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Doctoral Dissertation

A study on laser submicrometer machining by
photonic nanojet utilizing water medium

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Abstract

Since Feynman's 1959 groundbreaking vision of nanoscale technology, miniaturized products and their fabrication methods have become central in current engineering practice. This thesis investigates a novel laser-assisted machining method that employs photonic nanojet (PNJ) effects in a water medium. While ultrafast laser machining offers excellent spatial precision, it is constrained by the diffraction limit. The PNJ, formed by light interacting with dielectric microsphere, overcome this limitation by enabling sub-diffraction focusing. Although PNJ machining was first demonstrated in air, it suffered from debris redeposition and limited working distance and depth of focus. To address these challenges, this research develops PNJ machining in water, achieving submicrometer resolution and improved machining quality. Numerical simulations and experiments provide a comprehensive understanding of PNJ behavior in water. Finite-difference time-domain (FDTD) simulations showed that water medium increases the PNJ working distance and depth of focus while maintaining submicrometer beam diameters. Experiments confirmed ablation diameters as small as $0.57 \pm 0.19 \mu\text{m}$ in the x -direction and sub- $0.8 \mu\text{m}$ in the y -direction, with smoother surfaces compared from processing in air medium. Compared with conventional laser machining under identical parameters, $1.08 \pm 0.07 \mu\text{m}$ hole diameter in the x -direction was obtained. PNJ machining in water provides an extended working distance, deeper depth of focus, reduced redeposition, and cleaner features relative to air-based PNJ and conventional methods. Further enhancement was achieved using second-harmonic generation (SHG) illumination, which reduced the PNJ beam diameter to $0.43 \mu\text{m}$ and increased the working distance to $4.18 \mu\text{m}$. Experimental results showed sub- 600 nm , deeper ablation marks and an extended machining distance of up to $8 \mu\text{m}$. Combining SHG with controlled defocusing further improved uniformity and depth control, enabling high-quality 3D microfabrication. In summary, this work establishes PNJ machining in water as a high-precision, environmentally friendly approach for submicrometer fabrication, integrating ultrafast lasers, PNJ effects, and water immersion to improve feature size control, surface quality, and process versatility. The method shows strong potential for MEMS/NEMS, optoelectronics, and next-generation semiconductor manufacturing.

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Chapter 1

Introduction

1.1 Research background

Since Richard P. Feynman gave his influential lecture "There is Plenty of Room at the Bottom" at the American Physical Society Annual Meeting on 29 December 1959, there has been ongoing interest in developing technologies to create and handle smaller structures [1]. While people often associate this vision with nanotechnology, many practical applications have been realized through micrometer and submicrometer-scale fabrication methods, rather than solely at the nanoscale. In practice, research covers a wide range of length scales. This includes nanomaterials, nanomanufacturing, and micro- and submicrometer-scale systems as well as the integration of such structures into larger micro- or nanosystems [2, 3]. The work presented herein is situated within the submicrometer regime, wherein characteristic feature sizes are smaller than those of conventional microsystems but remain above the true nanoscale. This intermediate range enables enhanced device functionality while maintaining compatibility with established microfabrication techniques.

Systems operating across these length scales are commonly classified as Micro/Nano Electro-Mechanical Systems (MEMS/NEMS). MEMS combines electrical and mechanical components to create microscopic devices with characteristic lengths ranging from 100nm to 1 mm; devices below that are referred to as NEMS. The MEMS/NEMS terms are used in a broad sense, including electrical, mechanical, optical (MOEMS/NOEMS), and biological functions (bioMEMS/bioNEMS). Figure 1.1 shows the dimensional comparison of nanomaterials, MEMS/NEMS, and BioMEMS/NEMS. The examples shown in Figure 1.1 are a digital micromirror device with 12 μm in size for MEMS [4], a 100 nm molecular gear [5], and a 2 nm single-walled carbon nanotube (SWNT) chemical sensor [6] for NEMS. In comparison, a human hair is about 75000 nm (75 μm), red blood cells are about 8000 nm (8 μm) [7], and individual atoms are typically a fraction of a nanometer in diameter. Advances in micro- and submicrometer-scale science and engineering have played a key role in advancing miniaturization. This progress has enabled the creation of smaller, more efficient systems with wide-ranging technological effects.

The fabrication of MEMS/NEMS across micro-, submicrometer-, and nanoscale dimensions is generally achieved through either bottom-up or top-down manufacturing approaches. In the bottom-up approach, structures are constructed from their elements, like building a house from a set of

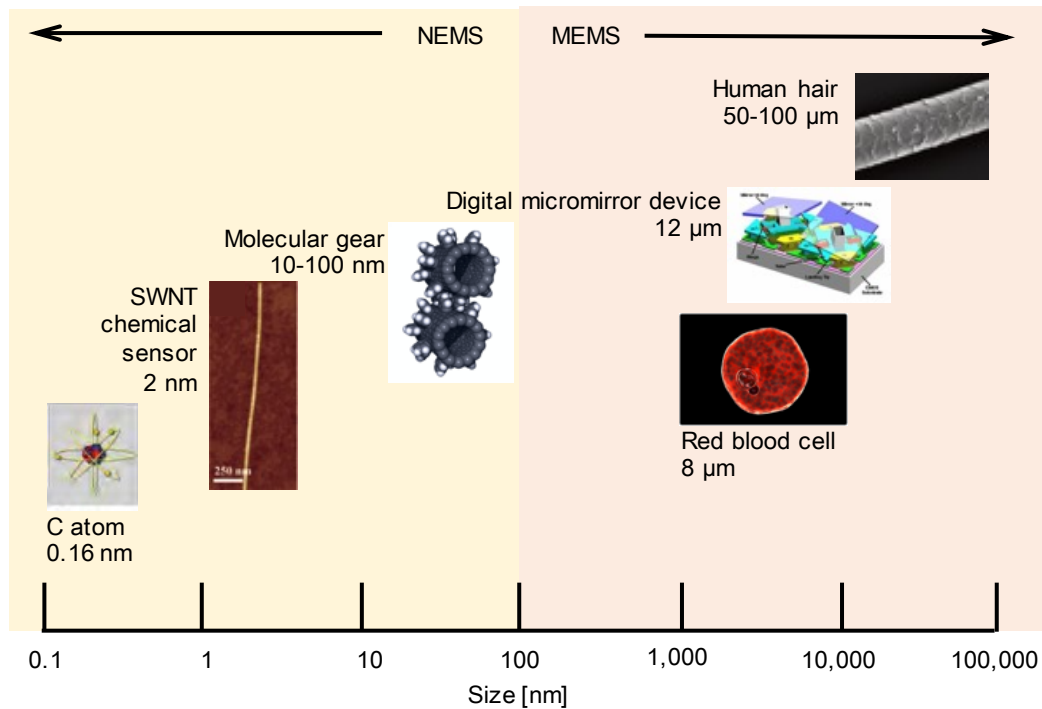


Fig. 1.1 Characteristics dimensions of nanomaterials, MEMS/NEMS, and BioNEMS in perspective [4–7].

Lego pieces. Those elemental constituents are combined using various manufacturing processes, including self-assembly, chemical synthesis, and nano-imprint lithography [8–10]. In contrast, the top-down approach mainly uses lithographic and non-lithographic fabrication technologies [11]. For non-lithographic techniques, thin-film deposition and doping are widely used [12].

In many MEMS/NEMS applications, partial or complete substrate removal is required to realize suspended mechanical structures. Among these techniques, bulk micro-/nanomachining is the oldest for fabricating MEMS/NEMS and, as such, is the most mature, making it the most commercially successful, helping to manufacture devices such as pressure sensors and inkjet printheads [13, 14]. Despite numerous variations, the fundamental principle of bulk micromachining is the selective removal of the substrate from a piece of raw material, making it a critical enabling technology for the continued development of micro- and submicrometer-scale systems relevant to nanoscale engineering.

1.1.1 Laser-assisted fabrication technologies

In recent years, the demand for three-dimensional (3D) micro- and nanostructures has risen significantly [15]. This change is due to their ability to enhance device performance and expand applications across photonics, microelectronics, biotechnology, and materials engineering [16, 17]. Creating these structures requires micro- and nanofabrication technologies that achieve submicrometer precision, offer high processing flexibility, and work with a wide range of materials. Among the available methods, laser-assisted fabrication has become a promising option due to its versatility, high spatial resolution, and ability to create both two-dimensional (2D) and 3D structures. Laser, an acronym for light amplification by stimulated emission of radiation, is essentially a coherent, convergent, and monochromatic beam of electromagnetic radiation with a wavelength (λ) ranging from the ultraviolet (UV) to the infrared [18]. A laser can deliver very low ($\sim$ mW) to incredibly high (1-100 kW) focused power with a precise spot size/dimension and interaction/pulse time (10^{-3} to 10^{-15}) on any substrate through any medium. This allows for flexible control over structure formation at the micro- and nanoscale. Figure 1.2 shows these laser-assisted fabrication technologies in terms of achievable resolution and free-form geometric structuring capability.

Historically, optical lithography has been the primary technique for microfabrication, especially in the semiconductor industry for producing integrated circuits [19]. This method transfers a pattern

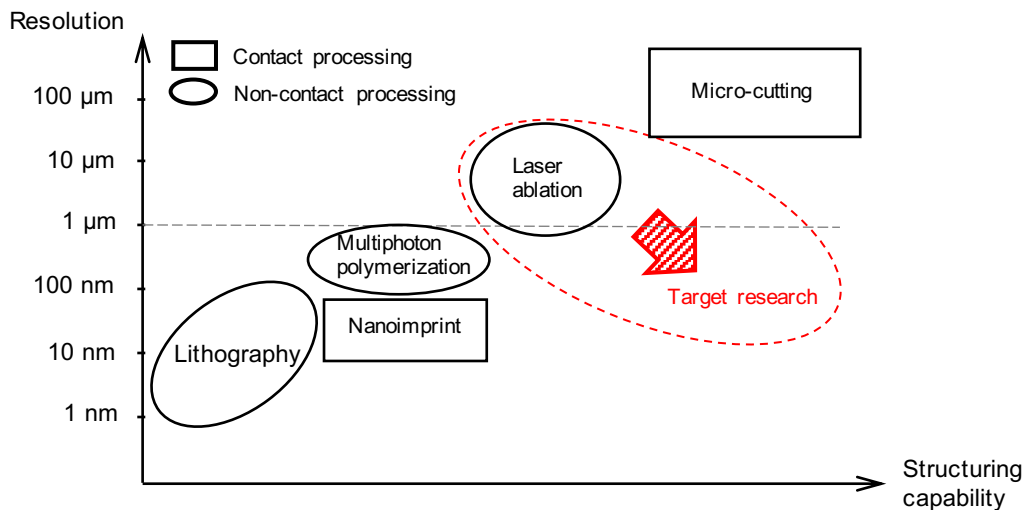


Fig. 1.2 Comparison of laser-assisted fabrication methods in terms of resolution and its structuring capabilities for fabricating 3D microstructures. FIB: Focused ion beam.

onto a photosensitive resist by exposing it through a photomask. The resolution of photolithography depends on the wavelength of the light used and the numerical aperture (NA) of the focusing optics. Shorter wavelengths and higher NA values allow for finer patterning [20, 21]. In recent decades, the introduction of deep ultraviolet (DUV) and extreme ultraviolet (EUV) lithography has significantly improved resolution, achieving feature sizes as small as 10 nm with EUV light at 13.5 nm [21]. Despite these improvements, optical lithography has inherent limitations for planar, surface-based patterning. Its working principle restricts fabrication to 2D or near-2.5D geometries, making it unsuitable for directly creating fully 3D structures. Additionally, the need for complex mask preparation and long processing times decreases design flexibility and production efficiency. This presents challenges for rapidly evolving applications that require customized or volumetric microstructures.

To overcome the two-dimensional limits of traditional photolithography, researchers developed nanoimprint lithography (NIL) as a technique for high-resolution pattern replication. In NIL, a transparent mold, usually made from quartz, presses against a photo-curable resin layer on the substrate [22]. UV light then cures the resin. After curing, the mold is removed, leaving an inverse copy of its nanostructure. This method can create highly detailed, intricate patterns with resolutions approaching 10 nm [23]. It offers a straightforward and cost-effective way to fabricate. However, its flexibility is limited because each pattern requires a new mold. The process is also constrained by the physical and chemical properties of photo-curable resins, which limit the materials that can be used. Therefore, while both photolithography and nanoimprint lithography are essential for mass production of nanoscale structures, their use in 3D microfabrication and design flexibility remains limited [24].

A more direct and flexible way to create 3D nanostructures is through two-photon, or multiphoton, polymerization (TPP). This technique exploits nonlinear optical absorption. Polymerization happens only at the focus of a tightly focused laser beam when two or more photons are absorbed at the same time. Since polymerization is restricted to the focal volume, structures with features smaller than the diffraction limit can be made directly in three dimensions by moving the laser within a photosensitive resin. TPP allows for the creation of freeform, curved, and tall structures that traditional planar lithographic methods cannot achieve [25, 26]. Using femtosecond pulsed lasers improves resolution and reduces thermal damage, enabling patterning with features as small as 100 nm [27]. However, the

technique often faces challenges due to a lack of suitable photosensitive materials and slow processing speeds, which limit its scalability for industrial use.

In parallel with these developments, laser ablation has become an important, non-contact method for micro- and nanomachining. Unlike lithographic processes that use photoresists or molds, laser ablation directly removes material through localized heating, melting, or vaporization. The energy of each photon is given by hc/λ , where λ represents the laser wavelength, typically ranging from 157 to 10,600 nm [16]. The wavelength determines the achievable spot size and directly affects the optical diffraction limit of the process [28]. The laser pulse duration is crucial for defining the precision and quality of the ablation. Long-pulse or continuous-wave lasers cause significant heat diffusion into the surrounding material, which results in large heat-affected zones (HAZ), recast layers, and reduced accuracy. In contrast, ultrashort-pulse lasers, such as femtosecond or picosecond sources, allow for “cold” ablation. In this case, material removal happens faster than thermal diffusion, minimizing collateral damage and improving spatial resolution [29].

The development of ultrafast lasers has transformed laser material processing, enabling laser-assisted micro- and nanofabrication. This technique is effective for creating 2D surface structures and 3D structures on materials. By reducing heat diffusion and exploiting nonlinear absorption processes, laser fabrication can achieve exceptionally high spatial resolution, reaching submicrometer scales and even tens of nanometers [1]. Among various laser-based methods, laser ablation stands out for its versatility and precision. It has been widely used in 2D tasks such as drilling, cutting, cleaning, marking, and scribing [30]. This technique is among the most flexible machining approaches, as it enables optimization of optical setups and works with a broad range of materials.

1.1.2 Laser-assisted machining in water medium

In some applications, the workpiece in laser processing is in contact with liquids (e.g., natural bodies of water, nuclear reactors, boreholes). The workpiece may contain the liquid in its normal state (e.g., moisture) or the liquid may be applied to the workpiece to enhance the processing or to achieve some other beneficial effect (e.g., improving surface quality, high aspect ratio holes, increasing Raman scattering signal, etc.) [31]. The liquid in the material processing can be used as a cleaning agent,

solvent, coolant, transport agent, and energy sources [31, 32]. Water or deionized (DI) water is typically used since it is cheap, harmless, and recyclable in terms for industrial aspect considerations.

Numerous studies have examined how a liquid environment, especially water, affects laser machining processes and material properties. These studies show that having a water layer on the workpiece is crucial for the laser ablation process [33–35]. Water, as a polar liquid, has strong intermolecular bonds and a high specific heat capacity ($c = 4184 \text{ J/kg}^\circ\text{C}$) compared to nonpolar solvents. As a result, during laser ablation, water helps cool both the plasma and the heated material surface. This cooling reduces the size of the heat-affected zone (HAZ).

Additionally, when the laser vaporizes the water, it creates a temporary vapor layer or cavitation bubbles. These bubbles help confine the plasma plume and improve ablation efficiency, leading to cleaner material removal. The surrounding liquid also effectively removes debris and prevents molten material from redepositing on the processed surface. This is shown in Fig. 1.3, where laser processing in water produces much cleaner features on silicon wafers and brittle materials, such as LCD glass (Figs. 1.3(b) and (d)), than processing in air (Figs. 1.3(a) and (c)). The benefits and drawbacks of liquid-assisted laser processing are listed in Table 1.1. While using a liquid medium can pose challenges, careful control of laser settings and processing conditions can help address them. Thus, submerging the workpiece in water offers a practical way to achieve more efficient, faster, and higher-quality laser patterning than processing in air.

1.1.3 Photonic nanojet

The major hurdle to the broader use of lasers in conventional materials-processing applications is the minimum beam size achievable with the respective optical setup. It is defined by the optical diffraction limit, which is given by

$$d_{\text{beam}} = \frac{\lambda}{2n \sin \alpha}, \quad (1.1)$$

where d_{beam} is the minimum obtained spot diameter, λ is the wavelength of the incident laser, n is the refractive index of the medium where the beam propagates, and α is the beam divergence angle. The term $n \sin \alpha$ in Equation (1.1) refers to the numerical aperture (NA) of the objective lens used to focus

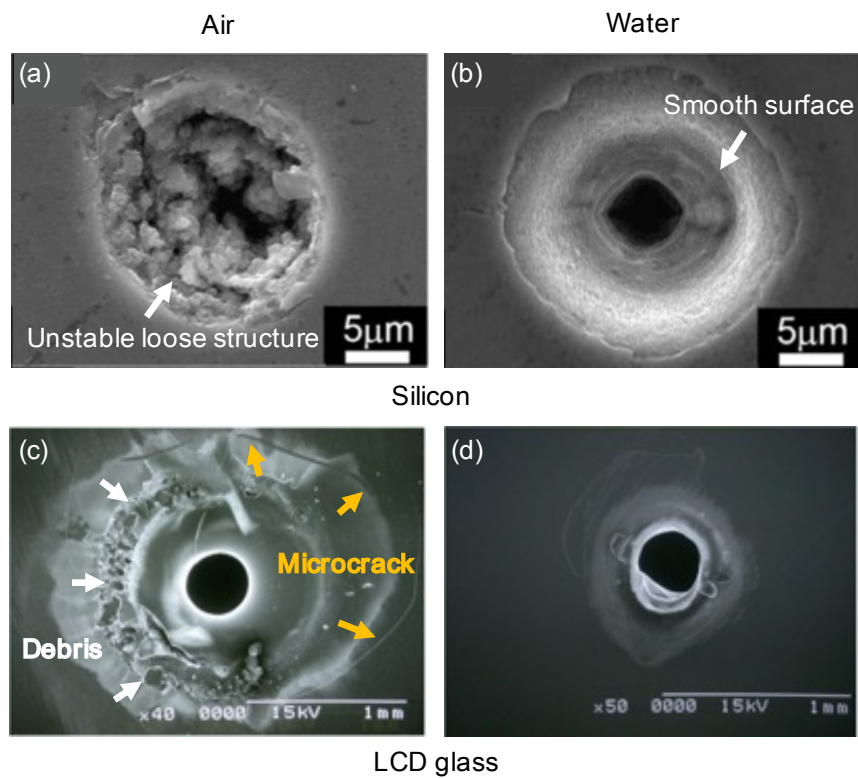


Fig. 1.3 SEM photos of silicon wafer drilled in (a) air and (b) water medium [36]; and SEM photos of LCD glass drilled in (c) air and (d) water medium [34].

Table 1.1 Advantages and disadvantages of laser processing in water compared in vacuum/gas medium [31].

Advantages	Disadvantages
• More effective cooling of workpiece and ejected material • Water convection/explosive evaporation carries debris away • Higher plasma pressure due to confinement • Useful chemical reactions with water for further analysis • Lower optical breakdown threshold than in air	• Power loss due to water cooling • Light might be scattered by the water surface, suspensions and bubbles • Slower processing due to the long relaxation time of the vaporized state of water

the irradiating laser beam [37]. In conventional laser machining, a focusing lens is typically used to focus the laser beam into the raw material or workpiece. However, there is a constraint on the working distance or depth of focus (DOF) in conventional laser machining; the DOF is relatively small, and if we miss this focusing spot, it would reduce the product's resolution. In micro- and nanomachining, one important factor is the ability to achieve a small beam diameter. It is usually done by applying a higher NA objective lens. There is also a limitation to the availability of this objective lens, so we need to develop another method to obtain a nanoscale laser beam.

Since it was introduced by Chen et al. [38] in 2004, the photonic nanojet has gained popularity among researchers for achieving a small beam diameter. It was demonstrated that its full width at half maximum (FWHM) could be less than the optical diffraction limit value. The term photonic nanojet, referred to as PNJ in this thesis from here on, stands for a fine high-intensity electromagnetic beam that is generated at the backside of an illuminated transparent dielectric microcylinder, microdisk, or microsphere of diameter greater than the illuminating wavelength, as shown in observation by Ferrand et al. [39] in Figure 1.4(a), and its 2D finite-difference time-domain (FDTD) calculation of PNJ is shown in Figure 1.4(b). In addition to a small beam diameter, a PNJ could propagate along a path that can extend for more than $\sim 2\lambda$ beyond the dielectric particle with minimum divergence [40, 41]. Due to their unique optical characteristics, PNJs are believed to have potential applications in nanoparticle sensing, sub-wavelength-resolution direct-write nanopatterning and nanolithography, broadband, low-loss waveguiding, ultramicroscopy, and ultrahigh-density optical data storage [40].

By applying a high-energy input laser to the transparent dielectric material, the generated PNJ would have an intensified light intensity that could reach the fluence value, or energy per area (mJ/cm^2), above the machining threshold of the working material, thereby initiating the ablation process on the material. The PNJ machining method in an air medium has been realized recently in various material [37, 42]. In air medium, the peak intensity of PNJ in the air medium was known to generate at $1\text{ }\mu\text{m}$ or less from the rear microsphere surface, which means one should approach the working material closely to acquire the best result. If it is not carefully aligned, the microsphere could damage the workpiece and reduce the resolution and stability of fabricated patterns. Therefore, it appears that controlling the position of the microsphere was difficult; the microspheres need to spread on the surface of the working material to accommodate the short peak intensity from the rear surface

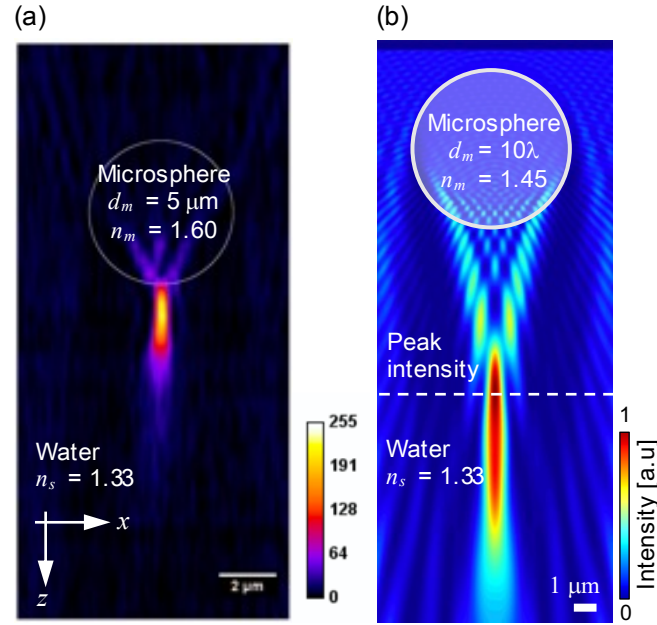


Fig. 1.4 Schematic diagram of PNJ shown in (a) observation [39] and 2D FDTD calculation.

of the microsphere. Those conditions will affect the flexibility of the PNJ to do the machining with complex shapes. Additionally, there is visible debris on the surface of the material, which needs to be addressed afterward; otherwise, it compromises the product's surface quality.

1.2 Research objectives

In response to the challenges faced in PNJ machining in air medium, this proposed study aims to establish a new machining method that uses water as the working medium. The overall goals of this proposed technique are illustrated in Fig. 1.5.

PNJ machining in water offers several potential advantages:

- **Sub-micrometer resolution:** This feature allows the method to achieve high-resolution fabrication, making it suitable for precision manufacturing.
- **Higher material removal:** Using a controlled PNJ beam in water medium extend the depth of focus and enhances the material removal of the machining process, allowing for the efficient processing.

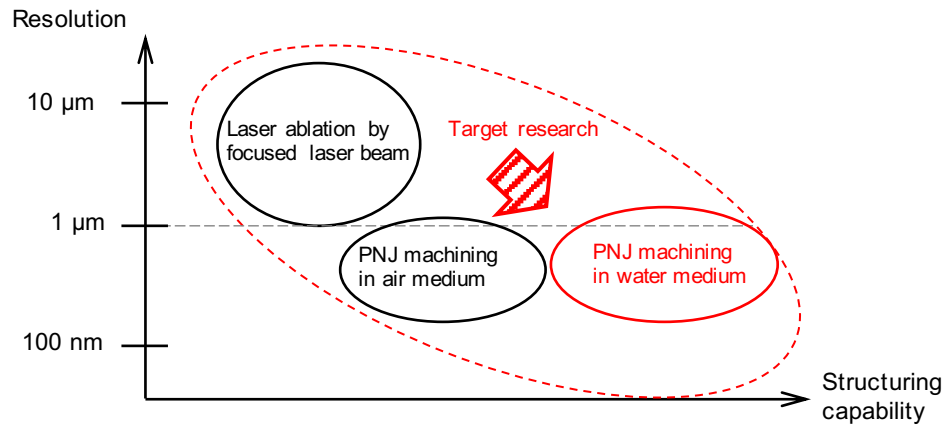


Fig. 1.5 Processing target of this proposed study.

- **Improved machining quality:** This method is expected to maintain the integrity of the work material by means of low level of oxidation and debris near the targeted fabrication area.

Based on these points, this study suggests using water as the machining medium for the PNJ technique. In this process, both the microsphere and the work material are submerged in water, and material removal occurs through PNJ machining. The choice of water as the medium is based on its simplicity and practicality; it allows for implementation with minimal changes to the existing optical system. Furthermore, water is eco-friendly, inexpensive, non-toxic, and recyclable, making it an excellent choice for sustainable manufacturing. Water also offers several benefits similar to those seen in traditional laser machining, including improved cooling and ablation mechanism, and reduced redeposition of ablated material.

In this research, we investigate the PNJ machining in a water medium using both numerical simulations and experimental methods. We use numerical simulations to analyze the PNJ intensity distribution in water under different parameter sets. This approach will help us examine effects that are hard to observe in experiments. These findings, along with controlled experimental validation, will deepen the understanding of the PNJ mechanism in water medium and establish this thesis as a key reference for PNJ machining in water.

1.3 Thesis organization

The overall structure of this thesis is illustrated in Fig. 1.6, and the content of each chapter is summarized as follows:

Chapter 2: Principle of Photonic Nanojet for Machining

In this chapter, the fundamental mechanism and optical characteristics of the PNJ are first described. This is followed by an analysis of the intensity distribution control of PNJ generated in a water medium. Finally, the principle of PNJ machining in a water medium is presented. Based on these discussions, a novel machining method utilizing PNJ in water is proposed.

Chapter 3: Numerical Simulation of Photonic Nanojet in Water Medium

Building upon the concepts introduced in Chapter 2, this chapter focuses on the numerical simulation of PNJ intensity distribution in a water medium. FDTD technique employed in this research is first

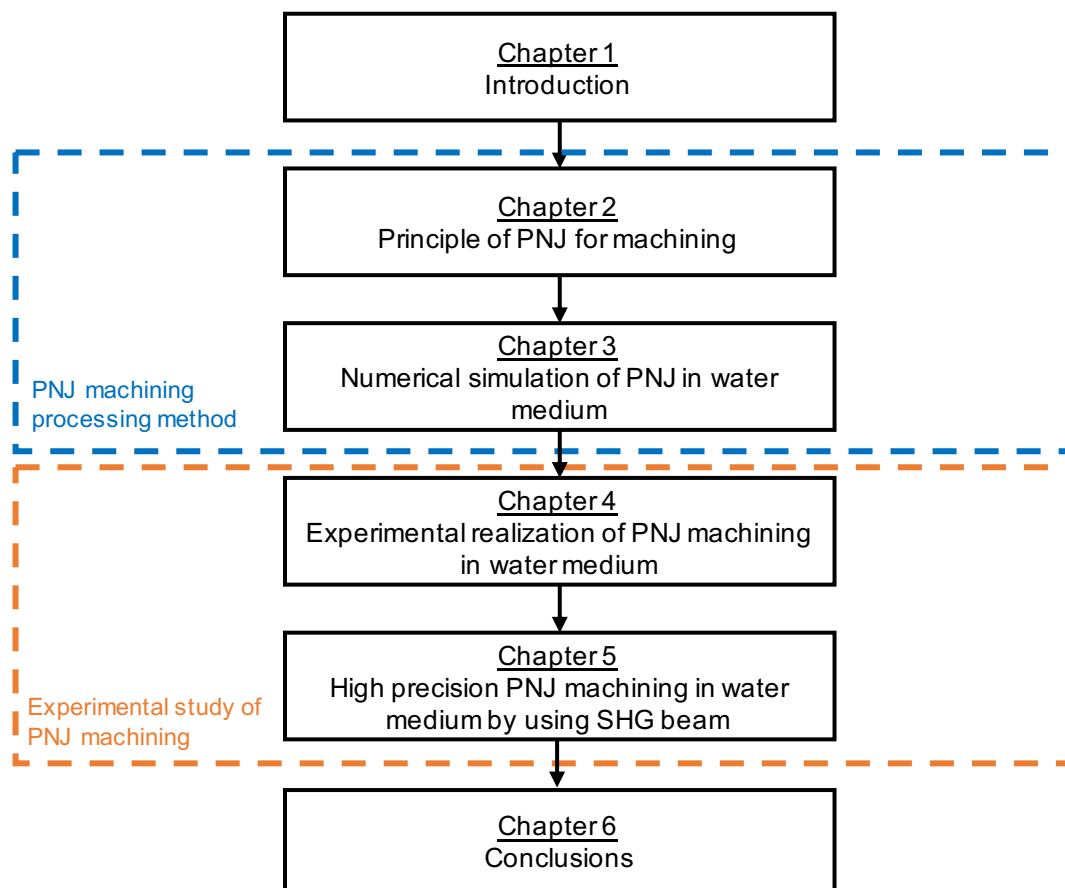


Fig. 1.6 Thesis structure.

described, followed by a detailed explanation of the simulation model. Subsequently, the results of the FDTD simulations are examined to evaluate the effects of different liquid media, microsphere materials, and input laser beam parameters on the PNJ intensity distribution.

Chapter 4: Experimental Realization of PNJ machining in Water Medium

This chapter presents the experimental investigation of PNJ machining in a water medium for the very first time. Initially, laser machining experiments are conducted using a focused laser beam in water, and the results are analyzed. Subsequently, PNJ-based machining experiments are performed under both uncontrolled and controlled parameters, and the outcomes are discussed. Finally, the machining results obtained from the focused laser beam and PNJ are compared, and the experimental findings are correlated with the numerical simulation results.

Chapter 5: High Precision PNJ Machining in Water Medium by Using SHG Beam

Chapter 5 explores the enhancement of PNJ machining in water through the use of a second-harmonic generation (SHG) beam. The chapter begins with the analysis of FDTD simulation results, which are then applied to guide PNJ machining experiments in water under controlled parameters. Finally, the results are comprehensively investigated in terms of surface morphology and material integrity.

Chapter 6: Conclusions

The final chapter synthesizes the findings of the entire study. It provides an integrated discussion linking all the preceding chapters, analyzes the implications of the results, and presents the overall conclusions of the research. The chapter also highlights the contributions of this study and discusses potential directions for future work.

Chapter 2

Principle of photonic nanojet for machining

This chapter examines the basic principles and processes underlying photonic nanojet-assisted laser machining in water medium. It provides a detailed theoretical framework that forms the foundation for the experiments discussed in later chapters. Understanding these principles is important for improving the machining process and achieving sub-micrometer resolution in material processing applications. The chapter is divided into three main sections, each focusing on key aspects of photonic nanojet machining. Section 2.1 covers laser ablation mechanisms in a water medium. It explains how water affects the interaction between the laser and material, along with heat transfer and debris removal, compared to conventional laser machining in air. Section 2.2 introduces photonic nanojet phenomena and their use in laser machining. This section discusses the generation and features of photonic nanojets and reviews recent improvements in photonic nanojet machining techniques. Lastly, Section 2.3 looks at methods for controlling the intensity distribution of photonic nanojets. It focuses on how photonic nanojets behave in water medium and on the specific control parameters that can be adjusted to optimize the intensity distribution in liquid environments.

2.1 Laser ablation in water medium

Laser ablation is an efficient method for creating high-resolution fabrication. Its benefits include precise resolution, minimal heat impact on the target material, and high flexibility [43, 44]. This technique is used across many fields, including electronics, semiconductor technology, medicine, and automotive manufacturing [45–47]. Laser ablation usually involves removing material with a pulsed laser. However, a continuous wave (CW) laser can also ablate, provided the laser intensity is high enough [48, 49]. When a laser beam focuses on the surface of a solid target, pulsed laser radiation can be absorbed through different energy processes. This absorption causes both thermal and non-thermal heating, melting, and ultimately, the removal of the target material.

In high-resolution laser machining, there are two significant constraints associated with laser use: wavelength and pulse width. Wavelength must be controlled to achieve small beam diameter to overcome the optical diffraction limit [50, 51]. Regarding pulse width, a longer pulse width (> picoseconds) can generate significant thermal effects and plasma formation, leading to a large heat-affected zone (HAZ), recast layers, and limiting achievable precision and quality [52].

The beneficial effects of ultrashort-pulse to material interaction can be explained by the fundamental changes in the laser/material interaction phenomena when the pulse width is shortened, as shown in Figure 2.1(a) when using a nanosecond (ns) pulsed laser and (b) for a femtosecond (fs) pulsed laser. In both ns and fs lasers, the laser radiation is mainly absorbed by the free-electron system over a time scale of 100 fs [53, 54]. The energetic electrons then transfer their energy to the lattice in picoseconds; e.g., the electron-phonon (quasi-state with lattice vibration) interaction times for aluminum and copper are 0.5 and 50.0 ps, respectively [50, 55]. Suppose the pulse width were made less than the electron-phonon interaction time. In that case, the high-intensity beam heats the electron to very high temperatures and transfers energy weakly to the ions and the lattice. The material in the interaction volume transforms into a vapor phase with high kinetic energy, resulting in a clean ablation process, as shown in Figure 2.1(b) by using a fs laser [56]. On the other hand, if the pulse width is much larger than the electron-phonon relaxation time, as in an ns-pulsed laser, the energy transfer from the electrons to the ions becomes significant. Therefore, heat diffusion occurs over a much larger volume than the focus volume as shown in Figure 2.1(a) [57].

The shorter pulse duration limits heat conduction and improves precision, especially in metals and semiconductors, because there is a large density of free and valence electrons with ionization potentials below the photon energy [58, 59]. When the absorption depth is smaller than the diffusion length ($\sqrt{Dt}$, where D is the thermal diffusivity, and t is the pulse width), the melt expulsion and

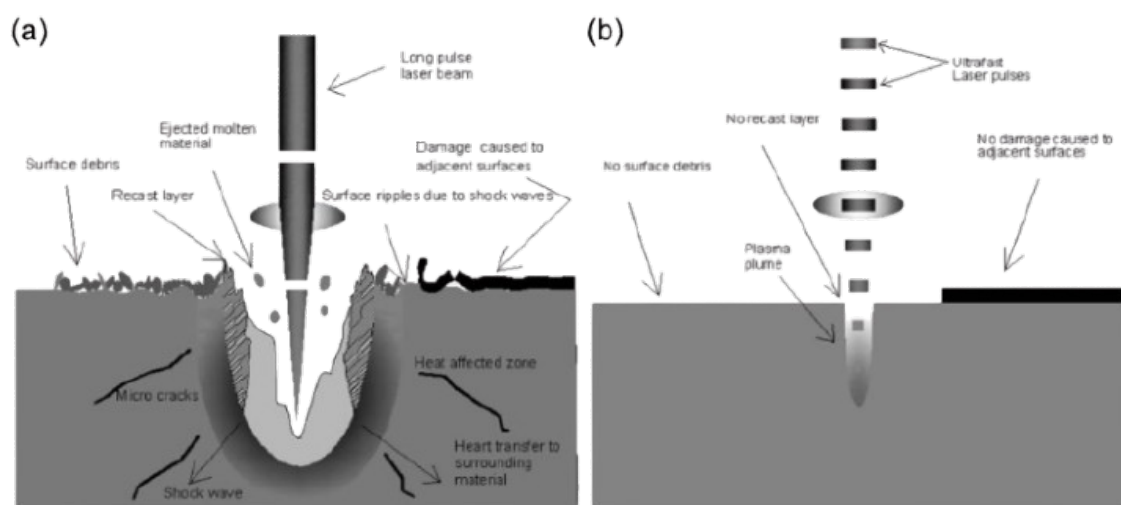


Fig. 2.1 Effects of laser ablation on the material with (a) ns pulses and (b) fs pulses [50].

associated fluid flow accompany the material removal, resulting in low precision. On the other hand, when the pulse length is reduced to ultrashort, the absorption depth exceeds the diffusion length, leading to energy localization within the depth and, consequently, direct vaporization, resulting in high precision, reduced HAZ, and improved surface quality of the product [60]. Thus, ultrashort-pulsed lasers have proved to be an effective tool for micromachining across a wide range of materials. Since this research aims to establish a sub-micrometer processing technology, the effect of heat must be minimized as much as possible. Therefore, in this research, an fs pulsed laser is preferable.

When we use liquid as a medium during the ultrashort ablation process, also known globally as Pulsed Laser Ablation in Liquid (PLAL), there is visible plasma emission, the production of shock waves, and the formation of cavitation bubbles [61, 62]. A schematic illustration of the fs laser ablation process in air and liquid medium over time is shown in Figure 2.2. As shown in Figure 2.2, after laser-matter interaction, laser-induced breakdown occurs, followed by the shock wave formation, plasma expansion, and cooling. Then, by the formation, expansion, and, lastly, collapse of cavitation bubbles. During the bubble collapse period, nanoparticles (NPs) are produced as plasma cools, then diffuse into the surrounding liquid and form a colloidal solution.

