

Photothermal generation of programmable microbubble array on nanoporous gold disks

JINGTING LI,^{1,2} FUSHENG ZHAO,^{1,3} YU DENG,⁴ DONG LIU,⁴ CHIA-HUNG CHEN,⁵ AND WEI-CHUAN SHIH^{1,*}

¹Department of Electrical and Computer Engineering, University of Houston, 4800 Calhoun Rd. Houston, TX 77204, USA

²Currently with VKanSee Ltd., Zhongguancun Software Park Bldg. 9-203, Haidian District, Beijing, China

³Currently with Academy of Opto-Electronics, Chinese Academy of Sciences, 9 Dengzhuangnan Rd., Haidian District, Beijing, China

⁴Department of Mechanical Engineering, University of Houston, 4800 Calhoun Rd. Houston, TX 77204, USA

³Department of Biomedical Engineering, National University of Singapore, Singapore *wshih@uh.edu

Abstract: We present a novel technique to generate microbubbles photothermally by continuous-wave laser irradiation of nanoporous gold disk (NPGD) array covered microfluidic channels. When a single laser spot is focused on the NPGDs, a microbubble can be generated with controlled size by adjusting the laser power. The dynamics of both bubble growth and shrinkage are studied. Using computer-generated holography on a spatial light modulator (SLM), simultaneous generation of multiple microbubbles at arbitrary locations with independent control is demonstrated. A potential application of flow manipulation is demonstrated using a microfluidic X-shaped junction. The advantages of this technique are flexible bubble generation locations, long bubble lifetimes, no need for light-adsorbing dyes, high controllability over bubble size, and relatively lower power consumption.

© 2018 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

OCIS codes: (350.5340) Photothermal effects; (240.6680) Surface plasmons.

References and links

- 1. R. Dijkink and C. D. Ohl, "Laser-induced cavitation based micropump," Lab Chip 8(10), 1676–1681 (2008).
- A. N. Hellman, K. R. Rau, H. H. Yoon, S. Bae, J. F. Palmer, K. S. Phillips, N. L. Allbritton, and V. Venugopalan, "Laser-induced mixing in microfluidic channels," Anal. Chem. 79(12), 4484–4492 (2007).
 W. H., K. S. Lhii, O. Far, and A. T. Oht, "Hubbergh microfluidic channels," Anal. Chem. 79(12), 4484–4492 (2007).
- W. Hu, K. S. Ishii, Q. Fan, and A. T. Ohta, "Hydrogel microrobots actuated by optically generated vapour bubbles," Lab Chip 12(19), 3821–3826 (2012).
- W. D. Song, M. H. Hong, B. Lukyanchuk, and T. C. Chong, "Laser-induced cavitation bubbles for cleaning of solid surfaces," J. Appl. Phys. 95(6), 2952–2956 (2004).
- C. D. Ohl, M. Arora, R. Dijkink, V. Janve, and D. Lohse, "Surface cleaning from laser-induced cavitation bubbles," Appl. Phys. Lett. 89(7), 074102 (2006).
- 6. P. Garstecki, M. J. Fuerstman, H. A. Stone, and G. M. Whitesides, "Formation of droplets and bubbles in a microfluidic T-junction-scaling and mechanism of break-up," Lab Chip **6**(3), 437–446 (2006).
- P. Garstecki, I. Gitlin, W. DiLuzio, G. M. Whitesides, E. Kumacheva, and H. A. Stone, "Formation of monodisperse bubbles in a microfluidic flow-focusing device," Appl. Phys. Lett. 85(13), 2649–2651 (2004).
- R. Xiong, M. Bai, and J. N. Chung, "Formation of bubbles in a simple co-flowing micro-channel," J. Micromech. Microeng. 17(5), 1002–1011 (2007).
- 9. Y. C. Tan, J. S. Fisher, A. I. Lee, V. Cristini, and A. P. Lee, "Design of microfluidic channel geometries for the control of droplet volume, chemical concentration, and sorting," Lab Chip 4(4), 292–298 (2004).
- J. P. Raven and P. Marmottant, "Periodic microfluidic bubbling oscillator: Insight into the stability of two-phase microflows," Phys. Rev. Lett. 97(15), 154501 (2006).
- 11. J.-H. Tsai and L. Lin, "Transient Thermal Bubble Formation on Polysilicon Micro-Resisters," J. Heat Transfer 124(2), 375 (2002).
- D. M. van den Broek and M. Elwenspoek, "Explosive micro-bubble actuator," Sensor Actuat a-Phys 145–146, 387–393 (2008).
- P. A. Quinto-Su, V. Venugopalan, and C. D. Ohl, "Generation of laser-induced cavitation bubbles with a digital hologram," Opt. Express 16(23), 18964–18969 (2008).

