SURFACE PLASMON-BASED MANIPULATION OF MICROPARTICLES AND DYNAMICS STUDY

A Thesis in Engineering Science

by

Wei Yan

© 2008 Wei Yan

Submitted in Partial Fulfillment of the Requirements for the Degree of

Master of Science

August 2008
The thesis of Wei Yan was reviewed and approved* by the following:

Tony Jun Huang  
James Henderson  Assistant Professor of Engineering Science and Mechanics  
Thesis Advisor

Bernhard R. Tittmann  
Schell Professor of Engineering Science and Mechanics

Sulin Zhang  
Assistant Professor of Engineering Science and Mechanics

Judith Todd  
Professor of Engineering Science and Mechanics  
P. B. Breneman Department Head of Engineer Science and Mechanics

*Signatures are on file in the Graduate School
The manipulation of micrometer or nanometer scale objects based on light-matter interaction has been a hot topic ever since Ashkin proposed in 1986 the ‘optical tweezers’, which realized optical trapping and manipulation of polystyrene latex micro-particles through focused laser beams. Notably, Kawata et al. manipulated micrometer scale spheres near the surface of a prism based on radiation pressure generated by total internal reflection-induced evanescent field. Righini et al. introduced “surface plasmon tweezers” by trapping single microparticles on patterned gold structures under surface plasmon excitation. There remains much work to be done, however, in areas including patterning techniques and dynamics study of manipulated objects.

The disclosed thesis contributes to the breadth of knowledge on optical manipulation. Key developments in the field were first reviewed. From this background ‘surface plasmon tweezers’ were devised to trap and arrange polystyrene beads. The aggregation of the beads was driven by the combination of force and thermal gradient, and the bulk migration of said beads by radiation pressure or scattering force along the light propagation direction. The dynamics of the beads were also found to be subject to thermal convection. Lastly, a tunable, surface plasmon-based patterning technique was proposed.
# TABLE OF CONTENTS

LIST OF FIGURES ..................................................................................................................... v

ACKNOWLEDGEMENTS ........................................................................................................... vii

Chapter 1  Introduction .............................................................................................................. 1

References.................................................................................................................................. 5

Chapter 2  Literature Review ................................................................................................... 7

References.................................................................................................................................. 12

Chapter 3  Surface Plasmon Tweezers System: Construction and Preliminary Tests ............... 14

3.1 Principles of surface plasmons ......................................................................................... 14
3.2 Optical forces in a surface plasmon excitation field ....................................................... 16
3.3 Surface plasmon tweezers system configuration ............................................................... 19
    a) Refractive index-matched prism-sample system ......................................................... 19
    b) Laser and incident angle tuning system ..................................................................... 20
    c) Microscopic CCD imaging system ........................................................................... 21
3.4 Surface plasmon tweezers preliminary experiments ....................................................... 21

References.................................................................................................................................. 26

Chapter 4  Dynamics Study of Microbeads in Surface Plasmon Manipulation ....................... 28

4.1 Microbeads velocity under different laser power ......................................................... 28
4.2 Single bead velocity calculation in three regimes ......................................................... 30
4.3 Thermal effects in surface plasmon excitation ............................................................... 32

References.................................................................................................................................. 39

Chapter 5  Future Work .......................................................................................................... 40

5.1 Dynamics study in surface plasmon manipulation of microbeads with different sizes .......... 40
5.2 Optimization for surface plasmon manipulation force study ......................................... 41
5.3 Tunable plasmonic patterning by controlling incident light ............................................ 42

References.................................................................................................................................. 45

Appendix  Nontechnical Abstract ........................................................................................... 46
LIST OF FIGURES

Figure 2-1: The principle of optical tweezers (courtesy of Reference 7). A strongly focused beam of light was used to trap objects. Intensity gradients in the converging beam drew small objects toward the focus while the radiation pressure of the beam drove them along the optical axis. Under conditions where the gradient force dominates, a particle can be trapped near the focal point [1]. ................................................................. 7

Figure 2-2: A schematic of particle movement in evanescent field produced by collimated laser illumination on a prism with incident angle larger than critical angle (courtesy of Reference 8)................................................................. 9

Figure 2-3: a: Schematic diagram of optical configuration of surface plasmon-based tweezers; b,c: Arrangement of polystyrene microbeads at a homogeneous (b) and a patterned (c) gold-water interface after laser illumination at the surface plasmon resonance (courtesy of Reference 14). ........ 10

Figure 2-4: Plots of (A) beads velocity, (B) reflectivity measurement and (C) trapping probability as a function of incident (courtesy of Reference 15). .......... 11

Figure 3-1: Principles of surface plasmons ........................................................................................................... 15

Figure 3-2: Illustration of optical forces on a particle in an evanescent field under total internal reflection .......................................................................................................................... 17

Figure 3-3: (a) Force vector and (b) module (circle), x component (squares) and z component (triangles) of the force versus distance form gold surface for a 2.0 μm bead. (c) Force on beads of different sizes at 500 nm from surface (courtesy of Reference 6)............................................................................................................ 18

Figure 3-4: A schematic of surface plasmon tweezers system configuration ........ 19

Figure 3-5: Plot of reflected laser intensity from 40 nm gold film versus incident angles. ........................................................................................................... 21

Figure 3-6: One of the earliest observation of surface plasmon trapping phenomenon on 40 nm thick gold film. The radiate stripes on the gold surface is the crinkling and peeling of gold film caused by the instability of gold in aqueous environment ........................................................................................................ 22

Figure 3-7: 2D and 3D plots of intensity profile of incident Gaussian laser beam. .... 23

Figure 3-8: Consecutive images recorded with interval Δt = 2 min in a surface plasmon manipulation experiment. Laser power was 500 mW. Dark cross
indicates the approximate position of beam center. Arrow shows the direction of wave vector $k$. .................................................................................................................24

Figure 4-1: Plot of instant velocities of beads 550 μm from beam center as laser power increased from 0 to 1800 mW. .................................................................................................................29

Figure 4-2: ImageJ interface for microbeads tracking and velocity calculation........30

Figure 4-3: Single bead velocity tracking under laser power 500, 1000 and 1500 mW. ........................................................................................................................................................................31

