Then, it persists for a few seconds. At this time, it may grow by itself due to the evaporation of molecules at the gas-liquid interface⁵⁰ and/or wait for another microbubble to join with it. After reaching a specific volume, it is attracted to the beam focus but finally floats up to the surface. The spatio-temporal evolution, pictured by the camera in Figure 2-1, is shown in Figure 6-4.



Figure 6-4: Left to right: 8 photographs taken at (increasing) time sequence, showing spatiotemporal evolution (type A) of the microbubble. The total elapsed time between the first and last photographs is approximately 14 secs. Light propagates from right to left in these photographs. The distance covered by the light shows the distance between the walls. For convenience, the entry wall is shown with white lines.

A tentative analysis based on the thermo-capillary force and other forces, viz. optical force (especially the gradient force) can be used for explaining the motions of microbubble after it has been generated. Around the location of the formation of the microbubble, the repulsive optical gradient force can be higher than the attractive thermocapillary force for the size of the bubble, as shown in Figure 6-5 (a) for a bubble radius of $R_0 = 50 \ \mu m$. As the bubble travels to the wall of the cuvette $z = -z_0$, its volume increases, and it is attracted to the beam focus because the attractive longitudinal thermocapillary force can now overcome the repulsive optical gradient force, as shown in Figure 6-5 (b). Additionally, the optical scattering force is always along the +z direction, and it also acts as an attractive force when z < 0. But the microbubble finally floats up to the surface due to the dominating buoyancy force which is larger than the attractive transverse thermo-capillary force. The longitudinal position where it floats upward is approximately determined by the balance of transverse thermo-capillary force and the buoyancy force.



Figure 6-5: The longitudinal on-axis optical gradient force and the thermo-capillary force *vs.* propagation distance around focus for microbubbles of the radius (a) $R_0 = 50 \ \mu m$ and (b) $R_0 = 100 \ \mu m$.

Type B: For $z_0 > 2 mm$ the microbubble is not attracted to the focus after attaching to the inner surface, instead, it floats upwards with a small angle from the vertical direction. The small angle indicates the existence of longitudinal thermo-capillary force.

Type C: For $z_0 < 1 \text{ mm}$, the microbubble keeps oscillating between the beam focus and the laser entry face of the cuvette and growing in volume, and finally floats up. This can be explained through the competition between the longitudinal thermo-capillary force and the optical force acting on the microbubble at the same time when the

microbubble is around the beam focus, in accordance with the simulation results of longitudinal thermo-capillary force and the optical gradient force. The oscillation process is shown in video 1.



Video 1: The oscillation of microbubble when laser focus is close to the cuvette wall $(z_0 < 1 \text{ } mm)$ at the entry face of the cuvette. (<u>https://youtu.be/dCF2ZF6PKdE</u>)

In all cases, the microbubble behavior shows the presence of transverse thermocapillary force because the microbubbles do not float to the surface immediately after being generated.

6.5 Conclusion

Experimental results showing the dynamics of the bubble for different positions of the focus of the laser beam are discussed. The far-field patterns show the results of SPM and TB followed by the effect of light scattering by the microbubble. For a range of distances over which the beam is focused, it is shown that the bubble is first pushed away from the focus due to the repulsive optical force, grows in size at the wall of the cuvette, then is attracted towards the focus due to the attractive longitudinal thermo-capillary force, and finally floats up when buoyancy overcomes the transverse thermo-capillary force. Dynamics of bubbles for other locations of the laser focus within the cuvette are also discussed.

CHAPTER VII

CONCLUSION AND FUTURE WORK

7.1 Conclusion

In this thesis, the observation of behaviors on thermally induced microbubbles in the absorbing liquid which is a mixture of colloidal red dye and 91% isopropanol alcohol with a CW Gaussian profile laser beam is analyzed and the exact solution for thermocapillary force has been determined. The experiments results show the existence of thermo-capillary force.