https://doi.org/10.1364/OE.26.016893

Research Article

Optics EXPRESS

- T.-H. Wu, L. Gao, Y. Chen, K. Wei, and P.-Y. Chiou, "Pulsed laser triggered high speed microfluidic switch," Appl. Phys. Lett. 93(14), 144102 (2008).
- A. Vogel, J. Noack, G. Huttman, and G. Paltauf, "Mechanisms of femtosecond laser nanosurgery of cells and tissues," Appl. Phys. B 81(8), 1015–1047 (2005).
- P. A. Quinto-Su, H. H. Lai, H. H. Yoon, C. E. Sims, N. L. Allbritton, and V. Venugopalan, "Examination of laser microbeam cell lysis in a PDMS microfluidic channel using time-resolved imaging," Lab Chip 8(3), 408– 414 (2008).
- S. L. Gac, E. Zwaan, A. van den Berg, and C. D. Ohl, "Sonoporation of suspension cells with a single cavitation bubble in a microfluidic confinement," Lab Chip 7(12), 1666–1672 (2007).
- C. Zhao, Y. Liu, Y. Zhao, N. Fang, and T. J. Huang, "A reconfigurable plasmofluidic lens," Nat. Commun. 4(1), 2305 (2013).
- K. Zhang, A. Jian, X. Zhang, Y. Wang, Z. Li, and H. Y. Tam, "Laser-induced thermal bubbles for microfluidic applications," Lab Chip 11(7), 1389–1395 (2011).
- A. Q. Jian, K. Zhang, Y. Wang, S. P. Lau, Y. H. Tsang, and X. M. Zhang, "Microfluidic flow direction control using continuous-wave laser," Sens. Actuators A Phys. 188, 329–334 (2012).
- Y. Xie, C. Zhao, Y. Zhao, S. Li, J. Rufo, S. Yang, F. Guo, and T. J. Huang, "Optoacoustic tweezers: a programmable, localized cell concentrator based on opto-thermally generated, acoustically activated, surface bubbles," Lab Chip 13(9), 1772–1779 (2013).
- C. Zhao, Y. Xie, Z. Mao, Y. Zhao, J. Rufo, S. Yang, F. Guo, J. D. Mai, and T. J. Huang, "Theory and experiment on particle trapping and manipulation via optothermally generated bubbles," Lab Chip 14(2), 384–391 (2014).
- Y.-C. Wu, T.-H. Wu, D. L. Clemens, B.-Y. Lee, X. Wen, M. A. Horwitz, M. A. Teitell, and P.-Y. Chiou, "Massively parallel delivery of large cargo into mammalian cells with light pulses," Nat. Methods 12(5), 439– 444 (2015).
- Y. Zheng, H. Liu, Y. Wang, C. Zhu, S. Wang, J. Cao, and S. Zhu, "Accumulating microparticles and directwriting micropatterns using a continuous-wave laser-induced vapor bubble," Lab Chip 11(22), 3816–3820 (2011).
- E. Y. Lukianova-Hleb, I. I. Koneva II, A. O. Oginsky, S. La Francesca, and D. O. Lapotko, "Selective and selfguided micro-ablation of tissue with plasmonic nanobubbles," J. Surg. Res. 166(1), e3–e13 (2011).
- E. Y. Hleb, Y. Hu, R. A. Drezek, J. H. Hafner, and D. O. Lapotko, "Photothermal bubbles as optical scattering probes for imaging living cells," Nanomedicine (Lond.) 3(6), 797–812 (2008).
- E. Y. Lukianova-Hleb, E. Y. Hanna, J. H. Hafner, and D. O. Lapotko, "Tunable plasmonic nanobubbles for cell theranostics," Nanotechnology 21(8), 085102 (2010).
- O. Neumann, A. S. Urban, J. Day, S. Lal, P. Nordlander, and N. J. Halas, "Solar vapor generation enabled by nanoparticles," ACS Nano 7(1), 42–49 (2013).
- Z. Fang, Y. R. Zhen, O. Neumann, A. Polman, F. J. García de Abajo, P. Nordlander, and N. J. Halas, "Evolution of light-induced vapor generation at a liquid-immersed metallic nanoparticle," Nano Lett. 13(4), 1736–1742 (2013).
