Femtosecond Laser Direct Writing of Optical Waveguides in Silicone Film

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For the first time to the authors' knowledge, optical waveguides have been inscribed in silicone ($[SiO(CH_3)_2]_n$) film by ultrafast laser radiation. The 522 nm femtosecond laser pulses with 3 nJ on target energy at 1 MHz repetition rate were focused into thin silicone film (Quantum Silicone, QSIL216) with a 0.55-NA aspherical lens to induce nonlinear photon absorption in the focal volume of the beam. By scanning the laser beam with respect to the silicone film, a permanent refractive index modification has been created inside the silicone. In the modification tracks, photo-dissociation of $SiCH_3$ and dangling bond of SiOH were observed by time-of-flight secondary ion mass spectrometry. The modification tracks showed multi mode at red (635 nm) wavelength and single mode at infrared (1550 nm) wavelength. Propagation losses were estimated to be < 2.2 dB/cm at 635 nm and < 2.8 dB/cm at 1550 nm by the cut back method.

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1. Introduction

Today, planar glass optical devices are routinely used for data distribution in telecommunication systems. These devices are extremely difficult to fabricate and thus very expensive. Polymeric materials, on the other hand, offer the potential to create low-cost highly complex optical interconnection circuitry. Moreover, flexible platform is attractive for the microelectronics industry because of lightweight and lamination layer packages. As a result, polymeric optical waveguides have attracted wide attention with potential applications as transceivers, optical interconnects, and other micro-optical systems in the access network field [1].

In terms of material, many kinds of polymers that have low optical loss in the visible and near-IR region from 0.5 to 1.0 μm have been developed. Amongst them, the optical waveguide fabricated using deuterated polymethyl methacrylate (PMMA) as a core material shows extremely low optical loss of 0.011 dB/cm at 0.68 μm wavelength [2]. Silicone (C_6H_5Si) also shows the propagation loss of 1 dB/cm in this wavelength region [3]. In the wavelength region from 1.0 to 1.7 μm for high-capacity optical communication, conventional polymer materials have high optical loss caused by carbon-hydrogen vibrational absorption. For improvement of the high optical loss, hydrogen in PMMA, polyimide and silicone replaced with deuterium or fluorine have been developed [4 - 6].

For manufacture method of polymeric optical waveguides, there are a number of ways - by selective polymerization, reactive ion etching combined with photolithography, direct photolithography, photo-bleaching and use of injection molding [3]. Amongst them, the reactive ion etching combined with photolithography is used to make single mode polymeric optical waveguides [5]. This fabrication technique provides processing accuracy, but the technique is rather complex and needs multi steps, which increases the cost of optical waveguide device and reduces its reliability.

Recently, Okoshi et al. reported direct writing of silica optical waveguides on the surface of silicone ($[SiO(CH_3)_2]_n$) rubber by F_2 laser induced photochemical reactions [7]. F_2 laser irradiation of silicone in the presence of oxygen can photochemically modify the surface into carbon-free silica, accompanied by swelling of the exposed area [8]. The high 7.9 eV photon energy of the F_2 laser was instrumental here in photochemically modifying silicone to produce carbon-free silica and thereby define a flexible optical circuit. The VUV photochemistry requires formation of reactive $O(^1D)$ from ambient oxygen and photo-dissociation of the silicone film [9, 10].

In this paper, we report an extension of this silicone optical waveguide fabrication technique to nonlinear femtosecond laser direct writing. The objectives of this study are to exploit multi-photon chemistry and internal waveguides writing in focal volume. A permanent modification of the refractive index inside the silicone that leads to the formation of multi-mode waveguides at red (635 nm) wavelength and single-mode waveguides at infrared (1550 nm) wavelength was formed by irradiating

with femtosecond laser light. Silicone films were irradiated at variable fluences, focal depths, and scan speeds followed by inspection under optical microscope, time-of-flight secondary ion mass spectrometry (TOF-SIMS), and waveguide-firing diagnostics to assess the formation of optical waveguides. This paper presents optimal femtosecond laser processing parameters for waveguide writing and assesses the underlying photochemical processes driving the refractive index changes.