In Chapter II, the experimental setup for generating and observing the microbubble is explained. Chapter III introduces the theory related to the generation of microbubbles in absorbing liquid with a laser beam. The nanoscale red dye particles serve as nucleation sites in the 91% isopropanol alcohol and help to absorb the heat which is generated by the incident green CW laser. When the laser power reaches the threshold power which makes the temperature around the red dye particles exceeds the evaporation temperature, thermal cavitation occurs, associated with gas bubble formation. It has been verified by experiments that the phase change and temperature change by thermal blooming are not enough to cause microbubble formation.

In Chapter IV, the steady state heat equation, which is of Poisson form, is used to derive typical transverse and longitudinal distributions of the temperature profiles, along with the transverse and longitudinal components of the thermo-capillary force for different bubble sizes. In this work, the thermo-capillary force on a microbubble is determined by first separately solving the transverse and longitudinal heat equations, and then by directly solving the 3D heat equation by 3D Fourier transform methods.

In Chapter V, other forces (viz., the buoyancy force, gravity, viscous force, friction, optical force, and surface tension force) acting on the microbubble and their estimated values are presented in detail.

In Chapter VI, the essential theory of laser propagation is explained. Far-field patterns of focused laser beam showing spatiotemporal evolution of regular self-phase modulation and thermal blooming affected by microbubbles are presented. Finally, the dynamics of bubbles for other locations of the laser focus within the cuvette are also discussed based on the thermo-capillary force and optical force competition.

This work was presented at the SPIE Annual Meeting in San Diego in August 2018⁶⁹ and the SPIE Photonics West in San Francisco in February 2019⁷⁰. A journal paper has been submitted to Optical Engineering⁷¹.

7.2 Future Work

The microbubble radius is only approximately estimated in this work; however, a more accurate imaging system should be used to characterize the microbubble size and the distance it moves. For an accurate estimate of the time intervals for microbubble travel, a high resolution/high-speed video should be recorded to get the precise time, and hence estimate the average velocity.

The microbubble size keeps getting larger with the continuous exposure under laser even though the time interval is short. An attenuator has been used in the experiment to decrease the laser power into the liquid after bubble formation; however, the balance of buoyancy force and transversal thermo-capillary force for controllable optical trapping could not be realized. This will be pursued in the future.

Finally, as for applications of microbubbles, nanoparticles can be introduced in the medium to analyze the trapping and manipulation of nanoparticles agglomerated around the microbubble. The temperature profile can be verified using a thermal camera experimentally and using a model building software like COMSOL[®] theoretically.

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APPENDIX

MATLAB® Codes Used in This Work

A1. 2D Solution of Thermo-capillary Force

A1.1 Transverse thermo-capillary force

```
clc
clear all;
lambda = 514.5e-9;%wavelength
R = 100e-6; % estimated bubble size
w0 = 4.91e-6; %beam waist
n iso = 1.3772;
beta=280;%/m, the absorption coefficient of liquid
k = 0.136;%
P0=0.1; %power on outter surface, W
P0=P0*0.9648;%P0 prime at the inner surface
Zr=pi*n iso*w0^2/lambda; %rayleigh range
z=1e-3; %axial position of bubbules
Wz=w0*sqrt(1+(z/Zr)^2); %beam waist at postion z
L = 10 * w0;
N=1024;
dx = L/N;
dy = dx;
x = -0.5*L:dx:0.5*L-dx;%integral range
y = x; %integral range
kx = (2*pi)*(-1/(2*dx):(1/(N*dx)):1/(2*dx)-(1/(N*dx)));
ky = kx;
[X,Y]=meshqrid(x,y);
[KX,KY]=meshgrid(kx,ky);
A=2*P0*exp(-beta*(z+2e-3))/pi/Wz^2;
G=A*exp(-2*(X.^2+Y.^2)/Wz^2);%intensity
GF = fftshift(fft2(beta/k.*G));
C=1e8;
g = 1./(KX.^{2+KY}.^{2+C});
TF = GF.*g;
TT= fftshift(TF);
T = ifft2(TT, 'symmetric');%temperature
% figure;mesh(X,Y,G); %intensity
% figure;mesh(X,Y,T); %temperature
% max(T,[],'all')
```

```
T0=T(514,:);
plot(y,T0,'linewidth',2);
grid on
set(gca, 'FontSize', 14);
xlabel('x[\mum]','fontsize',14)
ylabel('transversal temperature change?T(K)', 'fontsize', 14)
% hold on
sigmaT SI = -0.0789e-3; %surface tension coefficient of temprature
gradT0= gradient(real(T0),dy);
FT= -2*(pi*R*R)*sigmaT SI*gradT0;
figure
% plot(y*1e6,FT,'linewidth',2);
% plot(y*1e6,FT,':','linewidth',2);
plot(y*1e6,FT,'--','linewidth',2);
grid on
set(gca, 'FontSize', 14);
xlabel('x[\mum]', 'fontsize', 14)
ylabel('Transverse F {TC} (N)', 'fontsize',14)
legend('z=-1mm', 'z=0mm', 'z=1mm', 'Location', 'NorthEast')
hold on
```