Figure 4-4: Illustration of thermal effect in surface plasmon excitation. .................33

Figure 4-5: Fast movement of beads group driven by convection flow towards beam center. Interval between two frames is less than 1 s. .................................34

Figure 4-6: Images of a) end of 40 s laser illumination with power 1000 mW, b) about 1 min after the illumination ended.................................................................35

Figure 4-7: Images of the beginning of laser illumination under different power. The disappearing aggregation is due to dramatically increasing convection velocity that surpasses surface plasmon-based optical forces. .............................36

Figure 4-8: Beads driven from a 1000 mW laser beam center position (dark cross) to a higher plane above the gold surface, showing the convection effect. ..........37

Figure 5-1: A schematic of incident light manipulation system for tunable plasmonic trapping on a homogeneous gold film.........................................................43
ACKNOWLEDGEMENTS

I thank Professor Tony Jun Huang for serving as my thesis advisor, as well as Professors Bernhard Tittmann and Sulin Zhang for essential discussion. I also thank Dr. Vincent K.S. Hsiao and Yuebing Zheng for their tremendous help during my master’s study.

I dedicate this thesis to my parents and grandparents. I am grateful to them for nurturing me and for supporting my pursuit of studies.
Chapter 1

Introduction

As early as in 17th century, Johannes Kepler was intrigued by comet tails which always point towards the opposite direction as to the sun. Some speculation has brought up a concept that the sun is radiating a certain kind of force on the orbiting comets thus shapes the “pointing away” tails. In 1873, James Maxwell theoretically proved that light can exert force on matter [1]. While almost a century later, Arthur Ashkin reported in 1970 that micrometer scale particles can be accelerated and trapped in stable optical potential wells under the force of radiation pressure from two opposing equal Gaussian laser beams [2].

Ever since the experimental work by Ashkin was published, optical manipulation of objects with various dimensions has been under extensive research for nearly the last four decades. As the pioneer in exploring this exciting field, Ashkin demonstrated in 1986 the first “optical tweezers” that realized optical trapping and manipulation of polystyrene latex micro-particles by using continuous laser beams [3]. Since then the performance and applications of optical tweezers have been greatly improved and expanded, making optical tweezers a powerful technique in non-invasive manipulation of micrometer scale objects, especially in trapping, organizing and tracking various biological molecules [4]. Conventional optical tweezers requires high optical intensity hence strong gradient force in optical trapping, which is realized by tightly focusing light through high numeric aperture (NA) microscope objectives. This requirement not only
limits the further integration of optical tweezers into compact platforms, e.g. lab-on-chip
devices, due to necessary bulk optics, the highly focused light is likely to cause undesired
damage to trapped targets especially in biological studies.

To overcome the abovementioned limitation of optical tweezers, in 1992, Kawata
_etal._ reported in their work [5] of using total internal reflection (TIR)-induced evanescent
field to move micrometer scale polystyrene latex spheres and glass spheres near the
surface of a prism due to radiation pressure. Since evanescent wave is generated by TIR
in a surface parallel to the interface with small penetration depth of around several
hundreds nanometers, optical manipulation of sub-micrometer scale objects becomes
possible by taking advantage of the optical intensity gradient and radiation pressure
produced during evanescent wave generation [6-8].

Recently, there has been increasing interest in integrating surface plasmon field
into evanescent field-based optical manipulation. The surface plasmon near metal-
dielectric interface can be collectively oscillated when in resonance with the
electromagnetic filed of incident light. It has been shown that the forces induced by
surface plasmon on a single microscopic object near a homogeneous metal are almost
two orders of magnitude stronger than corresponding photonic forces [9]. While surface
plasmon field enables the enhanced magnitude of optical forces, it also renders promise
in nanometer scale optical manipulation [10, 11]. The work by Garcés-Chávez _etal._ [12]
has shown that a homogeneous metal surface illuminated by an unfocused laser beam
leads to a homogeneous in-plane optical potential, and later collaborators proposed that
the local trapping requires additional confinement in the surface plane which can be
achieved by patterning the metal surface. In 2007, Righini _etal._ reported a surface
plasmon-based optical tweezers and the trapping of single micrometer scale dielectric beads on predefined gold pattern [13]. This configuration provides the advantageous possibility of creating a large array of spots from an unfocused incident laser beam thus opening the field of sub-wavelength patterned optical trapping based on surface plasmon.

A year later, the same group brought up the new term “surface plasmon optical tweezers” in their report, stating the realization of tunable optical manipulation of micro-particles in the femtonewton range [14]. Using the technique photonic force microscopy, they assessed the surface plasmon forces in the range of a few tens of femtonewtons, and by adjusting the laser illumination parameters, selective trapping and tunable manipulation of micro-particles can be readily achieved. The pioneering work above has opened new perspectives for future optically driven lab-on-a-chip device.

Despite the original theoretical study and accurate metal structure engineering, most of the abovementioned research has been focused on the realization of stable trapping of targets, while relatively less effort has been devoted to the dynamics study of targets under surface plasmon forces generated at metal-dielectric interface. In this thesis work, we have both qualitatively and quantitatively studied the dynamics of micrometer scale polystyrene beads driven by surface plasmon forces generated at gold thin film and water interface as a result of unfocused laser beam illumination. First we built the platform for surface plasmon excitation with conventional Kretschmann configuration and realized effective trapping of 10.8 µm diameter polystyrene beads with distinctive results revealing the effect by both surface plasmon-based gradient force and scattering force. In order to verify that the existence of surface plasmon constitutes the main contribution to microbeads trapping, we then carried out a set of control experiments
under identical conditions with the only difference in that bare glass substrate was used instead of gold-coated glass substrate. The weak motion of microbeads proved that surface plasmon forces significantly facilitate the movement of targets compared to evanescent field without the enhancement by surface plasmon excitation. The dynamics study of microbeads under surface plasmon excitation was established by combining the adjustment of illumination parameters (laser power) and several groups of velocity calculation with theoretical analysis of microbeads movement based on recorded microscopic videos. Thermal effects including thermophoresis and convection are also considered in the analysis of microbeads dynamics.