2. Experimental setup

The laser used was an amplified Yb-doped fiber laser (IMRA America, Inc., FCPA μJewel D-400-VR) providing 280 fs pulses at 1045 nm wavelength with $M^2 < 1.5$ beam quality. Variable repetition rate with pulse energies of 2.5 μJ at 100 kHz to 400 nJ at 5 MHz allows the study of heat accumulation effects on refractive index modification and waveguide formation in silicone. To enhance nonlinear multi-photon absorption due to high band gap of silicone, the second harmonic output at 522 nm with 180 fs pulses in a Lorentzian profile was used for the exposure which was generated through non-critical phase matching with an LBO crystal.

Silicone film (Quantum Silicone, QSIL216) was uniformly spin-coated (~30 μm thick) on a glass substrate. The film thickness was controlled by the spinning speed, acceleration, deceleration, and time. The refractive index of silicone QSIL216 is 1.406. The laser beam was focused to a spot size of ~1 μm diameter into the silicone film at variable depth z = -10 μm to +4 μm with a 0.55-NA aspherical lens. The silicone sample was mounted on a 3-axis air-bearing translation stage (Aerotech ABL1000 motion stage) and scanning parallel to the laser polarization with speeds of 0.1 to 10 mm/s.

To assess insertion loss, 635 nm (Thorlabs, S1FC635) and 1550 nm (Photonetics, TUNICS-BT) laser diode lights were butt coupled from a silica-core fiber into the end facet of the modification track with index matching oil. A CCD camera (Panasonic, GP-KR222) was used to view the modification tracks to facilitate alignment. To explain mechanism of photochemical processes driving the refractive index changes, TOF-SIMS analysis was conducted.

3. Experimental results and discussion

3.1 Writing modification tracks

At first step, various pulse repetition rates, i.e., 100 kHz, 1 MHz and 5 MHz, were examined to write modification tracks in silicone film with second harmonic wave at 522 nm wavelength. As a result, any modification tracks and ablation were not observed at 100 kHz pulse repetition rate within the pulse energy of 0.5 - 4 nJ at 0.1 - 10 mm/s in scan speed. At 5 MHz pulse repetition rate, modification tracks with the pulse energy of 3 - 4 nJ were observed. However, ablation occurred easily at this repetition rate because of heat accumulation effects [11] and reproducibility of the modification tracks was very poor. The process window was very narrow at 5 MHz pulse repetition rate and we concluded this pulse repetition rate is not suitable for making modification tracks. On the other hand, some modification tracks could be observed at 1 MHz pulse repetition rate with appropriate pulse energy and scan speed. Thus our effort focused to write modification tracks at 1 MHz pulse repetition rate. At 1 MHz pulse repetition rate, we also tried to write the modification tracks by using fundamental wave, 1045 nm laser light, with the same experimental setup. But we could not make any modification tacks with the fundamental wave.

Figure 1 shows optical microscope images of top view of modification tracks inscribed in the silicone film with second harmonic wave, 522 nm laser light. The scan speed was 1.0 mm/s at 1 MHz pulse repetition rate and the beam scanning was repeated ten times in longitudinal direction. Therefore, the number of pulses per focal spot diameter is 10,000 pulses. Focal position was located at 6 μm below the silicone surface. As shown in Fig. 1 (a) to (c), modification tracks were successfully fabricated in the silicone film by irradiating 522 nm laser light. The optical contrast of modification tracks becomes more prominent as the pulse energy increases. But small ablation defects were observed on the modification tracks written with the pulse energy of 5 nJ (c) and continuous ablation occurs at the pulse energy more than 8 nJ (f).

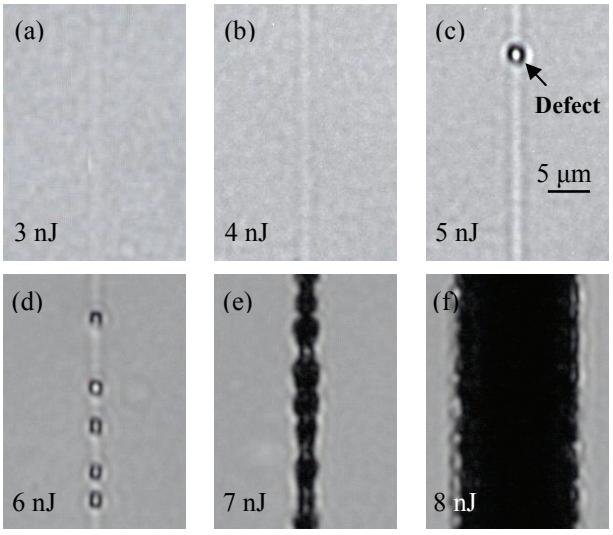


Fig. 1 Optical microscope images of silicone modification tracks written with 522 nm laser light at 1 MHz pulse repetition rate. Each line was formed with 10 scans at 1 mm/s speed, equivalent to 10,000 laser pulses per focal spot diameter. Focal position was 6 μm below surface. Single pulse energies are shown in each figure.