PLAL occurs when a high-intensity laser pulse, typically with an irradiance of $0.1\text{--}1\text{ GW/cm}^2$, is focused on a solid target immersed in a liquid. This starts a complex series of plasma and bubble dynamics as shown in Figure 2.3. When the laser pulse strikes the target surface, it ejects seed electrons through multiphoton ionization. These electrons then absorb more photon energy through inverse Bremsstrahlung. This process rapidly heats and ionizes the target material, forming a dense, hot plasma plume at the solid-liquid interface. The target material undergoes continuous phase change from solid to vapor, resulting in a significant drop in density and a supersonic plasma expansion that produces an intense shock wave in the surrounding liquid. As the plasma expands and cools, its pressure decreases, and the shock wave compresses the surrounding liquid until the plasma dissipates. While the fundamental processes of plasma formation in liquids are similar to those in air, the energy exchange mechanisms are quite different because liquids have a higher density and can better contain and absorb energy. After the plasma quenches, the quick energy transfer to the liquid creates a cavitation bubble that grows and collapses over several microseconds as shown in Figure 2.3. This bubble contains vaporized liquid, ablated NP, and gases. When it collapses, it creates microjets and

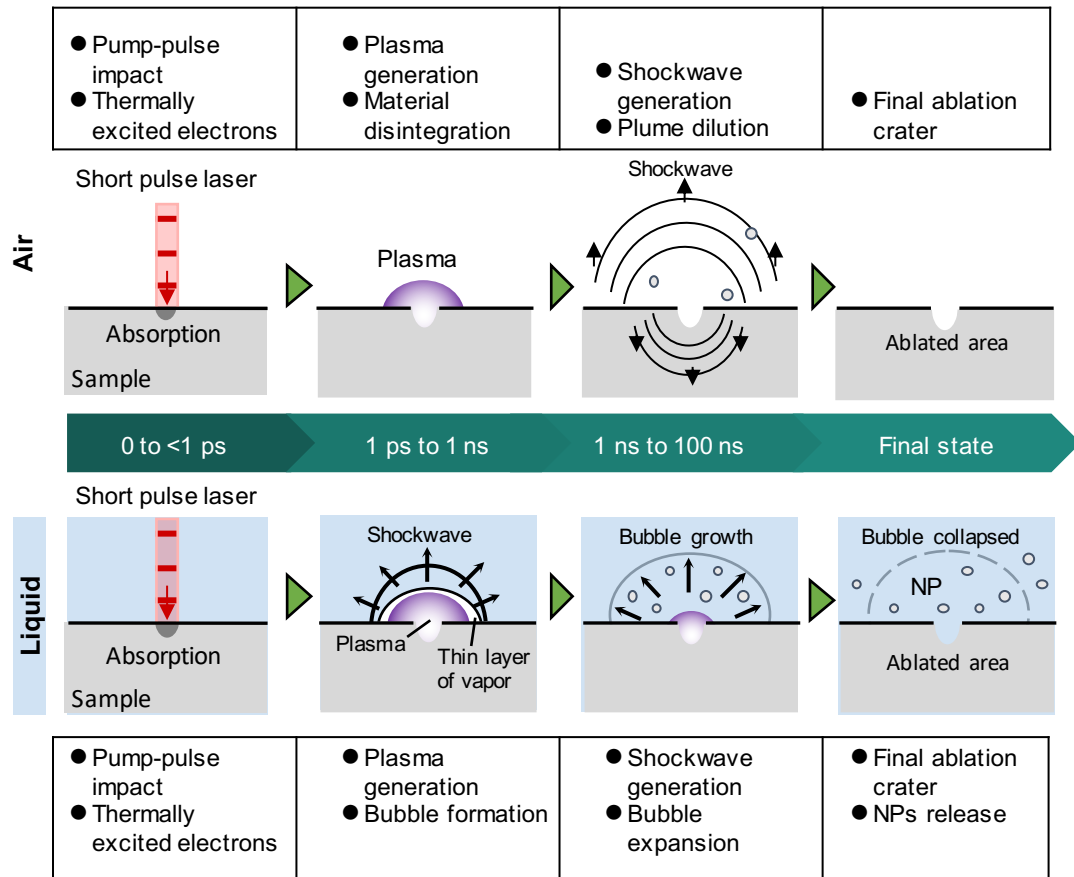


Fig. 2.2 Time sequence of the processes involved during the fs pulse laser ablation in air and liquid medium. NP: Nanoparticle, ns: nanosecond.

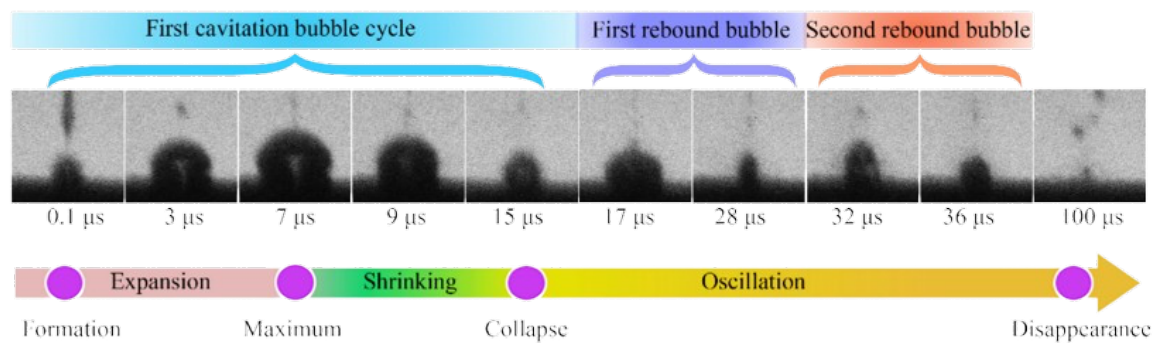


Fig. 2.3 Shadowgraph images of cavitation bubble generated after laser ablation of a silicon target in deionized water from 18 μ J pulse laser energy [32].

shock waves that effectively remove debris from the ablation site. As a result, laser ablation in liquid often yields cleaner, smoother surfaces with a smaller heat-affected zone than in air, making it better for precision micromachining and nanoparticle synthesis [32, 62].

Regarding the resolution achieved obtained by laser ablation, the machining region on the working material is determined by the relationship between the fluence distribution of the laser beam that irradiated the sample that has a specific value of the fluence threshold [63, 64]. In brief, material ablation occurs when the fluence of the input laser beam exceeds the material's fluence threshold. This fluence threshold is specific to the material and depends on the number of irradiating laser pulses and the surrounding medium. The enhancement of the ablation threshold fluence in liquid is related to the different optical properties at the interfaces between the transparent media and the silicon wafer. Liu et al. [36] performed a simple calculation and experiment, showing that for the same incident laser fluence, the reflection-induced energy loss in ambient air is 9% to 10% higher than in water or liquid environments. Thus, more energy could be injected into the sample surfaces due to the presence of liquids, thereby decreasing the ablation threshold fluence. Furthermore, the enhanced recoil pressures from laser-induced shockwaves and bubble-related mechanical forces under liquid confinement would cause additional damage to the sample, thereby reducing the threshold value [65].

The relationship between the fluence distribution and the laser machining resolution is shown in Figure 2.4. Suppose that we have a Gaussian beam with a beam radius r that has a peak fluence value F_0 , and such a fluence value F_0/e^2 is emitted to the sample. Then, the fluence distribution in the x -direction of the irradiated beam is given by

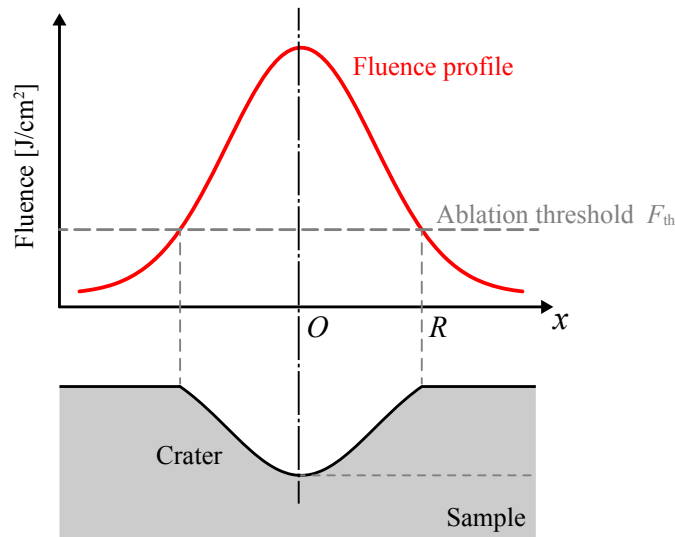


Fig. 2.4 Principle of laser ablation [66].

$$F(x) = F_0 \exp\left(-\frac{x^2}{r^2}\right). \quad (2.1)$$

Next, we assume the fluence threshold of the sample is given with F_{th} and the ablation radius at that time in R is given by

$$F(R) = F_0 \exp\left(-\frac{R^2}{r^2}\right) = F_{\text{th}}, \quad (2.2)$$

and from Equation (2.2), solving the R gives

$$R = r \sqrt{\ln\left(\frac{F_0}{F_{\text{th}}}\right)}. \quad (2.3)$$

Equation (2.3) shows that the machining radius R can be controlled by controlling the peak fluence F_0 and the beam radius r of the incident laser light. Therefore, the ablation diameter can be managed by controlling the fluence distribution of the incident laser to the fluence threshold of working material.

2.2 Photonic nanojet for laser machining applications

The photonic nanojet (PNJ) was initially defined by Chen et al. [38] and Li et al. [67] as a narrow, intense electromagnetic beam propagating into a background medium from the "shadow-side" surface of a plane-wave-illuminated, lossless dielectric microcylinder or microsphere of diameter greater than the illuminating wavelength. When the incident beam is focused or close to the shadow-side surface of the microelement, an interesting phenomenon occurs due to the near-field interactions of the sphere's boundary and the focused beam. A highly localized electromagnetic beam at the shadow-side surface of the sphere is created. This beam, called "photonic nanojet" due to its appearance is analogous to a high-speed gradient that is characteristic of a jet in fluid mechanism. [67, 68].

2.2.1 Photonic nanojet generation and characteristics

The PNJ generation from an incident light beam impinging on a dielectric microparticle can be explained using a ray-tracing method. Although the ray-tracing method from the geometrical optics

approach is not an accurate description of the PNJ generation mechanism, it provides essential insight into the focusing characteristics of microspheres and the location of PNJ. Geometrical optics approach is helpful to define the optical path of the light beam since the particle size is relatively big, a few micrometers in diameter (d_m) if using microsphere, comparing to the wavelength (λ) of the input laser beam, about 400 nm to 800 nm in the visible region, therefore $d_m \gg \lambda$. In the geometrical optics approach, light is treated as a set of rays rather than a wave. Therefore, ray tracing is a method for calculating the paths of waves or particles through a system with regions of varying propagation velocity, absorption characteristics, and reflecting surfaces. Ray tracing works by assuming that a particle or wave can be modeled as a large number of very narrow beams (rays). When there is some propagation distance in the same medium, even for a minimal distance, the ray is locally straight. The ray tracer will advance the ray over some distance, then use a local derivative of the medium to calculate the ray's new direction if it passes into another medium. From this location, a new ray was sent out, and the process was repeated until a complete path for the ray was generated.

Due to the utilization of microparticles to generate PNJ, the light beam propagates through a medium with a different refractive index value (n). When a light beam passes from one medium to another medium, there is a change in the direction of the ray at the interface between the two media. This phenomenon is called refraction. Figure 2.5 shows when light is propagating to a different medium. Figure 2.5(a) applies when the light passes a denser medium ($n_2 > n_1$), and Figure 2.5(b) is the other way around. The propagation direction of light from one medium with refractive index n_1 with an incident angle θ_1 to another medium with refractive index n_2 is obtained by Snell's law, which is given by

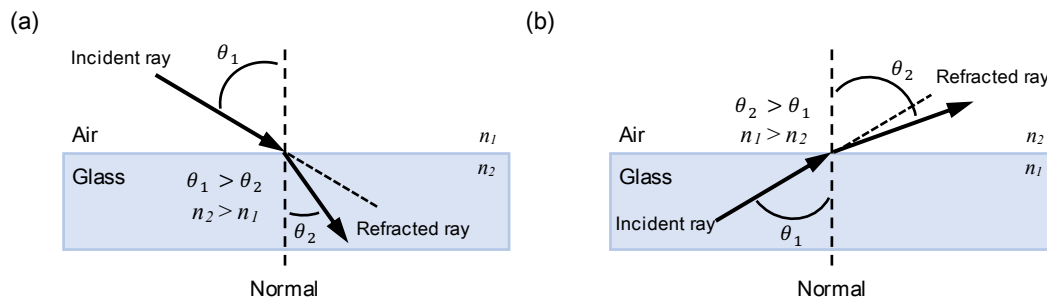


Fig. 2.5 Refraction phenomena when light going to (a) a denser medium (higher refractive index value) and (b) to a lesser dense medium (lower refractive index value).

$$n_1 \sin \theta_1 = n_2 \sin \theta_2. \quad (2.4)$$

Those simple refraction rules also apply in our PNJ generation mechanism. The PNJ generation mechanism is simpler to explain when we use a microsphere as the microparticle to focus the input laser beam. In order to get an appropriate PNJ form, one must use a microparticle that has a refractive index bigger than the surrounding medium ($n_m > n_s$) as shown in Figure 2.6 when we use a microsphere with $n_m = 1.4$ in (a) air medium ($n_s = 1.0$) and (b) in water medium ($n_s = 1.3$). On the other hand, if the rule does not apply, the PNJ is not generated as shown in Figure 2.6(c).

From the perspective of the ray angle, the central axis region of the microsphere functions as a low-NA objective lens. In contrast, the area far from the microsphere's central axis functions as an objective lens with a high NA [69]. The outer surface of the fully-immersed particle comprises the boundary of constant optical contrast. These causes geometrically similar refraction of peripheral and central rays on the sphere, resulting in the formation of one external focus along the propagation axis due to spherical aberrations, which is the one we referred to as PNJ [70]. Furthermore, Darafsheh

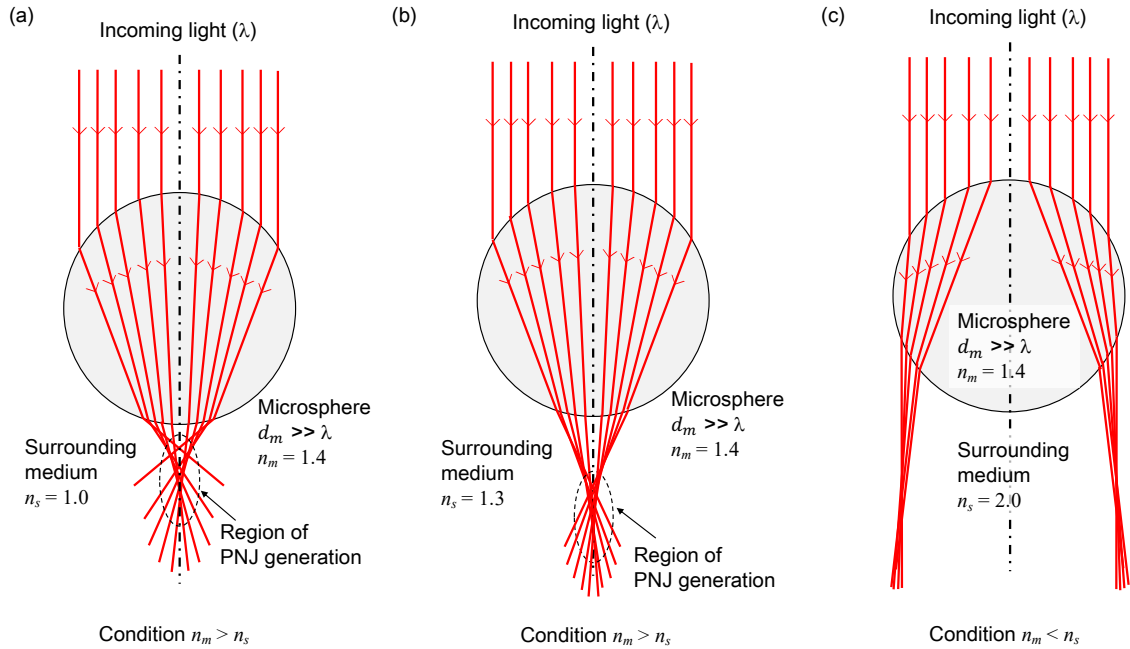


Fig. 2.6 Schematic diagram of an appropriate PNJ by ray tracing method when (a) $n_s = 1.0$, (b) $n_s = 1.3$, and (c) $n_s = 2.0$.

[68] stated that nanojet light confinement by microelements is a complex scattering and interference phenomenon, which is caused by constructive interference between the incident field and the scattered and diffracted light fields near the dielectric microparticle due to the near-field interactions at the microparticle's boundary.

The PNJ with jet-fluid-like shape has several key properties as follows [40, 70]:

1. PNJ attributed to a near-field optics phenomenon with a certain contribution of evanescent waves.
2. PNJ manifests itself as a relatively slowly-decaying propagating light beam by maintaining its sub-wavelength width along a path that can extend more than $\sim 2\lambda$ beyond the dielectric micro-cylinder or sphere.
3. The minimal full-width at half-maximum (FWHM) of this beam can be smaller than the classical far-field diffraction limit ($\approx 0.5\lambda$), in fact as small as $\sim 0.3\lambda$ for microspheres.
4. PNJ is a non-resonant diffraction phenomenon that can appear in a wide range of the dielectric microcylinder or microsphere sizes (from $\sim 2\lambda$ to more than 40λ) as long as the refractive index contrast (RIC) is less than 2.
5. The PNJ possesses a high intensity that can significantly exceed that of the illuminating wave by a factor of more than 100 spherical particles.

The key parameters of interest in PNJ machining are PNJ beam diameter, defined by its full width at half maximum (FWHM) in the lateral direction of PNJ propagation; the working distance (WD), which refers to the distance from the rear microsphere surface to the peak intensity of the generated PNJ on the propagating axis; and the depth of focus (DOF), defined by the FWHM along the propagation axis. All the PNJ parameters are schematically shown in Figure 2.7.

2.2.2 Recent photonic nanojet techniques for machining

To achieve finer machining resolutions, PNJ machining has emerged as an innovative alternative, reducing beam sizes compared with classical focusing via an objective lens in laser ablation. When subjected to high input laser energy, the dielectric microsphere generates PNJ with an enhanced

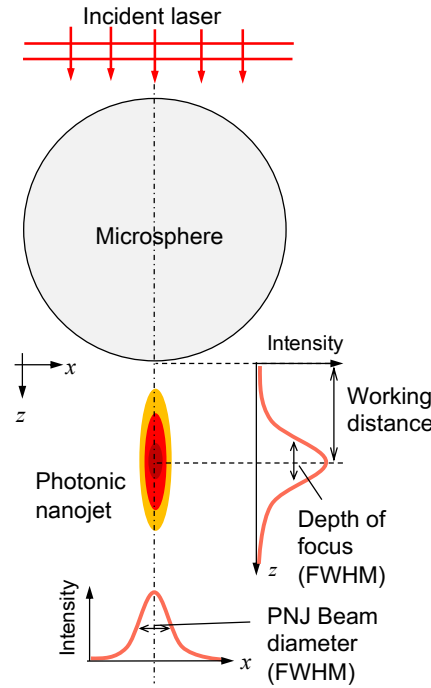


Fig. 2.7 Schematic diagram of generated PNJ from microsphere and its parameters.

fluence while maintaining its small beam size. This makes PNJ machining particularly advantageous for applications that require high levels of miniaturization and precision, such as semiconductor devices.

PNJ machining has been demonstrated in air-based environments for various materials using several input wavelengths. For instance, Abdurrochman et al. [42] successfully achieved 1300-nm features on a glass substrate using a 1064 nm input wavelength. Meanwhile, Uenohara et al. [37] fabricated 180 nm features on a silicon wafer with an 800 nm input wavelength. Elkarkri et al. [71] obtained 135 nm features on polystyrene film using a 532 nm input wavelength. Zhou et al. [72] realized 300 nm features on an indium tin oxide (ITO) glass substrate using a 355 nm input wavelength. Table 2.1 summarizes various state-of-the-art PNJ machining techniques and their simulation and achieved fabrication resolution.

For all previous works shown in Table 2.1, PNJ machining in air requires precise microsphere alignment to accommodate the short working distance from the generated PNJ. This constraint makes the process susceptible to resolution loss and potential damage to the workpiece if the alignment is not carefully maintained. To address these challenges, this study introduces a water medium for PNJ

Table 2.1 Established PNJ machining techniques with their respective simulation and experimental results.

Medium	Wavelength [nm]	Simulation			Experiment		Reference
		PNJ beam diameter [nm]	Working distance	Depth of focus [μm]	Smallest Resolution	Substrate	
Air	1064	700	3 μm	Not elaborated	1300 nm	Glass	[42]
	1064	700	150 nm	Not elaborated	1280 nm	Silicon	[42]
	800	437	1.1 μm	2	180 nm (up to 2.0 μm)	Silicon	[37]
	532	Not elaborated	1 μm	Not elaborated	369 nm	PS polymer	[71]
	355	260	Not elaborated	Not elaborated	300 nm	ITO glass	[72]
Water	1030	730	2.84 μm	3.16	920 nm (up to 4.5 μm)	Silicon	Our work
	800	580	2.94 μm	2.58	600 nm (up to 4.5 μm)	Silicon	Our work
	515	430	4.18 μm	2.51	550 nm (up to 8 μm)	Silicon	Our work

machining. As PNJs can propagate over longer distances in water, the microsphere does not need to be positioned near the workpiece, thereby reducing the risk of damage to the sample and alignment errors [73].

2.3 Control of photonic nanojet intensity distribution

The parameters of interest in PNJ are the beam waist size (here referred to as the PNJ beam diameter), the maximum beam intensity, and its location (WD). These parameters can be engineered primarily by modifying and modulating those related to the incident laser, the microsphere, and the surrounding medium. Complete parameters that could be controlled regarding the resulting PNJ are shown in Table 2.2. Parameters related to the incident laser are the wavelength, beam profile, polarization, and incident angle. In terms of the microsphere, the parameter that could be changed is the shape, size, and refractive index. Lastly, a parameter related to the surrounding medium is the refractive

Table 2.2 Parameters to control the PNJ intensity distribution.

Item	Parameter
Incident laser	Wavelength (λ)
	Beam profile
	Polarization direction
	Incident angle
Dielectric microparticle	Shape
	Size
	Refractive index (n_m)
Surrounding medium	Refractive index (n_s)

index. To the best of our knowledge, many researchers have clarified that the characteristics of the resulting PNJ can be controlled by adjusting parameters to obtain a PNJ that suits the intended application [68, 74, 75]. From the writer's perspective, it is better to first investigate the PNJ intensity distribution numerically, e.g., via electromagnetic field simulation using FDTD, before applying the PNJ mechanism in machining applications.

2.3.1 Photonic nanojet behavior in water medium

Due to the presence of a water with a refractive index greater than that of air in the surrounding medium, by applying the ray-tracing method shown in Figure 2.6(b), we can conclude that in the water medium, light is more confined farther from the rear microsphere surface. Thus, the value of WD becomes bigger. Figure 2.8 shows the Mie modeling results of the cross-sectional view of the normalized local field distribution ($|E|^2$) underneath a single $5.0\ \mu\text{m}$ particle in (a) air and (b) immersed in water medium, under an x -polarized plane wave excitation at 800 nm. As shown in Figure 2.8(a), the WD of generated PNJ is located at position $z/a = 1.1$, which is close (sub-wavelength distance) to the particle surface, a referring to particle diameter. As a near-field characteristic, the field enhancement lies within the range from $z/a = 1.1$ to $z/a = 2.2$. Moreover, the result decays almost exponentially. It means for efficient patterning, the sample must be kept in near-field contact with particles. On the other hand, the focusing properties of the particle change dramatically when a surrounding medium used. As shown in Figure 2.8(b), the presence of water medium greatly extends

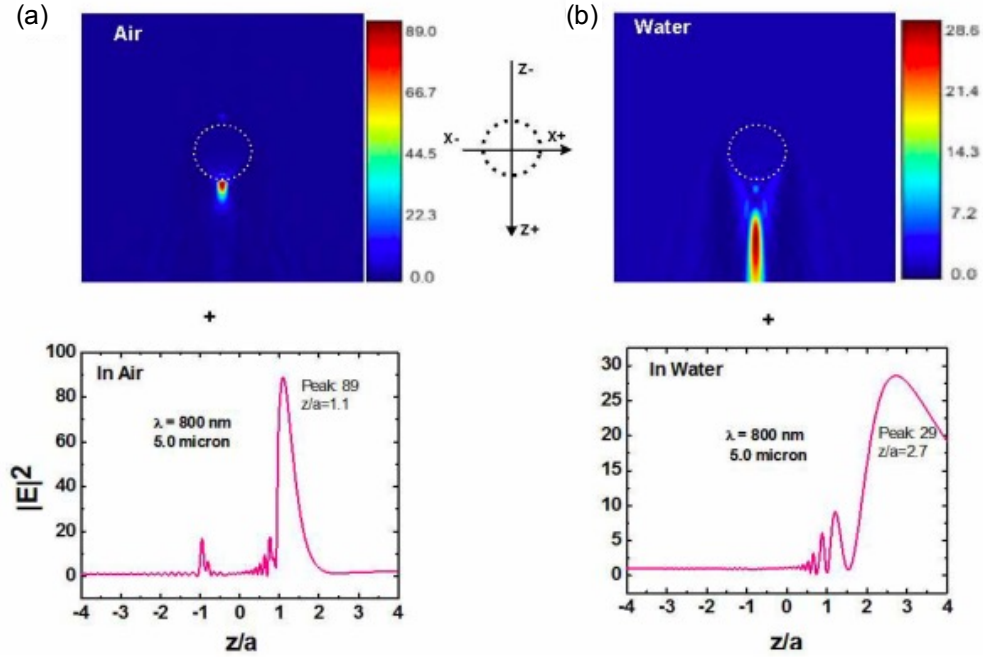


Fig. 2.8 PNJ intensity distribution by Mie modeling and cross-sectional view of the normalized local field distribution ($|E|^2$) (a) in air medium and (b) in water medium [78].

the WD from $z/a = 1.1$ to $z/a = 2.7$. Therefore, surrounding medium plays a big part in generated PNJ intensity distribution [76, 77].

2.3.2 Intensity distribution control in water medium

From the parameters of PNJ generation shown in Table 2.2, as water is going to be chosen as a liquid for the medium, the other parameter that could still play a part in the PNJ beam generation is the incident laser parameters and the morphology of the dielectric microparticle. Therefore, although PNJ comes with an extensive set of input variables, we must consider its application in real experimental conditions. From that mindset, we should investigate a resulting PNJ using readily available market parameters, as they make sense for the experiment [79, 80]. In terms of dielectric microparticles, the microsphere will be investigated, as it is readily available and could enable us to improve PNJ intensity distribution. For the microsphere, only the diameter and refractive index are considered, as they have a greater effect on the resulting PNJ. At a glance, a larger microsphere size gives us a larger PNJ beam diameter, a longer WD, and a longer DoF. Then, using a higher refractive index

microsphere, since the light will be more focused, the resulting PNJ will have a smaller PNJ beam diameter, a shorter WD, and a shorter DoF [81, 82]. Detailed numerical simulation will be discussed further in the chapter 3.

Regarding the incident laser beam, the most accessible parameter to change is the input wavelength. Lee et al. [83] suggested that using a shorter incident laser wavelength could yield a smaller PNJ beam diameter, a shorter WD, and a shorter DoF. However, although we can control the input laser wavelength, e.g., by using a tunable laser in the experiment, we need to tune the laser to its peak intensity, depending on the laser beam profile. Hence, it reaches the ablation threshold of the working material and could perform ablation if we use PNJ for machining. The other parameters, like beam profile, polarization, and incident angle, would also affect the resulting PNJ [84, 85]. The detailed explanation will be discussed in the chapter 3. However, changing those parameters is impractical in the experimental condition.

Chapter 3

Numerical simulation of photonic nanojet in water medium

This chapter is adapted partially from article titled "First step toward laser micromachining realization by photonic nanojet in water medium [73]"

In the manufacturing process, having predictive capabilities is crucial for simulating procedures before building the established manufacturing system. Numerical simulations play a key role in advancing the science of micro- and sub-microfabrication for predicting the laser ablation results. In this research, it is important to understand how laser beams forms a photonic nanojet (PNJ) predictably. The Finite-Difference Time-Domain (FDTD) method is a valuable tool for calculating the electromagnetic field generated by the PNJ for machining. Therefore, chapter 3 aims to quantitatively assess the intensity distribution of the PNJ by varying key input parameters and examining their effects on machining in a water medium. In Section 3.1, the FDTD method and the simulation models are introduced. The FDTD simulations in a various liquid medium are addressed in Section 3.2, and the results, along with evaluations for varied parameters, are discussed in Section 3.3. FDTD simulation results for machining are discussed in Section 3.4.

3.1 Numerical simulation methodology

3.1.1 FDTD method fundamentals

The finite-difference time-domain (FDTD) method is a widely used technique for analyzing electromagnetic fields. It is based on wave-optics principles. Unlike many other electromagnetic simulation methods that operate in the frequency domain, FDTD solves the optical equations in the time domain. K. Yee [86] originally proposed the method in the early 1970s, and since then, other researchers have made various improvements to it [87, 88].

In a brief step, the FDTD method starts from Maxwell's curl equation for free space, which is given by

$$\frac{\partial E}{\partial t} = \frac{1}{\epsilon_0} \times H, \quad (3.1)$$

$$\frac{\partial H}{\partial t} = \frac{1}{\mu_0} \times E, \quad (3.2)$$

with E and H are vectors in three dimensions, ϵ_0 and μ_0 are the permittivity and the magnetic permeability, respectively, and t is time. In general, Equations (3.1) and (3.2) represent three equations

representing three defined axes. To simplify the explanation, a simple one-dimensional case using only E_x and H_y so that Equation (3.1) and (3.2) become

$$\frac{\partial E_x}{\partial t} = \frac{1}{\epsilon_0} \frac{\partial H_y}{\partial z}, \quad (3.3)$$

$$\frac{\partial H_y}{\partial t} = \frac{1}{\mu_0} \frac{\partial E_x}{\partial z}. \quad (3.4)$$

Equation (3.3) and (3.4) are the equations of a plane wave with the electric field oriented in the x -direction, the magnetic field oriented in the y -direction, and the wave traveling in the z -direction. Next, by taking central difference approximations for both the temporal and spatial derivatives, it gives

$$\frac{E_x^{n+1/2}(k) - E_x^{n-1/2}(k)}{\Delta t} = \frac{1}{\epsilon_0} \frac{H_y^n(k + \frac{1}{2}) - H_y^n(k - \frac{1}{2})}{\Delta x}, \quad (3.5)$$

$$\frac{H_y^{n+1}(k + \frac{1}{2}) - H_y^n(k + \frac{1}{2})}{\Delta t} = -\frac{1}{\epsilon_0} \frac{E_x^{n+1/2}(k + 1) - E_x^{n+1/2}(k)}{\Delta x}. \quad (3.6)$$

In Equation (3.5) and (3.6), time is specified by the superscripts, " n " means a time step $t = \Delta t \cdot n$. The term $n + 1$ means a one-time step later. The term in parentheses represents distance, " k " means the distance $z = \Delta x \cdot k$. Since the FDTD is discrete in space and time, the Equation (3.5) and (3.6) assumes that the E and H fields are interleaved in both space and time. H uses the arguments $k + 1/2$ and $k - 1/2$ to indicate that the H field values are assumed to be located between the E field values. This step calculation is shown schematically in Figure 3.1. Figure 3.1 shows that in every step, three arrows are pointing towards it. Of these arrows, one comes from a half-step down and half-step to the right or left. This FDTD method is also referred to as the leapfrog method. Then, Equation (3.5) and (3.6) could be rearranged in an iterative algorithm as follows

$$E_x^{n+1/2}(k) = E_x^{n-1/2}(k) - \frac{\Delta t}{\epsilon_0 \cdot \Delta x} \left[H_y^n \left(k + \frac{1}{2} \right) - H_y^n \left(k - \frac{1}{2} \right) \right], \quad (3.7)$$

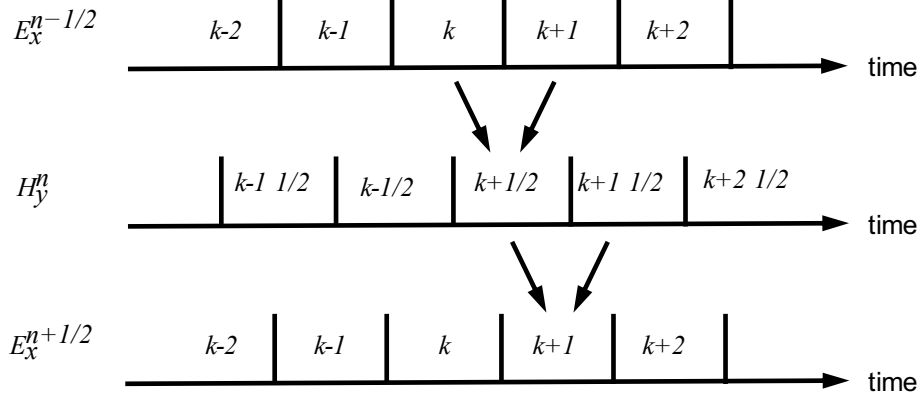


Fig. 3.1 Yee's one-dimensional scheme for interleaving of the E and H fields in space and time in the FDTD formulation.

$$H_y^{n+1} \left(k + \frac{1}{2} \right) = H_y^n \left(k + \frac{1}{2} \right) - \frac{\Delta t}{\mu_0 \cdot \Delta x} \left[E_x^{n+1/2}(k+1) - E_x^{n+1/2}(k) \right]. \quad (3.8)$$

Equations (3.7) and (3.8) are the fundamental paradigms of the FDTD method. Equations (3.7) and (3.8) are very similar, however, due to ϵ_0 and μ_0 differ by several orders of magnitude, E_x and H_y will differ by several orders of magnitude. This is circumvented by making the following change of variables introduced by Tavlofe [89] as follows

$$\tilde{E} = \sqrt{\frac{\epsilon_0}{\mu_0}} E. \quad (3.9)$$

Then, by substitute Equation (3.9) to Equation (3.7) and (3.8) and dropping the symbol " $\sim$ ", the equation become

$$E_x^{n+1/2}(k) = E_x^{n-1/2}(k) - \frac{1}{\sqrt{\mu_0 \epsilon_0}} \frac{\Delta t}{\Delta x} \left[H_y^n \left(k + \frac{1}{2} \right) - H_y^n \left(k - \frac{1}{2} \right) \right], \quad (3.10)$$

$$H_y^{n+1} \left(k + \frac{1}{2} \right) = H_y^n \left(k + \frac{1}{2} \right) - \frac{1}{\sqrt{\mu_0 \epsilon_0}} \frac{\Delta t}{\Delta x} \left[E_x^{n+1/2}(k+1) - E_x^{n+1/2}(k) \right]. \quad (3.11)$$

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Once the cell size Δx is chosen, then the time step Δt to calculate the equation is determined by

$$\Delta t = \frac{\Delta x}{2 \cdot c_0}, \quad (3.12)$$

where c_0 is the speed of light in free space. At last, Eq. (3.10) and (3.11) can be directly implemented in a computer code to do the calculation. As the number of problems grows, two-dimensional (2D) and three-dimensional (3D) FDTD calculations are necessary. The only changes are the definition of the cell since the 2D employs electromagnetic field in x and y direction as the cell for electric field and the magnetic field is shown in Figure 3.2(a) for 2D and (b) for 3D.

The FDTD method is typically used for a closed region in a specific space. However, the Yee algorithm cannot handle cases where the electromagnetic field is calculated at the boundary of the analysis region. Therefore, it is necessary to close the analysis region with a virtual boundary to prevent reflections and scattering. The Perfectly Matched Layer (PML) is preferred because it provides more accurate results than other absorbing boundary conditions.

The flowchart for calculating the electromagnetic field using the FDTD method is shown in Figure 3.3. Firstly, we create a simulation model by specifying the analysis area size, cell size,

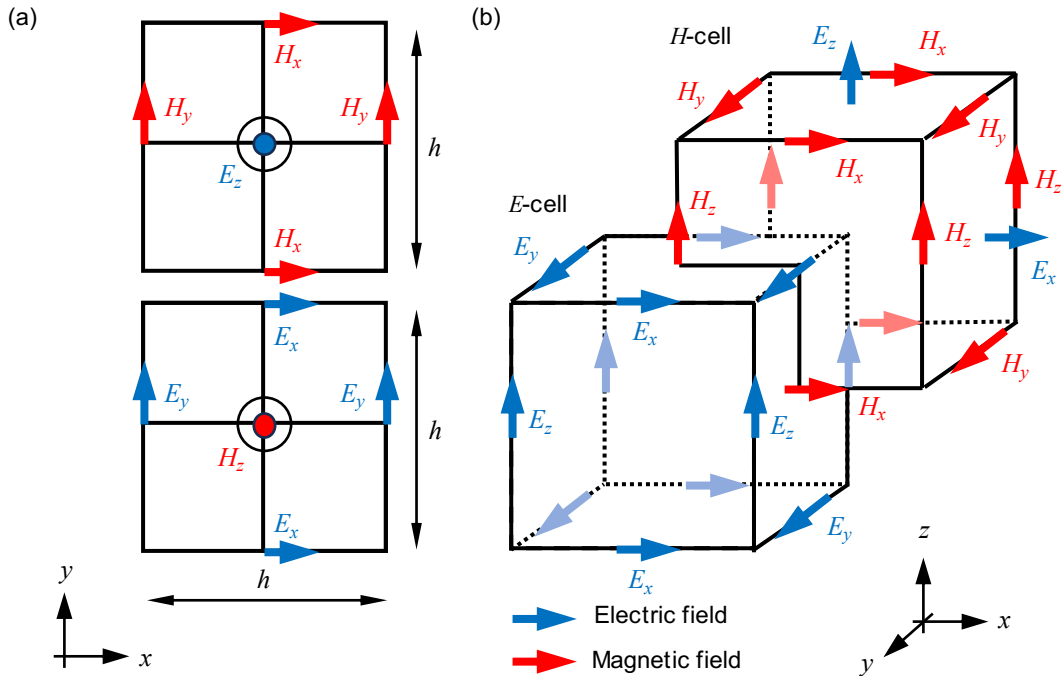


Fig. 3.2 Yee's arrangement of electric and magnetic fields in (a) 2D and (b) 3D.

incident-light parameters, the dielectric medium's physical constants, the maximum simulation time $t_{\max}$, and other parameters. Next, we calculate the electric field distribution E and apply the PML boundary conditions. Then, at the next half-step, we calculate the magnetic field distribution H and apply the PML boundary conditions for H . If the calculation time has not yet reached the preset $t_{\max}$, the calculation restarts to compute the electromagnetic field, and so on as it loops. The calculation process continues until time t exceeds the maximum preset time $t_{\max}$. This means that the calculation of electromagnetic field values proceeds in discrete time steps. One benefit of the time-domain approach is that it provides a broadband output from a single program execution, including not only the steady-state electromagnetic field distribution but also the transient changes. Furthermore, the main reason for using the FDTD approach is its excellent scaling performance as the problem size grows. As the number of unknowns increases, the FDTD method quickly outpaces other methods in efficiency.

