

# fs Laser-Triggered Dynamic Vapor Nanobubble Generation from Nanopillar Plasmonic Nanotransducer Arrays

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**Abstract:** Vapor nanobubble is generated on nanopillar plasmonic nanotransducer arrays using femtosecond laser. Its spatial-temporal dynamics is extracted and analyzed, facilitating the development of highly precise and efficient nanoscale cell manipulation techniques. © 2024 The Author(s)

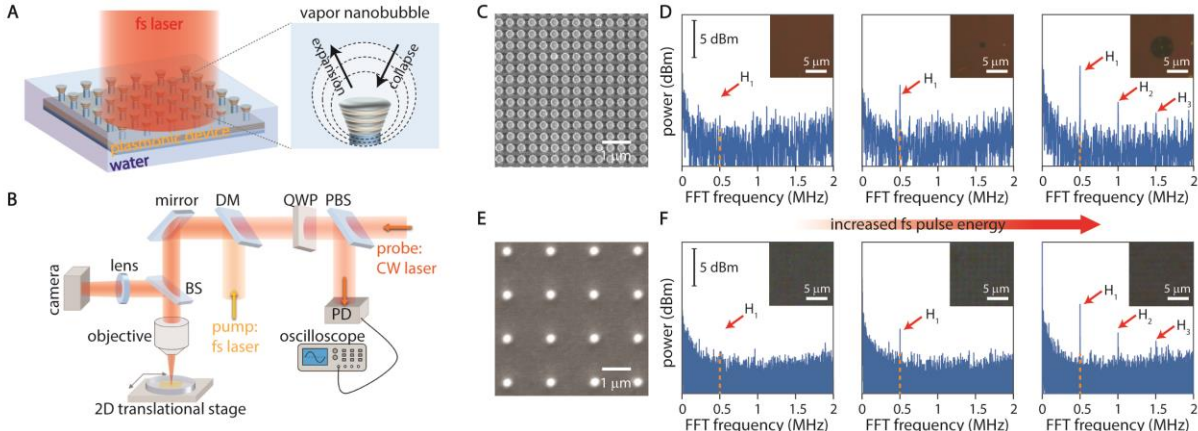
## 1. Introduction

Plasmonic nanotransducers are known for their exceptional photothermal conversion efficiency and substantial spatial-temporal thermal confinement when irradiated with ultrashort pulse lasers, which can enable the rapid formation of vapor bubbles in liquids, offering precision, selectivity, and localization [1]. This feature makes plasmonic nanotransducers advantageous for biological applications such as targeted drug delivery and photothermal therapy [2]. Past studies have predominantly focused on nanobubble/microbubble dynamics from noble metal nanoparticles [3]. However, the use of nanoparticles often faces challenges like random distribution, unstable nanobio interfacing, limited precision, potential biological toxicity, and issues in cellular clearance. In contrast, integrating plasmonic nanotransducers with nanopillar bio-nanodevices presents a promising avenue to achieve precise photothermal actuation and nanosurgery for cells that spontaneously engulf nanopillar bio-topology [4]. Therefore, investigating the spatial-temporal dynamics of vapor nanobubbles generated from nanopillar plasmonic nanotransducer arrays under femtosecond (fs) laser irradiation is of great importance, facilitating the development of precise and efficient nanoscale cell manipulation techniques with potential applications in diverse biomedical contexts.