The defects affect the propagation loss of the modification tracks. Hence we should decrease the defects if at all possible. Table 1 shows the number of defects on the modification tracks of 10 mm in length. The number of defects is strongly affected by the pulse energy and increases sharply beyond 6 nJ. Moreover, the ablation defects also depend on the focal position. The ablation

occurs more easily as the focal position approaches the silicone surface. The pulse energy of 3 nJ seems to be suitable for making the modification tracks from balance between the number of defects and the optical contrast of modification tracks.

Table 1 The number of defects on modification track of 10 mm in length. Focal position is 6 μm below surface.

3 nJ	4 nJ	5 nJ	6 nJ	7 nJ	8 nJ
0	3	50	> 1000	Ablation threshold	Ablation

The trade-off of having zero defects on the modification tracks, with the pulse energy of 3 nJ at ~10,000 pulses per focal spot diameter, is a weaker optical contrast as shown in Fig. 1. To obtain higher optical contrast, we increased the number of beam scanning, i.e., pulse number per focal spot diameter. The result is shown in Fig. 2. The optical contrast of modification tracks becomes more visible as the pulse number increases.

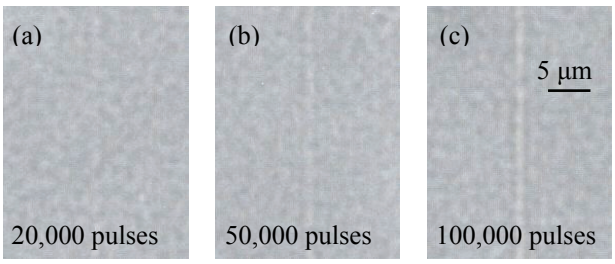


Fig. 2 Optical microscope images of silicone modification tracks written with 3 nJ of 522 nm laser light at 1 MHz pulse repetition rate. Focal position was 6 μm below surface. Pulse numbers per focal spot diameter are shown in each figure.

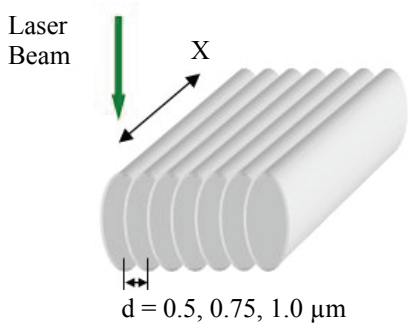


Fig. 3 Parallel line writing to widen modification tracks.

As observed in Fig. 2, widths of single modification tracks are very narrow due to the focal spot diameter of ~1 μm and a depth-of-focus of ~6 μm. To obtain wide modification tracks, we wrote seven parallel lines as shown in Fig. 3 with variable separation distances of 0.5 μm, 0.75 μm and 1 μm centre-to-centre. Optical microscope images of top view of modification tracks written with parallel line writing are shown in Fig. 4. We could obtain wide and uniform modification tracks with parallel line writing in Fig. 4 (a). However, we can see a clear separation between lines at the writing distance of (b) 0.75 μm and (c) 1.0 μm. Hence, we concluded that 0.5 μm in writing distance is suitable for making wide and uniform modification tracks.

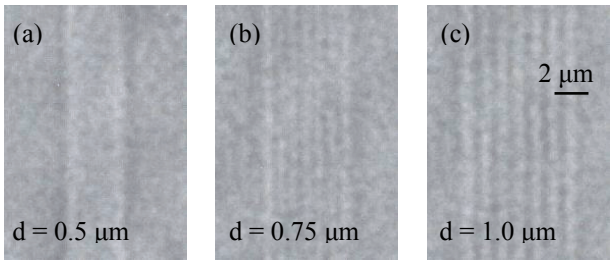


Fig. 4 Optical microscope images of silicone modification tracks written with 3 nJ of 522 nm laser light at 1 MHz pulse repetition rate. The number of parallel lines is seven and each line was formed with 100 scans at 1 mm/s speed, equivalent to 100,000 laser pulses per focal spot diameter. Focal position was 6 μm below surface.