A1.2 Longitudinal thermo-capillary force

```
clc
clear
lambda = 514.5e-9;
R = 100e-6;%estimated bubble size
w0 = 5.91e-6;%beam waist
n iso = 1.3772;
beta=280 ;%/m
k = 0.136;
n \text{ quartz} = 1.4585;
P0=0.1;%outter surface,W
P0=P0*0.9648;%P0 prime at the inner surface
Zr=pi*n iso*w0^2/lambda;
L = 5e-3;
N=512;
dz = L/N;
z = -1/2*L:dz:1/2*L-dz;%integral range
kz = (2*pi/dz)*(-1/2:(1/N):1/2-(1/N));
A=2*P0*Zr^2/pi/w0^2;
G=A./(Zr^2+z.^2).*exp(-beta*(z+2e-3));%intensity with decay
GF = fftshift(fft(beta/k*G));
C = 6.5e9;
g = 1./(kz.^{2+C});
TF = GF.*q;
TT=fftshift(TF);
T = ifft(TT);%temperature
figure;plot(z,T);
sigmaT_SI = -0.0789e-3;
gradT = gradient(T, dz);
FT = -2*(pi*R*R)*sigmaT SI*gradT;grid on
figure;plot(z*1e3,FT,'linewidth',2);grid on
set(gca, 'FontSize',14);
xlabel('x[mm]','fontsize',14)
ylabel('Longitudinal F_{TC} (N)', 'fontsize',14)
```