In the following chapters, we will first review related literatures in the field of surface plasmon-based optical tweezers, and then introduce the system established in our lab for surface plasmon excitation, followed by the presentation of results obtained in effective surface plasmon-based optical trapping, and finally we report the analysis of microbeads dynamics under both surface plasmon forces and thermal effects.
References


Chapter 2

Literature Review

Pioneering the field of laser-based optical trapping in the early 1970s, Arthur Ashkin demonstrated that optical forces could displace and levitate micrometer scale dielectric particles in both air and water [2], and with coworkers at AT&T Bell Laboratories, he developed a stable, three-dimensional trap by using two counter-propagating laser beams [3]. Their work published in 1986 [4] has brought the new term “optical tweezers” to optical manipulation field, and the following phenomenal work has

Figure 2-1: The principle of optical tweezers (courtesy of Reference 7). A strongly focused beam of light was used to trap objects. Intensity gradients in the converging beam drew small objects toward the focus while the radiation pressure of the beam drove them along the optical axis. Under conditions where the gradient force dominates, a particle can be trapped near the focal point [1].
stimulated tremendous interests in this field, including cooling and trapping of neutral atoms [5] and manipulating live bacteria and viruses [6, 7]. The basic principle of optical tweezers is shown in Figure 2-1 (courtesy of Reference 7). It uses forces produced by a strongly focused light beam to trap small objects which develop an electric dipole moment due to the light’s electric field, and it is drawn up intensity gradients in the electric field toward the focus. These optical gradient forces compete with radiation pressure generated from the momentum of the photons in the beam which blow particles along the optical axis. Stable trapping is possible when the axial gradient force dominates, and is achieved when the beam diverges rapidly enough away from the focal point, and this requirement entails the use microscope objectives with high numeric aperture[1].

To circumvent the limitation of conventional optical tweezers which require bulk optical system and utilize high power laser, in 1992, Kawata et al. proposed a novel optical manipulation mechanism which employs the evanescent field produced by illuminating a high refractive index prism with an incident angle larger than the critical angle to drive particles with diameters of 1-27 µm [8]. This movement is considered as a result of that the transverse exponential decay of the evanescent field intensity into water attracts the bead towards the interface while the scattering force pushes it along the in-plane projection of the incident k-vector [9], as shown in Figure 2-2 (courtesy of Reference 8). This configuration adopts the total internal reflection of a single collimated laser beam instead of a focused propagating one, largely reducing the laser power requirement compared to that in previous optical trapping by optical tweezers, thus encourages further investigation of stable optical trapping based on evanescent field. Cizmar et al. demonstrated the trapping of sub-micrometer scale dielectric objects in
evanescent standing wave pattern induced by the interference of two counter-propagating beam, both of which were totally reflected at a glass/water interface [10].

Figure 2-2: A schematic of particle movement in evanescent field produced by collimated laser illumination on a prism with incident angle larger than critical angle (courtesy of Reference 8)

In 2001, Song et al. suggested that, according to theoretical analysis, evanescent fields coupled with surface plasmon could enhance the magnitude of optical forces thus further reduce the laser power requirements in optical trapping [11]. Inspired by this work, Quidant et al. initiated the study on the radiation forces on a Rayleigh dielectric particle by a patterned optical near-field landscape at an interface on which resonant gold nanostructures were fabricated [12]. This work predicted that a large array of controllable sub-wavelength spots from a non-focused incident laser beam is possible for optical trapping. Later, the same group utilized a photonic force microscope to experimentally measure the surface plasmon radiation forces on different polymer beads as a function of distance form metal surface, and the results showed that the enhancement factor of the plasmonic force is about 40 with respect to that under non-resonant illumination [13]. The experimental implementation following their theoretical work was reported in 2007 [14]. In this work, a gold pattern which was lithographically fabricated at the surface of a
glass substrate is illuminated under total internal reflection by a linearly p-polarized laser beam (λ = 785 nm) through a hemicylindrical glass prism, with the power at the entrance of the prism at 500mW, corresponding to an intensity I = 5×10^7 Wm^−2. On top of the micro-fabricated sample, a chamber containing polystyrene microbeads solution is attached, and the dynamics of these colloids was monitored through a long working distance objective lens and recorded by a CCD camera. Figure 2-3 illustrates the optical configuration of this work and shows the microscopic image of microbeads at homogeneous/patterned gold-water interface after laser illumination at the surface plasmon resonance (courtesy of Reference 14). This demonstration not only considerably reduces the powered required for stable trapping compared to conventional optical tweezers, it also shows the ability of effectively sieving particles according to the bead’s size by controlling the corresponding size of the gold pattern.

![Figure 2-3](image)

**Figure 2-3:** a: Schematic diagram of optical configuration of surface plasmon-based tweezers; b,c: Arrangement of polystyrene microbeads at a homogeneous (b) and a patterned (c) gold-water interface after laser illumination at the surface plasmon resonance (courtesy of Reference 14).

In their latest work published (as of May 2008), Righini et al. continued to use photonic force microscopy to assess the surface plasmon traps properties including confinement and stiffness and observed stable trapping with forces in the range of a few tens of femtonewtons [15]. They carried out further theoretical investigation on the
selectivity of surface plasmon tweezers to the trapped particle size, and revealed that by tuning the polarization of the incident laser beam, different scales of microbeads confinement could be realized due to the coupling of incident light to the surface plasmon mode of gold pattern. Besides the study on different polarization, the effect of incident angle $\theta$ on the trapping properties of a single gold pad was well examined by observing the trapping probability while $\theta$ ranged from $64^\circ$ to $69^\circ$ (the surface plasmon resonance occurs at $\theta = 68^\circ$). The absence of trapping at $\theta \leq 65^\circ$ for both $p$ and $s$ polarization and stable trapping at $\theta \geq 67^\circ$ for both polarizations reveal the competition outcome between scattering force and gradient force, indicating the significance of surface plasmon coupling in generating large enough gradient force over scattering force, which decreases as $\theta$ increases, for stable trapping, as shown in Figure 2-4 (courtesy of Reference 15).

Figure 2-4: Plots of (A) beads velocity, (B) reflectivity measurement and (C) trapping probability as a function of incident (courtesy of Reference 15).


