Two-photon polymerization of cylinder microstructures by femtosecond Bessel beams

Liang Yang, Ayman El-Tamer, Ulf Hinze, Jiawen Li' , Yanlei Hu, Wenhao Huang, Jiaru Chu, and Boris N Chichkov

Citation: Appl. Phys. Lett. **105**, 041110 (2014); doi: 10.1063/1.4891841
View online: http://dx.doi.org/10.1063/1.4891841
View Table of Contents: http://aip.scitation.org/toc/apl/105/4
Published by the American Institute of Physics

APPLIED PHYSICS LETTERS 105, 041110 (2014)

Two-photon polymerization of cylinder microstructures by femtosecond Bessel beams

Liang Yang,[1,2] Ayman El-Tamer,[2] Ulf Hinze,[2] Jiawen Li,[1,a)] Yanlei Hu,[1] Wenhao Huang,[1] Jiaru Chu,[1] and Boris N Chichkov[2]
[1]Department of Precision Machinery and Precision Instrumentation, University of Science and Technology of China, Hefei 230026, People's Republic of China
[2]Laser Zentrum Hannover e.V., 30419 Hannover, Germany

(Received 1 July 2014; accepted 20 July 2014; published online 29 July 2014)

In this work, we present an approach to modulate femtosecond laser beams into Bessel beams with a spatial light modulator (SLM) for two-photon polymerization applications. Bessel beams with different parameters are generated and annular optical fields are produced at the focal plane of the objective. Uniform cylinder microstructures are fabricated by a single illumination during a few seconds without stage translation. By modulating the holograms encoded on the SLM, the diameters of the fabricated annular structures can be flexibly controlled in a wide range with no need of changing the optical elements and realignment of the optical path.
[http://dx.doi.org/10.1063/1.4891841]

Two-photon polymerization (2PP) technique, which has a scalable resolution from micrometers down to sub-100 nm,[1] is widely used for the fabrication of complex functional three dimensional micro- and nanodevices.[2,3] The common and conventional approach is to move the focus of an fs-laser pulse within a photoresist, producing a polymerized pattern. In this way, micro- and nanostructures are built by serially writing their contours and volume. However, this serial approach has the serious drawback that the processing time scales with the third power of the object size. For complex devices and objects with high polymerization volume, this can result in technically unrealistic processing times. Even the geometrical nature of an object may necessitate another processing approach. Namely, circular structures (e.g., nano- and microtubes) require permanent acceleration and deceleration of translation stages during processing. To reduce the processing time, the writing process was parallelized with optical elements that create multiple laser spots, namely, microlens arrays[4] and diffractive optical elements.[5] However, the position of the foci are fixed by the optical design of these optical elements. Winfield achieved parallel processing by shaping femtosecond laser beams into Bessel beams with an axicon.[6,7] Thus, annular structures were fabricated without stage translation. Similarly, Stankevicius *et al.* presented a method to shape a femtosecond laser beam into an optical vortex for 2PP with a holographic grating written in a chromium layer.[8] However, axicons or holographic gratings have to be changed and the optical path has to be realigned if cylinder structures with variable size are needed.

Spatial light modulators (SLMs) are versatile devices for beam profile reshaping by phase modulation. They were used in laser pulse shaping,[9] optical trapping,[10] and parallel ablation of metal surface.[11] Recently, SLMs were adopted to generate adjustable multi foci by loading computer generated holograms dynamically.[12,13] The position and quantity of multi foci could be well controlled for 2PP fabrication. But cylinder microstructures were still fabricated by the serial scanning of individual laser spots. SLMs were also used to shape a femtosecond laser beam into an optical vortex for microgear fabrication,[14] two dimensional patterns for efficient lithography of microstructures,[15] and non-Airy patterns for 2PP.[16]

In this Letter, we present our results on modulating a femtosecond laser beam into a Bessel beam, using a SLM, for fabrication of cylindrical structures. In contrast to laser beam shaping with axicon,[6,7] parameters of the Bessel beams can be flexibly controlled by loading different holograms to the SLM, without any modification of optical elements and optical path. Using this technique, uniform cylindrical structures with controllable diameters are fabricated in a single illumination step.