A2. 3D Solution of Thermo-capillary Force

A2.1 Transverse component

MaxT=max(T,[],'all');

```
clc
clear
%----- parameters
lambda = 514.5e-9;
w00 = 0.3e-3;% beam waist size of laser
f = 9e-3;% focal length of lens
R = 200e-6;%estimated bubble size
w0 = (lambda*f)/(pi*w00); %beam waist after M.O. in air
n iso = 1.3772;
alpha 1=280 ;%/m, absorption of liquid
k = 0.136;% conductivity
n quartz = 1.4585;
P0=0.1;%outter surface,W
P0=P0*0.9648;%P0 prime at the inner surface
Zr=pi*n iso*w0^2/lambda;
%-----z direction. spatical frequency
L = 4e-3;
N = 256;
dz = L/N;
z = -1/2*L:dz:1/2*L-dz;%integral range
kz = (2*pi/dz)*(-1/2:(1/N):1/2-(1/N));
%x and y direction, spacial frequency
Lp=10*w0;
dx = Lp/N;
dy = dx;
x = -0.5*Lp:dx:0.5*Lp-dx;%integral range
y = x;%integral range
kx = (2*pi)*(-1/(2*dx):(1/(N*dx)):1/(2*dx)-(1/(N*dx)));
ky = kx;
%-----3D vector: spatical frequency and position
[X,Y,Z]=meshgrid(x,y,z);
[KX,KY,KZ]=meshgrid(kx,ky,kz);
z0=2e-3;
%-----laser intensity
Wz=w0*sqrt(1+(Z./Zr).^2); %beam waist at postion z
A=2*P0*exp(-alpha l*(Z+z0))/pi./Wz.^2;
G=A.*exp(-2*(X.^2+Y.^2)./Wz.^2);%intensity with decay
GF = (fftn(alpha l/k.*G));
C=5e4;
q = 1./(KX.^{2+KY}.^{2+KZ}.^{2+C});
TF = GF.*q;
T = ifftn(TF);%temperature
```

```
sigmaT SI = -0.0789e-3; %surface tension coefficient of temprature,
N/(m*K)
% %%%-----transverse profile
% figure
% T01= T (:,128,1); %inner face r-temperature (z=-2mm)
% plot(x*1e6, real(T01), 'linewidth', 2);
% grid on
% set(gca, 'FontSize', 14);
% xlabel('x[\mum]')
% ylabel('Transverse temperature change ?T(K)')
2
% hold on
% T02= T(:,128,64); %z=-1mm
% plot(x*1e6, real(T02), ':', 'linewidth', 2);
2
% hold on
% T04= T(:,128,192);%z=1mm
% plot(x*1e6, real(T04), '-.', 'linewidth', 2);
8
% hold on
% T05= T(:,128,256);%z=2mm
% plot(x*1e6, real(T05), '--', 'linewidth', 2);
% legend('z=-2mm','z=-1mm','z=1mm','z=2mm','Location','NorthEast');
T03= T(:,128,128);%focal point r-T, z=0
% %GO= G(:,128,128);
% %figure
% %yyaxis left
% plot(x*1e6, real(T03), 'linewidth', 2);
% grid on
% set(gca, 'FontSize', 16);
% xlabel('x[\mum]')
% ylabel('Transverse temperature change ?T(K)')
% %yyaxis right
% %plot(x,real(G0),'linewidth',2)
% %ylabel('Beam intensity(W/m^2)')
%-----temperature gradient and TC force
gradT03= gradient(real(T03),dz);
FT = -2*(pi*R*R)*sigmaT SI*gradT03;
% figure
% plot(x*1e6,FT,'linewidth',2); %R=50um
% plot(x*1e6,FT,':','linewidth',2); %R=100um
% plot(x*1e6,FT,'-.','linewidth',2); %R=150um
plot(x*1e6,FT,'--','linewidth',2); %R=200um
grid on
set(gca, 'FontSize', 14);
xlabel('x[\mum]','fontsize',14)
ylabel('Transversal F_{TC} (N)', 'fontsize',14)
legend('R=50\mum', 'R=100\mum', 'R=150\mum', 'R=200\mum', 'Location', 'North
East')
hold on
```

A2.2 Longitudinal component

```
%GF = fftshift(fftn(beta./k.*G));
GF = fftn(alpha l./k.*G);
C=2e5;
g = 1./(KX.^{2+KY}.^{2+KZ}.^{2+C});
TF = GF.*q;
%TT = fftshift(TF);
T = ifftn(TF);%temperature
% GO= G(128,128,:);
% G0= real(squeeze(G0));
% GT= gradient(G0,dz);
% plot(z,GT)
%-----Axial temperature profile
TO= T(128,128,:);
T0= real(squeeze(T0));
% figure
% plot(z*1e3,T0,'linewidth',2)
% grid on
% set(gca, 'FontSize',16);
% xlabel('z[mm]','fontsize',16)
% ylabel('Longitudinal temperature change ?T(K)','fontsize',16)
%-----Axial temperature gradient and TC force
sigmaT SI = -0.0789e-3; %surface tension coefficient of temprature
gradTZ= gradient(T0,dz);
FT = -2*(pi*R*R)*sigmaT SI*gradTZ;
% figure
plot(z*1e3,FT,'linewidth',2) %R=50
% plot(z*1e3,FT,':','linewidth',2); %R=100um
% plot(z*1e3,FT,'-.','linewidth',2); %R=150um
% plot(z*1e3,FT,'--','linewidth',2); %R=200um
hold on
zz = [-20:0.2:20] *R;
OF = qz g(3,:)*nm*power/vc;
plot(zz*1e3,OF,':','linewidth',2)
grid on
set(gca, 'FontSize', 16);
xlabel('z[mm]','fontsize',16)
ylabel('Longitudinal F {TC}, F {og} (N)', 'fontsize', 16)
 legend('F {TC}','F {og}','Location','SouthWest')
% hold on
```