Chapter 3

Surface Plasmon Tweezers System: Construction and Preliminary Tests

In this chapter, we will first introduce the basic principles of surface plasmon, based on which our surface plasmon tweezers system was established. Then we will make detailed description of our system configuration. Next, the experiment procedures for surface plasmon-based manipulation of 10.8 μm diameter polystyrene beads will be explained, and finally the observed results will be presented and discussed.

3.1 Principles of surface plasmons

Surface plasmons are waves that are trapped on while propagate along the surface of a conductor, e.g. a metal, in which the free electrons of the metal respond collectively by oscillating in resonance with the light wave. They are produced from the resonance between the surface charge oscillation and the electromagnetic field of the light, which gives rise to the following two consequences [1]. First is the resulting momentum of the surface plasmon mode, \( \hbar k_{SP} \), which is greater than that of a free-space photon of the same frequency, \( \hbar k_0 \) (\( k_0 = \omega/c \), free-space wave-vector). By solving Maxwell’s equations under the appropriate boundary conditions, we obtain the surface plasmon dispersion relation [2] or the frequency-dependent surface plasmon wave-vector, \( k_{SP} \),

\[
k_{SP} = k_0 \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}
\]
Metals satisfy the conditions for surface plasmon excitation that their permittivity, \( \varepsilon_m \), is both negative and complex, making it as the opposite sign to that of the dielectric material, \( \varepsilon_d \). As indicated by Eq. 3.1, the increase in momentum is related to the confinement of surface plasmons to the metal surface, and the resulting momentum mismatch between light and surface plasmons of the same frequency must be compensated if surface plasmons are to be excited by the light. This coupling process can be achieved through several methods, and we have chosen to use the popular Kretschmann configuration by illuminating the metal layer under total reflection through a glass prism. Details of setup will be described later in this chapter.

The second consequence is that the field perpendicular to the metal surface decays exponentially with distance from the surface, which is usually named as evanescent or near field. The evanescent field is a result of the bound, non-radiative nature of surface plasmons, and it prevents power from propagating away from the surface while propagating along the surface, as shown in Figure 3-1. Due this unique property of
surface plasmon, it is possible to scale down the trapping range to sub-wavelength level as in the dielectric medium (typically air or water) above the metal, the decay length of the evanescent field is of the order of half the wavelength of the coupled light.

3.2 Optical forces in a surface plasmon excitation field

As mentioned in the previous chapters, the integration of surface plasmon into evanescent field could enhance the optical force by orders thus benefit the effective manipulation of target particles. When light field illuminates an object, the local electric and magnetic field $E(r,t)$ and $H(r,t)$ produce two elementary time-dependent forces [3]. One is Coulomb force, of which the amplitude depends on the charge density $\rho(r,t)$ of the material, while it will result in polarization forces due to energy potential relations. These conservative forces are categorized as gradient forces. The second is Laplace force that is related to local magnetic field amplitude $H(r,t)$ and will result in radiation pressure effects, or scattering forces). Taking the time-average of these two forces yields the total force exerted on the object. For excitation by frequency $\omega_0$, $E(r,t)$ and $H(r,t)$ can be written as:

$$E(r, t) = \frac{1}{2} \left\{ E\left(r, \omega_0\right) e^{-i\omega_0 t} + E^*\left(r, \omega_0\right) e^{i\omega_0 t} \right\}$$  \(3.2\)

and

$$H(r, t) = \frac{1}{2} \left\{ H\left(r, \omega_0\right) e^{-i\omega_0 t} + H^*\left(r, \omega_0\right) e^{i\omega_0 t} \right\}$$  \(3.3\)

From Eq. 3.2 and Eq. 3.3 we can obtain the time-average of both forces in terms of electric polarization $P(r, \omega_0)$ of the material:
In Eq. 3.4 and Eq. 3.5, \( P(r, \omega_0) \) is related to dielectric constant \( \varepsilon(\omega_0) \) of the material, and for particle in vacuum we can write:

\[
P(r, \omega_0) = \frac{\varepsilon(\omega_0) - 1}{4\pi} E(r, \omega_0)
\]

\[3.6\]

As for the forces on particles in an evanescent field, as shown in Figure 3-2, the electromagnetic forces on a particle is obtained from Maxwell’s stress tensor [4]:

\[
\langle F \rangle = \oint_{\mathcal{S}} \langle T \cdot n \rangle ds
\]

\[3.7\]

According to the model established by Arias-González et al. [5], the forces on a particle under the influence of an evanescent wave created under total internal reflection at an interface could be written as:

\[
\langle F_{\text{grad}}(t) \rangle = -\frac{1}{4} \int_{\mathcal{V}} d\mathbf{v} \left\{ \nabla \cdot P(r, \omega_0) E^*(r, \omega_0) + \left( \nabla \cdot P^*(r, \omega_0) E(r, \omega_0) \right) \right\}
\]

\[3.4\]

and

\[
\langle F_{\text{scat}}(t) \rangle = -\frac{1}{2} \int_{\mathcal{V}} d\mathbf{v} \Re \left\{ i\omega_0 P(r, \omega_0) \wedge H^*(r, \omega_0) \right\}
\]

\[3.5\]
\[
\langle F \rangle = \frac{1}{8\pi} \text{Re} \left( \int_{\Sigma} d^2 r \left\{ \varepsilon E(r, \omega) \cdot n \right. \nabla \times H(r, \omega) + \left[ \mu H(r, \omega) \cdot n \right] \nabla \times E(r, \omega) \right\} \right)
\]

where \( \Sigma \) is a surface enclosing the particle, \( n \) stands for local outward unit normal, \( \mu \) refers to permeability. The equation is written in Gaussian units.

Volpe et al. have furthered the experimental analysis of the forces induced by surface plasmons using photonic force microscopy [6]. By using an optically trapped microbead as a local probe whose position can be measured with high accuracy, they could obtain the information of the local forces the probe experiences through the analysis of its Brownian motion [7, 8]. Figure 3-3 depicts their results in calculating the force vector and properties of force versus distance from a 40 nm thick gold film for 2.0 \( \mu m \) bead and forces on beads of different sizes at fixed distance from gold surface. Consistent with evanescent wave theory, the bead experiences larger forces when

---

Figure 3-3: (a) Force vector and (b) module (circle), x component (squares) and z component (triangles) of the force versus distance form gold surface for a 2.0 \( \mu m \) bead. (c) Force on beads of different sizes at 500 nm from surface (courtesy of Reference 6).
approaching the metal surface, while the force modulus increases as the bead size decreases. Theoretically supported by the abovementioned work, we have built our surface plasmon tweezers system with essential apparatus and samples.