3.1.2 FDTD method for machining investigation

For machining applications, it is necessary to estimate the ablation area on the sample surface based on the relationship between the material's fluence threshold and the fluence of the generated PNJ. Therefore, in our research group, we built an electromagnetic field simulation using the FDTD method to estimate the ablation result on the sample surface, using the FDTD method by Uenohara [66]. This simulation could deepen our understanding of PNJ machining control. The algorithm flowchart of the calculation process is shown in Figure 3.4.

The simulation consists of two parts. The first one is used to calculate the electromagnetic field distribution for the PNJ generation. The second one is to determine the ablation result on the surface of the working material. For the former, it uses the scattering and diffraction principle described in Chapter 3.1.1 to generate the PNJ. Therefore, when analyzing the scattering and diffraction phenomena, the primary interest is the intensity distribution of the generated PNJ; the absolute value of the PNJ intensity is rarely used. In this simulation, we use the light intensity of the generated PNJ at the location where the sample is located to check whether it reaches the fluence threshold F_{th} of the working material, and then examine the ablation result on the surface of the working material.

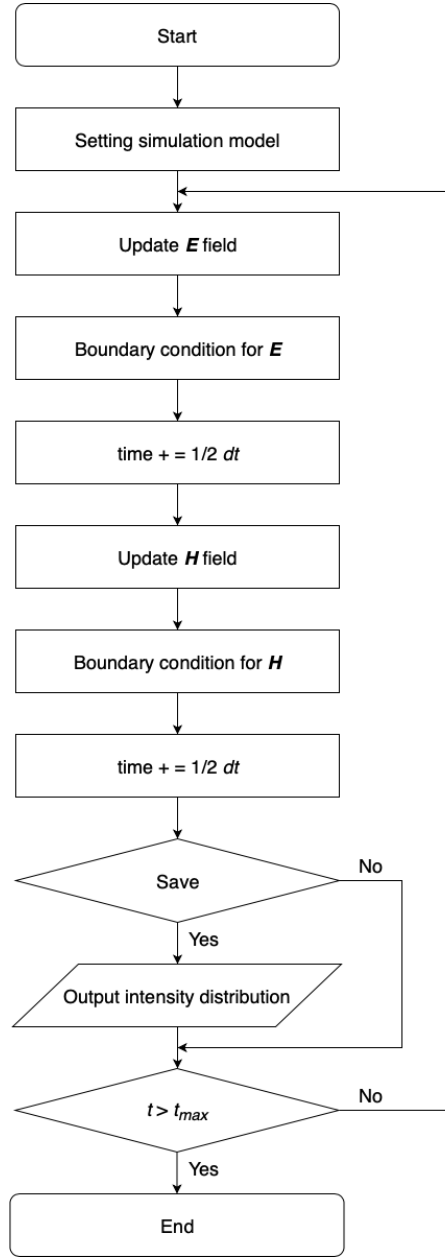


Fig. 3.3 Flowchart of FDTD calculation [66].

3.1.3 Simulation model

In order to calculate the PNJ intensity distribution by the FDTD method, firstly, we define the simulation model as shown in Figure 3.5(a) for 2D analysis and (b) for 3D analysis. In 2D analysis, the analysis region is in x_{range} and z_{range} . An assumption of the infinity range in the y -direction is used. As a result, the microsphere is represented as a microcylinder that extends in the y -direction, not as a

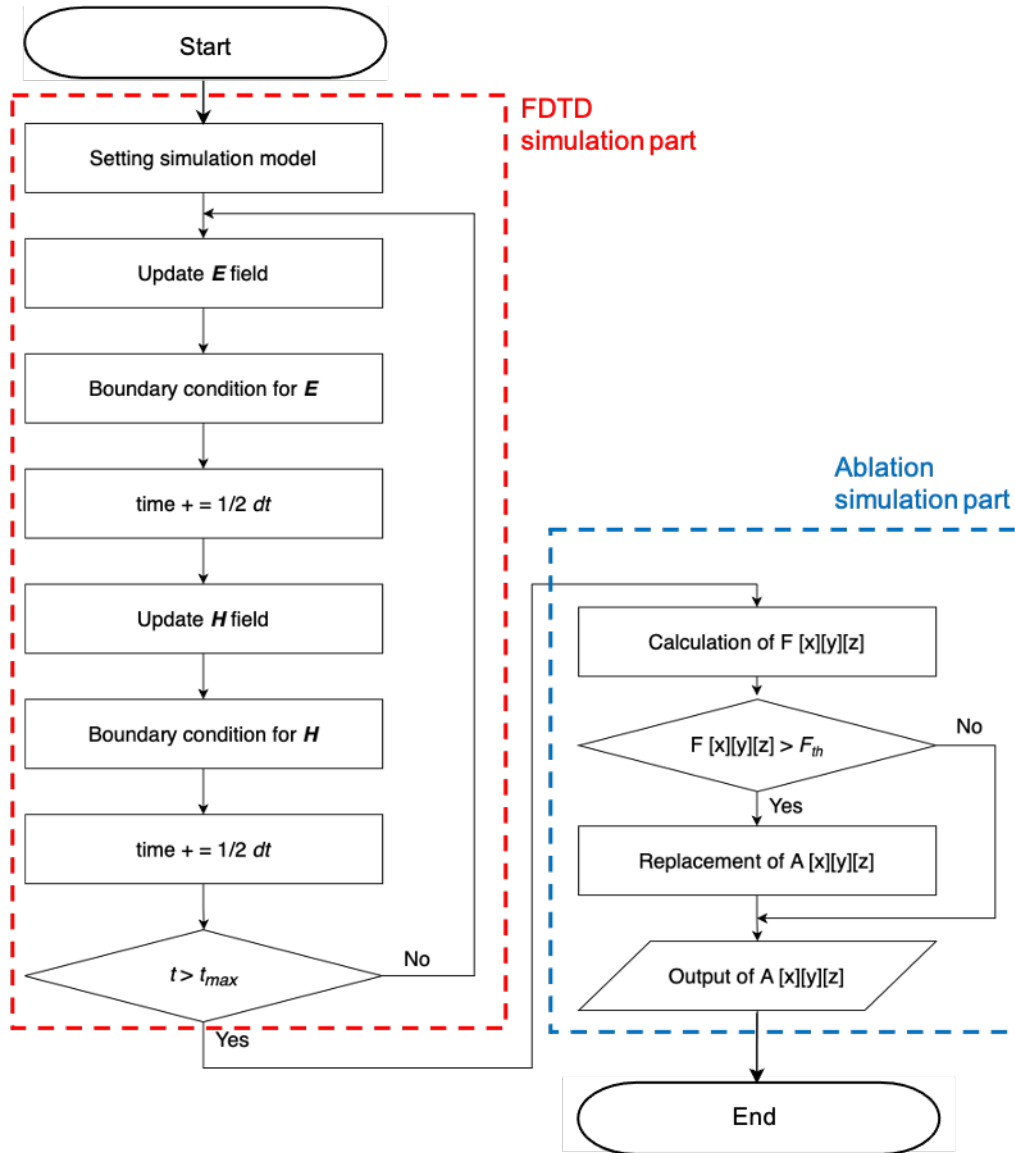


Fig. 3.4 Simulation algorithm of PNJ machining based on FDTD method. E : electric field, H : magnetic field, $t_{\max}$: maximum time, F : fluence, F_{th} : fluence threshold, and A : Physical constant [66].

sphere. On the other hand, the calculation time is faster since only two axes are actively calculating. Thus, a 2D FDTD calculation is more desirable for investigating the tendency of the PNJ intensity distribution with respect to various input parameters, although it does not yield exact results.

Simulation model of PNJ calculation is better explained with the 3D analysis model, as shown in Figure 3.5 (b). In the 3D analysis, the analysis region is defined with x_{range} , y_{range} , and z_{range} which consist of voxel, cell in 3D arrangement, in dx , dy , and dz for respective axis. The incident laser light is applied at $z = 0$ and propagates along the z -axis, irradiating the microsphere. Since PNJ is a light

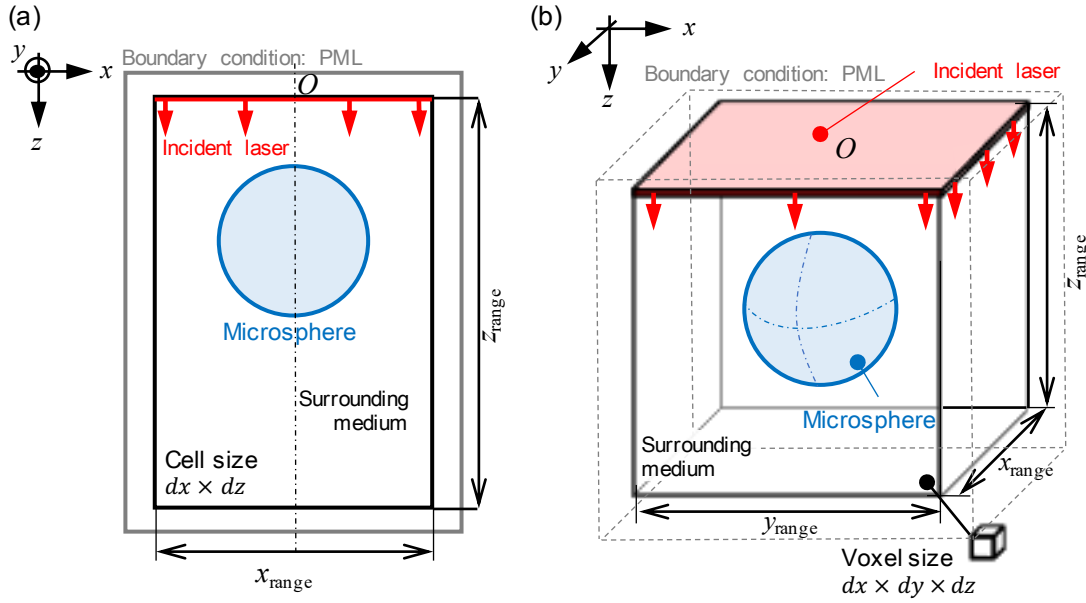


Fig. 3.5 FDTD simulation model for PNJ in (a) 2D and (b) 3D configuration.

scattering and diffraction phenomenon caused by the introduced dielectric microsphere, an absorbing boundary condition to suppress the reflection of the electromagnetic field at the boundary analysis region is required. A PML absorbing boundary is used to surround the analysis area for 2D and 3D FDTD calculations. 3D FDTD calculations require a very robust, high-memory system. Therefore, we run the FDTD program on OCTOPUS (Osaka University Cybermedia center's Over Petascale Universal Supercomputer) and SQUID (Supercomputer for Quest to Unsolved Interdisciplinary Datascience), supercomputer systems at the University of Osaka.

In the previous chapter, we already mentioned several parameters to control the intensity distribution. Therefore, to limit the discussion and scope of this thesis, we focused on the refractive index of the surrounding medium, the size and refractive index of the microsphere, the wavelength, and the beam profile for the laser input parameters. 2D FDTD calculations are used to evaluate the PNJ intensity as a function of changes in the medium, the microsphere, and the laser input wavelength. The 3D FDTD calculation is used to evaluate the intensity distribution from a defined beam profile and machining simulations to better approximate experimental results.

3.2 Photonic nanojet intensity distribution in liquid medium

In Chapter 2, we gain insight into the ray-tracing method and Mie simulation [78] that, in a liquid medium, PNJ tends to form farther from the rear microsphere surface than in the air. However, other parameters of PNJ have not yet been comprehensively evaluated. At first, we employ a 2D FDTD simulation with the analysis conditions shown in Table 3.1. In brief, an 800 nm plane wave illumination laser light is applied on an 8 μm SiO_2 microsphere in air ($n_s = 1.00$) and water medium ($n_s = 1.33$). Although the polarization of the incident laser beam cannot be fully represented in the 2D model, the Transverse Electric (TE) mode, in which the electric field oscillates in the plane of the analysis, is used.

Figure 3.6 shows the results of the simulation done in both air and water. The results are displayed side by side in panels (a) and (b). As seen in Figure 3.6(c) and (d), the PNJ has a larger beam diameter and a longer travel distance in water. It has nearly nine times the WD of the PNJ created in air. This longer WD enables effective surface patterning during PNJ machining in water. It removes the need for the sample to be close to the microsphere. This may make it easier to control the sample and lower the chance of damage from a nearby microsphere. In addition, a longer DOF was obtained in water to increase deep photon absorption and improve material removal in the machining application.

Additionally, during machining, we introduce liquid into the surrounding medium. Various types of liquids are used in industrial applications, including both organic and inorganic solvents, polar and nonpolar molecule. Each liquid has a specific refractive index, so it is important to study how the generated PNJ behaves with different liquid media. Table 3.2 shows the parameters used in

Table 3.1 Simulation parameters for PNJ generated in air and water.

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	(20, 40) μm
	(dx, dz)	(0.01, 0.01) μm
Incident laser	wavelength (λ)	800 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	diameter (d_m)	8 μm
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.00 and 1.33

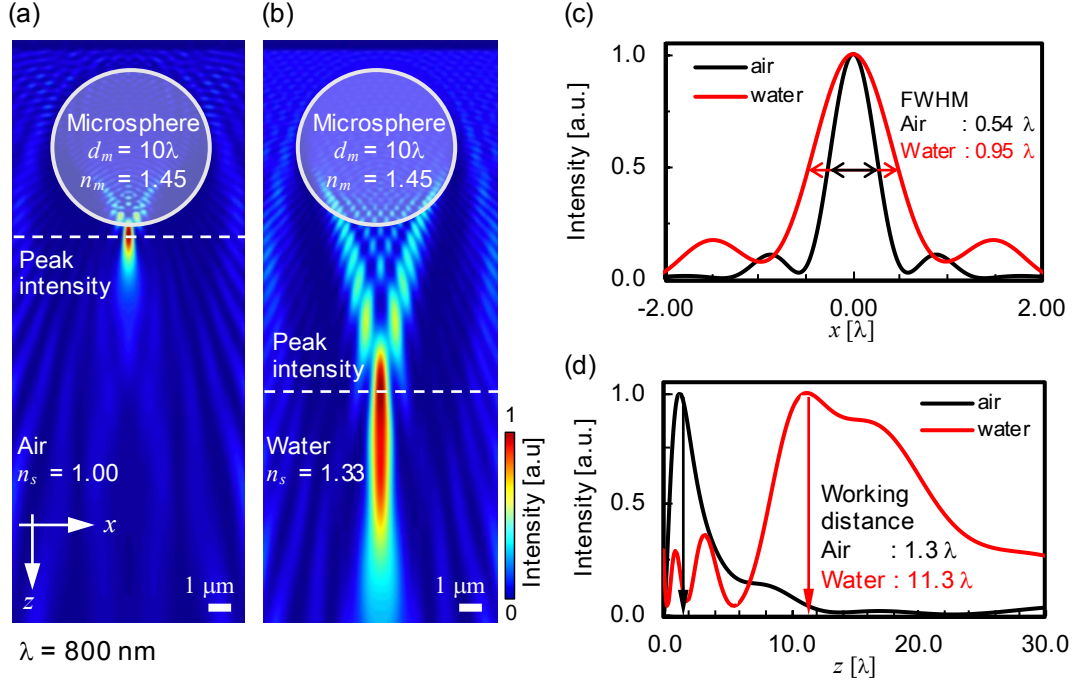


Fig. 3.6 PNJ intensity distribution in (a) air and (b) water medium. Intensity profile on (c) x -direction and (d) z -direction, $z = 0$ represents the microsphere surface.

Table 3.2 Simulation parameters for PNJ controlled by refractive index of surrounding medium.

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	$(20, 40)\ \mu\text{m}$
	(dx, dz)	$(0.01, 0.01)\ \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	diameter (d_m)	8 μm
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.25, 1.26, 1.27, 1.28, 1.29, 1.30, 1.31, 1.32, 1.33, 1.34, 1.35

the simulation based on the surrounding medium. For clarity, we will focus only on liquids with a refractive index (n_s) between 1.25 and 1.35. This range includes Chlorodifluoromethane (R-22), which has an n_s of 1.26, and Hexane (C_6H_{14}), which has an n_s of 1.37.

Figure 3.7 shows the simulated PNJ intensity distributions based on the refractive index of the surrounding medium, n_s . As shown in Fig. 3.7(a), changes in n_s affect the focusing characteristics of the PNJ formed beneath the microsphere. Figure 3.7(b) indicates that the PNJ beam diameter stays

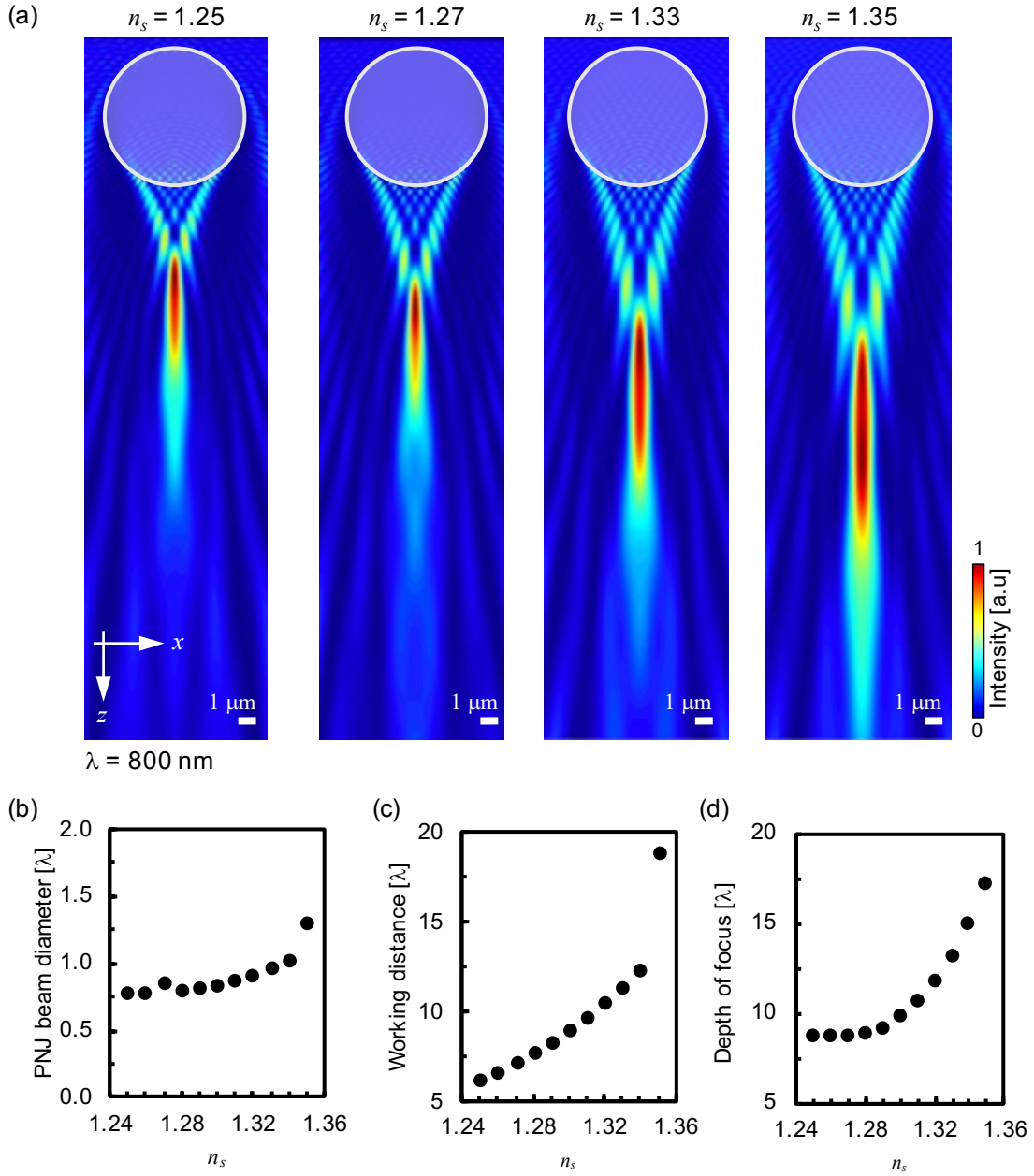


Fig. 3.7 PNJ intensity distribution controlled by refractive index of surrounding medium. (a) intensity distribution, (b) PNJ beam diameter, (c) working distance, and (d) depth of focus.

relatively constant across the range of surrounding refractive indices simulated. This suggests that the lateral confinement of the PNJ only slightly depends on n_s , meaning that the transverse resolution of PNJ-based machining is maintained chiefly even as the surrounding medium changes.

In contrast, Figs. 3.7(c) and (d) reveal a clear relationship between the axial PNJ characteristics and n_s . As the surrounding refractive index increases, both WD and DOF increase steadily. This change is due to the lower refractive index contrast (RIC) between the microsphere and the surrounding medium, which alters the refraction and interference conditions that affect PNJ formation. A higher n_s results in a smoother convergence of the PNJ, which extends the focal region along the direction of propagation. The extended WD and DOF is especially beneficial for PNJ-assisted machining and optical examination process. It eases alignment challenges and allows effective energy delivery from greater distances to the particle surface. This feature is particularly useful when processing or analyzing structures embedded in transparent materials, such as quartz, or when particles are separated from the substrate by a liquid layer [78]. Overall, these findings show that adjusting the surrounding refractive index offers a useful way to control the axial extent of the PNJ without affecting its lateral resolution.

3.3 Parametric analysis of photonic nanojet intensity distribution

3.3.1 Influence of microsphere parameters

In the PNJ machining technique, a preparatory step is needed to position and select the microsphere correctly before machining begins. These microspheres have specific shapes, sizes, and materials set by the manufacturer. Therefore, understanding how microsphere parameters affects the generated PNJ is vital before choosing the size or material for an experiment or application. In this analysis, we examine the PNJ intensity distribution as a function of microsphere parameters. We perform a 2D FDTD simulation to examine microspheres with different diameters and refractive index, thereby focusing our study. Although we consider the microsphere as a perfect circular cylinder in this 2D simulation, it still provides valuable insights into the generated PNJ distribution.

In summary, PNJ results from the scattering of light that hits the microsphere. The distribution of scattered light depends on the size of the scattering particle and the wavelength of the incoming light. To measure this relationship, we present the size of the microsphere as a ratio of its diameter (d_m) to the incident laser wavelength (λ). Table 3.3 shows the parameters used in the simulation for

Table 3.3 Simulation parameters for PNJ controlled by microsphere diameter.

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	$(20, 40) \mu\text{m}$
	(dx, dz)	$(0.01, 0.01) \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	d_m/λ	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.33

various microsphere sizes, with d_m/λ values ranging from 1 to 15. The incident light source is a laser operating at 800 nm, while all other parameters remain the same as in previous simulations.

Figure 3.8 shows the PNJ intensity distributions for microspheres of different diameters. The results indicate that a photonic nanojet forms when the normalized microsphere diameter d_m/λ equals or exceeds 2. Below this point, a clear PNJ is not seen.

As seen in Fig. 3.8(b), the PNJ beam diameter changes only slightly as the microsphere diameter increases. This means it relies only slightly on the microsphere's size. Similarly, Fig. 3.8(d) shows that the DOF changes modestly but stays within a narrow range across the examined diameters. These findings suggest that neither the PNJ beam diameter nor the DOF changes significantly with variations in microsphere size over the tested range. In contrast, Fig. 3.8(c) reveals a strong connection between microsphere size and WD. The WD steadily increases as the microsphere diameter larger. This means that larger microspheres create PNJs at greater distances from the microsphere surface. This feature is especially helpful for PNJ machining, as a longer working distance enables easier alignment control of the PNJ focal point relative to the target surface. From the viewpoint of stable retention of microsphere and availability in the, $8 \mu\text{m}$ microsphere diameter, $d_m/\lambda = 10$, is chosen in this study.

Furthermore, the refractive index of the microsphere (n_m) is an important parameter for light focusing when the microsphere is treated as a micro-lens. In order to generate PNJ, it is necessary to use a transparent dielectric particle, e.g., glass, quartz, or polystyrene, to transmit considerable light into the sample. Table 3.4 shows the parameters used in the simulation, with an $8 \mu\text{m}$ microsphere

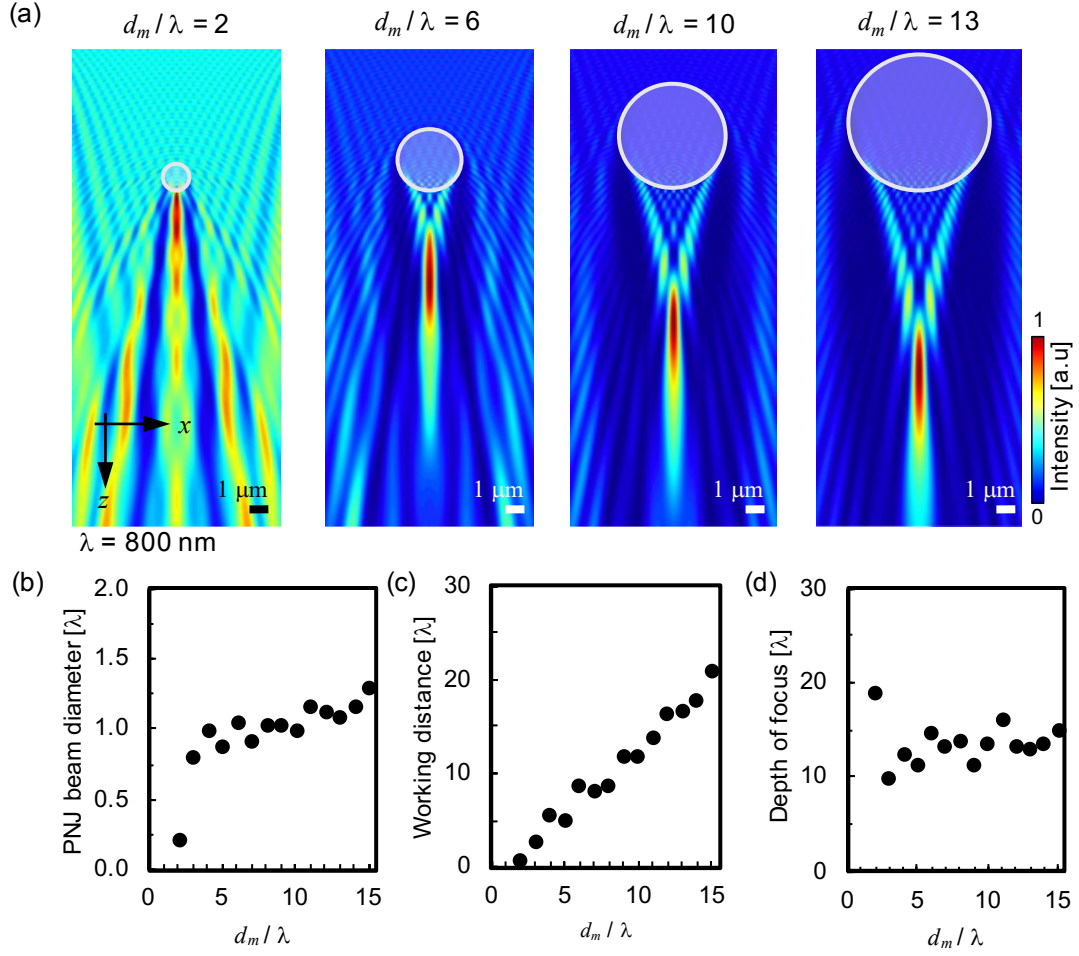


Fig. 3.8 PNJ intensity distribution controlled by microsphere diameter. (a) intensity distribution, (b) PNJ beam diameter, (c) working distance, and (d) depth of focus.

used and a refractive index of microsphere values varied from 1.39 to 1.51. The other parameters are the same as those used in the previous simulation.

Figure 3.9 shows the PNJ intensity distributions for microspheres with different refractive index. The results indicate that the optical properties of the PNJ depend heavily on the refractive index of the microsphere material. As seen in Fig. 3.9(b), the PNJ beam diameter gets smaller as the microsphere refractive index, n_m , increases. This narrowing suggests better light confinement due to the bigger refractive difference between the microsphere and its surroundings. A similar pattern is observed for the WD and DOF as n_m increases; both decrease, as shown in Figs. 3.9(c) and 3.9(d). These patterns imply that microspheres with a higher refractive index produce more focused PNJs near their surfaces.

Table 3.4 Simulation parameters for PNJ controlled by refractive index of microsphere.

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	$(20, 40) \mu\text{m}$
	(dx, dz)	$(0.01, 0.01) \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	diameter	$8.0 \mu\text{m}$
	refractive index (n_m)	1.39, 1.41, 1.43, 1.45, 1.47, 1.48, 1.51
Surrounding medium	refractive index (n_s)	1.33

Table 3.5 Simulation parameters for PNJ controlled by RIC and Δ .

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	$(20, 40) \mu\text{m}$
	(dx, dz)	$(0.01, 0.01) \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	diameter	$8.0 \mu\text{m}$
	refractive index (n_m)	$1.30 \sim 2.73$ (for $n_s = 1.30$) and $1.5 \sim 3.15$ (for $n_s = 1.50$)
Surrounding medium	refractive index (n_s)	1.30 and 1.50

In comparison, those with a lower index produce wider PNJs with longer WD and extended DOF. Therefore, the choice of microsphere material should be carefully matched to the specific application and machining conditions to balance spatial resolution with focal accessibility and process stability.

One interesting parameter that has yet to be comprehensively studied is Refractive Index Contrast (RIC), defined with n_m/n_s , and the difference of refractive index Δ , defined with $n_m - n_s$. Table 3.5 shows the parameters used in the simulation, where an $8 \mu\text{m}$ microsphere is used and has a refractive index of microsphere values varied to have RIC from 1.0 to 2.0 in the different surrounding medium that has refractive index values of 1.30 and 1.50.

Figures 3.10(a) to (f) show the PNJ intensity distributions for different RIC values. From Figs. 3.10(a) and 3.10(f), it is clear that RIC greater than 1 is necessary for PNJ formation. Without this condition, a distinct PNJ does not emerge. For a dielectric microsphere with RIC between 1

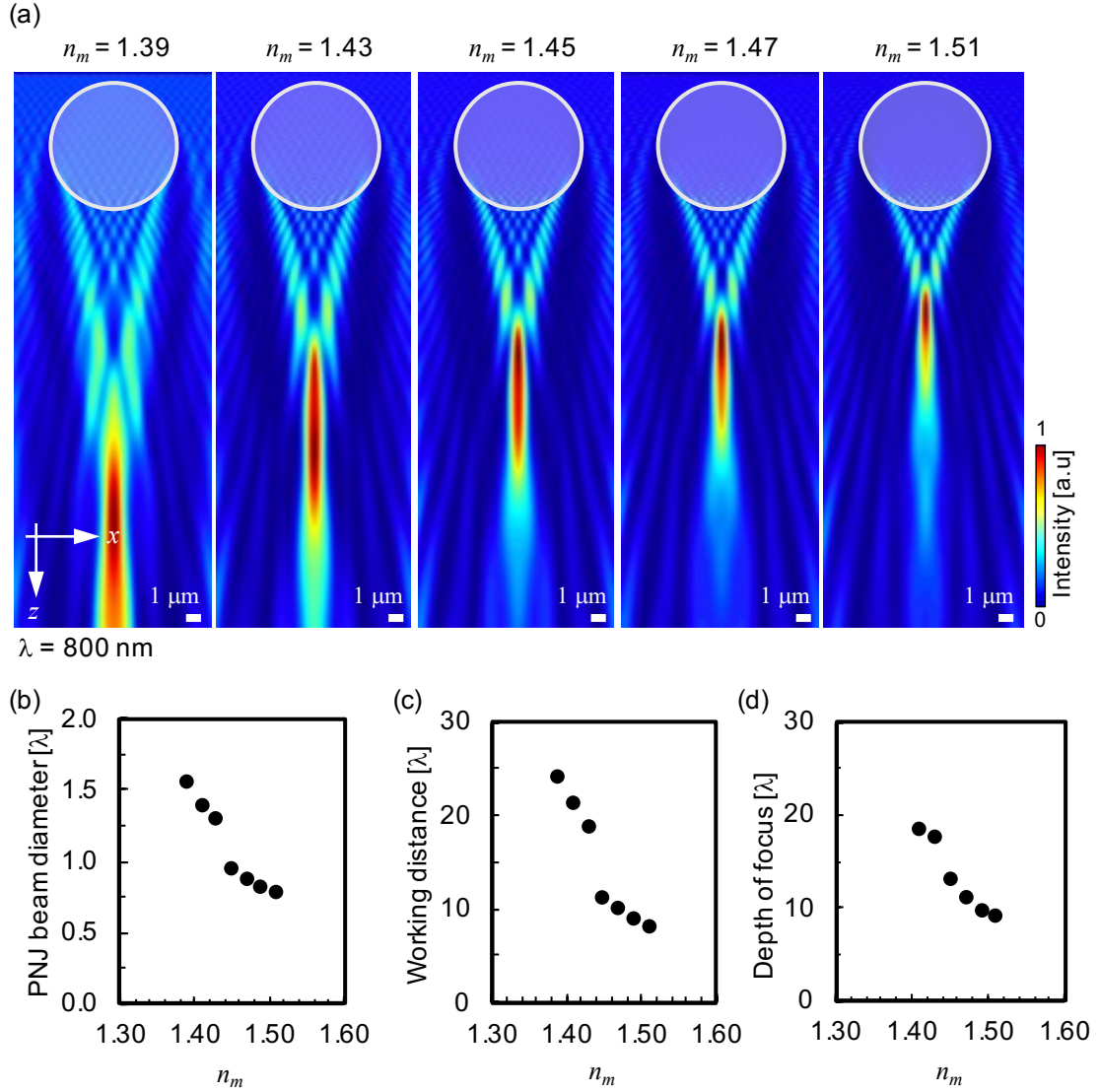


Fig. 3.9 PNJ intensity distribution controlled by refractive index of microsphere. (a) intensity distribution, (b) PNJ beam diameter, (c) working distance, and (d) depth of focus.

and 2 under plane-wave illumination, a PNJ appears at a finite distance from the rear surface of the microsphere. When RIC equals 2, the focal point is located directly at the shadow-side rim of the spherical lens. This is shown in Figs. 3.10(c) and 3.10(f) and aligns with previous reports [90]. As RIC increases, the characteristics of the PNJ change noticeably. Specifically, the PNJ beam diameter decreases quickly with rising RIC. This is alongside a significant drop in both the working distance (WD) and the depth of field (DOF), as shown in Figs. 3.10(g), 3.10(h), and 3.10(i). For RIC

values above 1.6, the DOF becomes hard to define. A considerable part of the PNJ is confined to the microsphere, rather than creating a clear PNJ.

Moreover, even with the same RIC value, the parameter Δ affects the PNJ properties. As shown in Fig. 3.10(g), larger Δ values result in a smaller PNJ beam diameter for a fixed RIC. This pattern matches the trends seen with increasing RIC. A similar relationship with Δ is noted for the DOF, which decreases as Δ increases. In contrast, the PNJ working distance does not show a clear relationship with Δ . These results emphasize the importance of considering both RIC and the relative refractive index difference between the microsphere and the surrounding medium when designing PNJ-based systems. Selecting the right microsphere material and immersion medium is crucial for achieving the desired PNJ characteristics for specific applications.

3.3.2 Influence of input laser beam parameters

Regarding the PNJ machining application, the laser beam is easily manipulated through changing wavelength, beam profile, polarization, and angle of incidence. In this study, we evaluate the PNJ intensity distribution only by varying the input wavelength and beam profile, as this is easiest to do during the machining process. In laser material processing, the wavelength is an important parameter because the material's absorption coefficient varies with incident wavelength. Moreover, the incident light wavelength can be easily adjusted using a tunable laser in the experiment. Table 3.6 shows the parameters used in the simulation with 8 μm SiO_2 submerged in a water medium. The wavelength varies from the visible light range to the near-infrared region, 400 nm $\sim$ 1000 nm. The other parameters are the same as those used in the previous simulation.

Figure 3.11 shows the PNJ intensity distributions for different incident light wavelengths. The results reveal that the properties of the generated PNJ are sensitive to the excitation wavelength, even when the microsphere shape and material are fixed. As shown in Fig. 3.11(b), the PNJ beam diameter gradually increases as the incident wavelength increases. On the other hand, both the WD and the DOF decrease with a longer wavelength, as illustrated in Figs. 3.11(c) and 3.11(d). These patterns suggest that shorter wavelengths create more tightly confined PNJs, but at positions further from the microsphere surface and with more axial reach. From a machining perspective, this behavior indicates a balance between spatial resolution and focal point accessibility. When high processing

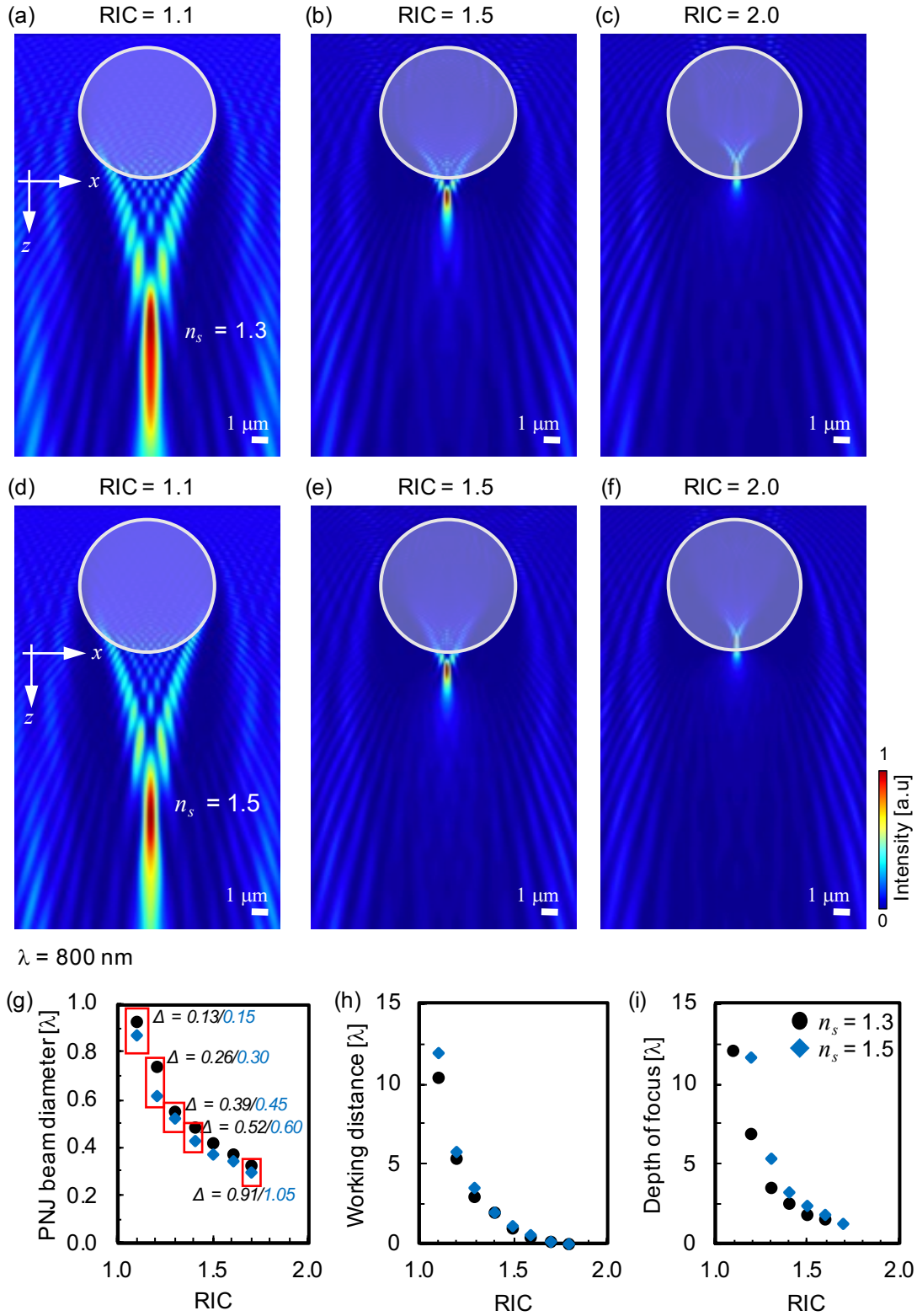


Fig. 3.10 PNJ intensity distribution controlled by RIC and Δ : When $n_s = 1.30$ with RIC (a) 1.1, (b) 1.5, and (c) 2.0; When $n_s = 1.50$ with RIC (a) 1.1, (b) 1.5, and (c) 2.0; (g) PNJ beam diameter; (h) working distance; and (i) depth of focus.