3.2 Measurement of beam profiles and propagation losses

Three modification tracks were fabricated and their waveguiding performances were compared. The results are shown in Fig. 5. The top row of Fig. 5 (a), (b) and (c) shows top view optical microscope images of the laser exposed modification tracks written with 3 nJ of 522 nm laser light at 1 MHz pulse repetition rate. Fig. 5 (a) shows single line track, Fig. 5 (b) shows a track written with three parallel lines separated by a distance d = 0.5 μm, and Fig. 5 (c) shows a track written with seven parallel lines at d = 0.5 μm. The width of modification tracks becomes wider as the number of parallel writing lines increases.

The second row of Fig. 5 (d), (e) and (f) shows optical cross sectional view microscope images of the laser exposed modification tracks, where the laser radiation was incident from the top and focused 6 μm below the sample surface. Bright elliptical shape can be observed in the cross sectional views and the increasing widths were (d) 1.1 μm, (e) 1.6 μm and (f) 3.6 μm according to the increasing number of parallel laser tracks.

The third row of Fig. 5 (g), (h) and (i) and the forth row of Fig. 5 (j), (k) and (l) show the near-field mode profiles of the modification track guiding at 635 nm and 1550 nm wavelengths, respectively. Light guiding became stronger as the width of modification track increased, and 635 nm light guiding was stronger than 1550 nm light at each modification track. The modification tracks showed multi mode at 635 nm wavelength and single mode at 1550 nm wavelength.

The propagation loss was measured by the cut-back method. Three 40 mm long straight waveguides were shortened by a sequence of 8 - 9 mm cuts, and insertion losses were measured. The loss for each waveguide was obtained from total waveguide transmission power, normalized against the direct fiber-to-fiber transmission.

Propagation losses were estimated to be < 2.2 dB/cm at 635 nm wavelength and < 2.8 dB/cm at 1550 nm wavelength.

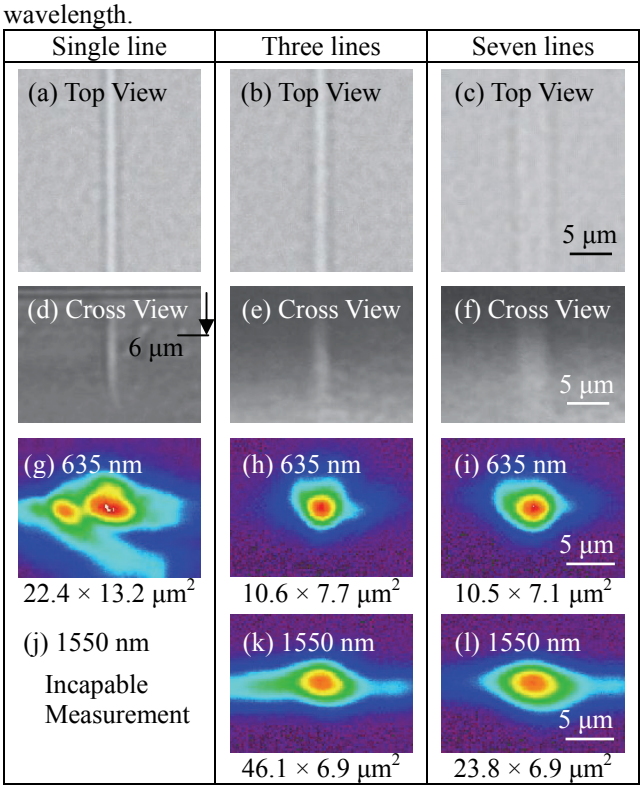


Fig. 5 Optical microscope images of (a) - (c) top and (d) - (f) cross sectional view of modification tracks written with 3 nJ of 522 nm laser light at 1 MHz pulse repetition rate and (g) - (i) the near-field mode profiles of the modification track guiding at 635 nm wavelength and (j) - (l) 1550 nm wavelength. The mode sizes ($1/e^2$) are given below the mode profiles. The numbers of parallel laser tracks are (a) one, (b) three and (c) seven and each line was formed with 100 scans at 1 mm/s speed, equivalent to ~100,000 laser pulses per focal spot diameter focused 6 μm below surface.