### 3.3 Surface plasmon tweezers system configuration

As is illustrated in Figure 3-4, our surface plasmon tweezers system consists of three sub-systems: a) refractive index matched prism-sample system, b) laser and incident angle tuning system and c) microscope CCD imaging system. The correct functioning of all three sub-systems is necessary to observe the manipulation of target particles.

![Figure 3-4: A schematic of surface plasmon tweezers system configuration.](image)

---

**a) Refractive index-matched prism-sample system**

A 1 in × 3 in glass slide was immersed in Nano-strip 2X (Cyantek) at 80 °C for 30 min before completely rinsed and dried. Then a layer of 40 nm thick gold film was
deposited on the glass substrate by thermal evaporation technique. The sample was then coupled to the cleaned top surface of a BK7 right angle prism \( (n \approx 1.52 \text{ at } \lambda = 514.5 \text{ nm, Thorlabs}) \) with index matching oil \( (n = 1.50) \). After fixing the sample-prism body in horizontal position, a polydimethylsiloxane (PDMS) spacer about 1 mm thick was attached on top of the gold surface. Next, sufficient amount of diluted 10 \( \mu \text{m} \) polystyrene bead solution was added in the chamber formed by the spacer, and a transparent cover slip was applied on the spacer to prevent evaporation of the solution.

b) Laser and incident angle tuning system

A p- polarized Argon ion laser (Innova 300, Coherent) with wavelength 514.5 nm and maximum power \( \sim 2 \text{ W} \) was used in surface plasmon excitation. A broadband dielectric mirror (400-750 nm, Thorlabs) fixed to a computer-controlled motorized goniometer (Thorlabs) was placed in the path of the laser beam, reflecting the beam into the prism for the purpose of illuminating the gold surface. In order to examine the status of surface plasmon excitation, a photodetector (DET10A, Thorlabs) together with a lock-in amplifier and an optical chopper (Stanford research systems) was used to measure the intensity of reflected beam, thus by manually tuning the inclination of the mirror meanwhile recording the reflected laser intensity, the surface plasmon resonance is supposed to occur when the observed reflectivity reaches minimum when the incident angle just goes beyond the critical angle.
c) Microscopic CCD imaging system

A 2-axis translation stage microscope (Microzoom, Bausch & Lomb) with 8×, 10× and 50× objectives was used to observe the surface plasmon tweezer experiment. The illumination of the sample is realized via the integrated reflection mode incandescent light source. A color microscope CCD camera (EM Scopes) was integrated to record images and real-time videos with resolution 640 × 480 pixels. A set of removable long pass filters (Edmund Optics) was used to filter out the 514.5 nm laser scattering light in order to facilitate manual observation as well as recorded image analysis.

3.4 Surface plasmon tweezer preliminary experiments

![Graph](image)

Figure 3-5: Plot of reflected laser intensity from 40 nm gold film versus incident angles.

To satisfy the conditions of surface plasmon excitation, the incident angle must be adjusted until it reaches surface plasmon resonance, or reflectivity minimum. Figure 3-5
shows the recorded reflected intensity form 40 nm thick gold film when tuning the incident angle $\theta$. We observed that the surface plasmon occurs as $\theta$ approaches 46.5°. For surface plasmon tweezers experiments, the angle tuning was carried out with the gold film immersed in aqueous solution of microbeads, which will yield a different angle for surface plasmon excitation due to the refractive index difference.

Figure 3-6 shows one of our earliest results of successful surface plasmon trapping. The microscope objective used for observation was 8×. The 10.8 μm polystyrene beads were randomly and evenly distributed before the laser beam with a power of 500 mW illuminated the 40 nm thick gold surface in surface plasmon angle.

Figure 3-6: One of the earliest observation of surface plasmon trapping phenomenon on 40 nm thick gold film. The radiate stripes on the gold surface is the crinkling and peeling of gold film caused by the instability of gold in aqueous environment.
Obvious centripetal movement of beads within and adjacent to the laser illumination region towards the illumination center was observed. The inset is the image taken with 50× objective and shows the tightly packed structure formed during the trapping process, which has also been reported by European researchers [9]. This extended organization of microbeads is consistent with Eq. 3.4 that the objects will be driven towards the highest local electrical field intensity region, in this case the center of the incident Gaussian beam.

Figure 3-7: 2D and 3D plots of intensity profile of incident Gaussian laser beam.

Figure 3-7 shows both two- and three-dimensional plot of the intensity profile of the incident laser beam. Considerable amount of radiate stripes on gold surface were observed in this image, and this phenomenon has been repeatedly occurring. These crinkling and peeling of gold film is because of the relative instability of gold film in aqueous environment, compared to some conventional gold deposition where a thin layer of Chromium was sandwiched between gold and substrate for adhesion purpose. This image was taken without using color filter and the laser intensity had been reduced to 5 mW, therefore the greenish scattering light remains.
Since the aforementioned preliminary observation of surface plasmon trapping, we have worked on the improvement of the surface plasmon tweezers system. Major work include the microbead concentration adjustment for optimal observation, gold

Figure 3-8: Consecutive images recorded with interval $\Delta t = 2$ min in a surface plasmon manipulation experiment. Laser power was 500 mW. Dark cross indicates the approximate position of beam center. Arrow shows the direction of wave vector $k$. 
sample selection, microscope CCD upgrade, laser intensity control investigation etc.