Table 3.6 Simulation parameters for PNJ controlled by incident wavelength.

Item	Parameter	Value
Model	$(x_{\text{range}}, z_{\text{range}})$	$(20, 40) \mu\text{m}$
	(dx, dz)	$(0.01, 0.01) \mu\text{m}$
Incident laser	wavelength (λ)	400, 450, 500, 550, 600, 650, 700, 750, 800, 850, 900, 950, 1000 nm
	wave mode	TE
	intensity distribution	plane wave
Microsphere	diameter	$8.0 \mu\text{m}$
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.33

resolution is required, using a shorter incident wavelength may help, given the smaller PNJ beam diameter. Additionally, the wavelength dependence of optical absorption in both the surrounding medium (e.g., water) and the microsphere material must be considered. Since absorption losses and thermal effects vary with wavelength, selecting the right excitation wavelength is crucial to deliver energy while minimizing unwanted heating or attenuation efficiently.

In the real laser processing experiment setup, an objective lens is typically used to focus the light into the sample, or, in the case of the PNJ system, the light is focused before irradiating the microsphere. Therefore, PNJ distribution analysis is more desirable to evaluate using a focused laser beam. Devilez et al. and Yousefi et al. [91, 92] suggested that using a focused laser beam could yield a smaller PNJ beam diameter. Figure 3.12 shows an illustration when a focused laser beam irradiates the microsphere with the optical axis of the focused laser beam aligned with the central axis of the microsphere and defocusing distance, z_{sp} , between the focal point of the laser beam and the microsphere center surface. Then, the PNJ intensity distribution is analyzed by varying the defocus distance from -20 to $+8 \mu\text{m}$, as shown in Table 3.7. The other parameters used are the same as those in the previous simulation for the microsphere and the surrounding medium. Since this simulation requires a good approximation of incident light, 3D FDTD is more suitable.

Figure 3.13 shows the result of PNJ intensity distribution when the amount of z_{sp} varied. As shown in Figure 3.13(a), when z_{sp} is 8 and $0 \mu\text{m}$, the peak intensity is generated before and inside the microsphere, respectively. In the case of $z_{sp} = 8 \mu\text{m}$, only a small aspect of a focused laser beam

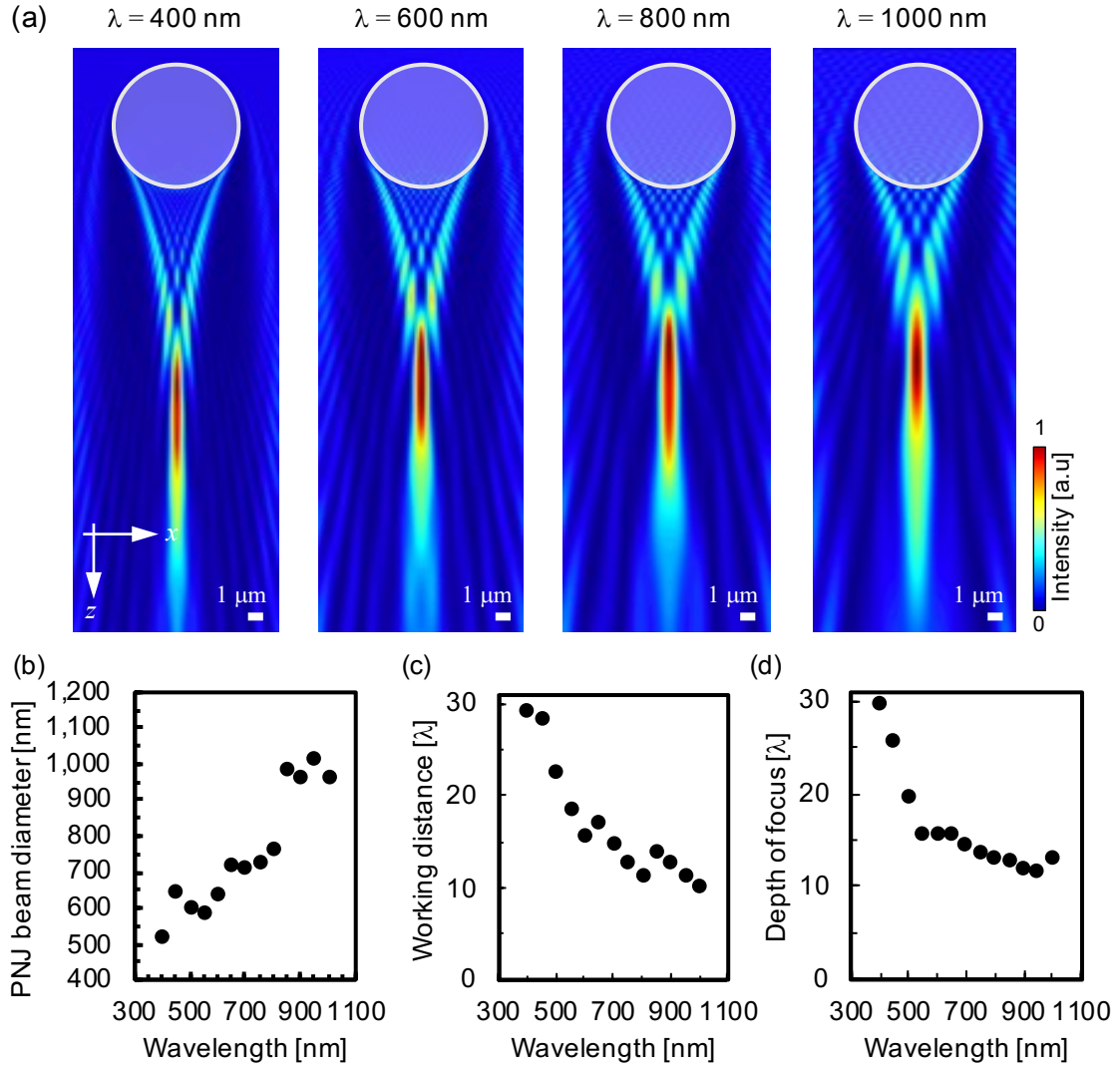


Fig. 3.11 PNJ intensity distribution controlled by laser light wavelength. (a) intensity distribution, (b) PNJ beam diameter, (c) working distance, and (d) depth of focus.

irradiates the microsphere. When the amount of z_{sp} decreases, at $4 \mu\text{m}$, unique phenomena occur when PNJ appears on the background medium. Then on, PNJ does not appear until $z_{sp} = -4 \mu\text{m}$. These unique phenomena are attributed to the Gaussian beam illuminating the microsphere. If the beam size value changes, this unique phenomenon might change into another position or even vanish. An appreciable PNJ starts when $z_{sp} = -6 \mu\text{m}$. Although some of the light is not yet transmitted to the background medium, it is still considered a formidable PNJ. However, the DOF value is difficult to determine because the light has not yet fully reached the background. When $z_{sp} = -14 \mu\text{m}$, the

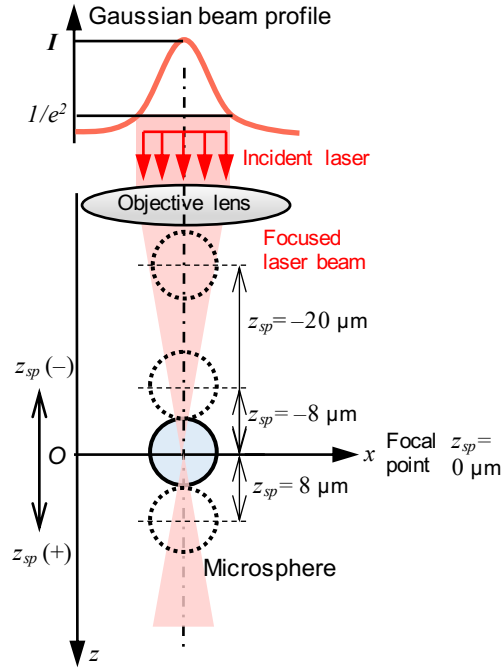


Fig. 3.12 Schematic diagram of focused laser beam irradiating the microsphere.

Table 3.7 Simulation parameters for PNJ controlled by focused laser beam.

Item	Parameter	Value
Model	$(x_{\text{range}}, y_{\text{range}}, z_{\text{range}})$	$(24, 24, 40) \mu\text{m}$
	(dx, dy, dz)	$(0.02, 0.02, 0.02) \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	beam profile	Gaussian beam with $1/e^2=16 \mu\text{m}$
	polarization	x linear
	intensity distribution	focused laser beam ($NA = 0.5$)
	z_{sp}	$-20, -18, -16, -14, -10, -8, -6, -4, -2, 0, 2, 4, 6, \text{ and } 8 \mu\text{m}$
Microsphere	diameter	$8.0 \mu\text{m}$
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.33

smallest PNJ beam diameter is shown as the trend, also shown in Figure 3.13(b). Then, at $z_{sp} = -20 \mu\text{m}$, some lights from an irradiating focused laser beam that is not fully covered with a microsphere are confined at some distance. Moreover, WD value and DOF value increased when the value of z_{sp} is decreasing, as shown in Figure 3.13(c) and (d). Therefore, this simulation also confirms that PNJ can

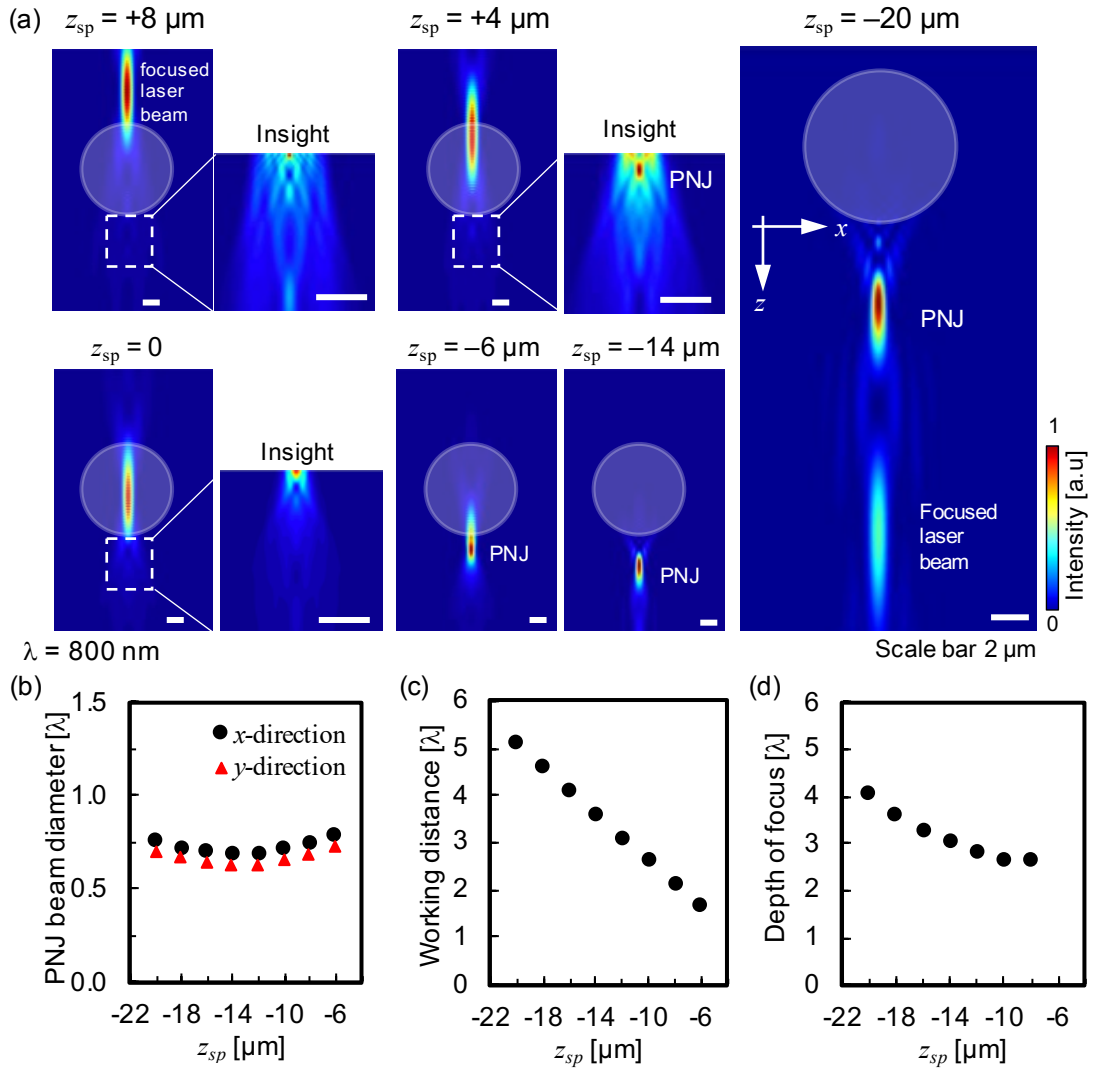


Fig. 3.13 PNJ intensity distribution controlled by z_{sp} amount of incident focused laser beam. (a) intensity distribution, (b) PNJ beam diameter, (c) working distance, and (d) depth of focus.

be generated with a smaller input beam diameter. However, the amount of z_{sp} must be controlled to acquire a sufficient PNJ beam to do the machining process [93].

3.4 Photonic nanojet machining analysis in water medium

In laser machining, we can control the ablation area by adjusting the laser intensity distribution. The area of ablation on the surface of the working material depends on how wide those intensities are when reaching the ablation threshold of the working material as shown in Fig. 2.4. Those principles

become critical when using the PNJ machining method, as we need to consider each parameter, not only the PNJ beam diameter, but also the WD and DOF values. Figure 3.14 shows the 3D simulation model for the PNJ machining investigation, based on 3D FDTD calculations. In this simulation, we focus on the machining results from a generated PNJ that uses $8\ \mu\text{m}$ silicon dioxide ($n_m = 1.45$) submerged in a water medium ($n_s = 1.33$). The generated PNJ would ablate the silicon wafer, which has a complex permittivity for light ($n = 3.692 + 0.006i$). The distance between the microsphere rear surface and the silicon surface (h) is subject to change with a $0.5\ \mu\text{m}$ pitch. The simulation runs from $h = 0$ (when the microsphere is on the silicon surface) to $7\ \mu\text{m}$ for both the polystyrene (PS) and SiO_2 microspheres to accommodate a longer WD. The complete simulation parameter is shown in Table 3.8.

Before the machining simulation begins, the generated PNJ (photonic nanojet) for the machining process exhibits the intensity distribution shown in Figure 3.15. The results are presented side by side in Figure 3.15(a) and (b). As illustrated in Figure 3.15(c) and (d), the PNJ generated from the SiO_2 microsphere has a beam diameter of $0.55\ \mu\text{m}$ at the peak intensity, located $2.86\ \mu\text{m}$ from the rear surface of the microsphere. In contrast, the PS microsphere produces a smaller PNJ beam diameter of $0.42\ \mu\text{m}$. This generated PNJ from the PS microsphere is also positioned closer to the microsphere,

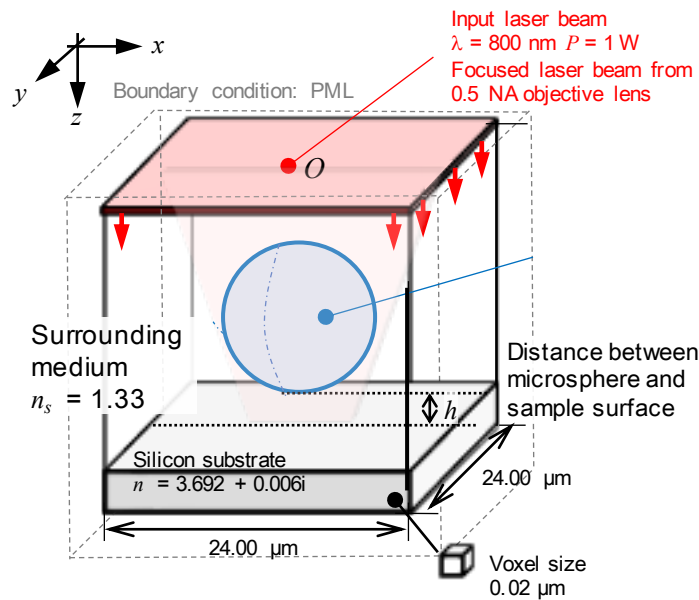


Fig. 3.14 Simulation model for machining investigation by PNJ machining on silicon surface.

Table 3.8 Simulation parameters for machining simulation using PNJ machining with controlled position.

Item	Parameter	Value
Model	$(x_{\text{range}}, y_{\text{range}}, z_{\text{range}})$	$(24, 24, 40) \mu\text{m}$
	(dx, dy, dz)	$(0.02, 0.02, 0.02) \mu\text{m}$
Incident laser	wavelength (λ)	800 nm
	repetition rate	80 MHz
	polarization	x-linear
	intensity distribution	focused laser beam ($\text{NA} = 0.5$)
	z_{sp}	$-14 \mu\text{m}$
	power	1 mW
Microsphere	diameter (d_m)	$8 \mu\text{m}$
	refractive index (n_m)	1.45 (SiO_2) and 1.55 (PS)
Surrounding medium	refractive index (n_s)	1.33
Silicon substrate	distance (h)	0.0 to $7.0 \mu\text{m}$ ($0.5 \mu\text{m}$ pitch)
	refractive index (n)	$3.692 + 0.006 i$
	fluence ablation threshold (F_{th})	190 mJ/cm^2 [36]

with a WD of $1.50 \mu\text{m}$. The confinement of the PNJ generated by the higher refractive index of the PS microsphere results in a greater fluence at the peak intensity compared to that of the SiO_2 microsphere.

Moreover, both PNJs demonstrate sufficient intensity to perform the ablation of a silicon wafer in a water medium, with an ablation threshold fluence of 190 mJ/cm^2 [36]. From panels Figure 3.15(c) and (d), we can conclude that the PNJ can effectively carry out the ablation process on the sample when the distance to the sample, h , is set between 0.5 and $5.0 \mu\text{m}$ using the PS microsphere, and between 1.5 and $6.0 \mu\text{m}$ when using the SiO_2 , as the intensity in the propagation direction reaches the fluence threshold value.

The intensity distribution on the silicon surface with a SiO_2 microsphere is shown in Figure 3.16(a) and (b). In comparison, the intensity distribution with a PS microsphere appears in Figure 3.17(a) and (b). Figure 3.18 presents the expected results for the ablation diameter on the silicon wafer. The figure shows that no ablation happened when the rear surface of the microsphere touched the silicon surface ($h = 0.0 \mu\text{m}$). We expect the ablation process to start at $h = 1.0 \mu\text{m}$ with the PS microsphere and at $h = 2.0 \mu\text{m}$ with the SiO_2 microsphere. Ablation is predicted to stop at $h = 4.5 \mu\text{m}$ for the polystyrene microsphere and at $h = 5.5 \mu\text{m}$ for the SiO_2 microsphere. We can also estimate these values using the known WD value, as shown in Figure 3.15(d).

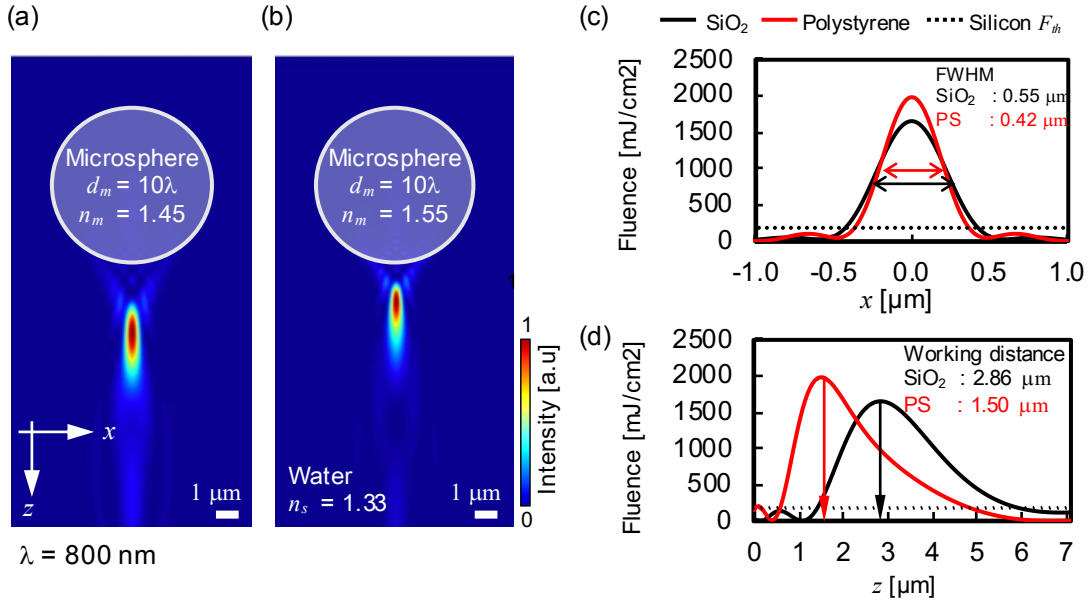


Fig. 3.15 PNJ machining intensity distribution using (a) SiO₂ and (b) Polystyrene. Intensity profile on (c) x-direction at the peak intensity and (d) z-direction, $z = 0$ represents the rear microsphere surface. PS: Polystyrene, F_{th} : fluence threshold.

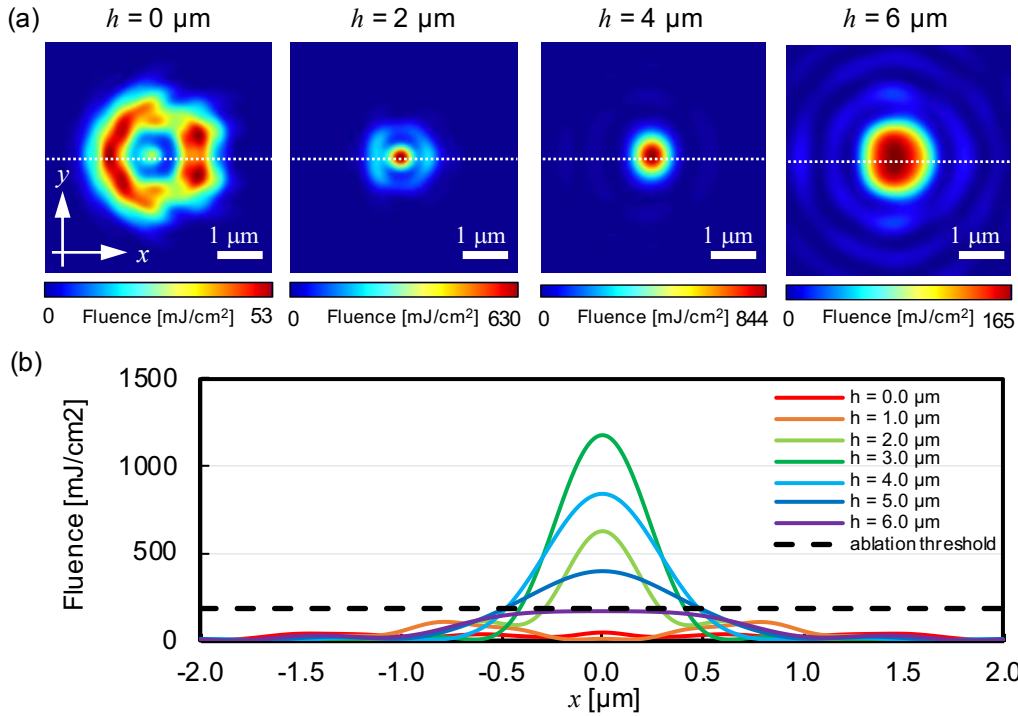


Fig. 3.16 Fluence distribution of the PNJ machining on the silicon substrate by SiO₂ microsphere in (a) xy-plane and (b) x-direction.

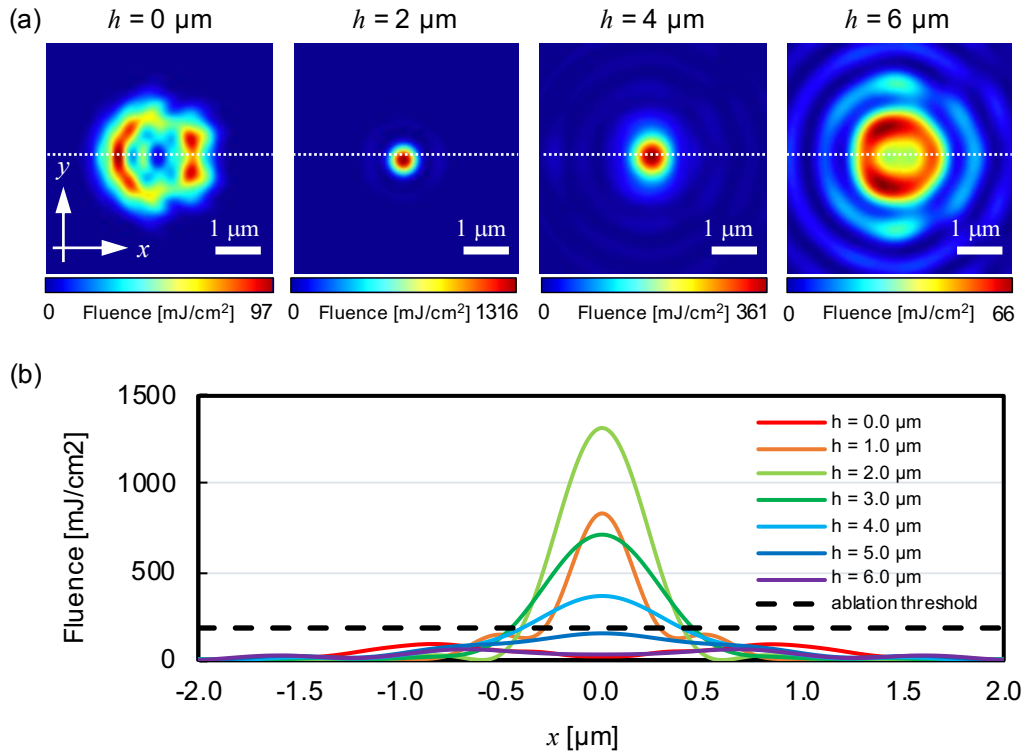


Fig. 3.17 Fluence distribution of the PNJ machining on the silicon substrate by polystyrene microsphere in (a) xy -plane and (b) x -direction.

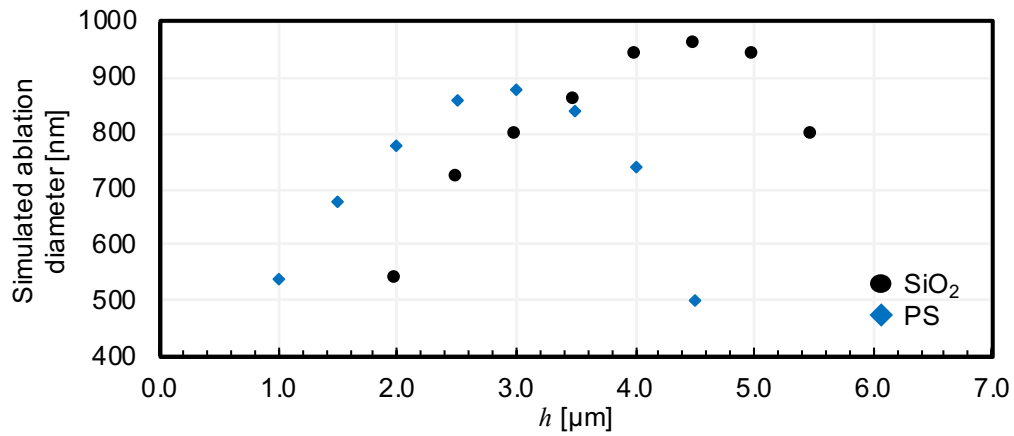


Fig. 3.18 Recap on simulated ablation diameter on silicon surface with different microsphere. PS: Polystyrene.

Regarding ablation diameter, we can achieve measurements as small as 500 nm with the PS microsphere at $h = 4.5 \mu\text{m}$ and 540 nm with the SiO_2 microsphere at $h = 2.0 \mu\text{m}$. For both microsphere types, as the distance h increases, the ablation diameter also grows. This pattern occurs due to a wider

intensity profile in the x -direction as h increases, which is illustrated in Figures 3.16(b) and 3.17(b). It is worth noting that while the PS microsphere offers a higher peak fluence value of 1380 mJ/cm^2 at $h = 1.5 \text{ }\mu\text{m}$ compared to the SiO_2 microsphere's 1158 mJ/cm^2 at $h = 3.0 \text{ }\mu\text{m}$, the PNJ produced by the SiO_2 microsphere is enough to meet the silicon ablation threshold in a water medium. We have shown that PNJ machining can achieve a pattern diameter of 500 nm on a silicon wafer, as predicted by 3D FDTD calculations using both PS and SiO_2 microsphere configurations.

3.5 Discussion

In this chapter, we conducted comprehensive numerical simulations using the FDTD method to analyze PNJ formation and machining capabilities in liquid media. The simulations revealed several key findings that establish the foundation for practical PNJ machining applications. Our analysis of PNJ in liquid media demonstrated that water as a surrounding medium ($n_s = 1.33$) produces significantly different PNJ characteristics compared to air, with approximately nine times longer WD. This extended WD eliminates the need for close proximity between the microsphere and the sample surface, simplifying experimental control and reducing the risk of damage.

Parametric analysis of microsphere properties showed that PNJ formation requires $d_m/\lambda \geq 2$, with larger microspheres producing longer WD values that facilitate position control. The refractive index contrast emerged as a critical parameter, where $1 < \text{RIC} < 2$ enables PNJ generation, with $\text{RIC} = 2$ positioning the focal point precisely at the microsphere's shadow rim. Higher RIC values and refractive index differences (Δ) produce smaller PNJ beam diameters but shorter WD and DOF. Investigation of laser parameters revealed that longer wavelengths generate slightly smaller PNJ beam diameters with reduced WD and DOF. Focused laser beam analysis demonstrated that defocusing distance (z_{sp}) critically influences PNJ formation, with $z_{sp} = -14 \text{ }\mu\text{m}$ producing the smallest beam diameter of approximately $0.55 \text{ }\mu\text{m}$ for SiO_2 microspheres. The summary of parameter effects on the generated PNJ characteristics is shown in Table 3.9. Therefore, to acquire PNJ with submicrometer-scale beam diameter with long depth of focus for higher material removal processing, we need to use a shorter input wavelength.

Table 3.9 Simulation parameters for machining simulation using PNJ machining with controlled position.

Parameter	PNJ beam diameter	Working distance	Depth of focus
Refractive index of surrounding medium (n_s)	Increasing refractive index increases beam diameter (wider PNJ)	Increasing refractive index increases working distance (moves further from microsphere)	Increasing refractive index increases depth of focus (longer PNJ)
Diameter of microsphere (d_m)	Increasing microsphere diameter increases beam diameter (wider PNJ)	Increasing microsphere diameter increases working distance (moves further from microsphere)	Increasing microsphere diameter increases depth of focus (longer PNJ)
Refractive index of microsphere (n_m)	Increasing refractive index decreases beam diameter (narrower PNJ)	Increasing refractive index decreases working distance (moves closer to microsphere)	Increasing refractive index decreases depth of focus (shorter PNJ)
Laser input wavelength (λ)	Increasing wavelength increases beam diameter (wider PNJ)	Increasing wavelength decreases working distance (moves closer to microsphere)	Increasing wavelength increases depth of focus (longer PNJ)

Machining simulations on silicon substrates predicted successful submicrometer-scale ablation, with minimum ablation diameters of 500 nm achievable using PS microspheres and 540 nm using SiO₂ microspheres. The simulations confirmed that both microsphere materials generate sufficient fluence to exceed silicon's ablation threshold (190 mJ/cm²) in water medium, with effective machining ranges of 0.5-5.0 μ m (PS) and 1.5-6.0 μ m (SiO₂) between the microsphere rear surface and substrate. These findings provide quantitative guidelines for selecting optimal parameters in PNJ machining applications and demonstrate the feasibility of achieving submicrometer resolution in water medium.

Chapter 4

Experimental realization of PNJ machining in water medium

This chapter is adapted from article titled "First step toward laser micromachining realization by photonic nanojet in water medium [73]"

4.1 Introduction

This chapter discusses the experimental investigation of laser machining using photonic nanojet phenomena in water. It builds on the theoretical analysis and numerical simulations from Chapter 3. The purpose of this experimental work is to validate performance and show the feasibility of photonic nanojet-assisted laser machining in a water medium [73]. A significant part of this study is the systematic comparison between conventional focused laser beam machining and photonic nanojet-assisted machining, both conducted in water. Conventional laser machining uses objective lenses to focus the laser beam to a spot size defined by the diffraction limit. In contrast, photonic nanojet machining employs dielectric microspheres to achieve sub-wavelength focusing and improve intensity. This comparison enables a direct evaluation of the benefits of the photonic nanojet approach, including improved spatial resolution, smaller feature sizes, and greater machining precision. This chapter is organized as follows. Section 4.2 describes the experimental setup, including the laser system and the optical components. Section 4.3 provides detailed specifications of the specimens used in the experiments. Section 4.4 presents the conventional laser-machining experiments conducted in air and water using a focused laser beam. Section 4.5 focuses on photonic nanojet-assisted machining in a water medium.

4.2 Experimental setup

In this section, we describe the laser machining system designed and constructed to realize the conventional laser machining and the proposed PNJ machining in a water medium. The experiments were all conducted in a clean room, a facility used for specialized industrial production or scientific research, with a constant, controlled temperature and humidity of 23 degrees Celsius and 30%, respectively. An experimental setup was constructed as shown in the schematic diagram, which is shown in Figure 4.1(a), with the insight of the laser machining part is shown in Figure 4.1(b) for focused laser beam experiment and (c) for PNJ machining, both of which are in the water medium. In addition, Figure 4.2 shows a photograph of the actual constructed optical setup in the laboratory. As shown in Figure 4.1(a) and 4.2, the optical setup consists of three components: a laser for machining, indicated with the red color; a position detection system using a confocal optical system, indicated

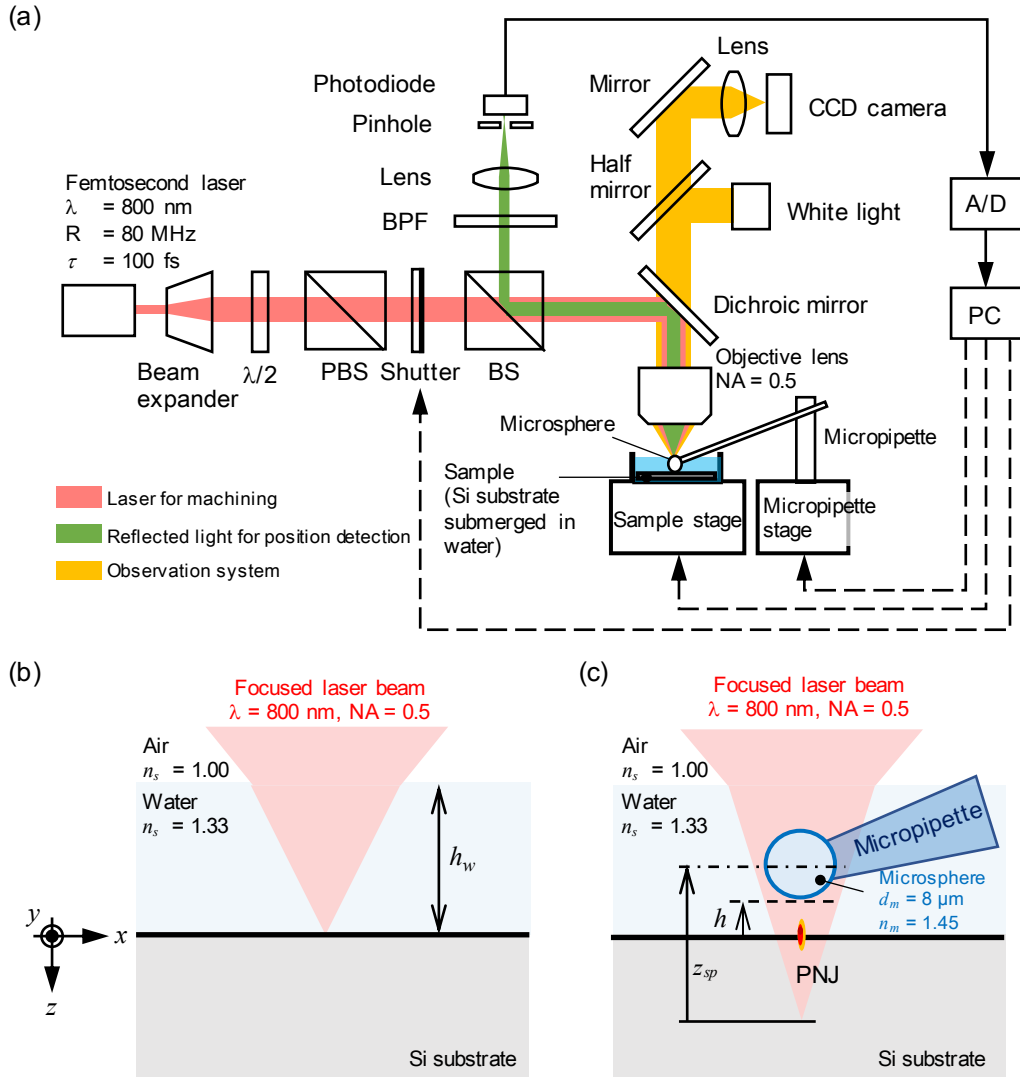


Fig. 4.1 Schematic diagram of (a) experimental setup with insight view of machining part for (b) focused laser beam machining and (c) PNJ machining in water medium. PBS: Polarizing beam splitter, BS: Beam splitter, and BPF: Band pass filter.

with the green color; and an observation system to observe the state of the machining experiment, indicating with the yellow color.