Chemical structure of silicone is shown in Fig. 6. Silicone has a polysiloxane backbone (-Si-O-) with side-chain methyl group (CH_3). C-H vibrational absorption causes high optical losses in the infrared region (1.0 - 1.6 μm). Energy of Si-O bond is 106 kcal/mol, C-H bond is 96 kcal/mol and Si-C bond is 71 kcal/mol. On the other hand, photon energy of second harmonic wave, 522 nm laser light (2.4 eV), equals to 55 kcal/mol. Therefore if non-linear absorption of more than two-photon occurs at focal volume, these bonds, especially Si-C bonds, might be dissociated by femtosecond laser irradiation. C-H bonds and Si-O main chains might also be photo-dissociated as the laser pulse energy or the number of laser pulses increases.

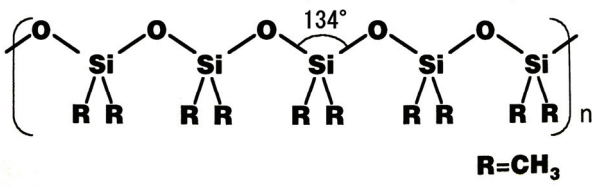
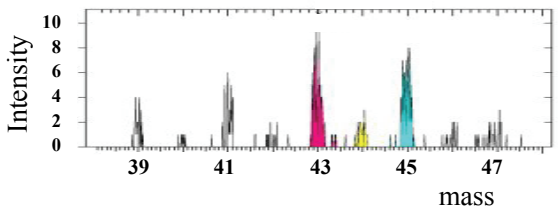


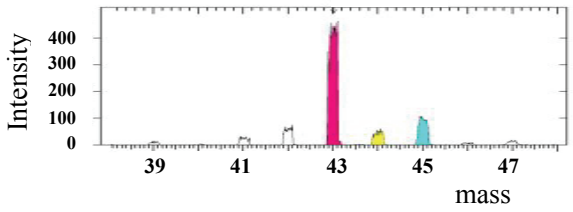
Fig. 6 Chemical structure of silicone, $[SiO(CH_3)_2]_n$.

At 157 nm F_2 laser direct writing of silica optical waveguides, Okoshi et al. reported F_2 laser irradiation of silicone in the present of oxygen can photochemically modify the surface into carbon-free silica [8]. To explain mechanism of photochemical processes driving the refractive index changes in the modification tracks written with femtosecond laser light, we analyzed laser irradiated region with TOF-SIMS. As shown in Fig. 5 (d) - (f), the modification tracks were inscribed inside the silicone film below the surface. Thus we conducted TOF-SIMS to the cross section of the modification track. The modification track was fabricated with 5 nJ of 522 nm laser light at 1 MHz pulse repetition rate. The number of pulses per focal spot was ~10,000 pulses. To obtain extra-wide modification track, 100 parallel laser tracks were written with a separation distance d = 0.5 μm. Under these conditions, we fabricated two extra-wide modification tracks with 50 μm width.

Figure 7 shows positive ion mass spectra of (a) laser irradiated region, i.e., modification track and (b) laser non-irradiated region in silicone. In both spectra, we can see $SiCH_3$ (43 mass), SiO (44 mass) and SiOH (45 mass) signals. The peak of $SiCH_3$ signal is highest in this mass region, but peak ratios of $SiCH_3$ to SiO signals and $SiCH_3$ to SiOH signals are different between the irradiated region and the non-irradiated region.