Bessel beams have an intensity profile that approximates Bessel functions with nondiffracting behavior. They can be generated by holograms.[17,18] The transmission function of the holograms can be written as

$$T_n(r,\theta) = \exp(in\theta)\exp(-i2\pi r/r_0), \tag{1}$$

where r and θ are coordinates in the hologram plane and r_0 is an adjustable constant parameter. The phase function has two terms in Eq. (1). The first term is the phase singularity of charge n associated with the azimuthal phase. Here, n denotes the nth order Bessel beam. The second term generates a zeroth order Bessel beam. To encode the holograms of Eq. (1) onto a SLM, we rewrite $r^2 = (i^2 + j^2)\Delta^2$, where i and j are integers indicating each pixel and Δ is the pixel pitch. The electric field formed at a distance z from the hologram plane is obtained as

$$E(\rho,\varphi,z) = \frac{\exp(ikz)}{ikz}\exp\left(\frac{ik\rho^2}{2z}\right)\exp\left[in\left(\varphi-\frac{\pi}{2}\right)\right] \times \int_0^R \exp\left(\frac{ikr^2}{2z}\right)\exp\left(\frac{-i2\pi r}{r_0}\right)J_n\left(\frac{kr\rho}{z}\right)rdr, \tag{2}$$

a)Author to whom correspondence should be addressed. Electronic mail: jwl@ustc.edu.cn