The optical setup is explained as follows. For the laser machining part, the output laser beam from a femtosecond laser device is first reflected by a couple of mirrors to adjust the optical path height. In this research, a femtosecond laser (Spectra-Physics, Maitai) with the specifications shown in Table 4.1 is used. Although this laser is tunable over 690 to 1040 nm, the output excitation laser power is maximized at 800 nm. Based on this, we used a wavelength of 800 nm for all the conducted

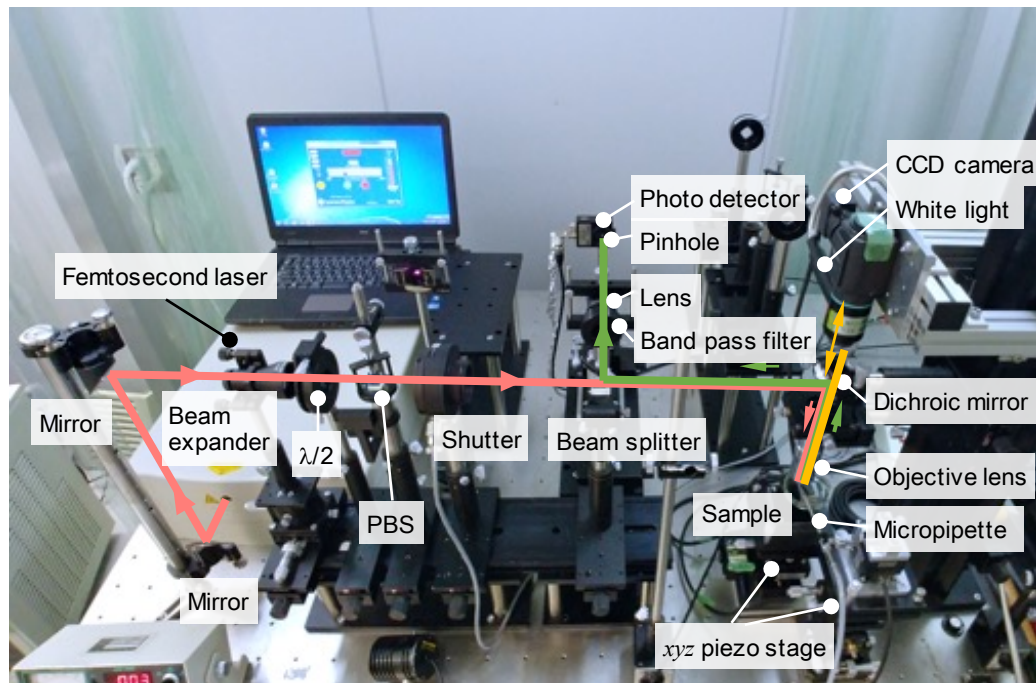


Fig. 4.2 Photograph of constructed experimental setup. PBS: Polarizing beam splitter.

Table 4.1 Specification of femtosecond laser.

Parameter	Value
Pulse duration (τ)	< 100 fs
Wavelength tuning range (λ)	690 - 1040 nm
Average power	> 2.5 W
Peak power	> 300 kW
Astigmatism	< 10 %
Repetition rate (R)	80 ± 1 MHz
Beam pointing stability	< 50 μ rad / 100 nm
Spatial mode	TEM ₀₀ , $M_2 < 1.1$
Polarization	> 500:1 horizontal
Beam divergence	< 1 mrad
Beam diameter	< 1.2 mm

experiments, despite differences in the PNJ intensity distribution obtained with different incident light wavelengths, as explained in the chapter 3.3.2 through numerical simulation. Next, the laser beam diameter is expanded by the beam expander ($\times 3$) to match the known value of objective lens pupil diameter for effectively accommodating the specification of the objective lens (Mitutoyo, M Plan Apo SL 80 $\times$), as shown in Table 4.2.

Table 4.2 Specification of objective lens.

Parameter	Value
Magnification	80
Numerical aperture	0.5
Focal length	2.5 mm
Working distance	15.0 mm
Aperture	2.5 mm

After that, the laser beam passes through an attenuator system comprising a half-wave plate and a polarizing beam splitter (PBS). This attenuator system controls the incident laser power by rotating the half-wave plate. The PBS also acts as a polarizer, and the schematic diagram of the constructed attenuator system is shown in Figure 4.3. The origin of laser light from the femtosecond laser has a linearly polarized profile. Therefore, the azimuthal angle of linearly polarized light can be adjusted by rotating the half-wave plate, which controls the proportions of p- and s-polarized light before they enter the PBS. Then, as shown in Figure 4.3, PBS transmits p-polarized light and reflects the s-polarized light. Then, by rotating the half-wave plate, we could adjust the intensity ratio of p-polarized to s-polarized light at the PBS and generate different amounts of laser power. Since the half-wave plate's rotation angle can easily adjust the power, the incident laser power can be continuously controlled, whether for measurement or a machining process. Figure 4.4(a) shows the result of measured laser power after transmission through the PBS when the half-wave plate was rotated from 0 to 360 degrees with 10-degree increments. From Figure 4.4(a), it can be seen that the laser power is periodically changed due to the azimuth of linearly polarized light changing by 2θ when the half-wave plate is rotated by a certain angle θ . Therefore, when the half-wave plate rotated by 90 degrees, the direction of linearly polarized light changed by 180 degrees, making the intensity ratio between p-polarized and s-polarized light periodic. For practical reasons, the half-wave plate is enough to be rotated from 0 to 45 degrees to accommodate a continuous change of laser power in the range from 0 to 2800 mW, as shown in 4.4(b).

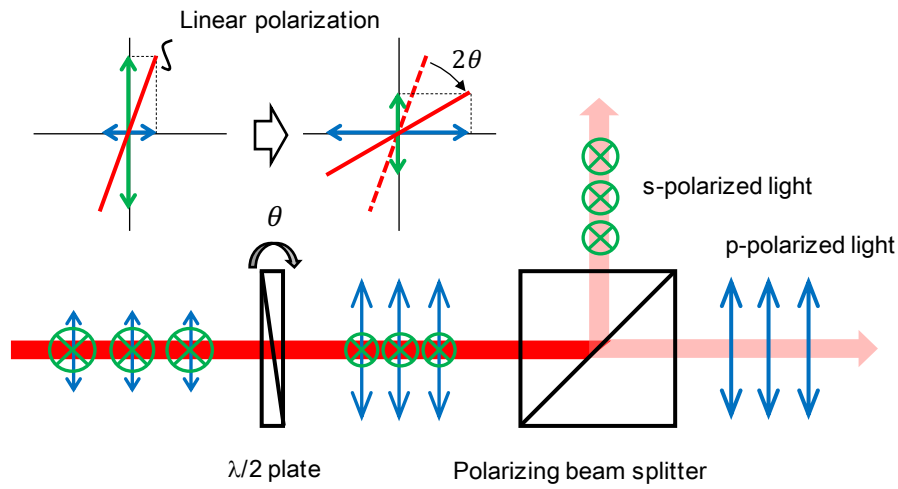


Fig. 4.3 Principle of attenuator system.

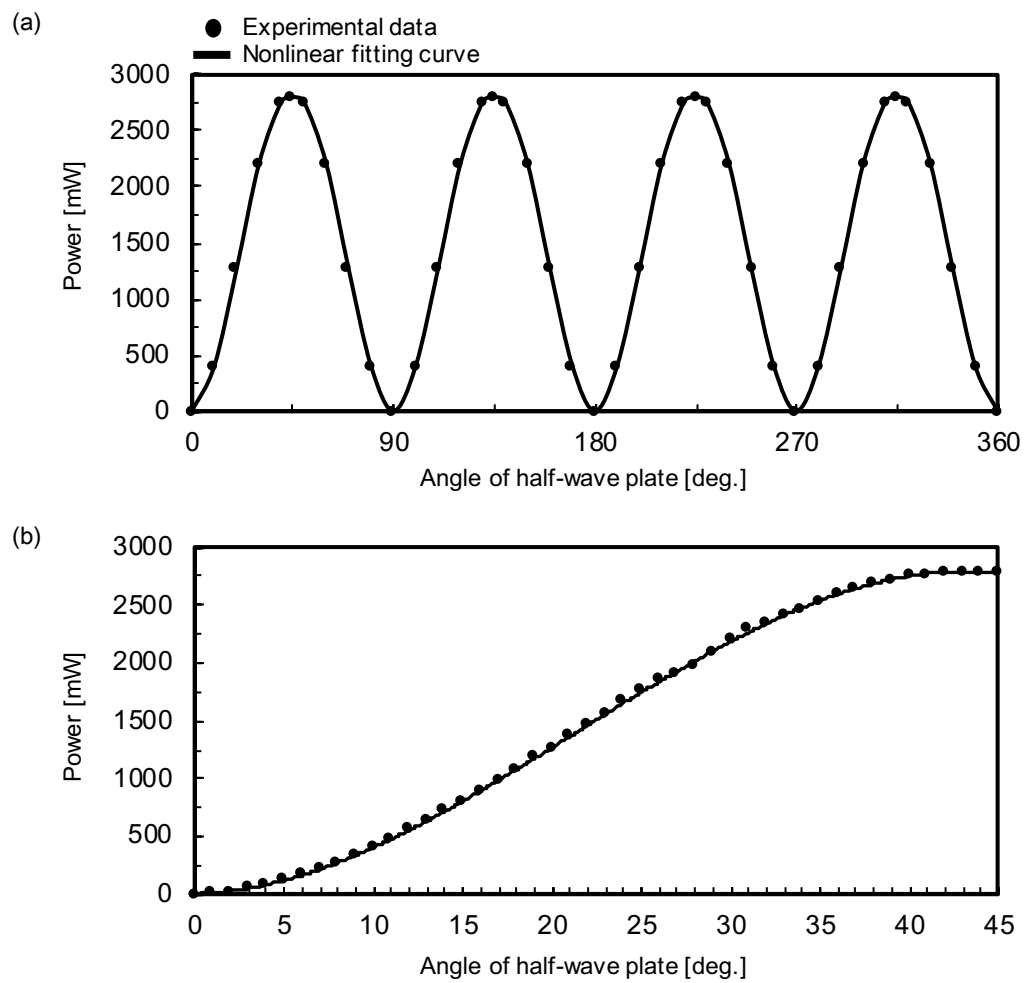


Fig. 4.4 Relationship between the angle of half-wave plate and power. (a) 0 to 360 degrees and (b) 0 to 45 degrees.

Then, the laser beam passes through a high-speed shutter and a beam splitter. In this research, the minimum shutter pulse number of 160,000 will be used. A particular beam splitter is used in this setup, with an R (Reflectance): T (Transmittance) ratio of 1:9 to conserve as much laser power as possible for the laser machining process. Next, the laser beam is reflected by a dichroic mirror (Sigma Koki Ltd., FLM1-25.4C05-800 series) and directed to the objective lens before irradiating the sample or microsphere during the machining process.

To control the irradiation power during machining, it is usually desirable to measure the power after it passes through the objective lens for experimental purposes. However, in the experimental setup, it was challenging to measure the laser power after the objective lens with a power meter because the sample was directly below the power meter. Therefore, to measure the laser power during the experiment, we insert the power meter head between the beam splitter and the dichroic mirror. By measuring the laser power before the objective lens, we estimate the incident irradiance on the sample using a variable called transmittance (T). Transmittance T refers to the power loss rate through the objective lens by merely dividing the power after passing through an objective lens, P_{out} , by the power before entering the objective lens, P_{in} . We calculated from 5 data sets, as shown in Table 4.3, and found the transmittance of the objective lens was 0.39.

Regarding the confocal optical system used to detect the sample surface, indicated by the green line, the explanation is as follows. The confocal system is required to position the sample accurately and the microsphere held by the micropipette relative to the incident laser beam so that the desired PNJ is generated and the ablation is performed on our sample. The reflected laser beam, usually lower in power than the power used for the machining process, from the sample or microsphere surface re-enters the objective lens, is reflected by a dichroic mirror, and propagates to the beam splitter in the

Table 4.3 Calculation of objective lens transmissibility.

Half-wave plate angle [deg.]	P_{in} [mW]	P_{out} [mW]	$T = P_{in}/P_{out}$
10	409	155	0.38
20	1253	502	0.40
30	2200	856	0.39
40	2740	1079	0.39
45	2790	1096	0.39
$T_{average} = 0.39$			

opposite direction of the laser for machining. In the beam splitter, the light is then reflected. Next, it passes through a bandpass filter with a transmission wavelength of 800 ± 5 nm, a lens with a focal length of 200 mm, and a pinhole with a diameter of 25 μm , before the laser light is detected by the photodiode (Thorlabs, DET36A/M). The electrical signal from the photodiode is then converted from analog to digital (A/D) using a data acquisition (DAQ) device (National Instruments, USB-6210) before being imported into the computer for data analysis.

Figure 4.5 shows the principle of sample surface detection using a confocal optical system. As shown in the schematic diagram, the confocal optical system comprises an objective lens, a half-mirror, an imaging lens, a pinhole, and a photodetector. In this confocal optical system, the pinhole is set at the focal point of the imaging lens, and the photodetector is placed behind it. When the sample surface is located at the focal point of the objective lens, as shown in Figure 4.5(b), the incident light that is focused by the objective lens (red color) is reflected by the sample and re-enters the objective lens (green color), where it is collimated. Since the light is at the focal point, it can pass through the pinhole and produce a high signal intensity at the photodetector.

On the other hand, as shown in Figure 4.5(a) and (c), if the sample is not located at the focal point of the incident laser, the light will not be collimated enough and not converge at the focal point of the imaging lens. Therefore, the light entering the pinhole is not at the optimal size, resulting in lower signal detection on the photodiode. By using this method, we could precisely adjust the sample position to the material's surface when the peak of the detected signal intensity, as shown in Figure 4.5(d), is obtained from the photodiode. The photodiode-detected signal is then converted from analog to digital using a DAQ device before being displayed on the PC for data analysis.

In this verification experiment, we analyze three samples: silicon substrates in air and submerged in water, with water thicknesses of 1.0 mm and 1.5 mm. For the silicon substrate in air, A/D conversion was set to 2 kHz, and 100 continuous signals were recorded at each z position. For the silicon substrate submerged in water, 400 continuous signals were set to minimize disturbance of the light-scattering signal by water. Figure 4.6 shows the result of silicon substrate surface detection by the confocal optical system. Using this optical confocal system, the peak signal in the air surrounding medium is around 8.0 V.

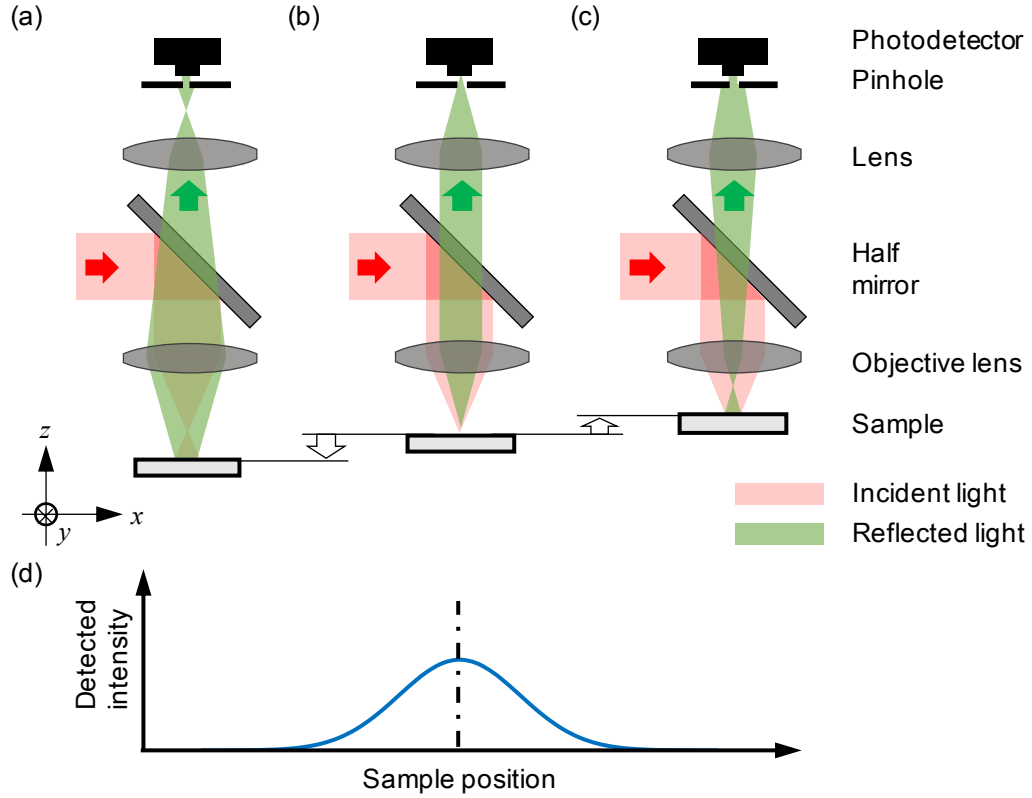


Fig. 4.5 Principle of surface position detection with a confocal optical system. The illustration when (a) minus defocus, (b) at focal point, and (c) plus defocus. (d) Confocal signal by scanning the sample in the z -direction.

In contrast, due to water absorption, the peak signal intensity at the water is around 1.0 V for water 1 mm thick and around 0.8 V for water 1.5 mm thick. The reason for the chosen water thickness will be explained in the next section. Due to this condition, valid representative data are better shown in normalized intensity, as shown in Figure 4.6(a). Although the profile of the detected signal is not as good when compared to the data acquired in air medium due to spherical aberration, this confocal system is considered good enough to acquire the sample position in water medium when we use a non-linear fitting curve to estimate the sample surface position, as shown in Figure 4.6(b) for close-range data.

Regarding top-microsphere detection by the confocal optical system, because the microsphere has a curved surface, the z -position of its surface changes as the x and y positions change, unlike the plane profile in a silicon substrate. Therefore, it is essential to detect the position of the microsphere's top surface. Figure 4.7 shows the detection principle of the top microsphere using a confocal optical

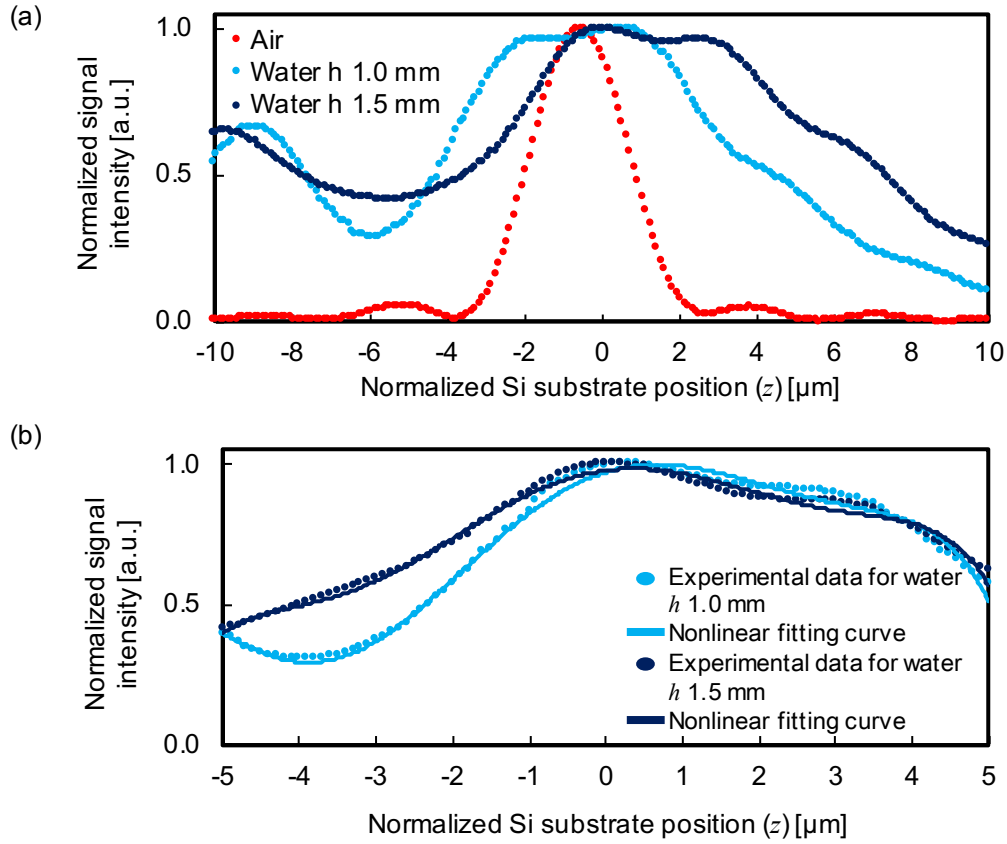


Fig. 4.6 Position detection of the silicon substrate surface with confocal optical system in (a) full range with normalized intensity and (b) near the peak signal in water with normalized intensity. $z = 0$ represents the center of data range.

system. As shown in the left side of Figure 4.7, the confocal system must work in the z -axis direction with the center of a focused laser beam that is aligned with the center of the microsphere. On the other hand, if the central axis of a focused laser beam is displaced as shown on the right side of Figure 4.7, the tangent plane is inclined from the horizontal perspective. Therefore, the reflected light is acquired without reaching maximum signal intensity, and the top of the microsphere cannot be accurately detected or defined. Then, by scanning the microsphere in the x and y -directions as shown in Figure 4.8, it is considered that the confocal signal is maximized at the position where the top of the microsphere is located.

Furthermore, to monitor the machining process, an observation system (indicated by the yellow line in the schematic diagram of the optical setup) is introduced into the optical system, which consists of a white light source, a half mirror, a lens, and a charge-coupled device (CCD) camera. This

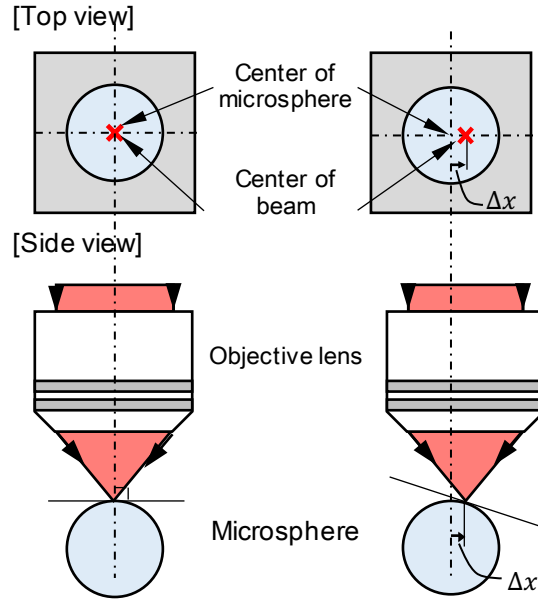


Fig. 4.7 Detection of the microsphere center with confocal optical system.

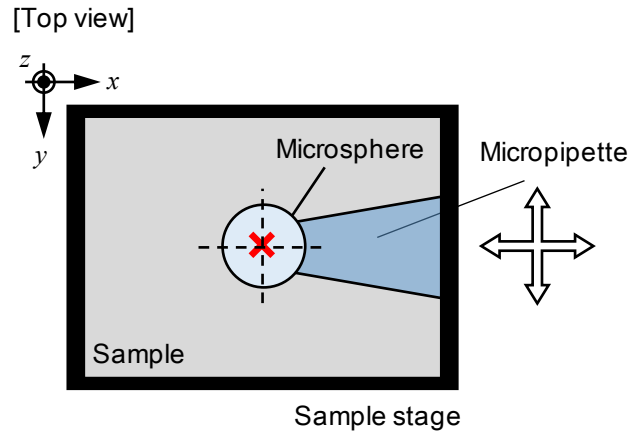


Fig. 4.8 Scanning the microsphere in x - and y -direction for microsphere center detection using confocal optical system.

observation system is only for rough measurements and sample and microsphere positioning during the process. Precise positioning is mainly done using the confocal optical system.

Regarding the sample, a single-crystal silicon substrate was submerged in deionized (DI) water inside a glass chamber. Detailed sample preparation will be explained in the next section. This sample is placed on a stage system that consists of a tilt stage for correcting the tilting angle, an xy -axes automatic stage for controlling the xy position of the sample, and a z -axis piezo stage for controlling

Table 4.4 Specification of piezo stage for Si substrate.

Parameter	Value
Active axes	X, Y, Z
Integrated sensor	Capacitive sensors
Closed-loop travel	100 μm /axis
Resolution (close)	0.4 nm
Repeatability	< 10 nm

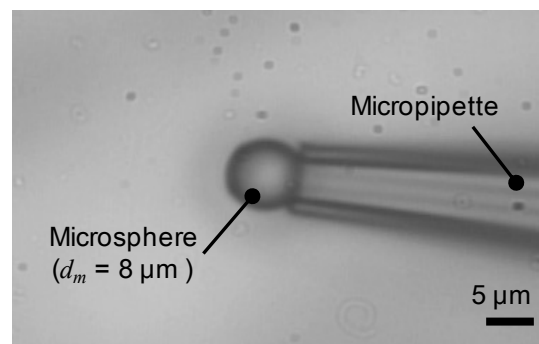


Fig. 4.9 Optical microscope image of the microsphere held by micropipette [66].

the sample position in the z -direction. The specification of the piezo stage is shown in Table 4.4. The xy -axes stage (Sigma Koki Co., Ltd., SGSP20-20 XY) has a resolution of 1 μm for each axis.

Two experiments were conducted in this research: the first, laser machining with a focused laser beam, and the second, PNJ machining. For the former one, the complete experimental setup is already explained in the previous paragraph. For the latter, the PNJ machining, a micropipette with an inner diameter of 5 μm (Micro Support Co., Ltd.), was introduced into the system to hold an 8 μm silicon dioxide microsphere (Ube Nitto Kasei Co., Ltd.). Figure 4.9 showed an optical microscope image when the micropipette held the microsphere. Therefore, by controlling the micropipette position, we can also adjust the microsphere's position relative to the focused laser beam and the sample during machining. Micropipette position is controlled using a micropipette stage system that consists of a manual xyz -axes configuration for rough positioning of micropipette and microsphere, a tilt stage for correcting the tilting angle, and a xyz -axes piezo stage for accurate position control of micropipette and microsphere. The specification of the micropipette stage is shown in Table 4.5. Therefore, by combining all the stage positions and the confocal optical system, we could control the positions of the sample and the microsphere, enabling PNJ machining with controlled positioning.

Table 4.5 Specification of piezo stage for micropipette.

Parameter	Value
Active axes	X, Y, Z
Integrated sensor	SGS
Closed-loop travel	100 μm /axis
Resolution (close)	1 nm
Repeatability	< 10 nm

4.3 Sample preparation and evaluation

In this study, a single-crystal silicon substrate was used as a machining sample. Silicon was chosen for its use as a substrate, widely utilized in electronics, photovoltaics, and high-density systems, owing to its excellent electrical and mechanical properties. Therefore, this PNJ machining technique could be applied to future MEMS/NEMS fabrication by directly applying it to the sample, which has also been used in an industrial application.

In this experiment, a square cross-sectional bar (19×16 mm on average) with 0.6 mm thickness of silicon substrate (h_{si}) was used to ablate the micro/nanoscale hole by a focused laser beam and PNJ machining. Next, the silicon substrate was placed in the glass chamber with a 55 mm inside diameter in such a way that the substrate can fully be submerged in DI water, as shown in the photograph shown in Figure 4.10(a) and their schematic illustration drawn in (b) for the ease of understanding. The water thickness above the workpiece, h_w , was obtained from a simple calculation using the total volume law as follows.

$$V_1 = V_{cb} - V_{si}, \quad (4.1)$$

$$V_2 = A_{ic} \times h_w, \quad (4.2)$$

$$V_t = V_1 + V_2, \quad (4.3)$$

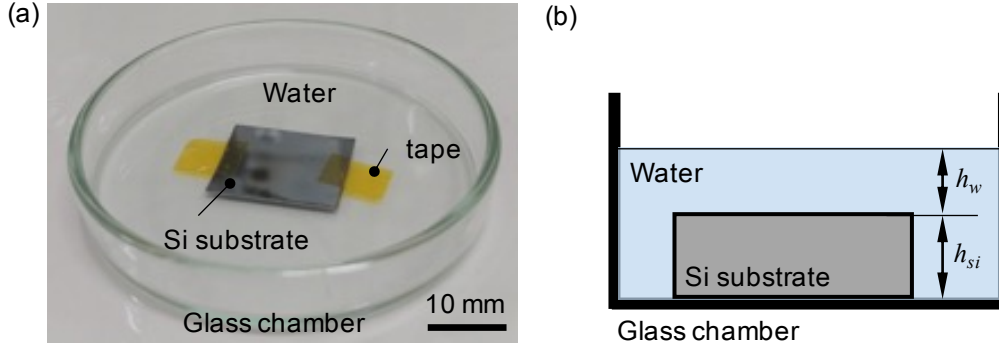


Fig. 4.10 Sample setup configuration. (a) Photographic view and (b) 2D schematic configuration of sample setup.

where V_1 is the volume of water at the level of the substrate, V_{cb} is the the volume of water container at silicon substrate height h_{si} , V_{si} is the volume of the silicon substrate, V_2 is the volume of water with the average film thickness h_w , A_{ic} is an area inside the glass chamber, h_w is average water film thickness, and V_t is the total volume of water added to the chamber of the desired h_w . By measuring the required values from the glass chamber and the silicon substrate with a vernier caliper, we could estimate the average water film thickness above the silicon substrate by injecting water using a pipette in milliliter (mL) increments, as shown in Figure 4.11. For water volumes below 4.0 mL, the surface tension was considered too high, preventing water from forming a layer on the silicon surface. In addition, the silicon substrate has hydrophobic properties that prevent water from forming a uniform layer when a smaller amount of water is added to the glass chamber. In the practical, if we are going to make 1.5 mm of water layer thickness above the silicon substrate by injecting 5 mL of water to the glass chamber from the data calculated from Figure 4.11, first we need to input 8 mL of water to break the hydrophobic properties of silicon substrates and then gradually decrease the amount of water thickness by taking out 3 mL from inside of glass chamber using a pipette. By doing so, 1.5 mm of water layer thickness could be uniformly applied to the sample.

Since a single-pass laser was used in both dry and underwater approaches in this study, the thickness of the water layer is also an essential parameter to consider. The thickness of the water layer during silicon wafer machining significantly influences surface quality, material removal, and thermal management. Thin water films are especially beneficial for flattening and securing wafers, while controlled water layers in laser and micromachining processes yield superior results [94]. We

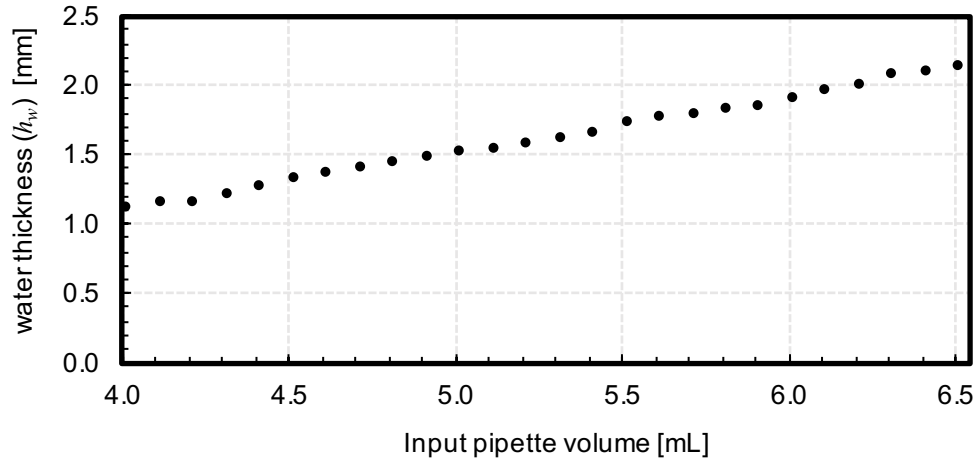


Fig. 4.11 Graph for estimating the average water layer thickness by injected some amount of water.

estimate the transmittance value of water at 800 nm by using quartz cuvette that has 10 x 10 mm in size that is filled by water as the schematic is shown in Fig. 4.12. Same as before, Transmittance T refers to the power loss rate through the cuvette by merely dividing the power after passing through the cuvette, P_{out} , by the power before entering the cuvette, P_{in} . We calculated from 5 data sets, as shown in Table 4.6, and found that the water's transmittance at 800 nm was 0.85. It is best to keep the water layer as thin as possible so our observation system is not affected by its absorbance [95]. Therefore, for the research conducted thereafter, the average film thickness was fixed at 1.0 and 1.5 mm, with a 0.1 mm uncertainty.

The result of the laser ablation process in the silicon substrate is shown in Figure 4.13(a) for the schematic illustration drawing from the side view. The hole diameter from the laser-machining process was examined using a scanning electron microscope (SEM) (JEOL, JSM-6010 Plus/LA) with

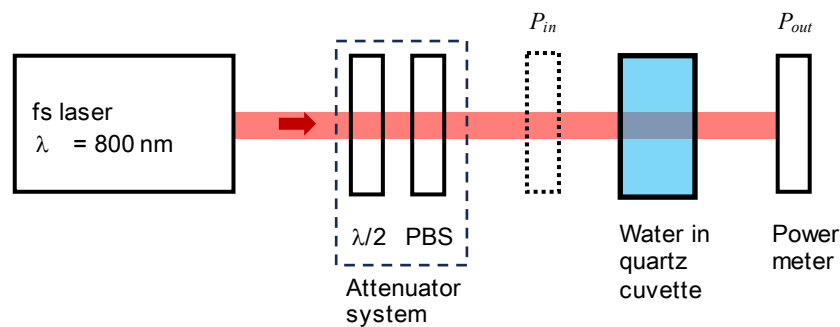


Fig. 4.12 Schematic drawing to measure transmittance of water.

Table 4.6 Calculation of water transmissibility at 800 nm of wavelength.

Half-wave plate angle [deg.]	P_{in} [mW]	P_{out} [mW]	$T = P_{in}/P_{out}$
2	16.09	14.05	0.87
3	31.23	27.76	0.89
4	46.48	39.71	0.85
5	62.54	53.47	0.86
6	79.70	68.10	0.85
			$T_{average} = 0.85$

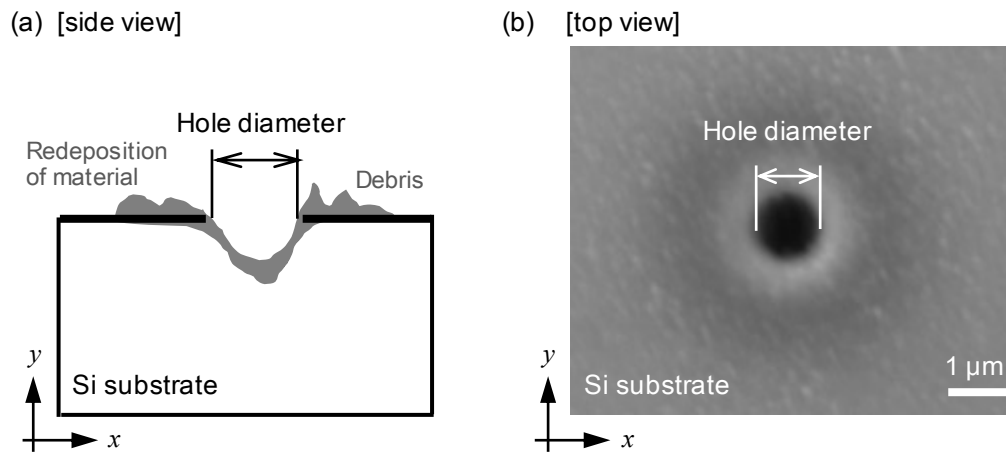


Fig. 4.13 Resulted laser ablation on silicon substrate.(a) illustration figure and (b) ablation result observed and measured by SEM.

the specifications shown in Table 4.7. Figure 4.13(b) shows an example from the laser ablation process observed by SEM, and the defined hole diameter size in the measurement that will be conducted afterward will follow these defined terms [96, 97].

4.4 Conventional laser machining

4.4.1 Experimental procedure

In this experiment, we utilized 585 mW, 663 mW, 741 mW, 819 mW, 897 mW, and 975 mW laser power. Due to the mechanical shutter specification, we used only 160,000 pulses. The laser pulses were then focused via an objective lens with 0.5 NA, as the schematic diagram of this experiment is

Table 4.7 Specification of scanning electron microscope.

Parameter	Value
Acceleration voltage	0.5 kV to 20.0 kV (43 steps)
Resolution	High vacuum mode 4 nm (20 kV), 8 nm (3 kV), and 15 nm (1 kV) Low vacuum mode 5 nm (20 kV)
Low vacuum pressure	10 to 100 Pa
Magnification	x5 to x300,000 (at image size of 128×96 mm)
Objective lens	Super conical mini lens
Image shift	$\pm 50 \mu\text{m}$
Image memory	$5,120 \times 3,840 \times 16$ bit for 1 sheet
Exhaust system	Fully automatic, TMP: 1 unit, RP: 1 unit

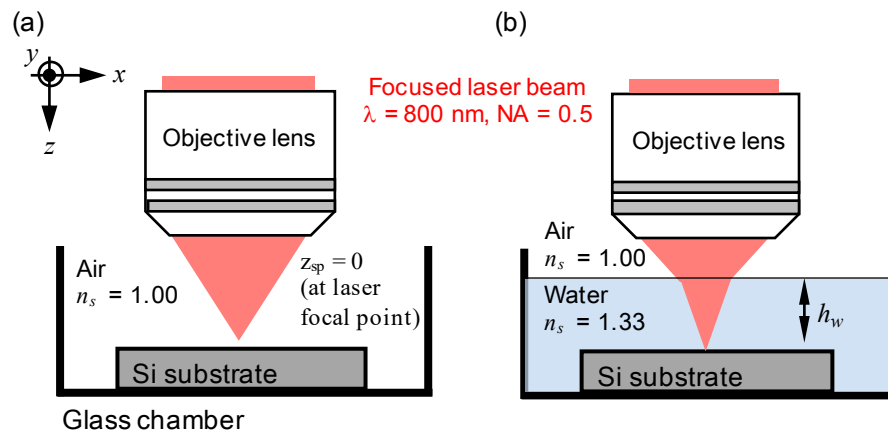


Fig. 4.14 Schematic drawing of laser machining by focused laser beam experiment in (a) air and (b) water medium.

shown in Figure 4.14(a) in air medium and (b) in water medium. For underwater experiments, the sample was placed at the bottom of a glass chamber mounted on a computer-controlled three-axis stage. When the glass chamber was filled with DI water, the thickness of DI water above the sample h_w was controlled at 1.0 and 1.5 mm by using our calculation in section 4.2. A complete experimental parameter used in this experiment is shown in Table 4.8.