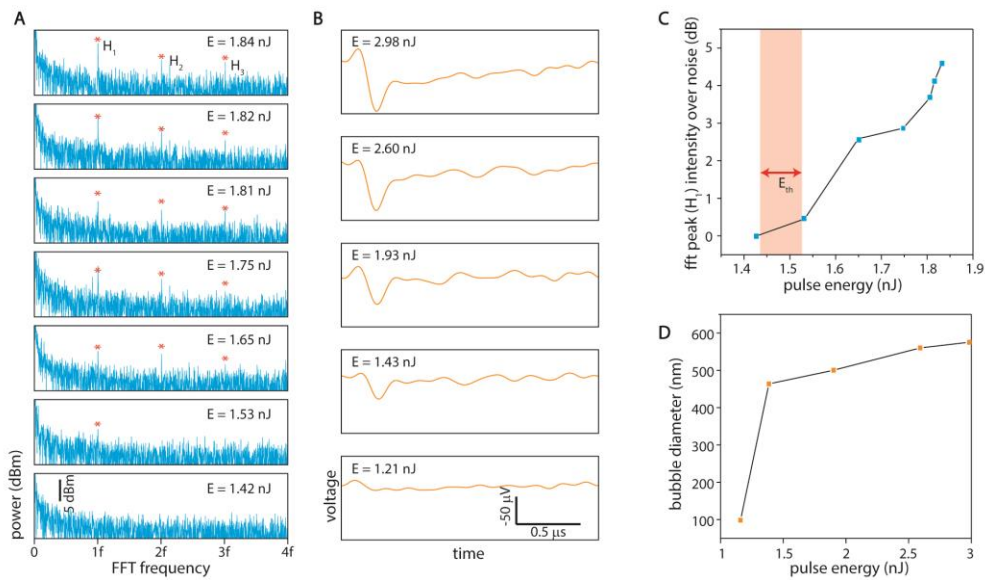
## 2. Results and discussion

Here, we experimentally investigate the spatial-temporal dynamics of vapor nanobubbles generated on a planar metal-insulator-metal (MIM) nanolaminate plasmonic nanotransducers under fs laser irradiation (Figure 1A). The MIM nanolaminate plasmonic substrate in water is irradiated by fs pump pulses (950 nm, 0.5 MHz) with a beam size of around  $13 \mu\text{m}^2$  through a  $20\times$  objective with an NA 0.4 (Figure 1B). A continuous wave (CW) probe laser (780 nm) is focused collinearly with the pump laser, and its back reflection is collected by the same objective and monitored with a photodetector (PD). First, we examine the MIM nanolaminate nanopillar (100 nm diameter) array with 400 nm pitch (Figure 1C). The fast Fourier transform (FFT) is used to analyze the PD signal in the frequency domain. With increasing the fs pulse energy, peaks at harmonics of the fundamental repetition rate appear, and their intensities increase (Figure 1D), revealing the cause by the scattering of the generated vapor bubbles on the plasmonic surface, and the appearance of the harmonics can serve as the criteria for generating vapor bubbles. However, it is straightforward to generate a microbubble instead of individual nanobubbles. Individual nanobubbles originate from the tens of nanotransducer within the beam spot of the pump laser, are of small spacing, and tend to merge into a microbubble. Therefore, to investigate the spatial-temporal dynamics of nanobubbles, a nanopillar array with a lower density is desired, and another MIM nanolaminate nanopillar (100 nm diameter) array with  $1.5 \mu\text{m}$  pitch (Figure 1E) is fabricated and applied, where increased fs pulse energy results in harmonics peaks, accompanied by an increase in their intensity (Figure 1F). In contrast, the microbubble is absent in the optical image. Observing the appearance of harmonic peaks and the absence of the microbubble manifests that the  $1.5 \mu\text{m}$  pitch substrate is suitable for investigating the spatial-temporal dynamics of vapor nanobubbles. A fs pulse energy sweeping is conducted using the  $1.5 \mu\text{m}$  pitch substrate to explore the dynamics of nanobubbles (Figure 2). The harmonic features indicate the existence of the vapor nanobubbles at higher pulse energies (Figure 2A). We apply an FFT shortpass filter and reconstruct the temporal signal through inverse fast Fourier transform (IFFT). By averaging and smoothing, the temporal signals are restored with an improved signal-to-noise ratio (Figure 2B). The signal drop originates from the nanobubbles' scattering loss and reflects the nanobubbles' lifetime, around 200 ns. Further, the threshold for nanobubble generation is estimated to be between 1.42 nJ and 1.53 nJ, as shown in Figure 2C. The bubble size is retrieved from the signal drop in Figure 2B and the experimental parameters by Mie theory (Figure 2D). A nanobubble over 400 nm can be formed over the threshold, consistent with our previous hypothesis that individual nanobubbles tend to merge into a microbubble in the 400 nm pitch substrate situation.

In conclusion, our experimental investigations have provided valuable insights into the spatial-temporal dynamics of vapor nanobubbles generated on nanopillar plasmonic nanotransducer arrays when subjected to fs laser irradiation. This study advances our understanding of vapor nanobubble behavior on such surfaces and contributes significantly to the broader field of plasmonic nanotechnology, particularly in its application in precision medical interventions like targeted drug delivery and nanosurgery.



**Figure 1.** (A) Schematic illustration of plasmonic-assisted vapor nanobubble generation under fs pulse irradiation with nanopillar plasmonic nanotransducer arrays. (B) The experimental setup for optical generation and detection of vapor bubbles. DM: dichroic mirror. QWP: quarter waveplate. PBS: polarized beam splitter. BS: beam splitter. PD: photodetector. Top-view SEM images of MIM nanolaminar nanopillar (100 nm diameter) array with (C) 400 nm pitch size and (E) 1.5 μm pitch size. FFT spectra of PD signals from (D) 400 nm pitch substrate and (F) 1.5 μm pitch substrate under increased fs pulse energy. Inserted are representative camera-captured images under each condition.



**Figure 2.** (A) FFT spectra of PD signals from 1.5 μm pitch substrate under different fs pulse energies. (B) Reconstructed temporal signals under different pulse energies. (C) Vapor nanobubble generation threshold estimation based on the first harmonic (H<sub>1</sub>) intensity. (D) Vapor nanobubble diameter estimation under different pulse energies.

### 3. References

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