- D. Hühn, A. Govorov, P. R. Gil, and W. J. Parak, "Photostimulated Au Nanoheaters in Polymer and Biological Media: Characterization of Mechanical Destruction and Boiling," Adv. Funct. Mater. 22(2), 294–303 (2012).
- G. Baffou, J. Polleux, H. Rigneault, and S. Monneret, "Super-Heating and Micro-Bubble Generation around Plasmonic Nanoparticles under cw Illumination," J. Phys. Chem. C 118(9), 4890–4898 (2014).
- E. Lukianova-Hleb, Y. Hu, L. Latterini, L. Tarpani, S. Lee, R. A. Drezek, J. H. Hafner, and D. O. Lapotko, "Plasmonic nanobubbles as transient vapor nanobubbles generated around plasmonic nanoparticles," ACS Nano 4(4), 2109–2123 (2010).
- M. M. P. Arnob, F. Zhao, J. Li, and W.-C. Shih, "EBL-Based Fabrication and Different Modeling Approaches for Nanoporous Gold Nanodisks," ACS Photonics 4(8), 1870–1878 (2017).
- 34. G. M. Santos, F. Zhao, J. Zeng, and W. C. Shih, "Characterization of nanoporous gold disks for photothermal light harvesting and light-gated molecular release," Nanoscale **6**(11), 5718–5724 (2014).
- S. Qiu, F. Zhao, O. Zenasni, J. Li, and W.-C. Shih, "Catalytic assembly of DNA nanostructures on a nanoporous gold array as 3D architectures for label-free telomerase activity sensing," Nanoscale Horiz 2(4), 217–224 (2017).
- W.-C. Shih, G. M. Santos, F. Zhao, O. Zenasni, and M. M. P. Arnob, "Simultaneous Chemical and Refractive Index Sensing in the 1-2.5 μm Near-Infrared Wavelength Range on Nanoporous Gold Disks," Nano Lett. 16(7), 4641–4647 (2016).
- G. M. Santos, F. I. S. Ferrara, F. Zhao, D. F. Rodrigues, and W.-C. Shih, "Photothermal inactivation of heatresistant bacteria on nanoporous gold disk arrays," Opt. Mater. Express 6(4), 1217–1229 (2016).
- J. Li, F. Zhao, and W. C. Shih, "Direct-write patterning of nanoporous gold microstructures by in situ laserassisted dealloying," Opt. Express 24(20), 23610–23617 (2016).
- 39. R. S. Taylor and C. Hnatovsky, "Growth and decay dynamics of a stable microbubble produced at the end of a near-field scanning optical microscopy fiber probe," J. Appl. Phys. **95**(12), 8444–8449 (2004).
- X. Liu, L. Bao, M. Dipalo, F. De Angelis, and X. Zhang, "Formation and dissolution of microbubbles on highlyordered plasmonic nanopillar arrays," Sci. Rep. 5(1), 18515 (2016).
- O. Shpak, L. Stricker, M. Versluis, and D. Lohse, "The role of gas in ultrasonically driven vapor bubble growth," Phys. Med. Biol. 58(8), 2523–2535 (2013).

1. Introduction

In microfluidic devices, microbubbles have been widely utilized as micro-pumps [1], micromixers [2], micro-robots [3] and surface cleaners [4,5]. Among various microbubble generation techniques, hydrodynamic method is one of the most commonly used, relying on shear force between liquid and gas flows in T-junction [6], flow-focusing [7] or co-focusing junction [8] microchannels. High-throughput production of bubbles with well-controlled size and frequency has been achieved [9,10]: generation speed of 10^6 bubbles per second, diameter ranging from 10 to 500 µm, and a standard deviation of bubble volume less than 2%. However, the location of bubble generation is limited to the position of junctions, and the generated bubbles cannot be fixed but flushed away with the continuous flow, making it not suitable for flow manipulation applications.