For the experiment, several procedures need to be followed. First, we prepare the sample and mount it on the sample stage. Next, we adjust the position of the laser focal point for machining and observation by moving the sample along the optical axis z -direction using the sample stage for rough positioning. Then, we utilized the confocal optical system to detect the sample surface position for accurate positioning. After that, we measure the laser power for the machining process using a power

Table 4.8 Experimental conditions for machining by a focused laser beam.

Parameter	Value
Power (P)	585, 663, 741, 819, 897, 975 mW
Pulse number (N)	160,000
z_{sp}	0 μm (at focal point)
Medium	air and water
Water thickness (h_w)	1.0 and 1.5 mm

meter. Next, for each chosen laser power, nine ablation spots were performed in each medium, and the machining process was then carried out. After machining, the sample is cleaned with ethanol to remove visible dust and excess water from its surface. Before SEM observation, we coated the sample with gold by sputtering to enhance its surface conductivity. Finally, the morphological characteristics and the hole diameter were then investigated by SEM.

4.4.2 Results and discussion

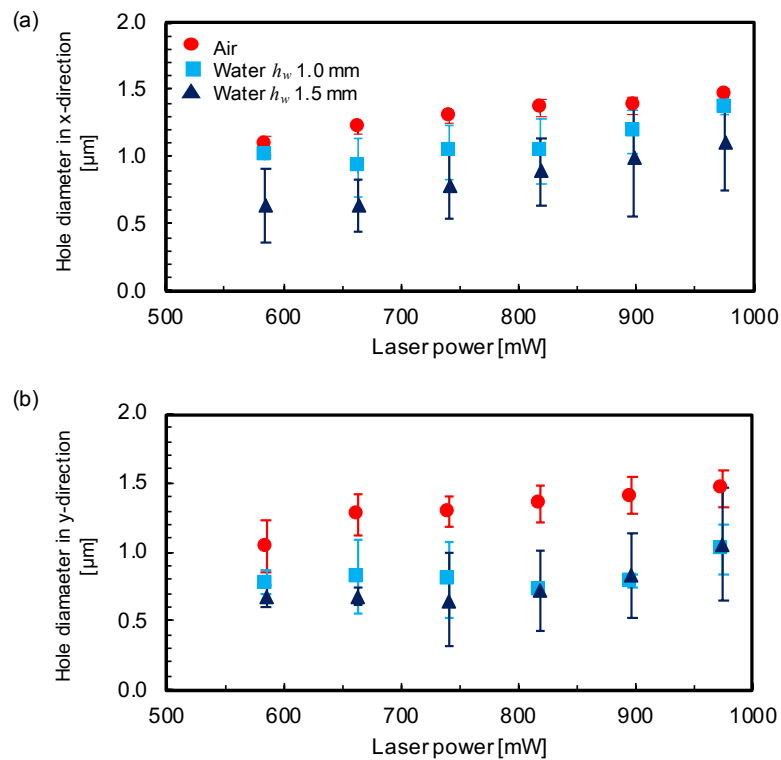
The SEM observation results of the ablated holes in the experiment are shown in Table 4.9 and Table 4.10. From a brief observation, the hole diameter increases with increasing laser power. Moreover, in water, the hole was smaller than in the surrounding air at the same laser power. Besides, when the water thickness increased, the size of the ablated hole became smaller, as the measurement evaluation results by SEM are shown in Figure 4.15, with the plotted graph being the average, and the error bar being the standard deviation from nine ablated holes.

In the air medium, we could obtain an average ablation diameter as small as $1.08 \pm 0.07 \mu\text{m}$ in the x -direction at 585 mW. On the other hand, in the water medium, we could achieve average diameters as small as $0.92 \pm 0.22 \mu\text{m}$ and $0.63 \pm 0.19 \mu\text{m}$ for 1.0 mm and 1.5 mm water thickness, respectively, at 663 mW. Therefore, from the evaluation results shown in Figure 4.15, a smaller ablation diameter could be obtained if we use a thicker water for ablation. During underwater laser beam machining, the material removal process mainly depends on the laser parameters (e.g., beam diameter and laser power) and fluid dynamics (particularly the liquid film thickness above the working surface) [98].

The water film thickness above the workpiece surface plays an essential role in the ablation mechanism. The thick water layer carries away the debris more efficiently but also reduces the

Table 4.9 Summary of conventional laser machining in different medium.

Power [mW]	Dia x -dir [μm]			Dia y -dir [μm]		
	Air	Water ($h_w=1.0$ mm)	Water ($h_w=1.5$ mm)	Air	Water ($h_w=1.0$ mm)	Water ($h_w=1.5$ mm)
585	1.08 $\pm$ 0.07	1.00 $\pm$ 0.01	0.64 $\pm$ 0.27	1.04 $\pm$ 0.19	0.78 $\pm$ 0.08	0.68 $\pm$ 0.08
663	1.21 $\pm$ 0.04	0.92 $\pm$ 0.22	0.63 $\pm$ 0.19	1.27 $\pm$ 0.14	0.82 $\pm$ 0.27	0.68 $\pm$ 0.06
741	1.29 $\pm$ 0.05	1.04 $\pm$ 0.20	0.78 $\pm$ 0.24	1.29 $\pm$ 0.11	0.80 $\pm$ 0.28	0.65 $\pm$ 0.34
819	1.36 $\pm$ 0.07	1.04 $\pm$ 0.24	0.89 $\pm$ 0.25	1.35 $\pm$ 0.13	0.73 $\pm$ 0.04	0.72 $\pm$ 0.29
897	1.38 $\pm$ 0.07	1.18 $\pm$ 0.16	0.99 $\pm$ 0.44	1.41 $\pm$ 0.14	0.79 $\pm$ 0.05	0.83 $\pm$ 0.31
975	1.45 $\pm$ 0.06	1.36 $\pm$ 0.05	1.10 $\pm$ 0.35	1.46 $\pm$ 0.13	1.02 $\pm$ 0.18	1.05 $\pm$ 0.41

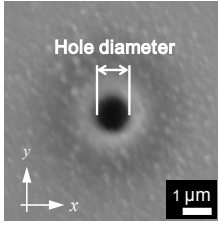
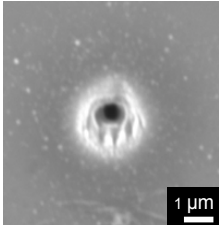
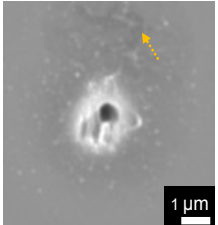
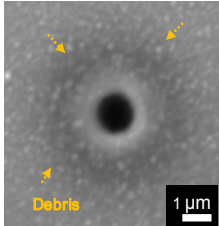
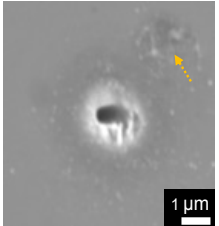
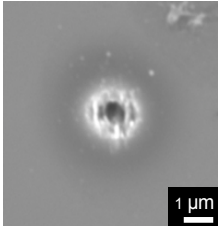
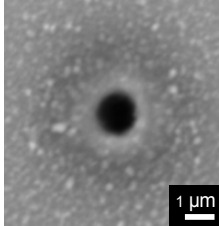
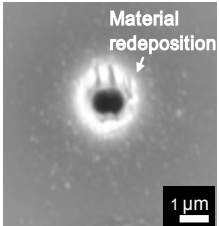
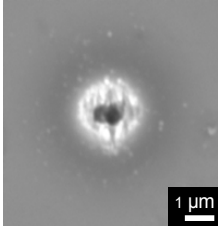
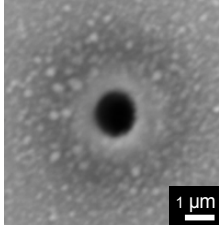
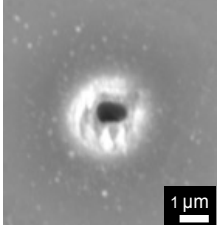
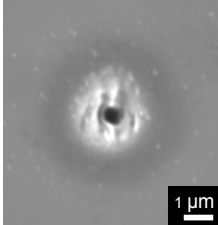
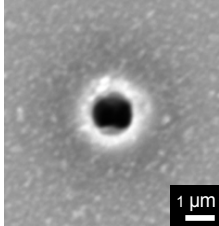
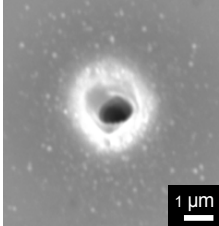
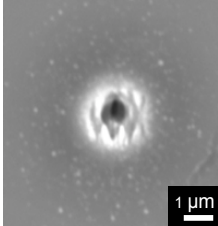
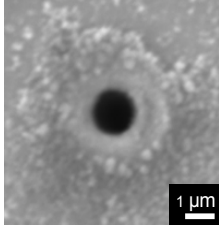
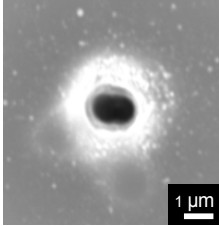
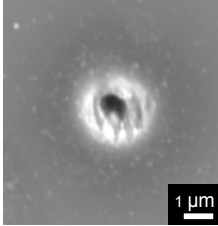
Fig. 4.15 Evaluation results of machined hole in (a) x -direction and (b) y -direction.

ablation rate. When a laser beam travels through a thick liquid film, the pulse shape is not uniform [99]. At the laser-work interface, a suspension of liquid, bubbles, and melt debris forms, confining the plasma above the target material. It scatters the laser light and distorts the laser pulse, thereby severely affecting the energy required to ablate the material. Besides, a high proportion of the energy is absorbed by the thick water film, thereby reducing the energy available to the workpiece material and resulting in lower ablation efficiency. Water splashing within the laser beam's closed premises is a common phenomenon that causes non-uniformity in the water layer thickness [100].

On the other hand, we can understand that the laser ablation process in an air medium is associated with various thermal-related processes, including laser-induced melting, surface tension, and re-solidification of ablated ejections [36]. When the pulse number is high and the pulse energy is relatively high, residual energy and ultrahigh-temperature plasmas can heat the sample surfaces, leading to the so-called thermal processes that enlarge the ablation diameter compared to a water medium.

Furthermore, as in Table 4.10, the debris was observed spreading near the ablated region. It in the air medium could degrade the product's surface quality. Since the amount of ejected material increases with laser density, debris becomes abundant near the ablation region. In water medium, the water could cool the sample surface, and bubbles could be removed by ejecting the ablated region. In the water medium, the ejected material accumulated at a single location where the cavity bubble diameter had been before it collapsed, and only a few debris particles were scattered near the ablation region. For example, the result is a hole ablated in a one mm-thick water sample at 663 mW input laser power. This result is consistent for all input laser power densities. The bubble size is observed to depend on the laser power density. The bubble size increased with increasing laser power density, allowing the ejected material to be flushed from the ablation region and preventing debris accumulation in the observation region. In the water medium, the amount of redeposited material is higher than in the air medium. This redeposited material must be treated using an ultrasonic bath or a further etching process so that the surface resulting from the focused-laser-ablation process has good surface quality. Using a water-based medium during machining can improve surface quality by removing debris near the ablated region. In addition, we could achieve a smaller ablation diameter when using a water medium at the same laser power density.

Table 4.10 SEM images of the ablated hole in different medium with different input laser power.

P [mW]	Air	Water $h = 1.0$ mm	Water $h = 1.5$ mm
585			
663			
741			
819			
897			
975			

4.5 Photonic nanojet-assisted machining

Using a focused laser beam in air and water, it was confirmed that the ablated diameter is approximately at the optical diffraction limit of the constructed optical setup. Although a smaller ablation size can be achieved with a thicker water layer, it becomes impractical to lower ablation efficiency. Therefore, using a PNJ machining system, we consistently broke this value by focusing the laser beam with a microsphere during the ablation process. In addition, we investigate our experimental results in simulation to provide a comprehensive understanding of PNJ for future machining techniques.

4.5.1 Experimental procedure

In the PNJ machining system, a microsphere is used to generate the desired PNJ, whose intensity distribution significantly affects the ablation result. From the result of the FDTD calculation described in Chapter 3, PNJ was generated at some distance from the rear microsphere surface in the liquid medium. Therefore, a micropipette, a tool for manipulating biological samples, was used to hold the microsphere and adjust its position relative to the sample surface and the input focusing laser beam. The micropipette was fixed to the stage by the holder, which was mounted on a computer-controlled three-axis stage. By controlling the microsphere's position, we investigate the effects of DOF and WD on PNJ machining in a liquid medium and the resulting hole diameter.

In this experiment, we use the same experimental setup as before, but in here, we added micropipette and microsphere to generate PNJ. There will be only one laser power utilized at 585 mW. However, the main variable is the distance between the rear microsphere surface and the sample surface h from 0.0 to 6.0 μm with 0.5 μm gap. $h = 0.0 \mu\text{m}$ means the rear microsphere surface is touching the sample surface. For the PNJ machining, the focused laser beam focal point is located at 14 μm below the microsphere center position. Table 4.11 shows a complete experiment setup used in this experiment. The position control between the sample, microsphere plus micropipette, and the laser beam is controlled using stage system and the confocal optical system.

In the constructed optical setup, since the objective lens position was fixed, the desired PNJ was generated by adjusting the sample and micropipette stage. The relative position between the objective lens and the micropipette stage is adjusted to set the beam irradiating condition at the desired z_{sp} value

Table 4.11 Experimental conditions for PNJ machining in water medium.

Parameter	Value
Power (P)	585 mW
Pulse number (N)	160,000
z_{sp}	$-14 \mu\text{m}$
Water thickness (h_w)	1.0 mm
h	0.0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, 5.5, and $6.0 \mu\text{m}$

to control the PNJ intensity distribution. The relative position between the micropipette stage and the sample stage is adjusted to set the distance between the rear microsphere surface at a desired h to examine the PNJ intensity distribution during the machining process. These positional controls are the steps for the conducted experiment, as shown in the schematic diagram in Figure 4.16. The experimental procedure is as follows:

- The initial position is the state where the microsphere is placed on the silicon substrate that is submerged in the water medium.
- Submerged the micropipette in the water and moved the micropipette close to the microsphere, and held the microsphere by the air suction mechanism. Then, move the microsphere that is already trapped by the micropipette in the positive z -axis direction, away from the silicon substrate to the upward direction. Next, we detect the sample surface position in the z -axis using the confocal optical system so that the silicon substrate coincides with the focal point of the focused laser beam.
- Move the sample stage in the negative z -axis (downward) direction by a_1 . In this experiment, we set $a_1 = 30 \mu\text{m}$. Then, we detect the top of the microsphere using the confocal optical system so that the laser beam's focal point is at its top.
- Move the microsphere in the positive z -axis (upward) direction by a_2 to set the focused laser beam irradiation condition so that it would match our chosen z_{sp} value, In here, a_2 is defined with

$$a_2 = z_{sp} + \frac{d}{2}, \quad (4.4)$$

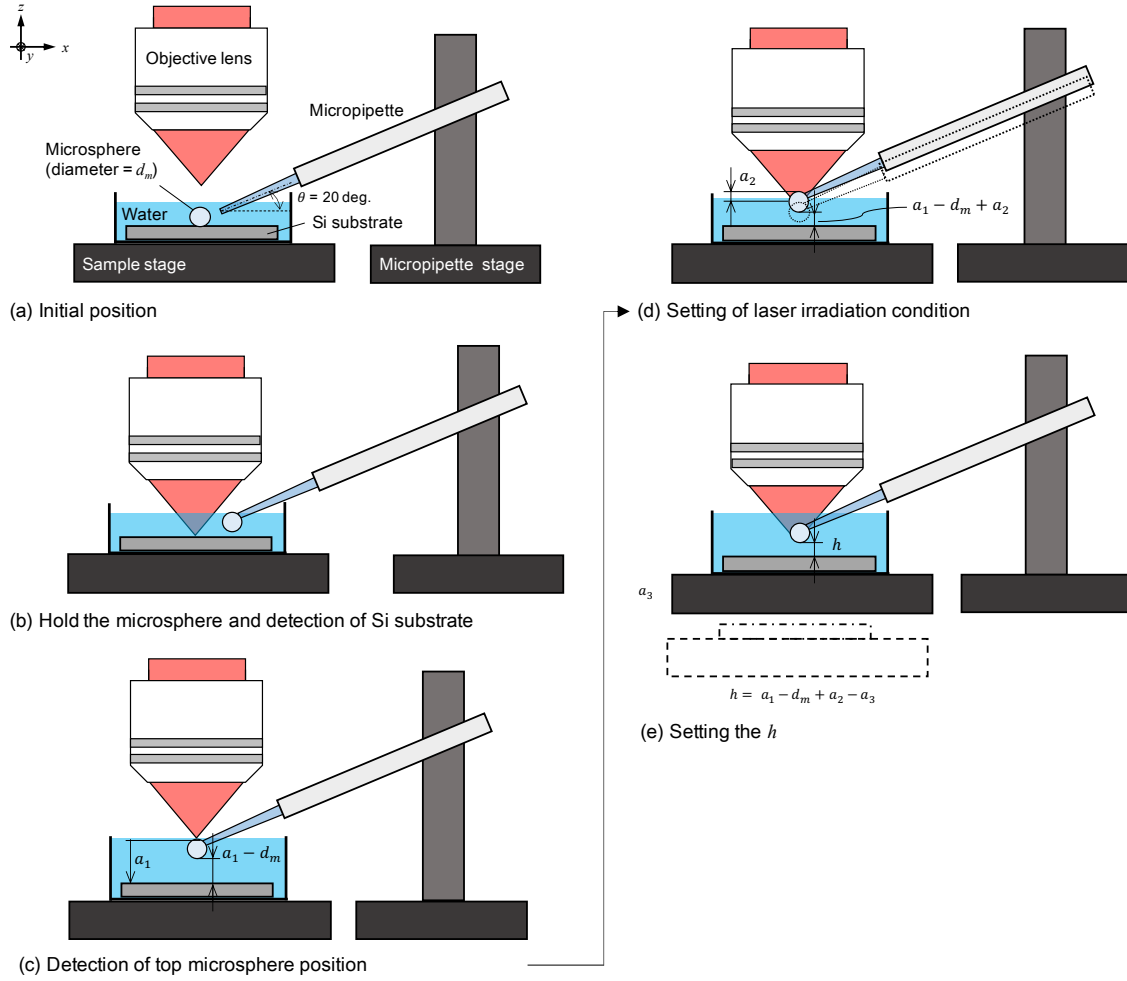


Fig. 4.16 Procedure of PNJ machining experiment: (a) initial position, (b) holding the microsphere and detection of silicon substrate, (c) detection of top microsphere, (d) setting the laser irradiation condition, and (e) setting the h .

where d is the known value of the microsphere diameter. In this experiment, we set $a_2 = 18 \mu\text{m}$.

- (e) Finally, we moved the sample stage in the positive z -axis (upward) direction by a_3 to set the distance between the rear microsphere surface and the silicon substrate h . In here, a_3 is defined with

$$a_3 = a_1 + a_2 - d - h. \quad (4.5)$$

A PNJ experiment was performed using the mentioned step, and there were nine spots of the ablated region for each h parameter. Next, after the PNJ machining process, the sample is cleaned with

ethanol to remove visible dust and excess water from the surface. The morphological characteristics and the hole diameter were then investigated by SEM.

4.5.2 Results and discussion

The results obtained from PNJ machining in the water medium experiment are shown in Figure 4.17. Figure 4.17(a) shows the SEM image of the hole obtained in the machining process with an increasing h value. Figure 4.17(b) and (c) show the measurement evaluation results in the x - and y -directions, respectively. During the experiment, no ablation was observed when $h = 0.0$ to $1.5 \mu\text{m}$. Ablation began at $h = 2.0 \mu\text{m}$ and continued to $h = 5.0 \mu\text{m}$.

However, the results obtained when $h = 2.0 \mu\text{m}$ and $2.5 \mu\text{m}$ show that the average hole diameter is $0.71 \pm 0.16 \mu\text{m}$ and $0.76 \pm 0.18 \mu\text{m}$ in the x -direction, respectively, which do not conform to a trend of the increased distance h . These phenomena are due to the presence of bubbles during machining, which could impact the resulting ablation results, as shown in Figure 4.18(a) for $h = 2.0 \mu\text{m}$ and Figure 4.18(b) for $h = 2.5 \mu\text{m}$. Dell’Aglia et al. [101] captured bubble generation during pulsed laser

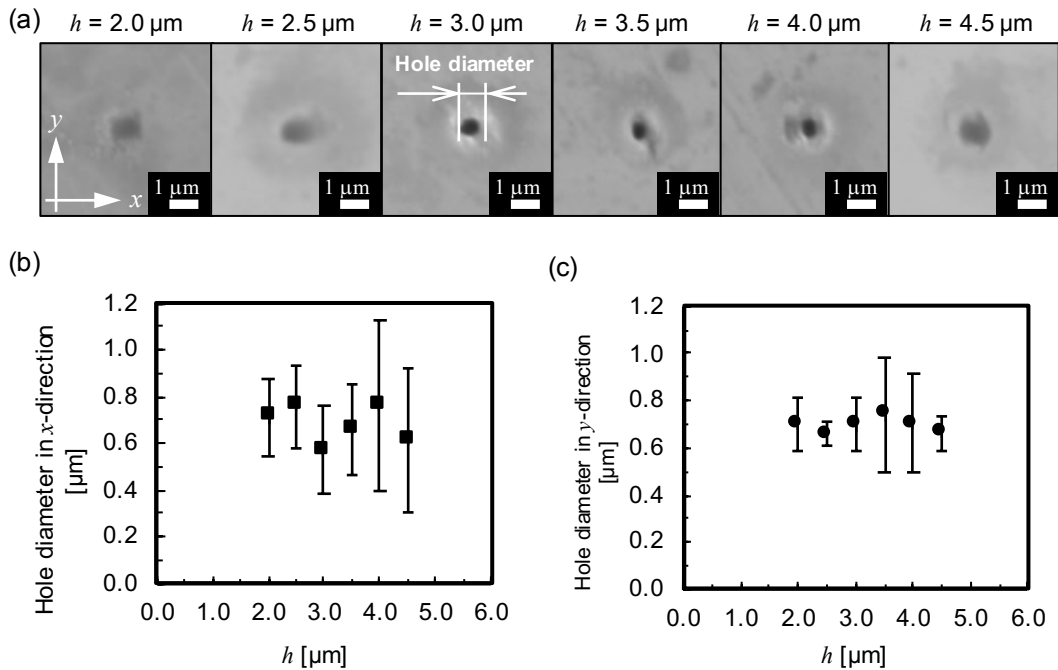


Fig. 4.17 Experiment results of hole diameter from PNJ machining (a) observed by SEM with controlled h position and the measurement evaluation in (a) x -direction and (b) y -direction.

ablation in a liquid using a fast camera, as shown in Figure 4.18(c). Based on this, we estimate the bubble size during PNJ machining in water to be approximately 2.0 to 2.5 μm . Further research is required to evaluate these specific conditions. In addition, this cavity bubble could affect the ablation result due to shockwave generation during its formation showing in a large error bar of the ablated hole.

When $h = 3.0 \mu\text{m}$, proper ablation by the PNJ machining starts to occur, as shown in 4.17(a) with an average hole diameter of $0.57 \pm 0.19 \mu\text{m}$ in the x -direction. The largest average hole diameter was obtained when h was set at $4.0 \mu\text{m}$. When $h = 5.0 \mu\text{m}$, the ablation shows a ripple structure, as shown in Figure 4.19. Therefore, we could not accurately measure the hole diameter. The evaluated results in the y -direction showed a consistent average hole diameter of approximately $0.7 \mu\text{m}$, because the laser light used in this experiment had x -linear polarization.

Through this experiment, we confirmed that PNJ machining in a water medium could be realized. This experiment also showed that the PNJ propagated farther in the water medium than in the air medium. Additionally, PNJ in the water medium has a long depth of focus (DOF) because the ablation occurs from $h = 2.0$ to $5.0 \mu\text{m}$. By achieving a greater DOF, higher removal material are expected

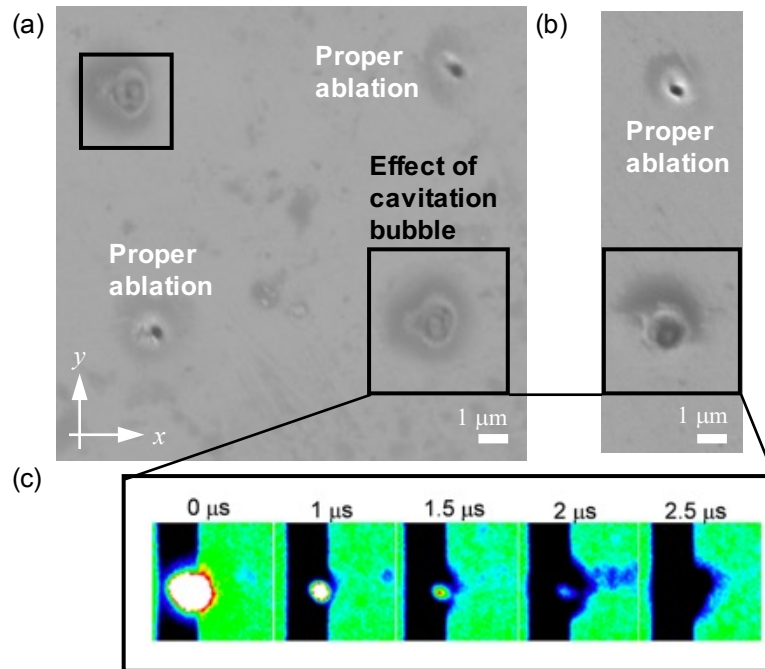


Fig. 4.18 Bubble effect on resulting ablation when (a) $h = 2.0 \mu\text{m}$, (b) $h = 2.0 \mu\text{m}$, and (c) captured bubble on laser-induced plasma on Pt-wire using fast camera [101].

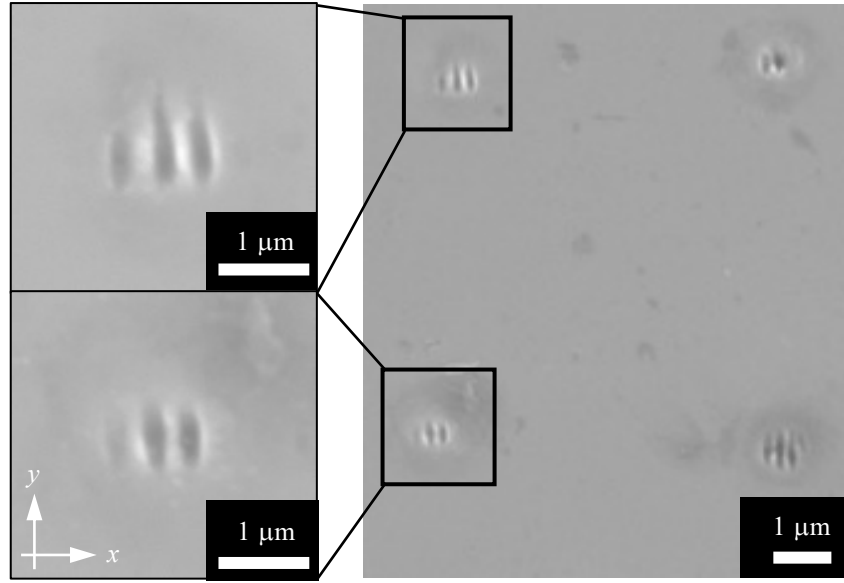


Fig. 4.19 Experiment results of hole diameter from PNJ machining observed by SEM when $h = 5.0 \mu\text{m}$.

using PNJ machining technique. Moreover, only a small amount of debris or redeposition of material is located near the ablated region compared to the case of PNJ in the air medium, as shown in Figure 4.20.

Conversely, ablation only occurs until $h = 2.0 \mu\text{m}$ in the PNJ machining in the air medium [37]. Because the PNJ is generated near the microsphere in air, the microsphere must be controlled near the sample. Therefore, a few gaps are required to discharge the ejected material. If the microsphere is close to the sample, debris can attach to it, affecting PNJ quality. However, because we used a

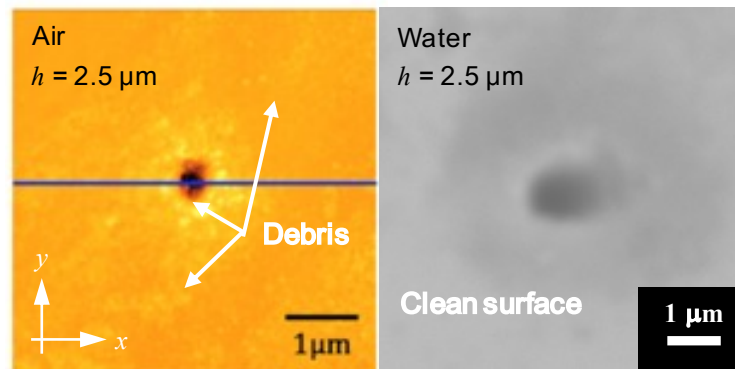


Fig. 4.20 Comparison of silicon surface obtained after PNJ machining in air medium [37] and water medium.

water medium, the PNJ formed at some distance from the microsphere. The effect of a closely located microsphere was not accounted for. However, bubbles generated during the ablation process could affect the PNJ machining results if the distance h is close to the sample.

4.5.3 Experimental validation of simulation results

To elucidate the change in the ablation size when the distance between the rear microsphere surface and the sample substrate h is controlled, it is necessary to estimate the ablation area on the sample surface based on the relationship between the fluence threshold of the material and the fluence value of the generated PNJ. Therefore, we conducted a 3D FDTD simulation with the model shown in Figure 3.14 and the same parameters used for the PNJ machining in the experiment, as shown in Table 4.12. The simulation model includes the micropipette, but it is not included to avoid complexity. The micropipette has an inner diameter of 5.0 μm and an outer diameter of 6.0 μm . This micropipette is made of glass with a refractive index of 1.45. The micropipette is holding the microsphere at 20° from the horizontal.

The generated PNJ for the machining process has the intensity distribution shown in Figure 4.21(a). As shown in Figure 4.21(b) and (c), the generated PNJ has 0.58 μm of beam diameter at

Table 4.12 Simulation parameters for machining simulation using PNJ machining in water medium with controlled position.

Item	Parameter	Value
Model	$(x_{\text{range}}, y_{\text{range}}, z_{\text{range}})$	(24, 24, 40) μm
	(dx, dy, dz)	(0.02, 0.02, 0.02) μm
Incident laser	wavelength (λ)	800 nm
	repetition rate	80 MHz
	polarization	x -linear
	intensity distribution	focused laser beam (NA = 0.5)
	z_{sp}	-14 μm
	power	585 mW
Microsphere	diameter (d_m)	8 μm
	refractive index (n_m)	1.45
Surrounding medium	refractive index (n_s)	1.33
Silicon substrate	distance (h)	0.0 to 6.0 μm (0.5 μm pitch)
	refractive index (n)	$3.692 + 0.006i$
	fluence ablation threshold (F_{th})	190 mJ/cm ² [36]

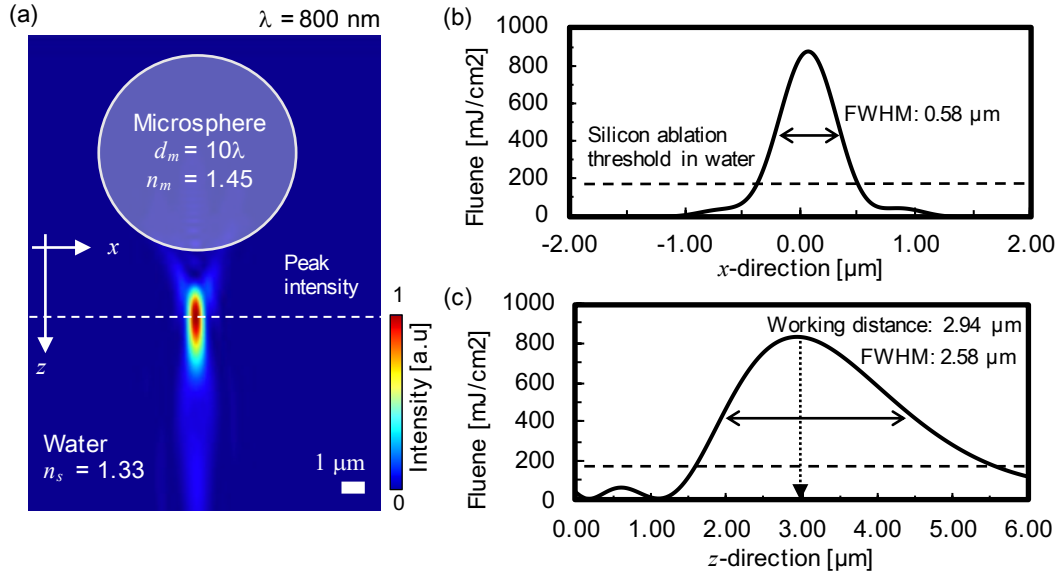


Fig. 4.21 PNJ intensity distribution based on experimental parameter (a) on xz -plane by 3D FDTD simulation. Intensity profile on (b) x -direction at the peak intensity and (c) z -direction, $z = 0$ represents the rear microsphere surface.

the peak intensity, which is located at $2.94 \mu\text{m}$ after the rear microsphere surface. The results from the simulation show that the generated PNJ has high enough intensity to do the ablation process of a silicon wafer in a water medium that has a fluence ablation threshold at 190 mJ/cm^2 [36]. From Figure 4.21(c), we can see that PNJ might do the ablation process on the sample when the distance to the sample h is set at 2.0 to $5.5 \mu\text{m}$, where the intensity in the propagation direction reaches the fluence threshold value.

Regarding the machining simulation, the results are shown in Figure 4.22. As indicated in Figure 4.21, the peak intensity of the PNJ is generated at $2.94 \mu\text{m}$ from the rear microsphere surface. Therefore, the highest fluence on the sample surface was obtained when h was set to $3.0 \mu\text{m}$, near the working distance of the PNJ. As the distance h increased, the fluence value decreased so that from $5.5 \mu\text{m}$ the fluence value will not reach the fluence ablation threshold of the silicon wafer, although from our result shown in Figure 4.21(c), at $h = 5.5 \mu\text{m}$ the intensity of the PNJ is on the ablation threshold. As h decreased, the fluence also decreased, reaching the ablation threshold at $h = 2.0 \mu\text{m}$. Below that value, there will be no ablation. Therefore, the ablation mechanism is expected to start when $h = 2.0 \mu\text{m}$ and stop at $h = 5.0 \mu\text{m}$ in this simulation.

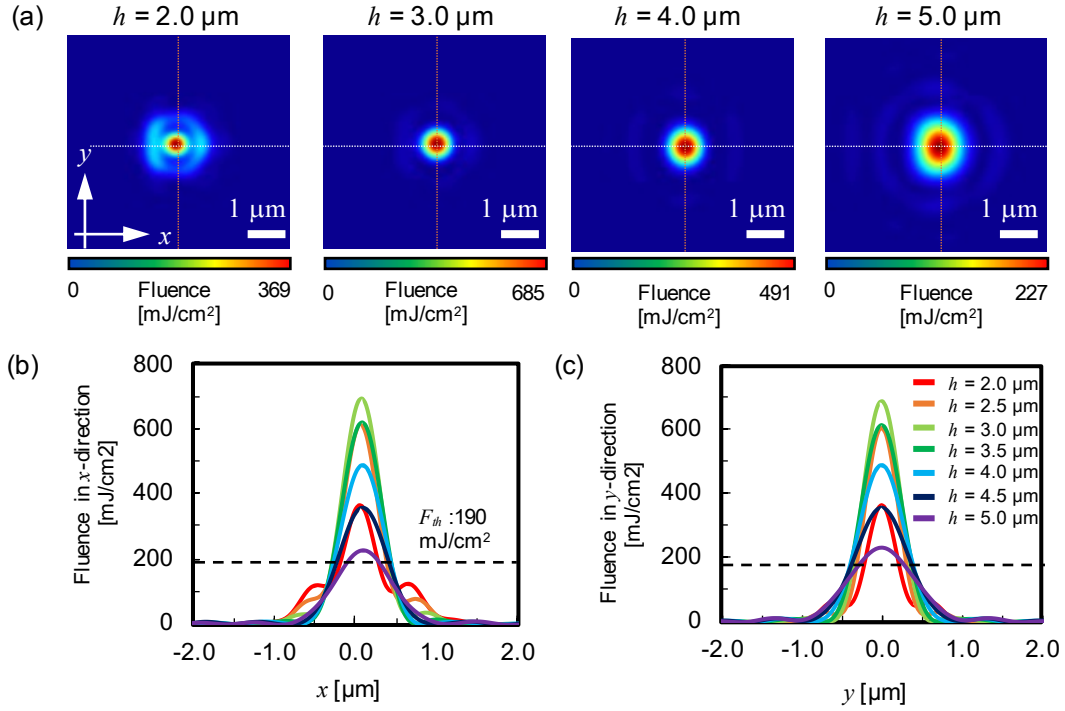


Fig. 4.22 Fluence distribution of the PNJ machining on the silicon substrate in (a) xy -plane, (b) x -direction, and (c) y -direction.

The simulation result is then compared with the experimental result, as shown in Table 4.13 for the SEM observation, Table 4.14 for results summary and in Figure 4.23 for the evaluated measurement. The results show a great compromise between the experiment and the simulation. In terms of ablation diameter, we could acquire as small as 0.38 μm ablation diameter in x -direction when $h = 5.0 \mu\text{m}$ and 0.4 μm ablation diameter in y -direction when $h = 2.0 \mu\text{m}$ as it shown in Figure 4.23(a) and (b) for x -direction and y -direction, respectively. However, a wider ablation diameter is obtained when the h value is near the working distance value of the generated PNJ. These trends are due to their intensity profiles in the x - and y -directions, as shown in Figure 4.22 for the h value used. This value is expected to vary when we utilize a different laser input power. Therefore, our simulation results could serve as a good guide for the experiment. The simulation results here act as an ideal result. From this result, it is possible to apply the more extended DOF characteristics in PNJ machining in a water medium by controlling the microsphere's position relative to the sample surface.

Table 4.13 Comparison of ablation results obtained from experiment observed by SEM and FDTD simulation with controlled distance.