Figure 3-8 shows a set of images recorded in a surface plasmon trapping experiment under interval $\Delta t = 2$ min. Still no filter was used. The illumination laser beam had a power of 500 mW and original beam diameter of 1.5 mm, corresponding to an intensity of $I = 2.83 \times 10^5$ Wm$^{-2}$, nearly two orders smaller than the work reported by Righini et al. that realized parallel and selective trapping in a pre-defined gold patterned glass substrate [10]. The entrance beam to the gold surface was reduced to a diameter of $\sim 800$ μm using an iris, and the beam center was tuned to the approximate center of the image field, as labeled with a dark cross. The arrow shows the wave vector $k$ direction of incident laser. The laser was blocked shortly before sub-image 3) was taken in order to reveal the distinct microbeads aggregation. In this experiment, we again observed that the beads within and in the vicinity of the illuminated region were driven towards the beam center immediately the laser was on, as 2) and 3) show. However, due to the existence of scattering force indicated by Eq. 3.5, the entire group of still aggregating microbeads will be guided along the $k$ direction as the laser continued to illuminate. Obviously, compared to the velocity that the microbeads move towards the intensity maximum, the translating velocity of the group was significantly smaller, proving that under surface plasmon excitation, intensity at the gold surface reaches maximum, and so does the gradient force, while the scattering force is minimum [3]. To our knowledge, this is the first reported direct observation revealing both gradient force and scattering force in gold thin film based surface plasmon manipulation of micrometer scale objects. Further quantitative analysis on the dynamics of microbeads in this mechanism will be discussed in the following chapter.
References


Chapter 4

Dynamics Study of Microbeads in Surface Plasmon Manipulation

In the previous chapter, we have demonstrated the movement of microbeads under optical forces originated from surface plasmon excitation. To further investigate in the relations among bead velocity, optical forces and incident laser power as well as to explore other possible effects that may influence the bead dynamics, we have worked on the quantitative analysis of bead dynamics based on real-time videos captured in surface plasmon manipulation process. We also took advantage of our chamber which has a relatively bigger thickness to look into the detailed thermal effects occurring during the surface plasmon excitation.

4.1 Microbeads velocity under different laser power

In order to better understand the influence of external optical energy source on the movement of microbeads, we recorded videos of beads movement under different laser powers. After fixing the system at the condition of surface plasmon excitation, we increased the laser power from 0 to 1800 mW with interval of 100 mW. At each power value, we recorded a 40 s length video with laser blocked for the first 1 s in order to observe the entire dynamic process of certain bead including the initial acceleration, which will be analyzed later. To be consistent in velocity calculation, the approximate instant velocities of beads which are 550 μm away from laser beam center at \( t = 2 \) s were
averaged for all 19 groups, since we found that single bead velocity became generally uniform within 2 s starting from laser illumination.

Figure 4-1: Plot of instant velocities of beads 550 μm from beam center as laser power increased from 0 to 1800 mW.

Figure 4-1 shows the evolution of beads velocity as laser power increases. We found the velocity-power curve could largely be divided into three regimes: a) laser power ≤ 600 mW, the curve shows linear characteristic, b) laser power between 600 and 1400 mW, there is sign of exponential growth and some uncertain trend, and c) laser power ≥ 1400 mW, the linear property appears again. This observation has led to our consideration that there might be more factors than purely surface plasmon forces that are affecting the dynamics of microbeads. We then started to analyze the details of videos recorded within these three regimes respectively.
4.2 Single bead velocity calculation in three regimes

The different characteristics in three regimes of microbeads velocity versus laser power curve has indicated there might be more complicated mechanisms involved in surface plasmon-based microbeads manipulation. In order to understand the specific bead dynamics in these regimes, we have selected three representative groups to track the velocity of a single bead: 500 mW, 1000 mW and 1500 mW. The image processing software ImageJ (NIH) was used to track a single bead about 450 μm away from beam center from zero velocity until not available for tracking due to overlapping with other beads or moving out of focus. Figure 4-2 shows the interface of ImageJ and its plug-in used to track single bead and calculate approximate instant velocity.

![ImageJ interface for microbeads tracking and velocity calculation.](image)

Figure 4-2: ImageJ interface for microbeads tracking and velocity calculation.

The single bead instant velocity tracking results for 500, 1000 and 1500 mW are
Figure 4-3: Single bead velocity tracking under laser power 500, 1000 and 1500 mW.
shown in Figure 4-3. Interestingly, three distinctly different velocity profile were obtained. For a) laser power = 500 mW, the bead velocity dramatically increased during the first second, and then started to increase about linearly, while kept about constant when approaching the beam center. For b) laser power = 1000 mW, while similar with a) in the accelerating, the bead velocity began to drop after reaching maximum at about 10 s, and finally it was mixed in the large aggregation of beads thus lost track. For c) laser power = 1500 mW, the bead velocity kept increasing even when arriving at the beam center but it moved out of focused plane immediately.

The fairly diverse behaviors of bead movement in the three regimes have revealed that some force with direction perpendicular to the gold surface drives the beads up and away from the surface. During the laser power increase from 500 mW to 1000 mW, the beads started to massively aggregate at the beam center, occupying large area thus preventing the continued acceleration of beads. While the continued increase of laser power has enhanced some force which drove the aggregated beads away from the plane, and due to the clearance of movement path, the beads under 1500 mW laser power could keep accelerating partly by the new force that only prevails under high laser power. This can be well explained by convection and thermophoresis [1], as will be discussed in the following section.

### 4.3 Thermal effects in surface plasmon excitation

The excitation of surface plasmon, especially under high laser power, will generate heating and thermal gradients in surrounding medium. At microscopic scale, the thermal gradients can induce convection, or heat transfer, and thermophoresis, or thermal
diffusion. Convection circulates fluid around a volume to transfer heat to adjacent medium, while thermophoresis drives fluid (in this case with microbeads) from hot to cold region. According to Rusconi et al., the convection velocity $U$ in a chamber of thickness $l$ that contains lateral temperature gradient could be written as:

$$U \approx g\alpha_0 l^2 \Delta T / \nu$$  \hspace{1cm} 4.1

where $g$ is gravity, $\alpha_0$ is the thermal expansion coefficient of medium, $\Delta T$ is the vertical temperature gradient and $\nu$ is the kinematic viscosity [2]. Eq. 4.1 suggests that the convection velocity is more readily to be observed when chamber thickness is larger. This explains the phenomenon observed in our experiments, where chamber thickness is about 1 mm, making the convection occur even with very small temperature gradient, as is illustrated in Figure 4-4.

Figure 4-4: Illustration of thermal effect in surface plasmon excitation.