High heat-flux pulse heating is another technique for bubble generation, where the bubble grows on the surface of resistive heaters due to the localized nucleate boiling effect [11,12]. It has been employed in thermal-bubble ink-jet printers. Although this technique features low cost and easy integration with microfluidic systems, the bubble generation is limited to the position of resistive heaters, which requires electrical connections. Additionally, the generated bubbles provide short lifetime and collapse immediately after current pulses.

Another technique for bubble generation utilizes lasers to enable localized photothermal heating. In contrast to other techniques, there is great flexibility in the location of bubble generation by repositioning the laser spot. Additionally, without the need for resistive heaters and interconnects embedded in the microchannel, lithography is not required. Low-cost device fabrication techniques such as soft lithography can be employed. However, light-based generation techniques have been mostly limited to high power laser pulses and single bubble generation. Cavitation bubbles have been generated by a highly focused laser spot inside a microscale gap, causing the liquid to vaporize rapidly [13]. Such bubbles have been demonstrated for fluid actuation [1,2,14], cell surgery [15], cell lysis [16], and cell membrane poration [17]. Dyes are usually utilized for cavitation bubbles collapse quickly due to the flow. The need of dyes can also limit applications where they are either not allowed or difficult to incorporate into the flow with high uniformity.

Metal films integrated in microchannels can act as photothermal heat sources when illuminated. Because of the high thermal conductivity of metal, the converted heat is effectively transferred to the surrounding liquid and causes it to reach a temperature above the boiling point. The heated liquid vaporizes rapidly, which leads to the explosive expansion of hot steam and generation of vapor bubbles. Those bubbles can reach hundreds of micrometers in diameter in tens of seconds, and remain in a steady state as long as the irradiation is kept on. Laser-induced photothermal bubbles generated on the surface of metal films have been utilized in microfluidic devices for altering the propagation of surface plasmon polariton (SPPs) [18], fluid pumping and valving [19,20], concentrating and manipulating particles/cells [21–23], and direct-writing micro-patterns [24]. While it requires significant less laser power compared to other laser-based techniques, hundreds of milliwatts are still needed to generate photothermal microbubbles on metal films.

As an alternative, noble metal nanoparticles of Au or Ag hold great promise for the role as photothermal heaters. They are excellent light absorbers when illuminated at the plasmonic resonance wavelength, which is tunable by nanostructure design. The photothermal bubbles generated around colloidal plasmonic nanoparticles under laser irradiation have been explored for various biomedical applications, such as ablation of biological tissues [25], cell imaging [26], and cell theranostics [27]. For example, Halas's group explored the generation of nanobubbles around gold nanoparticles (AuNP) dispersed in a liquid under solar [28] and resonant excitation [29]. However, most experimental and theoretical studies are carried out under pulsed laser illumination, in which the bubbles collapse immediately after generation, having a life cycle of nanoseconds. The irradiated nanoparticles also risk shape modification

or even thermal destruction from the laser pulses, limiting their reusability and reliability. Additionally, it is challenging to form a robust, uniform layer of solution-synthesized colloidal nanoparticles in a microfluidic channel, thus limiting its practical use for flow manipulation. Huhn *et al.* [30] observed bubbles generated around agglomerated AuNP clusters embedded in polyelectrolyte films under continuous illumination, demonstrating laser-mediated superheating. Baffou *et al.* [31] studied superheating and bubble generation dynamics around continuous-wave (CW) laser-activated uniform arrays of AuNPs. Representative techniques for laser-based microbubble generation are summarized in Table 1.