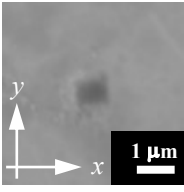
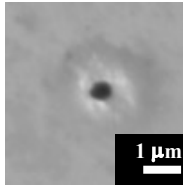
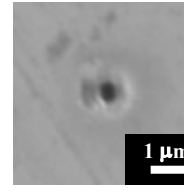
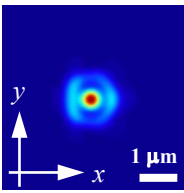
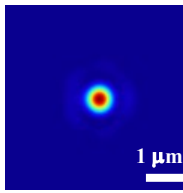
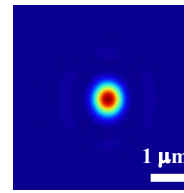
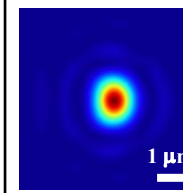
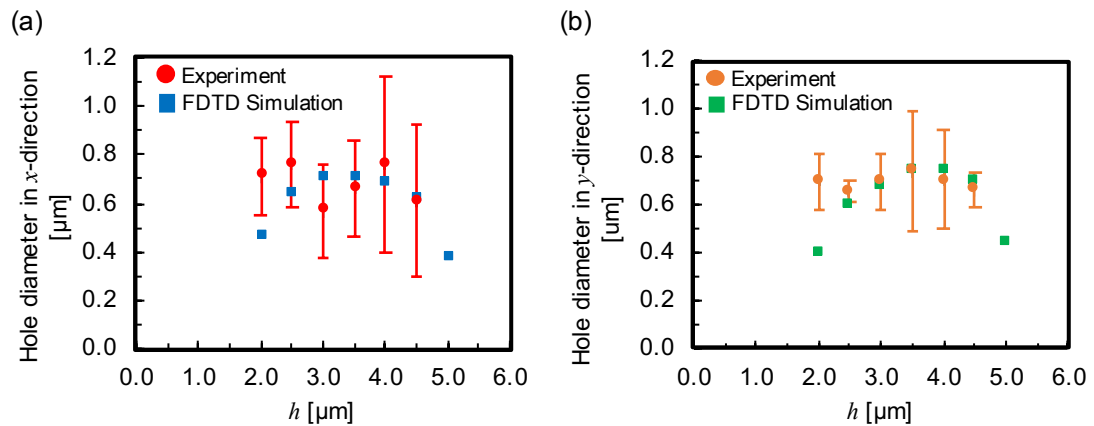
h	2.0 μm	3.0 μm	4.0 μm	5.0 μm
Experiment				
FDTD simulation				

Table 4.14 Summary of PNJ machining results compared between 3D FDTD and experiment.

h [μm]	Dia x -dir [μm]		Dia y -dir [μm]	
	Simulation	Experiment	Simulation	Experiment
2.0	0.46	0.71 ± 0.16	0.40	0.70 ± 0.11
2.5	0.64	0.76 ± 0.18	0.60	0.66 ± 0.05
3.0	0.70	0.57 ± 0.19	0.68	0.70 ± 0.12
3.5	0.70	0.66 ± 0.20	0.74	0.74 ± 0.25
4.0	0.68	0.76 ± 0.36	0.74	0.70 ± 0.21
4.5	0.62	0.61 ± 0.31	0.70	0.66 ± 0.07
5.0	0.38		0.44	

Fig. 4.23 Comparison between machining experiment and FDTD simulation with $P = 585$ mW in (b) x -direction, and (c) y -direction.

In this study, we successfully performed novel laser micromachining using a PNJ in a water medium with a controlled position and comprehensively confirmed it with simulation results. We consistently achieved submicrometer ablation diameter with an ablation diameter as small as $0.57 \pm 0.19 \mu\text{m}$ in the x -direction and sub- $0.8 \mu\text{m}$ in the y -direction was obtained. These values are smaller than the optical diffraction limit of the constructed optical setup. Additionally, a suitable surface quality of the ablation results, with less debris and material redeposition, is achieved when using PNJ machining in a water medium. The proposed technique match our objetices to provide a potential method for future micromachining techniques.

Chapter 5

High precision photonic nanojet machining by using second-harmonic generation beam

This chapter is adapted from articles titled "Second harmonic generation-enhanced photonic nanojet machining in water medium with position control for submicrometer ablation [102]"

5.1 Introduction

Chapter 4 reveals that PNJ machining in water has been realized for the first time, showing great promise. However, an enhancement is needed to achieve a smaller machining resolution and an extended working distance (WD) and depth of focus (DOF) for greater machining flexibility. PNJ can be optimized by adjusting incident laser properties, microsphere characteristics, and surrounding medium [68, 103]. To achieve high spatial resolution while maintaining a high material removal rate, we found in Chapter 3 that precise control of the illumination wavelength is particularly effective, as it enables the generation of a PNJ with a reduced beam diameter and an extended DOF.

In laser-based material processing, the input wavelength is a crucial parameter because the absorption coefficient of a material varies with the incident laser wavelength [104–106]. As the microsphere and surrounding medium parameters remain constant in this research, we focus on modulating incident laser properties. Therefore, it is essential to choose an appropriate wavelength that takes into account the wavelength dependence on the medium and the working material. A direct comparative analysis of PNJ machining using different input wavelengths on the same material has yet to be conducted. Therefore, this study investigates the impact of varying input wavelengths on the PNJ machining of silicon wafers in a water-based medium.

5.2 PNJ mechanism at different input wavelengths

The evaluation of the PNJ intensity distribution is essential for understanding its effect on PNJ machining. This requires systematically varying critical input parameters and quantitatively assessing their impact on the outcome. Thus, we employed the established 3D-FDTD method based on the Maxwell equations in the time domain. According to Wang et al. [78] and Lee and Wang [83], a shorter incident wavelength leads to a smaller PNJ beam diameter and an extended working distance. Hence, when a high-resolution PNJ machining is required, short wavelengths are more suitable. Furthermore, Devilez et al. [91] suggested that a smaller PNJ beam diameter can be achieved using a focused laser beam. In the case of the PNJ system, light is focused as it irradiates the microsphere. The focused light either converges or diverges from the central position of the microsphere. Defocusing

influences PNJ formation and its properties in the air medium [92], but the effect of the focusing light on the PNJ in a water medium has not yet been elaborated.

We conducted 3D FDTD simulations using a focused laser beam at the fundamental wavelength ($\lambda = 1030$ nm) and its SHG wavelength ($\lambda = 515$ nm) as input sources. The laser was focused through an objective lens with a NA of 0.5, and the defocusing distance (z_{sp}) was varied from 0 to -20 μm . The laser was incident on a readily available silica microsphere with a diameter (d_m) of 8.00 μm . The refractive index of the silica microsphere (n_m) was set to 1.45 at the fundamental wavelength and 1.46 at the SHG wavelength, based on dispersion values reported by Malitson [107]. The microsphere was fully submerged in water, which had a constant refractive index of $n_s = 1.33$ for both wavelengths. Figure 5.1 illustrates the 3D-FDTD simulation models for the fundamental and SHG wavelengths. Table 5.1 shows the complete simulation parameters set similarly to the experimental conditions to ensure consistency and accuracy.

The results of the 3D-FDTD simulations are shown in Figure 5.2(a). When $z_{sp} = 0$ μm , no PNJ was generated, while converging light can form a PNJ in a water medium. This phenomenon occurs in water due to its refractive index, which causes light to propagate farther. When using the fundamental wavelength, PNJ started to form outside of the microsphere when $z_{sp} = -9$ μm . Meanwhile, when

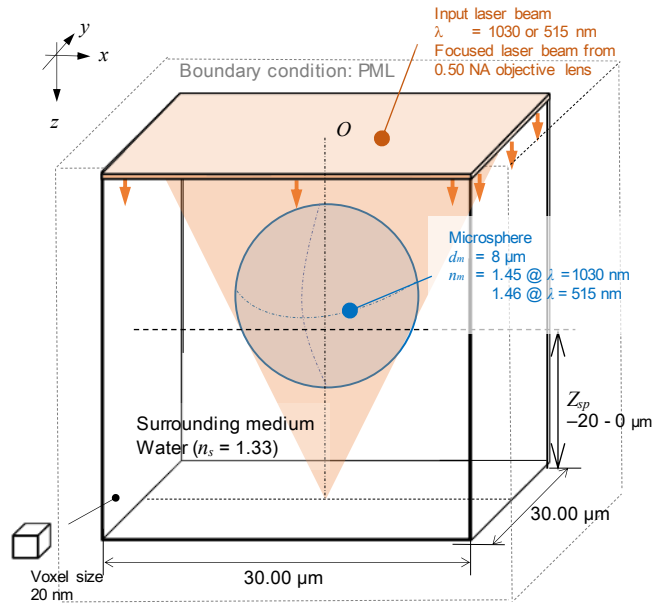


Fig. 5.1 Simulation model for PNJ investigation on SHG beam.

Table 5.1 3D FDTD simulation parameter.

Item	Parameter	Value
Model	$(x_{\text{range}}, y_{\text{range}}, z_{\text{range}})$	$(30, 30, 40) \mu\text{m}$
	(dx, dy, dz)	$(0.02, 0.02, 0.02) \mu\text{m}$
Incident laser	wavelength (λ)	515 and 1030 nm
	beam profile	Gaussian beam with $1/e^2=2.5 \text{ mm}$
	polarization	x linear
	intensity distribution	focused laser beam ($NA = 0.5$)
	z_{sp}	$-20 - 0 \mu\text{m}$ (with $1 \mu\text{m}$ pitch)
Microsphere	diameter (d_m)	$8.0 \mu\text{m}$
	refractive index (n_m)	1.46 ($\lambda = 515 \text{ nm}$), 1.45 ($\lambda = 1030 \text{ nm}$)
Surrounding medium	refractive index (n_s)	1.33 (both wavelengths)

using the SHG, PNJ started to form when $z_{sp} = -8 \mu\text{m}$. As shown in Figure 5.2(b), the PNJ beam diameter became wider when we utilized a longer input wavelength. Figure 5.2(b) also shows that a change in defocusing distance only had a slight influence on the PNJ beam diameter at both input wavelengths.

Furthermore, a shorter working distance and a greater DOF were obtained when a longer input wavelength was used, as shown in the Figures. (c) and (d), respectively. These figures also show that a further converging distance produced a longer working distance and a more extended DOF. Shorter distances ($h < 3.0 \mu\text{m}$) were avoided to prevent bubble formation and induced shockwave, which could interfere with the PNJ machining process in the water medium [32, 73]. Hence, the chosen z_{sp} values were $-14 \mu\text{m}$ for the fundamental wavelength and $-19 \mu\text{m}$ for the SHG beam. These values were chosen to accommodate a small PNJ beam diameter, long WD, and extended DOF.

Figures 5.2(e) and (f) depict the intensity distribution at the peak intensity of the PNJ with the chosen z_{sp} in the x - and z -directions, respectively. The generated PNJ from the fundamental wavelength at $z_{sp} = -14 \mu\text{m}$ had a $0.73 \mu\text{m}$ PNJ beam diameter, a $2.84 \mu\text{m}$ working distance, and a $3.16 \mu\text{m}$ depth of focus. Meanwhile, the PNJ from SHG at $z_{sp} = -19 \mu\text{m}$ had a $0.43 \mu\text{m}$ PNJ beam diameter, a $4.18 \mu\text{m}$ working distance, and a $2.51 \mu\text{m}$ depth of focus. Therefore, as can be observed in Figures 5.2(e) and (f), a smaller PNJ beam diameter was achieved using SHG, and the PNJ peak intensity was positioned further from the rear microsphere surface, making it more effective for

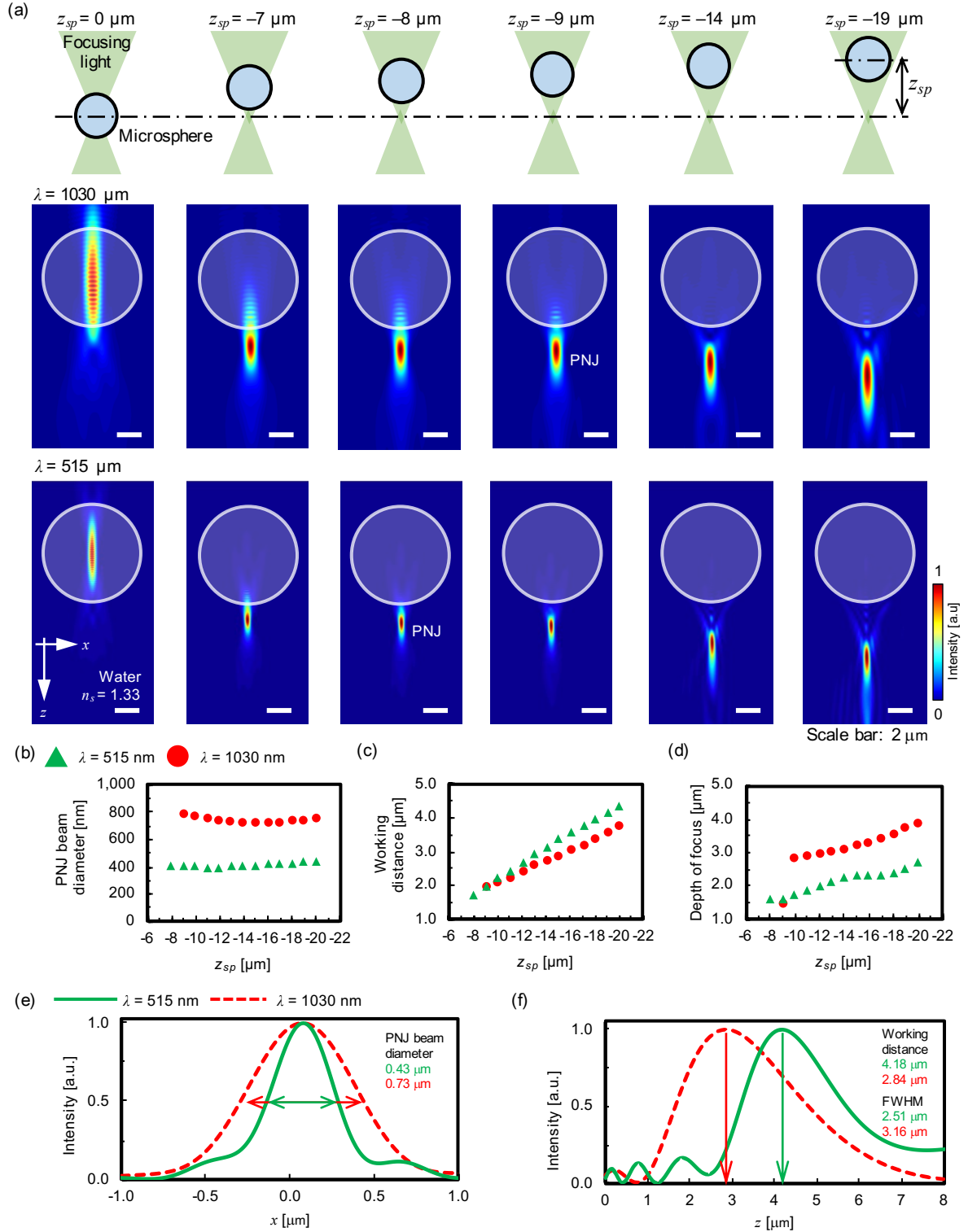


Fig. 5.2 PNJ intensity distribution controlled by different laser focus distances at the fundamental wavelength and its SHG. (a) Intensity distribution; (b) PNJ beam diameter; (c) working distance; and (d) depth of focus. PNJ intensity distribution of the experimental parameter (e) in the x -direction at the peak intensity and (f) in the z -direction..

PNJ machining. These numerical simulation results provide valuable insights into the experimental conditions in PNJ machining.

5.3 Experimental setup

In this section, we describe the laser machining system constructed to enhance the PNJ machining in a water medium using the SHG beam. The experiments were all conducted in a clean room with controlled temperature and humidity of 23 degrees Celsius and 30%, respectively. A hole machining experiment was conducted to investigate the impact of the SHG-generated PNJ machining. The complete experimental conditions are listed in Table 5.2.

The schematic of the experimental setup is shown in Figure 5.3(a), with a detailed view of the laser machining process depicted in Figure 5.3(b). Figure 5.4(a) shows a photograph of the actual constructed optical setup in the laboratory and its (b) PNJ machining part. As shown in Figure 5.3(a), the optical setup consists of three components: laser machining part, confocal system, and observation system.

For the laser machining part, a single-shot 1030 nm femtosecond (fs) laser (NKT Photonics, OneFive ORIGAMI 10XP) with a pulse duration of 321fs was used. The specification of the laser is shown in Table 5.3. The fs laser had a Gaussian beam profile and a diameter of 2.5 mm at $1/e^2$

Table 5.2 General SHG-enhanced PNJ machining experimental parameter.

Item	Parameter	Value
Incident laser	wavelength	Fundamental ($\lambda = 1030$ nm) SHG ($\lambda = 515$ nm)
	pulse width	321 fs
	beam profile	Gaussian beam with $1/e^2 = 2.5$ mm
	polarization	x linear
Sample	material	silicon
	orientation	[111]
	size	10×10
	thickness	0.5 mm
Liquid	substrate	pure water (H_2O)
	thickness	2 mm
	condition	static
	temperature	300 K (Room temp.)

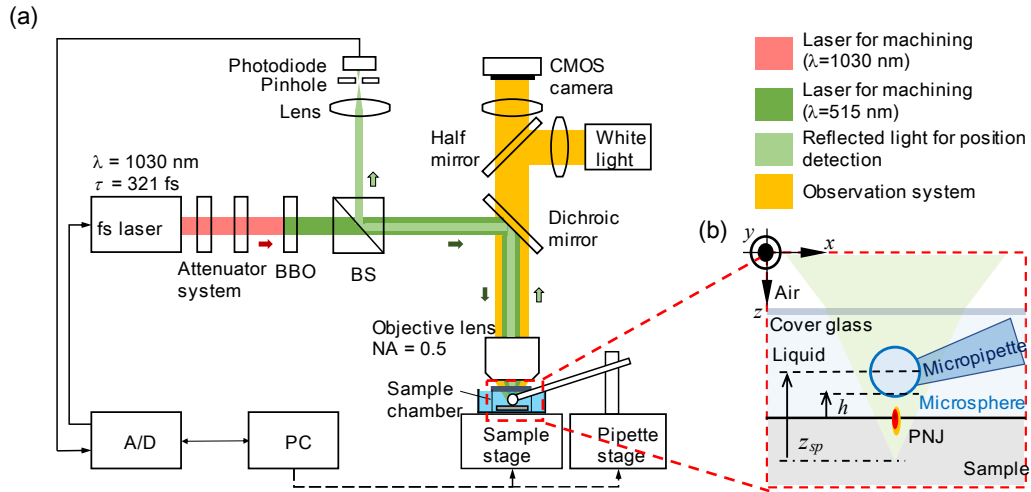


Fig. 5.3 Schematic diagram of (a) experimental setup with (b) insight view of machining part for PNJ machining in water medium. Fs: Femtosecond, BBO: Barium Borate crystal, BS: Beam splitter, A/D: Analog to digital converter.

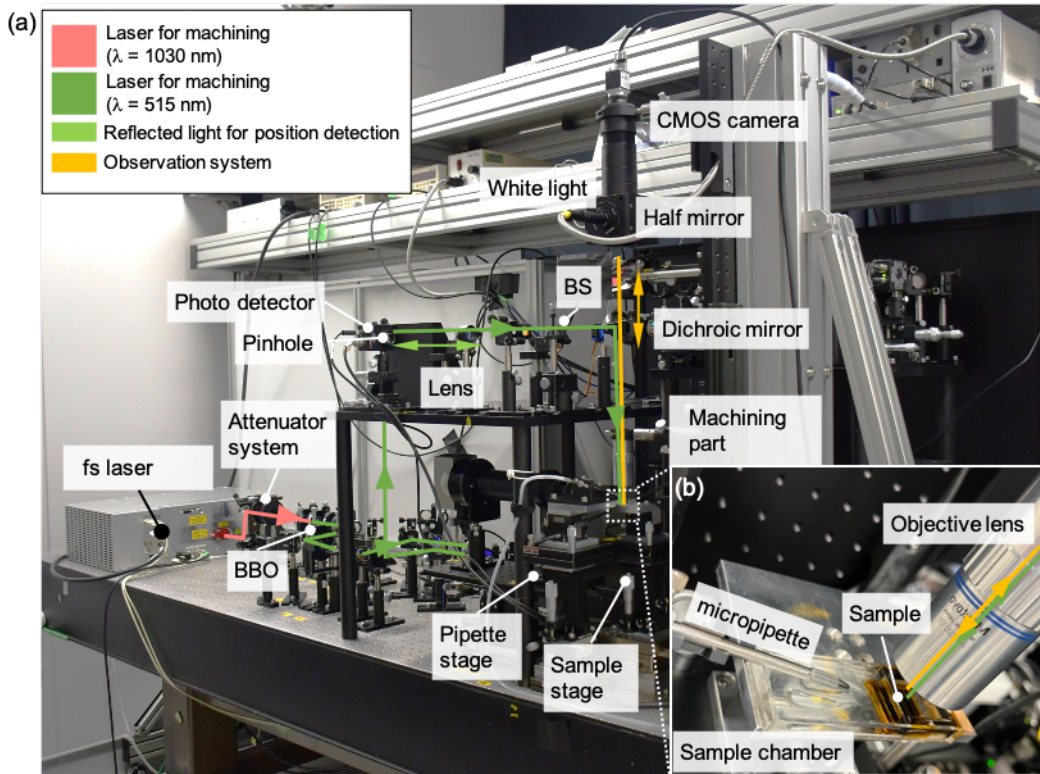


Fig. 5.4 Photograph of (a) constructed experimental setup with (b) insight view of machining part for PNJ machining in water medium. BBO: Barium Borate crystal, BS: Beam splitter.

Table 5.3 Specification of femtosecond laser.

Parameter	Value
Pulse duration (τ)	<321 fs
Center wavelength (λ)	1030 nm
Spectral bandwidth	<5 nm
Average power	<4 W
Peak energy	<40 μ J
Peak power	>100 MW
Repetition rate (R)	50 kHz $\sim$ 1 MHz
Beam quality	$M^2 < 1.2$
Beam diameter	± 2.5 mm

as shown in Figure 5.5. The laser beam then passed through an attenuator system, which consisted of a half-wave plate (Edmund Optics, 39-177) and a polarizer (Thorlabs, GL10-B). This attenuator system controlled the incident laser power by rotating the angle of the half-wave plate. The fs laser then passed through a BBO crystal (Newlight, THG1030(I)-A05-400fs) to obtain SHG at a 515 nm wavelength.

To assess the SHG onversion efficiency of a BBO crystal, we used a femtosecond laser source. We varied the average power and repetition frequency of the laser to compare the BBO crystal's performance before and after harmonic generation. After separating the wavelengths, we measured the powers of the fundamental and SHG individually using a calibrated power meter (Thorlabs, PM400). We calculated the conversion efficiency (η) as the ratio of the SHG output power to the incident fundamental power on the BBO crystal, taking into account the transmission losses from the two harmonic separators, each with a reflectivity of 0.999 for the SHG.

The relationship between SHG conversion efficiency and pulse energy is shown in Figure 5.6. We found that the maximum measured conversion efficiency of the BBO crystal for SHG was about 0.4. Additionally, we observed that the conversion efficiency does not depend on the laser repetition rate but is strongly affected by pulse energy. This finding suggests that the SHG conversion efficiency in the BBO crystal is mainly determined by the instantaneous intensity (or pulse energy) of the fundamental wave, highlighting the nonlinear optical behavior of the BBO crystal. In this respect, SHG is a practical approach for obtaining different input wavelengths in the PNJ optical system, allowing precise control over beam characteristics. The wavelength of incident light could easily be

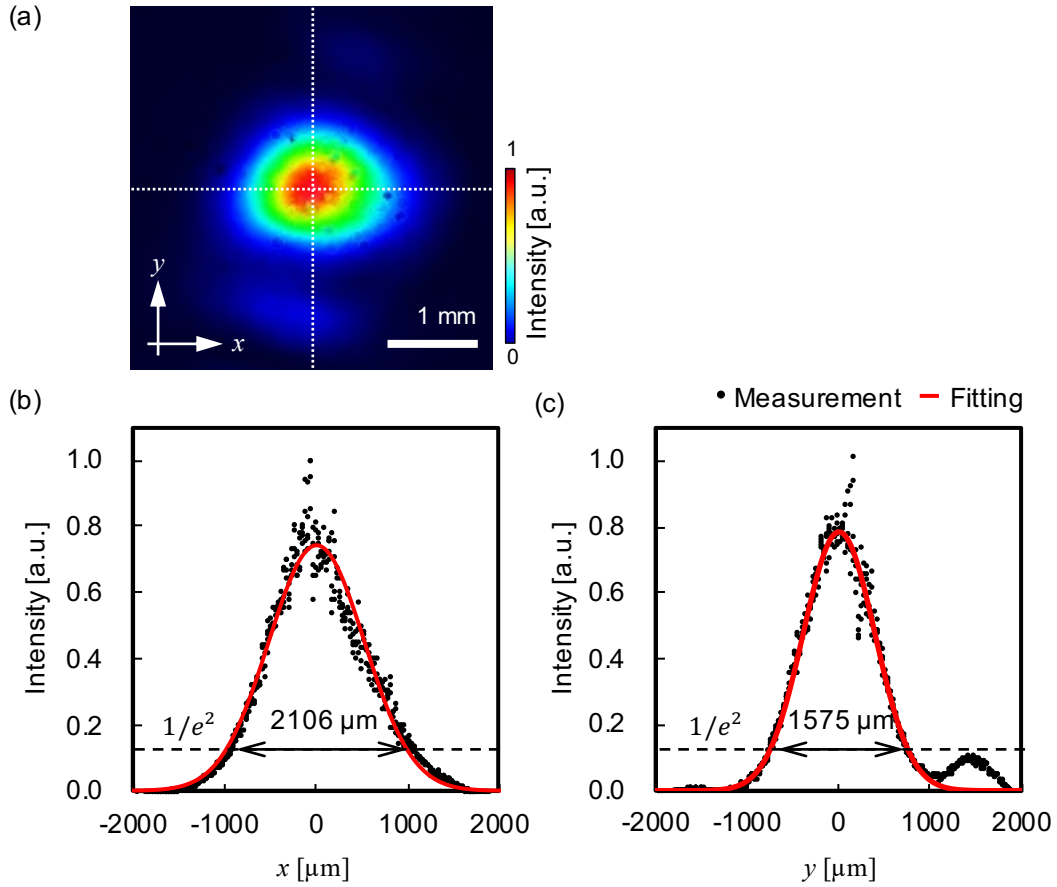


Fig. 5.5 Beam profile of the laser machining: (a) Intensity distribution, intensity profile in the (b) x -direction and (c) y -direction.

changed using a tunable laser or by setting some optical parts in the optical setup. The introduction of BBO crystal to obtain an SHG beam from the fundamental wavelength is one of the practical ways to obtain different incident input wavelengths in the optical system.

Subsequently, the laser beam passed through a beam splitter (Thorlabs, BS037). The beam splitter used in this setup had an $R:T$ (reflectance:transmittance) ratio of 10:90 to conserve as much laser power as possible for laser machining. After that, the transmitted laser beam was reflected by a dichroic mirror and passed through the objective lens with an NA of 0.5 (Mitutoyo, M Plan Apo SL 80 $\times$) before being irradiated by the laser beam onto the microsphere for the PNJ machining process. The specification of the objective lens is the same as shown in Table 4.2.

For the PNJ machining experiment, a micropipette with an inner diameter of 5 μm (Micro Support, MP-005) was introduced to the system to hold an 8 μm SiO_2 microsphere (Ube Nitto Chemical),

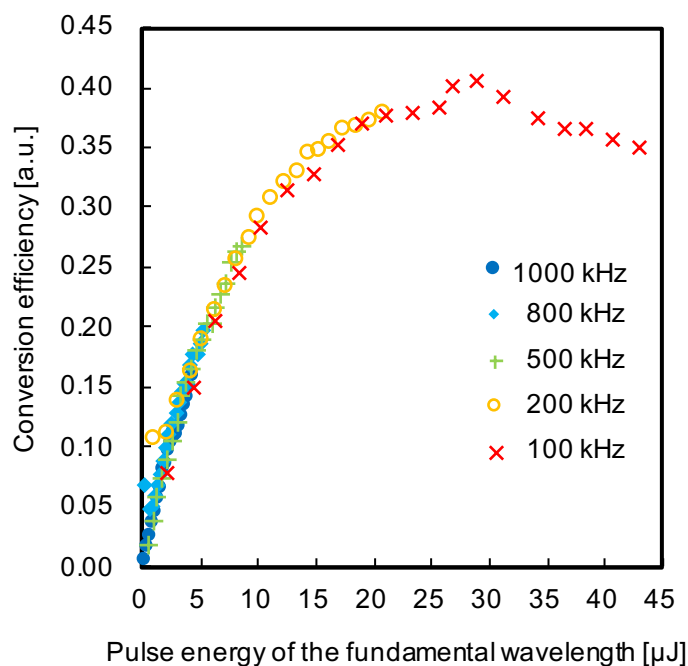


Fig. 5.6 BBO crystal conversion efficiency.

as shown in Fig. 5.3(b). The micropipette position was controlled by a micropipette stage system, which consisted of a manual xyz -axis configuration for the rough positioning of the micropipette and microsphere, a tilt stage for correcting the tilting angle, and an xyz -axis piezo stage (PI, P-616.3s) for accurate position control of the micropipette and microsphere.

A single-shot laser was used to irradiate the microsphere, producing PNJ. The laser energy was controlled the same for all wavelengths, even though water has a transmission of 0.74 at the fundamental wavelength and 0.96 at the SHG beam, as we measured using a simple transmittance experiment as shown in Figure 4.12. The PNJ irradiated a single-crystal undoped silicon (orientation [111]) with a size of 10 mm×10 mm, a thickness of 500 μm , and a natural oxide layer (2–3 nm). Figure 5.4(b) shows the sample setup with a special chamber used to immerse the sample in pure water at room temperature and control the thickness of the liquid layer above the sample surface to ~ 2 mm in static conditions. The sample was then placed above the sample stage system, which consisted of a tilt stage for correcting the tilting angle, an xy -axis automatic stage (THK precision, LS1L120-25B-06) for controlling the xy -position of the sample, and a z -axis piezo stage (PI, P-611.3c)

for controlling the sample position in the z -direction. The pipette and sample control systems could operate independently to ensure accurate positioning during PNJ machining.

A confocal system was introduced to detect the sample surface and the top position of the microsphere to precisely control the microsphere's position relative to both the focal point of the laser beam (z_{sp}) and the sample position (h) during PNJ machining. The confocal principle is same as described in Figure 4.5. The low-power reflected light beam from the sample surface (or microsphere surface) re-entered the objective lens before it was reflected by a dichroic mirror and propagated to the beam splitter in the opposite direction from laser machining. The beam splitter then reflected the light. Next, it passed through a bandpass filter (Thorlabs, FBH515-10), a lens ($f = 200$ mm), and a pinhole (Thorlabs, P40K) before being detected by a photodiode (Thorlabs, APD440A2). The electrical signal obtained by the photodiode was then converted from analog to digital using a data acquisition(analog–digital [A/D]) device (National Instruments, USB-6210) before it was imported into a computer for data analysis.

The procedure for controlling the distance h and z_{sp} using the confocal system is illustrated in Figure 5.7(a). First, the Si substrate was scanned in the z -direction, and its surface was estimated using Gaussian fitting as shown in Figure 5.7(b). Next, the Si substrate was moved downward by a specific distance (D) by adjusting the sample stage position with micrometer precision. The microsphere was then inserted between the focused laser beam spot and the Si substrate, and its top surface was scanned using the micropipette stage. To ensure accurate alignment, the microsphere's center was correlated with the optical axis of the laser beam by scanning it in the x - and y -directions. The top microsphere was also estimated using Gaussian fitting, as shown in Figure 5.7(c). The introduced confocal system achieved accuracies of $0.073\ \mu\text{m}$ for sample surface detection and $0.898\ \mu\text{m}$ for top microsphere detection, as indicated by our repeatability measurements shown in Figures 5.7(d) and (e), respectively. Thereafter, z_{sp} was adjusted by moving the pipette stage to the chosen value. Then, h was calculated as $h = D - d_m$. Finally, h was controlled by moving the sample stage to the calculated distance. For each machining process, the confocal procedure was repeated to ensure the accuracy of the positioning before the machining process was done.

Furthermore, to monitor the state of the machining process, an observation system was integrated into the optical system, which consisted of a white light source, a half-mirror, a lens, and a CMOS

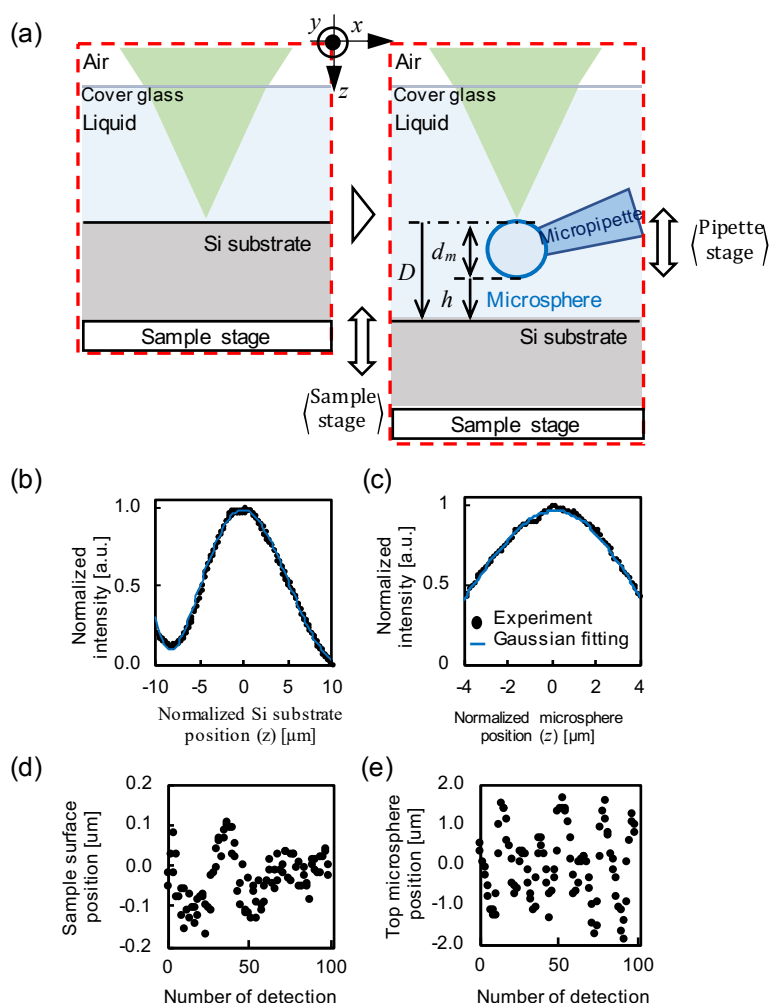


Fig. 5.7 Schematic diagram of the position control for PNJ machining. Confocal signals when the following are scanned: (b) Si substrate and (c) top microsphere held by a micropipette. Confocal repeatability when scanning (d) the sample and (e) the top microsphere position.

camera (Basler, a2A2840-48umBAS). This system was only used for rough measurement and rough positioning of the sample and microsphere during the process. Precise positioning was still mainly performed using the confocal optical system. Each PNJ machining experiment was conducted five times under identical conditions to ensure repeatability. After PNJ machining, the samples were treated with an ultrasonic bath for 5 min in both deionized water and acetone to clean the residual ejected material off the surface [36]. The geometry of the machined holes was then examined using an atomic force microscope (AFM, Bruker Innova). The AFM scanning lateral resolution was 40 nm.

5.4 Influence of wavelength on PNJ machining

In this set of experiments, a single-shot laser with 50-nJ energy levels was used to irradiate the microsphere, producing PNJ. z_{sp} was set to $-14\text{ }\mu\text{m}$ for 1030 nm and $-19\text{ }\mu\text{m}$ for SHG (515 nm). h was controlled from 3.0 to 8.0 μm with a 0.5 μm -pitch. A complete list of experimental parameters in this experiment is shown in Table 5.4 as an addition from Table 5.2.

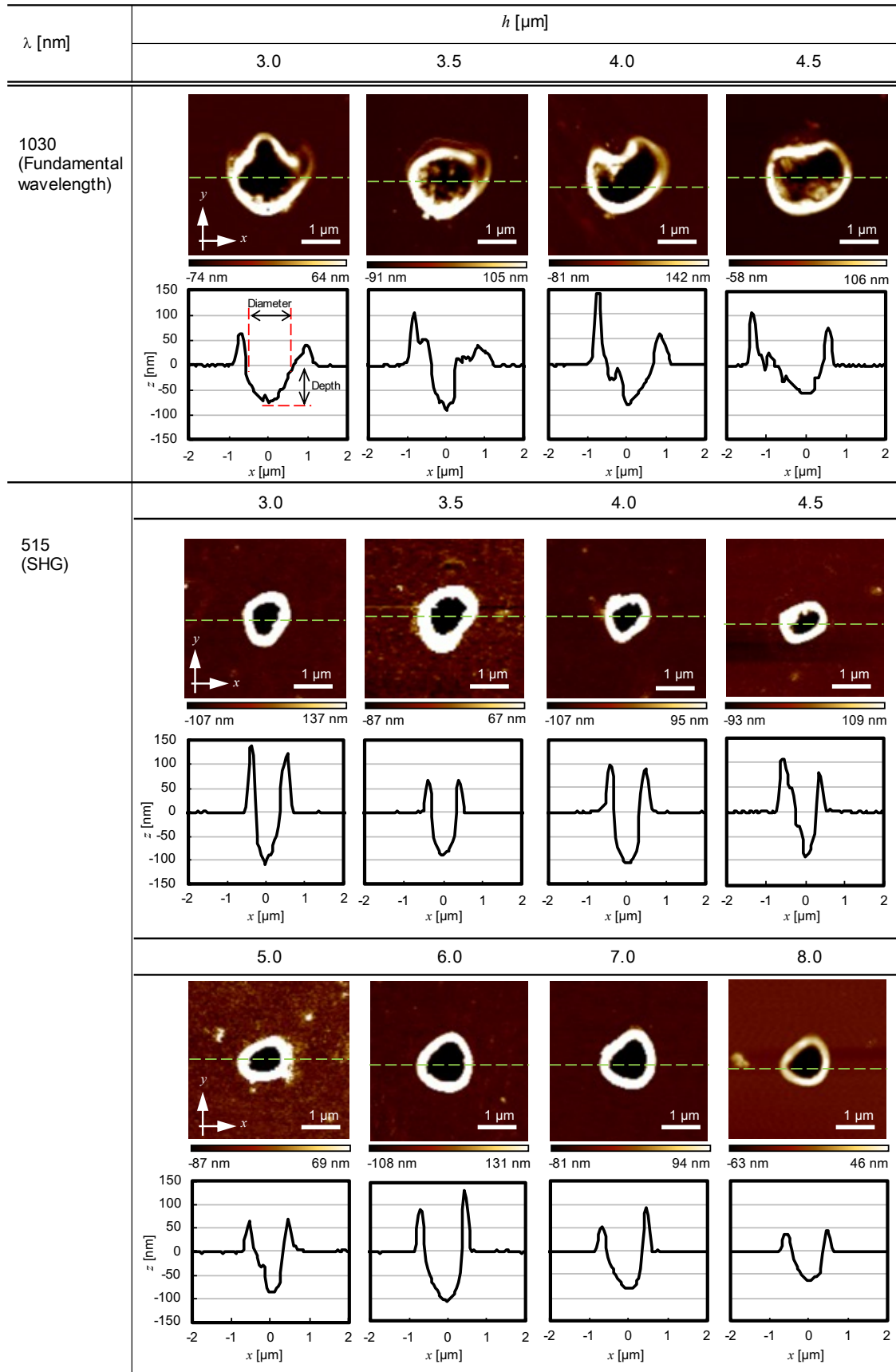
The single-shot craters obtained from PNJ machining in a water medium with the fundamental wavelength ($\lambda = 1030\text{ nm}$) and its SHG beam ($\lambda = 515\text{ nm}$) were characterized using AFM. Their cross-sectional profiles are presented in Table 5.5. Near the ablated holes, most of the machined holes appeared free of many bumpy structures containing small debris on the sample surface, compared with previous PNJ machining results in an air medium [37]. This indicates the beneficial effect of water as a flushing medium to remove ejected materials.