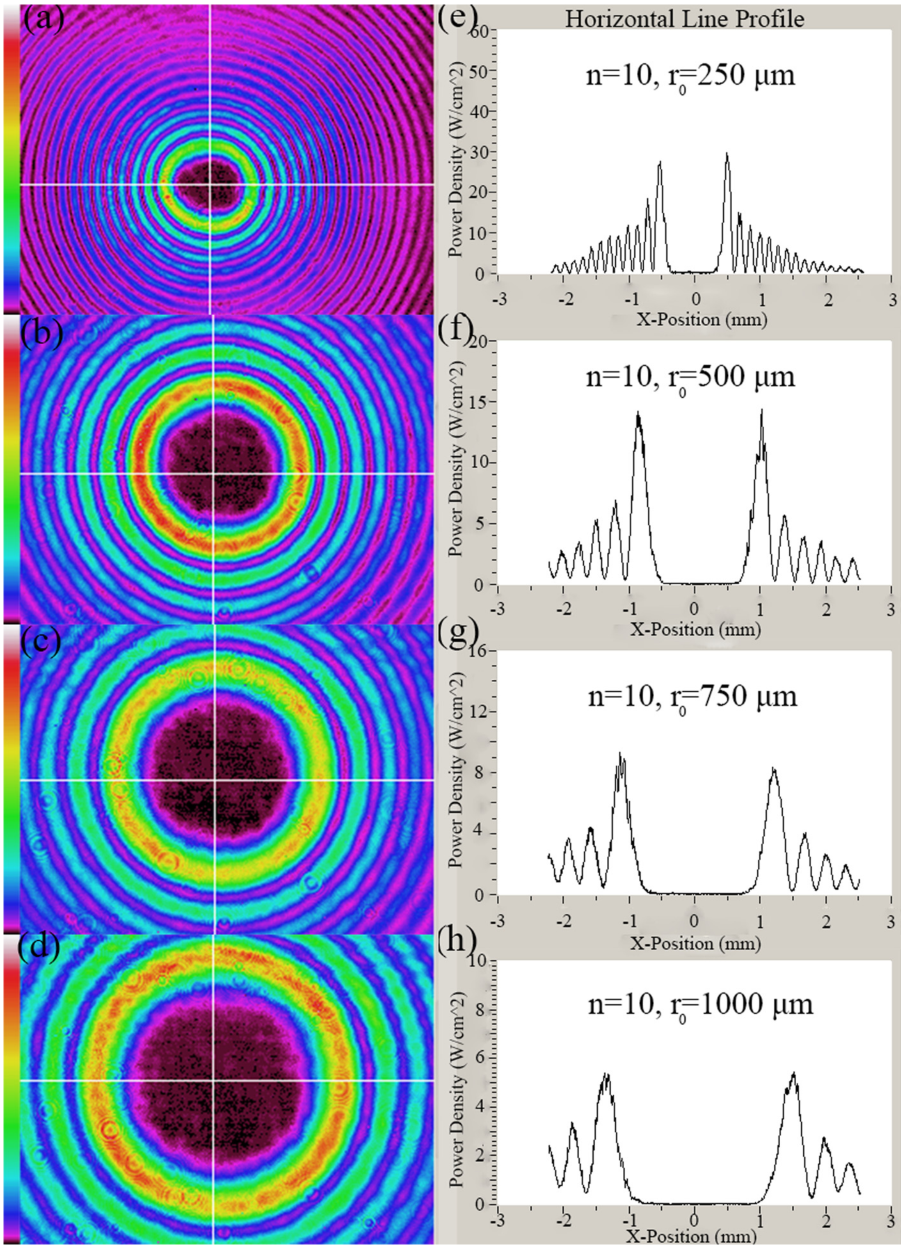

FIG. 1. Bessel beams with different parameters generated by SLM. (a)–(d) The transversal intensity distributions measured with a beam profiler (Ophir Photonics) at a distance of 1500 mm from the reflecting surface of the SLM. (e)–(h) The intensity profiles of (a)–(d) in the x direction, respectively.

where ρ and φ are polar coordinates in the observation plane and J_n is the nth order Bessel function of the first kind. With a spatial light modulator, Bessel beams with arbitrary order can be generated. Bessel beams of zero to 20th order provide suitable intensity profiles for the 2PP production of cylindrical microstructures, and any of them could be used. Fig. 1 shows the beam profile of the 10th order Bessel beam generated with different r_0, proving the ability of the SLM to modulate the laser beam even with this complex high order mathematical function. The intensity profiles of the generated Bessel beams keep unchanged over the distance $L = Rr_0/\lambda$,[17] where R is the diameter of phase function and λ is the wavelength of laser source.

The experimental setup is schematically illustrated in Fig. 2(a). A high-repetition rate femtosecond laser source with a wavelength of 780 nm (Chameleon Ultra, Coherent GmbH; repetition rate: 80 MHz, pulse width <140 fs) was used for the 2PP process. After passing through a pulse energy controlling unit and a beam expander, the laser beam

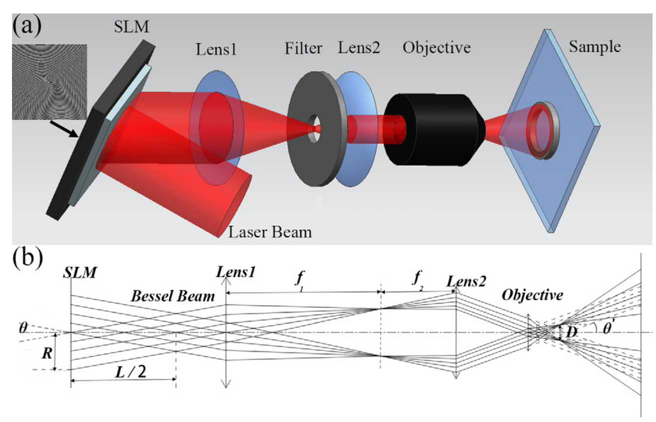

FIG. 2. (a) Diagram of the experimental setup. (b) Parameters of the optical path. The incident Gaussian beam is transformed into a Bessel beam propagating over a distance of L. The Bessel beam is scaled down with Lens1 and Lens2 and then focused into the resist with an objective for 2PP.

illuminates a reflection type liquid crystal SLM (X10468, Hamamatsu, 800 × 600 pixels, 256 grey levels, pixel pitch of 20 μm), on which calculated phase modulation pattern are encoded. To achieve better modulation, the expanded laser beam is slightly larger than the SLM display and only the central 600 × 600 pixels are used for Bessel beam generation. The Bessel beam is scaled down with a 4f optical system made of Lens1 and Lens2 and then focused into the sample with an objective (63×, NA = 1.4, focal length 2.6 mm, Zeiss) for 2PP processing. A filter is placed at the Fourier plane of Lens1 to block the zeroth order beam, which is shifted from the Bessel beam. A cylinder pattern is generated in the focal region of the objective. A full cylinder is polymerized by a single exposure of a spin coated sample with photoresist (Ormosil). Ormosil is suitable for the fabricating of functional micro and nano devices because of its good mechanical stability, high optical quality, and good post-processing inertness. More importantly, it shows low shrinkage which makes it suitable for the fabrication of structures that are sensitive to deformations.[19] The process of fabrication is monitored *in situ* with a charge coupled device (CCD) camera.