irradiation	light absorber	power/energy	bubble size	bubble lifetime	ref.
ns pulsed laser	light absorbing dye	44-56 μJ	up to 50 µm	cycle <10 µs	[13]
CW	metal film	35-250 mW	up to 570 µm	tens of seconds in growth	[18,19,21]
CW	AuNP array	25-1000 mW	a few micrometers	-	[29]
CW	AuNP array	15-300 mW	a few micrometers	seconds to hours	[31]
ns pulsed laser	AuNP suspension	0.6-1.5 μJ	up to 500 nm	50-500 ns	[32]
CW	NPGD array	5-50 mW	a few micrometers	seconds to minutes	this work

Table 1. Summary of buddle generation technicit

In this paper, we report microbubble generation in microfluidic devices using photothermal effect aronound nanoporous gold disk (NPGD) array under CW laser illumination. NPGD arrays recently developed by our group provide tunable plasmonics with high-density electric field localization [33]. We have demonstrated their applications for photothermal conversion and light-gated molecular delivery [34], surface-enhanced Raman scattering [35], fluorescence, near-infrared absorption [36], integration with microfluidic devices, and as a surface coating for photothermal inactivation of pathogens [37]. NPGD arrays have been fabricated directly on a glass substrate via a monolithic process, thus providing uniform coverage that are essential for bubble generation at arbitrary locations. Owing to the porous nature, NPGDs exhibit higher photothermal conversion efficiency compared to AuNP of similar size [34]. In addition, the fabrication of NPGD substrates do not require additional immobilization of the nanoparticles. Using a computer-generated holography realized on a phase-only spatial light modulator (SLM), simultaneous and independent generation of multiple microbubbles can be achieved at arbitrary locations. In the rest of the paper, we first provide the microfluidic design and fabrication steps. We then show the dynamics of bubble formation and growth under different power density and the bubble shrinkage when the laser is turned off. Finally, we show parallel microbubble generation and independent control of individual bubbles inside the microfluidic channel and the use of the microbubbles as multifunctional light-gated microvalves for dynamic flow manipulation.

2. Materials and methods

Figure 1(a) shows the experimental platform based on a projection microscopy system described in our previous work [38] and briefly outlined here. The laser source was a 532 nm CW laser (Spectra-Physics Millennia X) with its output incident on an SLM (Boulder Nonlinear Systems, Inc.) for phase modulation. The modulated beam forms a laser illumination pattern, which was directed towards the sample via the back port and the objective of an inverted microscope (Olympus IX70). The focused laser beam has a beam width of ~2.5 μ m on the sample plane. A tungsten-halogen lamp on top of the microscope was employed for bright-field observation. The translucency of the NPGD substrate enables simultaneous imaging and heating from either top or bottom side of the channel. The

Research Article

Optics EXPRESS

transmitted light was then relayed via the microscope side port to a charge coupled device (CCD) camera (PIXIS 400, Princeton Instruments) for image recording at 0.3 s intervals.

A schematic of the microfluidic device is shown in Fig. 1(b). The microfluidic channel was formed by a double-sided adhesive sheet sandwiched between the plasmonic substrate and a piece of cover glass. The channel structure was formed on the adhesive sheet via laser cutting (VLS 3.50, Versa Laser System, Universal Laser System Ltd.) and the resulting channel has a width of 200 µm and a depth of 170 µm. Inlets and outlets were drilled on the cover glass for fluidic introduction. The plasmonic substrate consists of monolithic uniform arrays of NPGD fabricated by nanosphere masking technique, which was described in a previous publication [36]. Thus fabricated nanoparticle arrays are highly reproducible and their optical properties can be fine-tuned in the visible to near-infrared range by changing the design dimensions in order to improve light-particle interaction. The NPGDs, as shown in Fig. 1(c), are on average 350 nm in diameter, 75 nm in thickness, and 14 nm in pore size. The device assembly allows for high flexibility in its property customization since the design of the channel shape and the fabrication of plasmonic substrate are completely independent of each other. The assembled channel was filled with DI water at a constant flow rate of 3 μ L/min driven by a syringe pump. When the laser beam was focused on the NPGD, water adjacent to the nanoparticles can be sufficiently heated to initiate explosive vaporization that formed a microbubble at the glass-water interface, as shown in Fig. 1(d).