To prove this deduction, we have focused on the examination of convection flow occurring during laser power adjusting, and we also scanned the different focus planes of microscope in order to find proof of thermal effect. Our observation of the videos
positively supported this assumption. When laser power $\leq 500$ mW, almost all beads that were driven by optical forces maintained uniform velocity after reaching force equilibrium, indicating that convection flow has not shown observable effect. From 600 mW above, considerable amount of blurred (indicating out of focus) beads groups started to be driven by convection flow towards beam center region in velocities much higher than optically driven beads. Figure 4-5 shows the fast movement of some blurred beads group under 1000 mW laser power (frame interval $< 1$ s).

![Figure 4-5: Fast movement of beads group driven by convection flow towards beam center. Interval between two frames is less than 1 s.](image)

Meanwhile the beam center region began to show the stacking of aggregated beads due to the direction of convection at beam center region is vertical to the gold surface. The “stacking” phenomenon is accompanied with the reducing diameter of beads aggregation area, since as the convection continues, the liquid flow with beads from the non-illuminated region will carry the aggregated beads away from their previously stably trapped positions. However, as soon as the laser is off, the beads flushed up from the beam center region will deposit again on gold surface due to gravity, hence restoring the
aggregation area. Figure 4-6 shows the images at a) end of 40 s long 1000 mW laser illumination, b) about 1 min after the illumination ended, revealing both “stacking” and “restoring” phenomena.

Figure 4-6: Images of a) end of 40 s laser illumination with power 1000 mW, b) about 1 min after the illumination ended.
Another interesting observation is that the bead aggregation will finally disappear as the laser power increases. Figure 4-7 shows the beginning of laser illumination under different high laser powers. This is because according to Eq. 4.1, the convection transports beads in increasing velocity $v$ as temperature gradients $\Delta T$ increase, disabling the stable trapping by surface plasmon forces and even restoring of aggregation that requires lower velocity hence sufficiently quantity of beads right above the region.

![Figure 4-7: Images of the beginning of laser illumination under different power. The disappearing aggregation is due to dramatically increasing convection velocity that surpasses surface plasmon-based optical forces.](image-url)
Finally, the vertical scanning of microscope focal planes directly proved and characterized the convection all along the depth of the chamber. By vertically lifting up the microscope focal plane, we have observed that the moving directions of beads started to transit from centripetal to centrifugal, showing the beads were following the convection pattern as depicted in Figure 4-4. Figure 4-8 are images of a plane higher above the gold surface plane in the chamber taken with 1 s interval. The beads gushed from the 1000 mW laser beam center (labeled with dark cross) which maintains highest temperature towards the rest part of chamber due to convection.

![Figure 4-8: Beads driven from a 1000 mW laser beam center position (dark cross) to a higher plane above the gold surface, showing the convection effect.](image)

It is worth noticing that although convection dominates the microbeads dynamics under the relatively high laser power, the majority of the beads that are driven near the gold surface still maintain generally uniform velocity when moving towards the beam center. This velocity is much smaller than that of the beads carried by convection flow. Considering the linearity of the velocity curve fit at high laser power range in Figure 4-1, we still believe that surface plasmon-based gradient forces are mainly responsible for the
moving of the beads near the gold surface while convection has relatively less influence on them. Therefore, further understanding and clarifying the influence of convection on particles at different location in the chamber would be an interesting topic to continue.
References


Chapter 5

Future Work

As a newly emerged field, plasmonics holds tremendous potential in exploring the field of sub-wavelength optics. Based on the current setup and results, this chapter will give a discussion on the work planned to accomplish in the near future.

5.1 Dynamics study in surface plasmon manipulation of microbeads with different sizes

In this thesis work, only 10.8μm diameter beads were used in the experiments since the topics focused on requires the consistent target conditions. And to our knowledge, no systematic work has been published on the influence of different laser powers on different sizes of beads. Righini and colleagues have studied the selectivity of surface plasmon trapping and angle dependence velocity of targets by using two sizes of polystyrene beads: 3.55 μm and 4.88 μm [1, 2], both showing necessary in supporting their theoretical claims.

Particle size is one of the key factors in evanescent field force or surface plasmon force research, because when it scales down to sub-wavelength level or dipole level, the unique properties of plasmons make it possible for different exciting applications including data storage, light generation, microscopy and bio-photonics [3]. In this sense, to study from the fundamentals about particle size would benefit the better understanding of critical cornerstone of plasmonics.
The experiment will utilize similar setup as the current one in our lab except to use polystyrene beads with 5.01 μm or fewer diameters, and it may include some improvement to be mentioned in the following section. By tuning laser power and also incident light size within proper range, it is anticipated that the dynamics study results would able to compensate the explanation for the results observed using the current 10.8 μm beads. And it is also possible to solve certain equations that are not soluble by single group of data.

5.2 Optimization for surface plasmon manipulation force study

As was discussed in this thesis work, thermal effects including convection and thermophoresis play an important role in driving polystyrene microbeads. One can choose to fully understand the interaction and coexistent properties of optical and thermal forces, thus master both, or can circumvent the non-mainstream to attack the more intriguing. I believe most people would choose the latter path.

Quidant et al. reported using chambers of 10-20 μm thick to prevent the dominance of thermal effects [2, 4]. This method is effective in confining the conditions that facilitate the convection generation. We could minimize the interference of thermal effects in an alternative way, by carefully controlling laser intensity. Since in this work, we have observed that the approximate threshold for obvious convection flow, we could continue to find a laser power that serves as effective tweezers energy level and modest in thermal gradient generation.

Currently we do not have equipment that can directly measure the generated forces and related properties of surface plasmons, but as this work has underlined that
employing indirect means to solve problems would also be an option. We are considering by integrating certain type of “gauge” into the surface plasmon system, and relate it with the pursued unknown, e.g. the stiffness of DNA strands and the minimum laser power used for surface plasmon force to break them. This is also the way we learned to solve many other unknowns.

5.3 Tunable plasmonic patterning by controlling incident light

Despite the merits in low laser power requirement compared to conventional optical tweezers, the surface plasmon tweezers proposed by Righini et al. is largely limited in tunability of trapping patterns which relies exclusively on the pre-defined pattern of fabricated metal structures, making it less preferable in applications where real-time manipulation of targets is necessary.