Geometrical evolution of the decreasing and focusing of the Bessel beam is shown in Fig. 2(b). The Bessel beam generated behind the SLM can be expanded in two plane waves whose wave vectors have a semi angle of θ.[20] The nondiffracting distance of the Bessel beam is $L = Rr_0/\lambda$. After a 4f optical system constituted of Lens1 and Lens2, the beam is transformed to a shorter Bessel beam, which can be expanded in two plane waves with a semi angle of θ'. Focused with an objective, a sharp annular pattern with diameter D is generated at the focal region. The parameters are calculated as

$$\theta = \arctan(R/L) \approx R/L = \lambda/r_0; \quad \sin\theta' = \sin\theta \times f_1/f_2; \\ D = 2f_{objective}\tan\theta', \tag{4}$$

where f_1, f_2, and $f_{objective}$ are the focal length of Lens1, Lens2, and objective, respectively. The semi angle θ' is calculated to be $\theta' = \lambda f_1/(r_0 f_2)$. The diameter of the annular pattern formed at the focal region is calculated to be $D \approx 2f_{objective} f_1 \lambda/(f_2 r_0)$. We can see that $D \propto 1/r_0$ and does not depend on the order of Bessel beam.

Scanning electron microscope (SEM) images of the fabricated cylinder structure are shown in Fig. 3. The Bessel beam used here has a parameter of $n = 0$ and $r_0 = 300$ μm. The structure was fabricated by a single illumination with an exposure time of 5 s at a laser power of 210 mW (measured behind the objective). The diameter is 45 μm, which is in good agreement with the design calculation of 45.1 μm. The measured thickness of cylinder is ~1.25 μm. If the exposure time is increased from 5 s to 10 s, the thickness slightly increases from ~1.25 μm to ~1.56 μm. The cylinder has a very smooth surface; the height of the cylinder corresponds to the thickness of the photoresist. With 10 s acceleration to 3000 rpm and keeping this speed for 50 s, the thickness of spin-coated photoresist is 11.2 μm.

The thickness of the cylinder wall is given by the intrinsic divergence of the focused Bessel beam and threshold behavior of the 2PP process. In this study, the intensity uniformity of the Bessel beams, which is a significant parameter affecting the thickness of the cylinder wall, was greatly improved by fine tuning of optical path and the peak of Gaussian beam to the center of SLM display. However, due to the pixilation of the SLM, imperfection of Gaussian beam and small residual technical deviations, the intensity is not perfectly uniform, which, results in a slight variation of the cylinder wall thickness. The uniformity of wall thickness is 87% and it could be improved by increasing the exposure power progressively. In this study, the exposure time is limited by the power of the laser source of our experimental setup. If the usable power is increased, the exposure time can be further decreased.

The most remarkable advantage of this approach is that the parameters of the Bessel beam can be adjusted without any change of the optical path. By controlling r_0 in Eq. (1), Bessel beams with various intensity distributions were generated and thus cylinder microstructures with various diameters were obtained as shown in Fig. 4. Fig. 4(a) shows the holograms used to generate the Bessel beams. Since, the order of the Bessel beams has no effect on the diameter of the fabricated cylinder structures, the order of Bessel beam is fixed as $n = 10$, while r_0 varies from 250 μm to 1000 μm. Figs. 4(b) and 4(c) show fabricated cylinder microstructures captured from top view and at 45°. The measured diameters of cylinder microstructures are 55, 46, 35, 28, 24, 21, 18, 16.7, and 15.5 μm, which are in perfect agreement with the design calculation. With the decreasing diameter, both the laser power and the exposure time were decreased. Theoretically, the diameter may vary in a very wide range