Fig. 1. (a) Schematic of the experimental platform. (b) Schematic illustration of bubble generation around NPGD nanoparticles embedded in a microfluidic channel. (c) A scanning electron microscope (SEM) image of a substrate patterned with a monolithic uniform array of nanoporous gold particles. (d) An optical image showing a single microbubble.

3. Results and discussion

Microbubble growth dynamics

With sufficiently high laser power, a microbubble could be initiated immediately upon laser irradiation. The power threshold for microbubble initiation within 0.3 s was 5 mW for DI water at a constant flow rate of 3 μ L/min. The photothermally generated microbubble undergoes rapid expansion following its initiation and continues to grow at a lower rate as the laser was held at a constant power. The expansion rate decreased as the bubble volume gradually increased towards a steady state, where an equilibrium was reached between the expansion of the vapor and gas-filled bubble cavity and the condensation of vapor and permeation of gasses at the liquid interface.



Fig. 2. (a) Consecutive images of bubble under 16 mW laser power. (b) Bubble growth curve under different illumination laser powers: 16 mW, 12 mW, 8 mW in normal DI water and 8 mW in degassed water. (c) Bubble shrinkage curve under different illumination laser powers: 16 mW, 12 mW, 8 mW in normal DI water and 8 mW in degassed water. Inset: Enlarged view of the shrinkage curve indicated by the dotted area in (c).

Consecutive images at intervals of 3 s with 16 mW laser power at the sample plane were acquired, as shown in Fig. 2(a). In the presence of the substrate, thus generated microbubbles were not spherical or suspended in the liquid, but were spherical-cap-shaped, bound to the plasmonic substrate around the heated spot, and not affected by the motion of the flow at this flow rate. This allows precise control of the bubble size and position by the laser power and position.

To quantify the dynamics of bubble growth, bubble diameter was measured during the expansion at laser powers of 8, 12, and 16 mW. Figure 2(b) shows the increase of bubble diameter over time. As shown in Fig. 2(b) black curve, the initial rate of growth is as high as 94.9 μ m/s, which decreased significantly after 1 sec, and decreased furthermore as the bubble size increased. Generally, a higher power can result in a higher initial growth rate and contribute to a larger final bubble size. Within the first 3 s of bubble formation, the average bubble growth rate was 7.6, 10.7 and 13.3 μ m/s, and the bubble sizes at 29 sec were 35, 49 and 66 μ m in diameter for 8, 12 and 16 mW, respectively. The fitted curves for the bubble diameter as a function of time are $19.83|t|^{0.179}$, $27.99|t|^{0.173}$, and $35.37|t|^{0.182}$ for 8, 12, and 16 mW, respectively. The results are reproducible on different areas across the substrate with up to 3% size variation. With higher laser power and sufficiently long time, bubbles up to 200 μ m in diameter were generated to completely block the width of the channel.

In order to compare the contribution from the vaporization of water and degassing of air molecules during bubble formation, microbubbles were generated in DI water degassed via boiling and cooled to room temperature. The threshold for immediate bubble initiation upon irradiation remains at 5 mW, which indicates that the explosive expansion of superheated liquid around the nanoparticles was the dominant mechanism for the formation of a microbubble [31,39,40]. After the bubble initiation, however, the driving force of bubble expansion was attributed to both vaporization and degassing. Unlike cavitation bubbles generated by high energy pulsed laser with lifetime of only microseconds, one cannot assume that the bubble was made of pure water vapor and consider the lifetime too short for air molecules to diffuse from the liquid to fill the bubble cavity. Under the same laser power (8

mW), the bubble diameter during its growth in degassed water was traced and plotted against time, as shown in Fig. 2(b). In comparison, the fitted curve for 8 mW in degassed water is $15.5|t|^{0.152}$, where bubble generation in degassed water had a lower expansion rate, and the bubble size was smaller compared to normal DI water.

The minimum power required for a single microbubble generation on NPGD substrate was 5 mW, equivalent to 1×10^5 W/cm² power density. In comparison, with AuNP arrays under CW laser irradiation at the plasmonic resonance wavelength, the power threshold for microbubble generation with focused laser beam exceeds 20 mW [29,31]. The performance of microbubble generation with NPGD as the heat absorber showed excellent heat transfer capability and relatively lower power consumption.