(a) Laser irradiated region

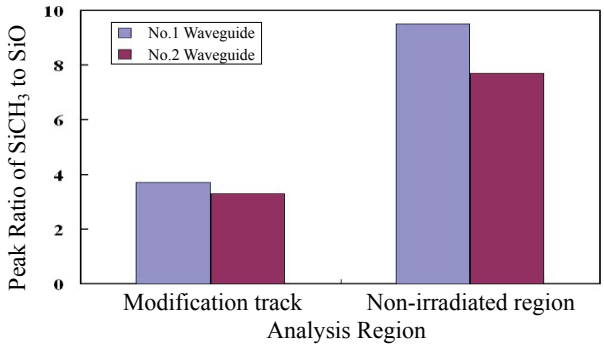


(b) Laser non-irradiated region

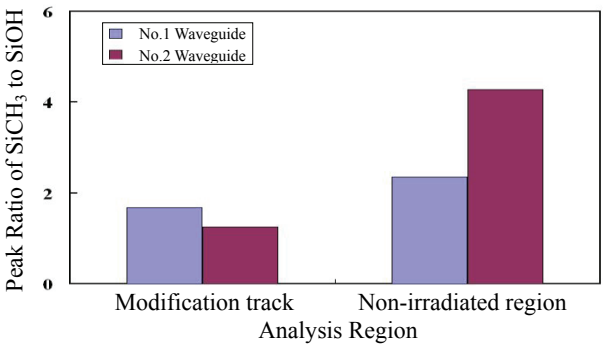
Fig. 7 Positive ion mass spectra of (a) laser irradiated region and (b) laser non-irradiated region in silicone. The modification tracks were fabricated with 5 nJ of 522 nm laser light at 1 MHz pulse repetition rate. The number of parallel laser tracks was increased to 100 at a separation distance d = 0.5 μm to obtain extra-wide modification track with 50 μm width. Pulse numbers per focal spot was ~10,000 pulses.

Fig. 8 (a) shows peak ratio of $SiCH_3$ to SiO signals obtained from TOF-SIMS spectra. As shown in this figure, peak ratio of $SiCH_3$ to SiO in the modification tracks is

half the number in the non-irradiated region. This shows that the concentration of $SiCH_3$ decreases as a result of femtosecond laser irradiation, i.e., photo-dissociation of $SiCH_3$ processes by the laser irradiation. Fig. 8 (b) shows peak ratio of $SiCH_3$ to SiOH signals. The peak ratio of $SiCH_3$ to SiOH in the modification tracks is smaller than that in the non-irradiated region. This means that the concentration of $SiCH_3$ decreases or that of SiOH increases by the laser irradiation. Okoshi et al. reported dissociation of $SiCH_3$ processes in silicone by irradiating 157 nm F_2 laser light [10]. They also said main chains Si-O of silicone is photo-dissociated, then H and O atoms are combined with the dangling bonds of Si-O- and -Si-O to produce O-H bonds and the number of O-H bonds increases with increasing laser irradiation period. Anderson et al. also reported dissociation of $SiCH_3$ by UV (290 - 320 nm) irradiation to the product Si 200, a mixture of short-chain dimethylsiloxanes with a chain length of n = 2 to 15, in D_2O solution [12]. Therefore, we consider that nonlinear femtosecond laser irradiation drives similar photochemical processes, namely silicone photo-dissociation, as the 157 nm F_2 laser. However, there was no remarkable change of SiO_2 peak in TOF-SIMS spectra. We consider that modification of silicone into silica might be occurred by femtosecond laser irradiation but its degree was very small and we could not obtain remarkable SiO_2 peak in TOF-SIMS spectra. Finally, we conclude that the refractive index is changed and waveguides are inscribed in the silicone film as results of photo-dissociation of $SiCH_3$ and photo-production of SiOH by nonlinear femtosecond laser irradiation. Further, thermal modification is suspected, and the modification of silicone seems to be caused by hybrid of the photo-dissociation and pyrolysis.



(a) Peak ratio of $SiCH_3$ to SiO signals



(b) Peak ratio of $SiCH_3$ to SiOH signals

Fig. 8 Peak ratio of (a) $SiCH_3$ to SiO signals and (b) $SiCH_3$ to SiOH signals obtained with TOF-SIMS on laser modification tracks with 5 nJ of 522 nm laser light at 1 MHz pulse repetition rate with ~10,000 pulses per focal spot.

4. Conclusions

A new approach to waveguide writing was demonstrated for the first time to produce waveguides in silicone film with 1 MHz femtosecond laser pulses. Optical waveguides were successfully fabricated in the silicone film by nonlinear multi-photon absorption. Laser processing conditions were optimized for fabricating silicone waveguides. The mechanism of photochemical processes driving the refractive index changes was explained. The results demonstrate a new opportunity for fabricating micro-optical components, optical circuits, and sensors on flexible substrates.

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