As an alternative to surface plasmon-based optical trapping by metal surface patterning [2], we have proposed to construct an incident light manipulation (ILM) system to achieve tunable plasmonic patterning, circumventing the complicated fabrication of different pre-defined gold substrates. The ILM system renders manually or computer-dependently tunable light patterning on Au film by controlling the reflective optical elements immediately before the light enters the prism. Two categories of ILM system components are proposed here: (1) Manually designed and fabricated micromirror pattern on glass substrate. (2) Commercially available light modulation products, e.g. digital micromirror device (DMD), which consists of a large array of micromirrors and all pixels are individually controllable via microelectromechanical systems (MEMS) technology.
Figure 5-1 shows the principle of tunable plasmonic patterning based on ILM system. First, a optical system will be used to generate nearly flat-top laser beam, instead of Gaussian profile since the targets will be driven towards the highest intensity. A spatial filter might be helpful but for ideal case an aspheric lens is preferred. Then, selective reflection of incident light is enabled by (1) pre-defined micromirror pattern deposited on transparent glass substrate, reflecting only the light of patterned portion; (2) computer-generated effective micromirror array pattern in DMD, reflecting only the patterned portion of light in the effective angle. Incident angle tuning to obtain surface plasmon excitation is implemented for both setups before experiments.

![Diagram](image)

**Figure 5-1**: A schematic of incident light manipulation system for tunable plasmonic trapping on a homogeneous gold film.

Next, selectively reflected light passes the prism and glass substrate, forming corresponding light pattern on gold film. Due to the dramatic gradient pressure difference between the light-active and inactive region on gold film, micrometer scale polystyrene...
beads will be driven from low intensity towards the high intensity region. The majority of polystyrene beads in the vicinity of patterned light region are supposed to be aggregated to form the pattern as the ILM system defines. The effective control over the balance of gradient force and scattering force in both setups is to be investigated based on specific light patterns.

Tunable plasmonic patterning of micrometer scale polystyrene beads can be achieved by (1) micrometer scale strict parallel translation or rotation of micromirror substrate without changing the incident angle and (2) DMD pattern transition by computer programming. The proposed DMD setup has several attractive advantages in accurate and fast surface plasmon-based manipulation: no movement of equipment is required during the optical pattern tuning, making it a very stable candidate for ILM system; all micromirror pixels are uniform in dimensions of micrometer scale (1024 × 768 mirrors each with 13.68 μm × 13.68 μm dimension for Discovery 3000 Chipset, Texas Instrument), making it highly accurate even for single micrometer scale object manipulation; instant micromirror pattern change through computer control, making it possible for real-time fast plasmonic patterning. Compared to conventional optical tweezers and recent surface plasmon-based manipulation, our proposed tunable plasmonic patterning technique has significantly lowered the necessary laser power for trapping objects, taken the initiative in tunable plasmonic patterning while avoiding complicated metal substrate fabrication, and enabled the possibility in highly stable, accurate and fast real-time surface plasmon-based tunable manipulation of micrometer scale objects.
References


Appendix

Nontechnical Abstract

Chopsticks were devised long ago to pick up and move food. Similarly, a recent invention called ‘optical tweezers’ entails the use of a force gradient caused by a laser beam to manipulate microscale and nanoscale objects. An optical tweezers setup that is ‘state of the art’ nonetheless requires the use of bulky optics and intense laser beams. These requirements prohibit the use of the invention in delicate biological applications like the ‘lab-on-a-chip’, a burgeoning microscale setup for the real-time analysis of ailments.

In 1992, Satoshi Kawata developed a novel mechanism for optical manipulation that uses a thin evanescent wave to propel microscale particles toward a material interface, because of the rapidly decaying intensity of the field. Yang Gon Song and Romain Quidant, respectively, did theoretical and experimental work on using surface plasmons (the collectively oscillating electrons at a metal surface due to the resonance with an incident light wave), to enhance the trapping effect of evanescent waves. By introducing surface plasmon enhancement, microscale particles can be successfully trapped on existing metal structures with illumination of much lower laser power.

The author has realized the surface plasmon based manipulation of micrometer scale polystyrene beads on a gold surface. The dynamics of the microbeads have been observed and further analyzed. The development of optical manipulation tools from optical tweezers to surface plasmon tweezers was briefly reviewed. From the introduction
to surface plasmon principle, the author generalized optical forces generated in surface plasmon excitation. Following a description of experiment details for sample preparation and surface plasmon excitation, an angle-dependent reflectivity plot showed the condition which enables surface plasmon based manipulation. Microbeads aggregated at the center of a spot illuminated by a laser beam due to the force and thermal gradient; the aggregate was pushed along the direction in which the surface plasmons propagated.

To further understand the effect of the optical forces on microbeads movement, the dynamics of microbeads were analyzed by investigating the relationship between incident laser power and the mean velocity of beads. Two different stages which were respectively dominated by surface plasmon-induced forces and convection effect were observed when the laser power increased from 0 to 1800 mW, and by tracking the velocity of single beads from peripheral to beam center, different dynamic profiles are also discovered. By microscopic characterization, convection due to laser thermal effect was found to play an important role in affecting microbeads movement at high laser powers larger than 600 mW. The occurrence, relaxation and recovery of aggregation were realized when enabling and disabling laser illumination are alternated. The distribution of microbeads on gold surface at different laser power was then characterized by optical microscopy, revealing thermal convection.

A progression of the described work entails a) equivalent studies on the dynamics of microbeads; of many diameters; b) optimization of optical manipulation by circumventing convection dominance; and c) tunable patterning of microparticles.
One may actuate nanoscale matter with a surface plasmon tweezers setup. Its range from tens of nanometers to hundreds of micrometers, and from femtonewtons to nanonewtons covers many physiologic processes responsible for human body operation. For physics and chemistry, this range also reaches the classical and quantum mechanics interface. Engineering nanometer scale actuators are also promising in exploring atomic level mechanical behavior. The better understanding of surface plasmon based optical forces facilitates the subwavelength scale research in abovementioned fields thus plays a significant rule in the future optical manipulation realm.