Microbubble shrinkage dynamics

After bubble generation, once the laser is turned off, the microbubble starts to shrink in size due to the removal of heat source. Depending on the initial size of bubble shrinkage, the bubble volume decreased gradually over a few seconds to a few minutes, until it finally disappeared. The shrinkage dynamics were traced for microbubble generated with 16 mW, 12 mW, and 8 mW in normal DI water and 8 mW in degassed DI water. The variation of bubble diameter over time is plotted in Fig. 2(c), where the time origin (i.e. t = 0) was arbitrarily chosen at the moment when the bubble disappeared. Instead of a sudden decrease in size as with cavitation bubbles, the shrinkage curve resembles the inverse of the growth curve, but spans over a longer time scale. The fitted curves for bubble diameter as a function of time are $8.22|t|^{0.34}$, $8.49|t|^{0.34}$, $8.55|t|^{0.33}$ and $8.92|t|^{0.34}$ for 16 mW, 12 mW, 8 mW in normal DI water and 8 mW in degassed DI water, respectively. As shown in Fig. 2(c) inset with the plot zoomed in to the last 15 s of the shrinking process, regardless of the initial bubble size, the curves are almost overlapping. During the shrinking process, the bubble diameter appeared to be linearly dependent on $\sim t^{1/3}$. By investigating microbubbles with various sizes, we found that the dynamics was highly reproducible, despite the starting size of the shrinkage process, which is consistent with previous findings involving microbubbles generated in a similar fashion [31]. The vapor and gas could be considered as ideal gases under saturated conditions [39,41]. Under a constant dissolution rate, the bubble lifetime τ_0 and initial radius r_0 satisfies the relationship $r_0 \propto \tau_0^{1/3}$ [31]. The required laser power and maximum bubble size for a specific response time of bubble generation/collapse cycle could be estimated by the above calculations for fine control during applications.

Generation and control of microbubble arrays

Since the microbubble generation site is dependent on the position of laser beam instead of the patterning of nanoparticles inside the channel, we demonstrate the function of programmable bubble generation by the creation of arrays of microbubbles in designed patterns. The location of microbubbles could be arbitrarily chosen within the field of view, which provides flexibility in the manipulation of flow or particles in the flow. As shown in Fig. 3(a-b), arrays of microbubbles in patterns consisting of 4 and 8 bubbles were generated simultaneously by their pre-defined laser illumination patterns. The slight size difference among individual bubbles is due to the possible variation of NPGD coverage throughout the whole field of view. The convective flow induced by the temperature gradient from the bubble surface towards the surrounding liquid, as well as the motion of flow actuated by the growth and shrinkage dynamics of the microbubbles, have potential applications of flow mixing.

Besides the parallel generation of microbubbles, the programmable patterning function is also capable of controlling individual microbubbles within a pre-existing microbubble array. By dynamically refreshing the illumination pattern that includes or excludes specific laser

spots within a pattern, individual bubbles can be removed in sequence while others remain. Figure 3(c-d) shows the parallel generation of four microbubbles, and Fig. 3(e-h) shows their removal one after another at time 17 s, 24 s, 30 s and 36 s, respectively.



Fig. 3. Parallel microbubble generation with illumination patterns. (a) Four microbubbles with 60 μ m center-to-center distance. (b) Eight microbubbles, with 30 μ m center-to-center distance for adjacent microbubbles. (a-b) share the scale bar. (c-h) The generation and removal of four microbubbles. (c-d) depict the generation of bubbles at t = 1s and 9s. (e-h) depict the removal of bubbles one by one at t = 17 s, 24 s, 30 s, and 36 s. (c-h) share the scale bar.

Photothermal microbubbles as microvalves

The photothermal microbubbles generated around NPGDs under CW laser illumination provide long lifetime, are producible at any location of interest, and can be held at a fixed location. It allows the generated microbubbles to be used as an active flow-manipulation element in microfluidic devices. Herein, we demonstrate the functionality of microbubbles serving as microvalves in a multi-inlet multi-outlet (MIMO) microchannel for flows. Figure 4 illustrates the X-shaped MIMO junction having two inlets (left) and two outlets (right), all of which are 200 μ m in width and 170 μ m in height. Inlet 1 and Inlet 2 were injected with DI water and red dye (Rhodamine 6G, R6G), respectively. The flow rates at both inlets were set to 3 μ L/min. The flow direction and the blockage were depicted in the figure by arrows and Xs, respectively, with white color as water and red color as dye for clarity. Without laser illumination, we observed the boundary of laminar flow of R6G and water at the junction. The boundary is not precisely centered due to slight pressure difference in the outlets.