Furthermore, smooth ablation results were obtained because water's high density and thermal conductivity caused the plasma generated during machining to be highly confined and quickly quenched. Therefore, the confinement of the liquid resulted in a smaller plasma plume with a shorter duration, thereby reducing the plasma shielding effect [32, 108]. Consequently, more laser energy was directed at the target material. Combined with the flushing effect, this contributed to the overall improvement in machining quality and surface finish [108, 109]. Upon PNJ machining with the fundamental wavelength, concentric periodic patterns appeared on the periphery of the crater, which indicated surface melting [110]. This appearance was associated with plume- or bubble-driven shock waves, which subsequently caused surface capillary waves in the silicon melt when irradiated with a

Table 5.4 PNJ machining with different input wavelength experimental parameter.

Item	Parameter	Value
Incident laser	wavelength	Fundamental ($\lambda = 1030\text{ nm}$) SHG ($\lambda = 515\text{ nm}$)
	polarization	x linear
	energy	50 nJ
	number of pulses	1 shot
Machining parameter	defocusing distance (z_{sp})	$z_{sp} = -14\text{ }\mu\text{m}$ ($\lambda = 1030\text{ nm}$) $z_{sp} = -19\text{ }\mu\text{m}$ ($\lambda = 515\text{ nm}$)
	distance (h)	3.0–8.0 μm (0.5 μm pitch)

Table 5.5 Comparison of ablation results using position-controlled PNJ and cross-sectional profile observed using AFM.



near-infrared laser beam [111]. Table 5.5 also shows that the machined holes had a conical shape. A mound that appeared at the edge of the machined holes was caused by material expansion during the laser processing of the materials [112]. The hole diameter and hole depth were defined based on the reference height of the measured area. The distance between the intersection of the reference height and the cross-sectional profile was defined as the diameter of the hole. The distance from the reference height to the deepest position in the z -direction was defined as the hole depth.

A summary of the machined hole characteristics is shown in Figure 5.8 and Table 5.6. The error bar in Figure 5.8 indicates the consistency of the machined holes. When using a 1030 nm laser input, ablation was limited to $h = 4.5 \mu\text{m}$, whereas ablation was observed up to $h = 8.0 \mu\text{m}$ with SHG. In comparison, ablation was only observed when the microsphere was in the proximity of the sample ($h = 0 \mu\text{m}$) when using the 1030-nm input wavelength by Abdurrochman et al. [42]. Up to a distance $h = 2 \mu\text{m}$ when using the 800 nm input wavelength by Uenohara et al. [37] when PNJ machining was done in an air medium, as shown in Table 2.1. Furthermore, hole machining could not be performed outside the distance h because the fluence of the PNJ at this position was below the ablation threshold of the Si substrate. In addition, the DOF of the PNJ indicated how well the laser intensity was maintained over a long distance. Thus, the more extended propagation profile in the SHG-generated PNJ yielded a longer depth of focus, making it more effective for fabrication at longer distances.

In terms of the hole diameter, Figures 5.8(a) and (b) show that ablated holes were consistently smaller when using SHG. This confirmed that a smaller PNJ beam diameter was successfully achieved and utilized for machining with SHG, aligning with the numerical calculations shown in Figure 5.2. The smallest hole diameter in the x -direction was $0.55 \pm 0.16 \mu\text{m}$ at $h = 4.0 \mu\text{m}$ with SHG, compared with $0.92 \pm 0.38 \mu\text{m}$ at $h = 4.5 \mu\text{m}$ with the fundamental wavelength. In addition, the smallest hole diameter in the y -direction was $0.55 \pm 0.15 \mu\text{m}$ at $h = 4.5 \mu\text{m}$ with SHG, compared with $0.57 \pm 0.15 \mu\text{m}$ at $h = 4.5 \mu\text{m}$ with the fundamental wavelength. Compared with the established PNJ machining in an air medium, the measured hole diameter was $1.28 \mu\text{m}$ when using the fundamental wavelength [42]. The advantage of SHG-based PNJ machining for ultraprecise applications was demonstrated by its consistent ability to obtain submicrometer hole diameters.

As shown in Fig. 5.8(c), all ablated holes were consistently deeper when using SHG for h values between 3.0 and 4.5 μm . This confirmed that the higher PNJ intensity at 515 nm enhances

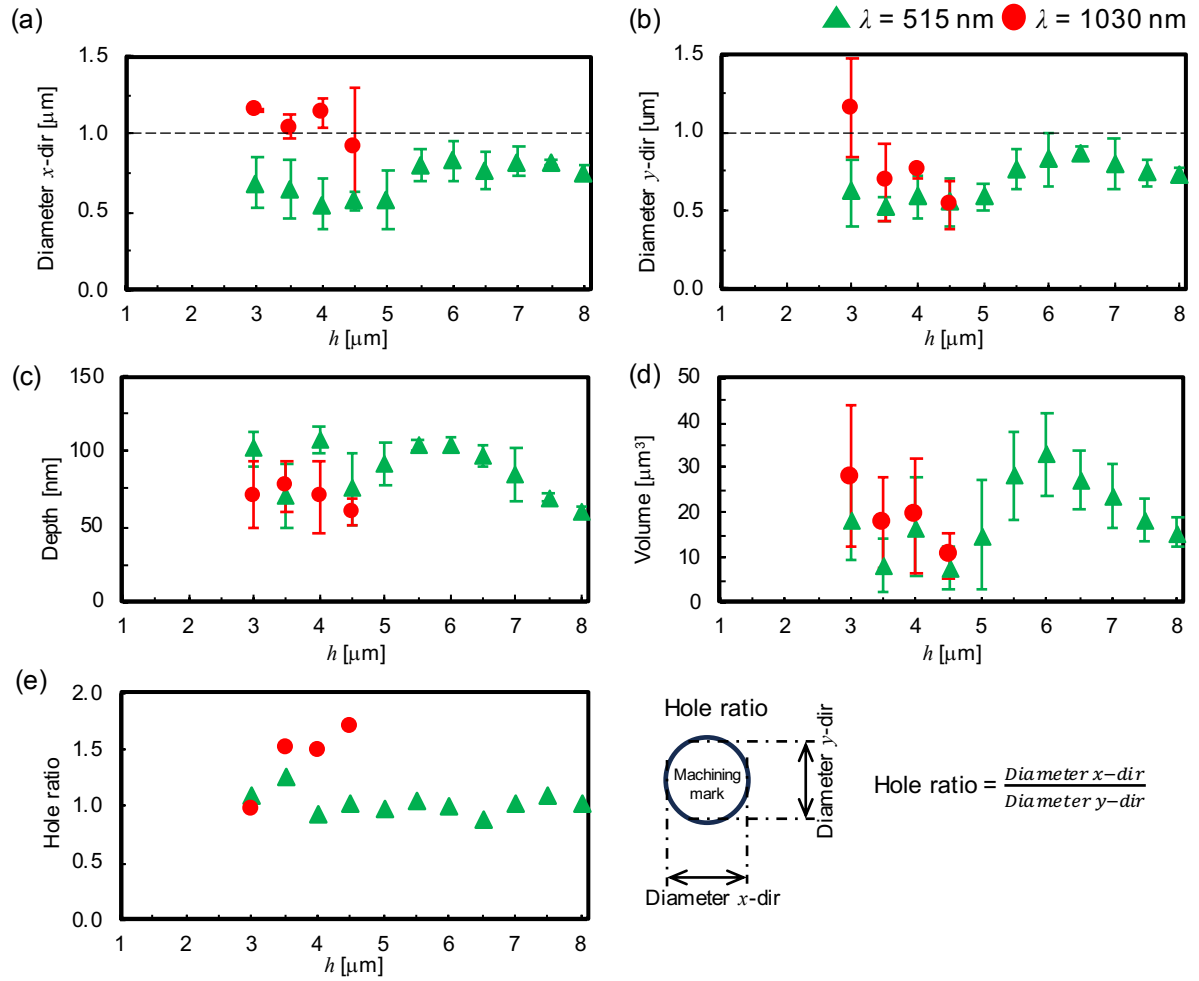


Fig. 5.8 Machined hole diameter in the (a) x -direction and (b) y -direction; (c) hole depth; (d) volume of the evaluated ablated holes; and (e) hole ratio. Vertical error bars represent the standard deviation in the measured machining spot size.

material removal. The increased intensity enabled photons to penetrate deeper into the sample, thereby producing more substantial ablation. The deepest holes were evaluated at $108 \pm 9 \text{ nm}$ for $h = 4.0 \mu\text{m}$ (SHG), compared with $77 \pm 17 \text{ nm}$ for $h = 3.5 \mu\text{m}$ (1030 nm). Compared with the PNJ machining in an air medium done by Abdurrochman et al. [42] at the fundamental wavelength, only a 23 nm hole depth was obtained. These results suggest that silicon is more responsive to SHG-enhanced PNJ machining because of the improved photon–material interaction at this wavelength [113].

The ablation diameter and depth fluctuations over the distance h were related to the PNJ intensity distribution in the propagation direction, as shown in Figure 5.8(f). The different focusing distances, h , correspond to different PNJ beam diameters and the spatial distribution of the laser intensity.

Table 5.6 Summary of ablation results by SHG-enhanced PNJ machining in a water medium.

h [μm]	$\lambda = 1030$ nm (Fundamental)				$\lambda = 515$ nm (SHG)			
	Dia x -dir [μm]	Dia y -dir [μm]	Depth [nm]	Volume [μm^3]	Dia x -dir [μm]	Dia y -dir [μm]	Depth [nm]	Volume [μm^3]
3.0	1.15 ± 0.02	1.16 ± 0.32	71 ± 23	28 ± 16	0.69 ± 0.17	0.62 ± 0.21	102 ± 11	18 ± 9
3.5	1.04 ± 0.08	0.68 ± 0.24	77 ± 17	18 ± 10	0.65 ± 0.19	0.56 ± 0.07	70 ± 21	8 ± 6
4.0	1.14 ± 0.10	0.75 ± 0.04	70 ± 24	19 ± 13	0.55 ± 0.16	0.58 ± 0.14	108 ± 9	17 ± 11
4.5	0.92 ± 0.38	0.57 ± 0.15	59 ± 9	10 ± 5	0.57 ± 0.06	0.55 ± 0.15	75 ± 25	8 ± 5
5.0					0.58 ± 0.18	0.59 ± 0.09	92 ± 14	15 ± 12
5.5					0.80 ± 0.10	0.77 ± 0.12	105 ± 4	28 ± 10
6.0					0.83 ± 0.13	0.83 ± 0.18	105 ± 4	33 ± 9
6.5					0.77 ± 0.11	0.86 ± 0.04	97 ± 7	27 ± 7
7.0					0.82 ± 0.10	0.80 ± 0.16	84 ± 18	24 ± 7
7.5					0.82 ± 0.02	0.74 ± 0.08	69 ± 3	18 ± 5
8.0					0.75 ± 0.04	0.73 ± 0.05	60 ± 4	16 ± 3

The working distance of the PNJ was the position where the highest PNJ intensity was generated. Therefore, when h was set near the working distance, the machined holes were ablated broadly and deeply due to the high impact energy on the workpiece, resulting in a larger ablated volume. Meanwhile, smaller and shallower holes were obtained when the distance h was set far from the working distance because of the lower PNJ intensity. According to Tian et al. [109], a positive defocusing condition, where the highest laser intensity is set above the Si surface, influences the instability of the machined region because of induced plasma and microbubbles in the region between the highest laser intensity and the sample surface. In the case of PNJ, a positive defocusing condition indicated that h was set beyond the working distance. Therefore, the machined holes exhibited

instability. Other factors, such as pressurized liquid, flowing liquid, and higher liquid temperatures, can improve machining stability in a liquid medium [108, 114, 115].

The ablated volume of the machined holes, as shown in Fig. 5.8(d), indicated that the fundamental input wavelength resulted in slightly higher total material removal in a comparable processing distance ($h = 3.0\text{--}4.5\text{ }\mu\text{m}$). This was primarily due to the larger PNJ beam diameter obtained using the fundamental wavelength, despite the shallow ablation depth. However, the difference in total material removal between the fundamental wavelength and SHG was relatively small, with the highest material removal volume evaluated at $33 \pm 9\text{ }\mu\text{m}^3$ ($h = 6.0\text{ }\mu\text{m}$) for SHG being only slightly higher than that using a 1030 nm input wavelength at $28 \pm 16\text{ }\mu\text{m}^3$ ($h = 3.0\text{ }\mu\text{m}$). This result confirms that SHG can achieve a balance between high precision and effective material removal.

The hole shape was also evaluated, and the results are shown in Fig. 5.8(e). The hole shape was defined by the hole ratio, where the hole diameter in the x -direction was divided by the hole diameter in the y -direction. Therefore, when perfectly circular machining marks were obtained, the hole ratio was equal to one. Accordingly, circular machining marks were obtained at all distances when PNJ machining was done using the SHG beam with a Gaussian profile. Meanwhile, a slightly elliptical shape with a wider diameter in the x -direction was obtained when using fundamental wavelength input. The x -linear polarization of the input laser caused a slight change in the hole diameter in the y -direction because nonlinear absorption mechanisms were more effective in the x -direction, in addition to the lower photon–material interaction using the fundamental wavelength [116, 117]. Uenohara et al. [69] suggested that radially polarized light could improve the machining shape. In addition, controlling cavitation bubbles during machining could improve the results, although this is a challenging task during water-assisted machining [32, 111].

Surface oxidation, a processing defect in post-processing laser ablation, is undesirable because it can compromise material integrity while increasing surface roughness due to the formation of oxide layers during laser processing [118, 119]. Therefore, low oxidation levels in laser processing must be controlled. Here, postablative chemical modification on the silicon surface was analyzed using energy-dispersive X-ray spectroscopy (EDS). Figure 5.9 shows the results of EDS measurement using a scanning electron microscope (JEOL JSM-F100). According to Figure 5.9, SHG-enhanced PNJ machining in a water medium resulted in a low oxygen peak at all controlled distances h . Figures

5.9(a) and (b) show the EDS results at the near working distance ($h = 3.0 \mu\text{m}$), whereas Figures 5.9(c) and (d) show the EDS results at the furthest distance from the microsphere ($h = 8.0 \mu\text{m}$). These aligned with the experimental results of Goodarzi and Hajiesmaeilbaigi [111] and Saraeva et al. [120], where the machining process in a liquid medium resulted in a minimal oxygen peak compared with machining in air. In a water medium, the content of dissolved oxygen in water was far less than that in air. The change in the source of oxygen elements caused the significantly lower oxygen content of materials in the water environment compared to that in the air. The cooling effect of the water medium alleviated heat accumulation during the machining process, reducing the material's temperature [118, 120].

According to these findings, PNJ machining in water significantly improved the surface quality of silicon holes with minimal damage. These findings collectively demonstrate that SHG-enhanced PNJ machining in a water medium not only allows for deeper and more precise ablation but also minimizes thermal effects and machining debris, making it a promising method for future high-resolution laser microfabrication applications.

5.5 Influence on focus position for PNJ machining

In order to investigate the effect of laser defocusing for PNJ machining, a hole machining experiment was conducted with the experimental setup shown in Figure 5.3 and parameters shown in Table 5.7, in addition to Table 5.2. In this set of experiments, a single-shot laser with 50-nJ energy levels was used to irradiate the microsphere, producing PNJ. z_{sp} was set to -9 and $-19 \mu\text{m}$ for SHG (515 nm). h was controlled from 2.0 to 8.0 μm with a 0.5 μm -pitch. A complete list of experimental parameters in this experiment is shown in Table 5.7 as an addition to Table 5.2.

The single-shot craters obtained from PNJ machining in a water medium with different laser defocusing states were characterized using AFM. Their cross-sectional profiles are shown in Figure 5.10(a) at the closest distance from the rear microsphere surface ($h = 2.0 \mu\text{m}$). The furthest one ($h = 8.0 \mu\text{m}$), showing that PNJ has a very long DOF if we could adjust the defocusing distance during the processing. Additionally, clean ablation results were observed near the ablation area. A summary of the machined hole characteristics is shown in Table 5.8. Figures 5.10(b) and (c) show the recap of

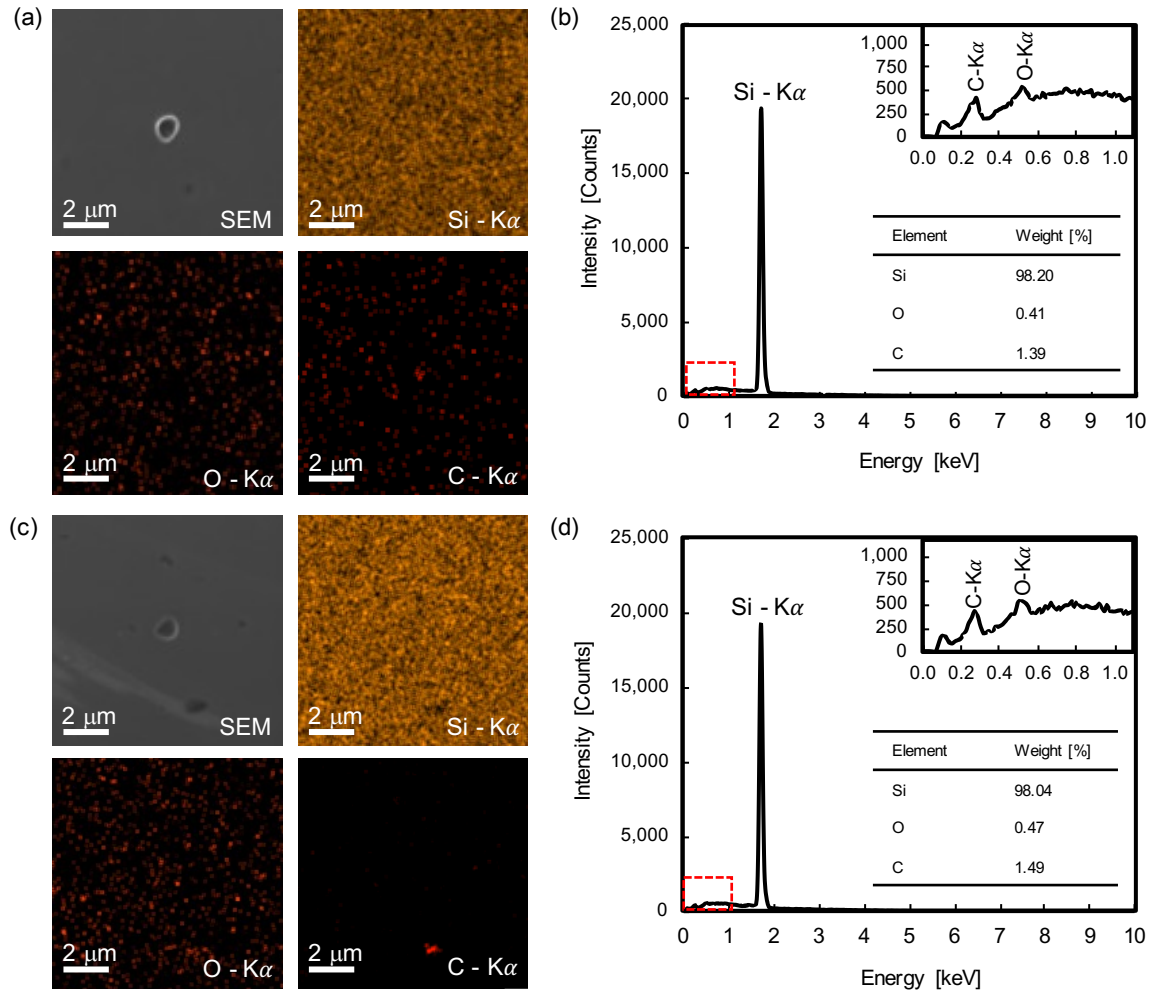


Fig. 5.9 EDS analysis on the silicon surface after PNJ machining with controlled distance at $h = 3.0 \mu\text{m}$: (a) elemental mapping and (b) spectra, and $h = 8.0 \mu\text{m}$: (c) elemental mapping and (d) spectra, including detailed insights and elemental weight percentages.

Table 5.7 PNJ machining with different input wavelength experimental parameter.

Item	Parameter	Value
Incident laser	wavelength	SHG ($\lambda = 515 \text{ nm}$)
	polarization	x linear
	energy	50 nJ
	number of pulses	1 shot
Machining parameter	defocusing distance (z_{sp})	$z_{sp} = -9$ and $-19 \mu\text{m}$
	distance (h)	2.0–8.0 μm (0.5 μm pitch)

hole diameter in x -direction and depth with different h , respectively. As shown in Figure 5.10(b), the smallest diameter obtained when $z_{sp} = -9 \mu\text{m}$ was $0.42 \pm 0.01 \mu\text{m}$ for $h = 6.0 \mu\text{m}$ while at $z_{sp} = -19$

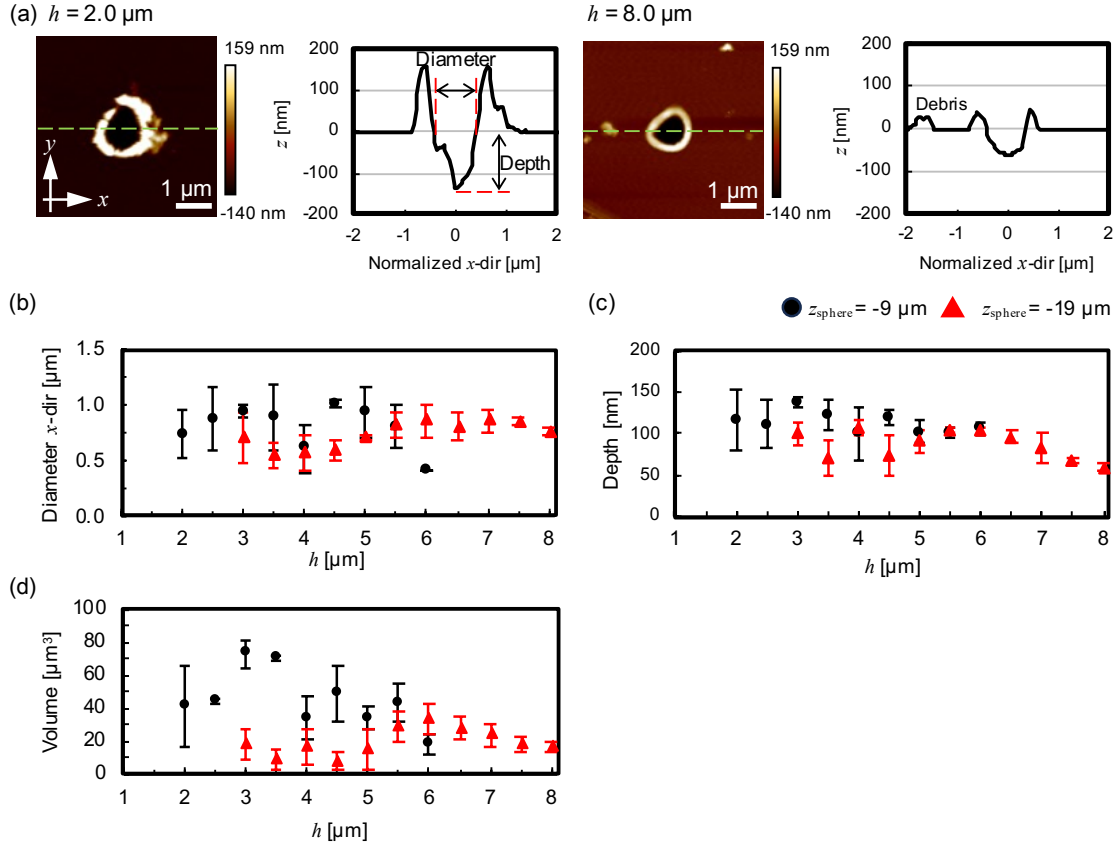


Fig. 5.10 PNJ machining results measured by AFM and (a) the cross-section profile at the closest and furthest h . Results of hole (b) diameter in x -direction, (c) depth, and (d) ablated volume with different h .

μm was $0.55 \pm 0.16 \mu\text{m}$ for $h = 3.5 \mu\text{m}$. When the PNJ is at peak intensity, it has the smallest beam diameter, resulting in the smallest hole diameter. From the experiment, sub-micrometer holes were obtained at all defocusing distances. Therefore, we could obtain a very long processing distance if we could control the defocusing parameter during the processing.

In terms of hole depth, when $z_{sp} = -9 \mu\text{m}$, the hole depth is at 100 nm on average, while $z_{sp} = -19 \mu\text{m}$ produces a hole depth of 75 nm on average. The deepest holes were evaluated at 139 ± 5 nm for $h = 2.5 \mu\text{m}$ at $z_{sp} = -9 \mu\text{m}$, compared with 108 ± 9 for $h = 4.0 \mu\text{m}$ nm at $z_{sp} = -19 \mu\text{m}$. From these results, the deepest holes were obtained at near working distance, $1.96 \mu\text{m}$ at $z_{sp} = -9 \mu\text{m}$ and $2.51 \mu\text{m}$ at $z_{sp} = -19 \mu\text{m}$, showing a relationship between intensity and ablation depth. When PNJ is at a working intensity, it is known that PNJ has its peak intensity along the propagation path. From these results, we could also suggest that a shorter defocusing distance will result in a deeper ablation,

Table 5.8 Summary of ablation results by SHG-enhanced PNJ machining with different defocusing condition in a water medium.

h [μm]	$z_{sp} = -9 \mu\text{m}$				$z_{sp} = -19 \mu\text{m}$			
	Dia x -dir [μm]	Dia y -dir [μm]	Depth [nm]	Volume [μm^3]	Dia x -dir [μm]	Dia y -dir [μm]	Depth [nm]	Volume [μm^3]
2.0	0.79 ± 0.17	0.91 ± 0.28	124 ± 42	41 ± 24				
2.5	1.04 ± 0.05	0.79 ± 0.13	131 ± 2	45 ± 2				
3.0	0.92 ± 0.06	1.20 ± 0.05	139 ± 5	73 ± 9	0.69 ± 0.17	0.62 ± 0.21	102 ± 11	18 ± 9
3.5	1.00 ± 0.15	1.13 ± 0.07	131 ± 0	70 ± 2	0.65 ± 0.19	0.56 ± 0.07	70 ± 21	8 ± 6
4.0	0.71 ± 0.09	0.94 ± 0.27	119 ± 8	34 ± 13	0.55 ± 0.16	0.58 ± 0.14	108 ± 9	17 ± 11
4.5	0.97 ± 0.04	1.08 ± 0.12	121 ± 8	49 ± 18	0.57 ± 0.06	0.55 ± 0.15	75 ± 25	8 ± 5
5.0	0.99 ± 0.21	0.88 ± 0.21	109 ± 14	34 ± 7	0.58 ± 0.18	0.59 ± 0.09	92 ± 14	15 ± 12
5.5	0.80 ± 0.19	1.22 ± 0.23	103 ± 6	43 ± 12	0.80 ± 0.10	0.77 ± 0.12	105 ± 4	28 ± 10
6.0	0.42 ± 0.01	0.78 ± 0.24	107 ± 9	17 ± 6	0.83 ± 0.13	0.83 ± 0.18	105 ± 4	33 ± 9
6.5					0.77 ± 0.11	0.86 ± 0.04	97 ± 7	27 ± 7
7.0					0.82 ± 0.10	0.80 ± 0.16	84 ± 18	24 ± 7
7.5					0.82 ± 0.02	0.74 ± 0.08	69 ± 3	18 ± 5
8.0					0.75 ± 0.04	0.73 ± 0.05	60 ± 4	16 ± 3

showing a higher confinement effect compared to a longer defocusing distance. However, it comes with a trade-off that the DOF is shorter when using a shorter defocusing distance. Interestingly, the hole diameter and hole depth showed the same tendency with the PNJ profile on the z -direction as shown in Figure 5.2.

Regarding material removal, highest material removal were evaluated at $73 \pm 9 \mu\text{m}^3$ for $h = 3.0 \mu\text{m}$ at $z_{sp} = -9 \mu\text{m}$, compared with 33 ± 9 for $h = 6.0 \mu\text{m}$ nm at $z_{sp} = -19 \mu\text{m}$. These results show that the highest material removal is obtained at a near working distance, indicating that a higher PNJ

intensity is achieved near the working distance. These experiments demonstrate that by adjusting the defocusing distance, sub-micrometer ablation can be achieved with a very long processing distance using PNJ machining in a water medium.

5.6 Discussion

In this study, we showed the clear benefits of using SHG in PNJ machining in a water medium. By using the SHG wavelength of 515 nm instead of the fundamental wavelength of 1030 nm, we achieved significant improvements in machining characteristics. These improvements include a notable reduction in machining mark diameters to below 600 nm, deeper ablation, a longer processing distance of up to 8 μm , and better machining uniformity and repeatability. Using water as the surrounding medium also enhanced the quality of the fabricated structures. It helped reduce oxidation and debris formation at the ablation surface. The combined effects of SHG illumination and the water-assisted environment enable cleaner, finer, and more consistent laser machining. This demonstrates clear advantages over PNJ machining in air medium.

The other experimental results showed that laser defocusing has a significant impact on the PNJ machining process. Defocusing changes the spatial intensity distribution and focal position relative to the microsphere. This affects both the nanojet confinement and the resulting ablation profile. This effect is particularly significant in applications that require maintaining high precision at greater distances from the microsphere or when creating structures with varying topographies. By carefully controlling the defocusing during laser processing, we can create a PNJ with a more extended DOF using the SHG beam. This provides more flexible and adjustable microfabrication conditions. This capability broadens the functional range of PNJ laser processing, allowing for the creation of complex 3D structures with nanometric precision.

Overall, our findings highlight the strong potential of SHG-enhanced PNJ machining in a water medium for improving high-precision laser microfabrication technologies. The observed improvements in resolution, depth control, and surface quality suggest valuable applications in areas where submicrometer-scale accuracy and minimal thermal damage are crucial. These areas include the fabrication of MEMS, optoelectronic components, and semiconductor devices. Additionally, the

combination of SHG input, water-assisted processing, and controlled defocusing offers a flexible approach for achieving stable, reproducible, and high-quality microstructuring. This study lays the foundation for future advancements in PNJ-assisted laser processing systems that can meet the stringent demands of next-generation micro- and nanofabrication applications.

Chapter 6

Conclusions

In this chapter, we consolidate the thesis's essence by providing a concise yet comprehensive overview of each preceding chapter. This segment serves as a culmination of the research journey, offering brief discussions and conclusions drawn from Chapters 2 through 5. Each chapter explored distinct methodologies and applications, ranging from the principles and theoretical foundations of proposed analysis techniques to their practical implementations in various contexts. By synthesizing the key findings and insights gleaned from individual chapters, this section aims to provide a holistic perspective on the thesis's contributions to the field of laser ablation techniques.

Chapter 2:

Chapter 2 explains the theoretical framework for using PNJ-assisted in laser machining within a water medium. This serves as the basis for the experiments discussed in later chapters. It describes how ultrashort-pulse laser ablation works. This method can achieve sub-micrometer precision by reducing thermal effects and minimizing the heat-affected zone. When this process happens in a liquid environment, called Pulsed Laser Ablation in Liquid (PLAL), it benefits from better plasma confinement, effective debris removal, and smoother surface formation compared to ablation in air. These features highlight the advantages of liquid-assisted ultrafast laser processing for high-resolution and thermally stable microfabrication.

The chapter also discusses the photonic nanojet phenomenon. In this case, a focused laser beam interacts with dielectric microspheres. This interaction creates a highly localized electromagnetic beam that is smaller than the diffraction limit. This near-field optical effect enables submicrometer to nanometer-scale machining with high accuracy. The main properties of PNJs, including PNJ beam diameter, working distance (WD), and depth of focus (DOF), depend on factors like refractive index contrast, microsphere shape, and laser settings. In a water medium, PNJs can provide small beam diameter for high resolution machining, longer working distances for improved alignment tolerance, longer depth of focus for increasing material removal, and less surface damage. By adjusting the intensity distribution through careful parameter selection, machining performance can be optimized. Therefore, Chapter 2 establishes the important theoretical link between ultrafast laser ablation and PNJ effects in a water medium.

Chapter 3:

Chapter 3 presents a detailed numerical investigation of PNJ formation and behavior in a water

medium using the FDTD method based on Maxwell's equations. The study reveals that PNJs formed in water have a WD nearly nine times longer than those in air, highlighting the advantages of a water medium for increasing alignment tolerance. As the surrounding medium refractive index, n_s , increases, both the WD and DOF extend while the beam diameter remains nearly constant, making water an ideal medium for stable PNJ formation. The analysis further shows that larger microspheres produce longer WDs, whereas higher microsphere refractive indices enhance optical confinement but reduce both WD and DOF. Effective PNJ generation occurs when the refractive index contrast (RIC = n_m/n_s) ranges between 1 and 2; beyond this range, excessive confinement limits external focusing.

Laser parameters also play a significant role in shaping PNJ characteristics. Varying the wavelength from 400 to 1000 nm increase the beam diameter but decreases WD and DOF. Furthermore, adjusting the defocusing distance (z_{sp}) shows that at $z_{sp} = -14 \mu\text{m}$, the beam diameter reaches its minimum ($\sim 0.6 \mu\text{m}$) for SiO_2 microspheres, emphasizing the importance of precise focusing conditions. 3D FDTD simulations of PNJ-assisted laser ablation on silicon substrates submerged in water confirm the capability for submicrometer machining. Specifically, PS and SiO_2 microspheres yield ablation diameters of approximately 500 nm and 540 nm, respectively, with optimal microsphere–substrate distances of 1.0–4.5 μm for PS and 2.0–5.5 μm for SiO_2 . Altogether, these findings establish a coherent framework for optimizing PNJ formation in water, demonstrating that careful control of microsphere and laser parameters enables stable, precise, and thermally efficient submicrometer laser machining.

Chapter 4:

Chapter 4 shows the results of experiment on PNJ assisted machining in a water medium. It compares this method to conventional laser machining, which involves a focused laser beam. A femtosecond laser ($\lambda = 800 \text{ nm}$, $<100 \text{ fs}$, 80 MHz) and a 0.5 NA objective lens were used. A confocal optical system ensured precise nanometer-scale alignment. Single-crystal silicon substrates were processed in air and in deionized water layers that were 1.0 mm and 1.5 mm thick. In conventional laser machining, the ablation diameters increased with power. However, in water, the diameters were smaller and cleaner due to better cooling and debris removal. Average ablation sizes were about 1.0 μm in air, 0.8 μm in 1.0 mm water, and 0.7 μm in 1.5 mm water. In the air, the larger ablation area was affected

by thermal melting and redeposition. In contrast, machining in water produced smoother surfaces. However, using thicker water layers slightly lowered the ablation rate due to scattering and absorption.

For PNJ-assisted machining, an 8 μm SiO_2 microsphere held by a micropipette created sub-wavelength focusing in water at 585 mW and 160,000 pulses. By changing the distance from the microsphere to the sample ($h = 0 \sim 6 \mu\text{m}$), ablation was seen between $h = 2.0$ and $5.0 \mu\text{m}$. The smallest holes, 0.6 μm , were achieved around $h = 3.0 \mu\text{m}$, which is approximately half the size obtained by traditional machining and well below the diffraction limit. Unlike PNJ machining in air, where ablation occurred only up to $h = 2 \mu\text{m}$ and debris often stuck to the microsphere, PNJ machining in water provided a more extended DOF (3 μm), less redeposition, and cleaner features. 3D FDTD simulations supported these findings, showing a PNJ beam diameter of 0.58 μm located 2.94 μm behind the microsphere and DOF of 2.58 μm . The effective ablation range matched the experiments. Overall, PNJ machining in water provided finer resolution, less thermal damage, and superior surface quality compared to both traditional and air-based processes. This illustrates its strong potential for future MEMS/NEMS microfabrication.

Chapter 5

This chapter presents a study on improving PNJ machining by using second-harmonic generation (SHG) in water. The investigation builds on earlier research that showed the feasibility of PNJ machining. It aims to improve machining resolution, WD, and DOF. We analyzed PNJ behavior under different input wavelengths: 1030 nm (fundamental) and 515 nm (SHG). We employed 3D-FDTD simulations for this purpose. The results indicated that the shorter SHG wavelength produced a smaller PNJ beam diameter of 0.43 μm and a longer WD of 4.18 μm compared to the fundamental wavelength. This meant better light control and more efficient energy use. These findings confirmed that SHG illumination can significantly enhance PNJ performance for submicrometer-scale machining.

We validated the results using a femtosecond laser system, which used a barium borate (BBO) crystal to generate the SHG beam. This setup allowed a direct comparison of PNJ machining at both wavelengths on silicon substrates immersed in water. The SHG-enhanced PNJ machining resulted in smaller and deeper ablation marks, with diameters of less than 600 nm. It increased the effective machining distance to 8 μm . The water medium improved machining precision and surface quality by reducing oxidation, debris, and thermal damage through effective cooling and flushing. The results

showed that combining SHG illumination with water immersion leads to better performance than traditional air-based PNJ machining. It achieves cleaner, finer, and more consistent ablation results.

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- [1] Reza Aulia Rahman, Tsutomu Uenohara, Yasuhiro Mizutani, and Yasuhiro Takaya. First step toward laser micromachining realization by photonic nanojet in water medium. *International Journal of Automation Technology*, 15(4):492–502, 2021.
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Award List

Award title:

[1] Best Presentation Award

Presentation title: Study on In-Liquid Laser Nanomachining by Photonic Nanojet in Optical Tweezers

Configuration (1st report) –Investigation on machining realization by numerical simulations–

Conference name: 2019 JSPE Autumn Conference

Date: 4-6 September 2019

[2] Best Paper Award.

Presentation title: Position controlled photonic nanojet for machining in water medium by second harmonic generation

Conference name: 16th International Symposium on Measurement Technology and Intelligent Instruments (ISMTII) 2025

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