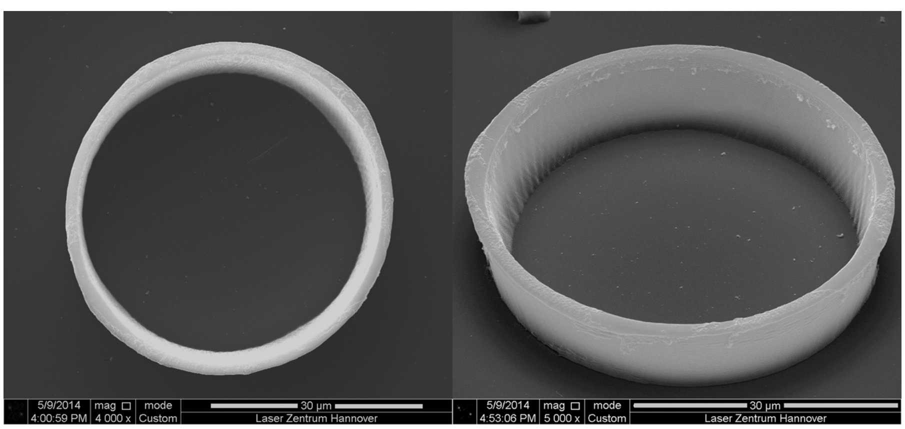

FIG. 3. Cylinder microstructure fabricated with a 0th order Bessel beam in a single illumination step. (a) Top view of the cylinder. (b) Cylinder captured at 45°. The Scale bars correspond to 30 μm.

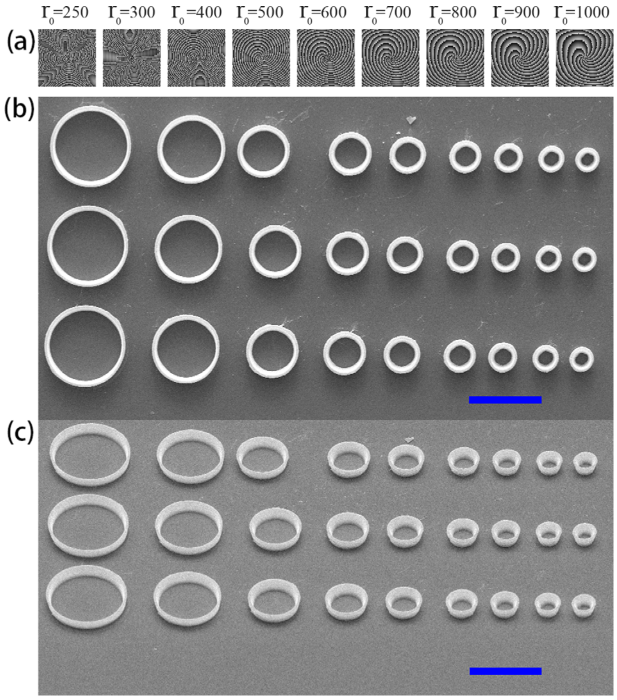

FIG. 4. Cylinder structures with various diameters fabricated with Bessel beams with different parameters. (a) Holograms used to generate Bessel beams. $n = 10$, r_0 varies from 250 μm to 1000 μm. (b) Top view of the fabricated cylinders. (c) 45° captured images of the fabricated cylinders. The height of the cylinders is 11.2 μm. The scale bars correspond to 50 μm.

from 2.25 μm to 676 μm continuously, as the parameter r_0 varies from 20 μm (pitch of pixels on SLM) to 6000 μm (dimension of the shorter size of the SLM display) if the limitation of apertures of the optical elements meets the requirements. Such cylinder microstructures are of interest for the fabrication of bio-scaffolds,[2,13] drug delivery,[21] and other microfluidic applications.[22]

The walls of the cylinder structures in Figs. 4(b) and 4(c) have a small slant angle, which agrees with the geometrical analysis in Fig. 2(b). While r_0 varies from 250 μm to 1000 μm, the angle increases from 6.3° to 13.6°. The angle is found to increase with the decreasing r_0 and is much larger than θ'; in fact, it is related to intrinsic parameters of the Bessel beams and the objective. By optimal modulation of optical parameters (e.g., parameter r_0, focal length of Lens1 and Lens2, and magnification of objective), the slant angle can be controlled.

In a conventional 2PP process, cylinder microstructures are processed by serial scanning. In the demonstrated approach, a full cylinder is fabricated by a single illumination in seconds. Compared with Bessel beam fabrication based on an axicon lens, the parameters of the Bessel beams can be flexibly adjusted by the holograms encoded on the SLM without any modification of the optical path.

In conclusion, it has been demonstrated that femtosecond laser beams can be modulated into Bessel beams with a SLM for fabrication of cylindrical microstructures by 2PP. Cylinder microstructures were fabricated by a single illumination in seconds. The diameters of the fabricated cylinders could be flexibly controlled in a wide range by changing the holograms on the SLM without any change of the optical path. Note that this technique is not restricted to a single Bessel beam. Bessel beam arrays and complex beam patterns can be used for efficient 2PP fabrication.