First, the control of the flow direction is demonstrated. As shown in Fig. 4(a), a bubble was generated at the top outlet. With the growth of the bubble, the boundary of the laminar flow was pushed towards the bottom outlet. When the upper outlet was completely closed by the microbubble valve, both liquids exited from the bottom outlet, as evidenced by the equal splitting between the two liquids within the outlet. Then, by adding an additional microbubble valve into one of the inlets, we demonstrate the control over both the type and the direction of the liquid flow. As shown in Fig. 4(b), Inlet 2 with the R6G was closed and only water flowed inside the channel. Moreover, the direction of the flow was well controlled by the location of the bubble valve in the outlets. Finally, we demonstrate the possibility of creating a temporarily isolated chamber at the center of the X-junction by blocking all of the inlets and outlets. As shown in Fig. 4(c) i-iii, before the expansion of the bubble completely covers the width of the channels, the flow was partially obstructed but not completely stopped and the fluctuation of the laminar flow boundary was observed due to the unbalanced pressure in the channel during the bubble growth. When the bubbles expanded enough to stop the flow, an isolated chamber was formed, where the flow boundary blurred and the liquids mixed gradually. The chamber provided a temporary yet flexible reaction site inside the device, allowing for further measurements of the liquid located in the closed space, and convenient

Research Article

Optics EXPRESS

disassembly by increasing the flow rate to more than 20 μ L/min to flush away the generated bubbles while the laser was blocked.

However, the current setup showed disadvantages in applications that require a fast response in terms of flow manipulation, due to the relative long bubble lifetime. The channel size could be reduced to adapt to the bubble size for a shorter lifetime in order to achieve faster flow manipulation response. With low cost photolithography patterning techniques, channel width of a few micrometers could be readily obtained. For instance, when the channel is 1 μ m in width and depth, the cycle of a complete bubble valve generation and collapsing could be as short as 2 ms at laser power of 8 mW, according to previously calculated bubble growth rate.



Fig. 4. Timed sequences of photothermal microbubbles generated inside the X-shaped channels for the individual as well as simultaneous controls of inlets and outlets. The flow direction is depicted by arrows, and blockage is depicted by Xs. (a) Flow mixing by blocking one outlet. (b) Selection between two outlets for one incoming flow. (c) Temporary mixing chamber by blocking all inlets and outlets. The images share the scale bar.

4. Conclusions

In this work, we have reported a novel technique to generate microbubbles upon irradiation of NPGD nanoparticles with CW laser illumination. This technique is capable of instantaneously producing long lifetime microbubbles at arbitrary locations, without the requirement of lightabsorbing dyes or electrical interconnects. The property of the plasmonic substrate and the design of channel shape are independent processes which allow for convenient customization and easy design and assembly. The NPGD as the heat absorber showed excellent heat transfer capability and relatively lower power consumption. The focused laser beam maintains tight heat localization while requires power as little as 5 mW. The experimental results indicate that the size of the generated bubble varies with respect to the duration, power density of laser illumination, and air content in the liquids. Moreover, the lifetime of generated bubble could range from a few seconds to a few minutes, depending on its initial size, and maintains a constant relationship $r_0 \propto \tau_0^{1/3}$. Coupled with computer-generated holography on an SLM, multi-point microbubble generation and independent control of individual microbubbles has been demonstrated. We have also demonstrated the application of the microbubble as multifunctional light-gated microvalves in a microfluidic X-shaped junction with parallel control of multiple inlets and outlets, providing potential applications in flow manipulation and creating temporary reaction chambers inside the device.

Vol. 26, No. 13 | 25 Jun 2018 | OPTICS EXPRESS 16902

Optics EXPRESS

Funding

National Science Foundation (NSF) (1151154, 1605683, and 1643391).