This work was supported by National Science Foundation of China (Nos. 51275502, 91223203 and 51205375), Anhui Provincial Natural Science Foundation (No.1408085ME104), National Basic Research Program of China (No. 2011CB302100), China Postdoctoral Science Foundation Funded Project (Nos. 2012M511416 and 2012M521245), the Fundamental Research Funds for the Central Universities (KB2090090001), BMBF Remedis B2 (FKZ 03IS2081I), and Biofabrication for NIFE.

[1]V. F. Paz, M. Emons, K. Obata, A. Ovsianikov, S. Peterhansel, K. Frenner, C. Reinhardt, B. Chichkov, U. Morgner, and W. Osten, J. Laser Appl. **24**, 042004 (2012).
[2]A. Ovsianikov, S. Muhleder, J. Torgersen, Z. Q. Li, X. H. Qin, S. Van Vlierberghe, P. Dubruel, W. Holnthoner, H. Redl, R. Liska, and J. Stampfl, Langmuir **30**, 3787 (2014).
[3]S. H. Park, D. Y. Yang, and K. S. Lee, Laser Photonics Rev. **3**, 1 (2009).
[4]J. Kato, N. Takeyasu, Y. Adachi, H. B. Sun, and S. Kawata, Appl. Phys. Lett. **86**, 044102 (2005).
[5]X. Z. Dong, Z. S. Zhao, and X. M. Duan, Appl. Phys. Lett. **91**, 124103 (2007).
[6]R. J. Winfield, B. Bhuian, S. O'Brien, and G. M. Crean, Appl. Phys. Lett. **90**, 111115 (2007).
[7]B. Bhuian, R. J. Winfield, S. O'Brien, and G. M. Crean, Appl. Surf. Sci. **254**, 841 (2007).
[8]E. Stankevicius, T. Gertus, M. Rutkauskas, M. Gedvilas, G. Raciukaitis, R. Gadonas, V. Smilgevicius, and M. Malinauskas, J. Micromech. Microeng. **22**, 065022 (2012).
[9]P. S. Salter, A. Jesacher, J. B. Spring, B. J. Metcalf, N. Thomas-Peter, R. D. Simmonds, N. K. Langford, I. A. Walmsley, and M. J. Booth, Opt. Lett. **37**, 470 (2012).
[10]J. E. Curtis, B. A. Koss, and D. G. Grier, Opt. Commun. **207**, 169 (2002).
[11]Z. Kuang, W. Perrie, D. Liu, P. Fitzsimons, S. P. Edwardson, E. Fearon, G. Dearden, and K. G. Watkins, Appl. Surf. Sci. **258**, 7601 (2012).
[12]Y. L. Hu, Y. H. Chen, J. Q. Ma, J. W. Li, W. H. Huang, and J. R. Chu, Appl. Phys. Lett. **103**, 141112 (2013).
[13]S. D. Gittard, A. Nguyen, K. Obata, A. Koroleva, R. J. Narayan, and B. N. Chichkov, Biomed. Opt. Express **2**, 3167 (2011).
[14]G. Bautista, M. J. Romero, G. Tapang, and V. R. Daria, Opt. Commun. **282**, 3746 (2009).
[15]N. J. Jenness, R. T. Hill, A. Hucknall, A. Chilkoti, and R. L. Clark, Opt. Express **18**, 11754 (2010).
[16]H. Lin and M. Gu, Appl. Phys. Lett. **102**, 084103 (2013).
[17]N. Chattrapiban, E. A. Rogers, D. Cofield, W. T. Hill, and R. Roy, Opt. Lett. **28**, 2183 (2003).
[18]J. A. Davis, E. Carcole, and D. M. Cottrell, Appl. Opt. **35**, 593 (1996).
[19]A. Ovsianikov, J. Viertl, B. Chichkov, M. Oubaha, B. MacCraith, I. Sakellari, A. Giakoumaki, D. Gray, M. Vamvakaki, M. Farsari, and C. Fotakis, ACS Nano **2**, 2257 (2008).
[20]M. D. Wei, W. L. Shiao, and Y. T. Lin, Opt. Commun. **248**, 7 (2005).
[21]K. Takei, T. Kawashima, T. Kawano, H. Kaneko, K. Sawada, and M. Ishida, Biomed. Microdevices **11**, 539 (2009).
[22]D. J. Thurmer, C. Deneke, Y. F. Mei, and O. G. Schmidt, Appl. Phys. Lett. **89**, 223507 (2006).