

## Fabrication of quasi-periodic micro-voids in fused silica by single femtosecond laser pulse

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**Abstract** We demonstrate that quasi-periodic void structure can be self-formed in transparent materials by single femtosecond laser pulse. Compared to the multiple-pulse induced structures, the single-pulse induced void structures are very short and may contain absent voids. The formation mechanisms have been discussed comparatively in detail. Based on this, a technique for high-speed and large-area fabrication of micro-void arrays in transparent materials has been presented. The experimental results show that 3D micro-void structures which contain over several hundred thousand voids in micrometer scales are produced in areas of square millimeters within a few minutes, and the periods of micro-void structures can be easily varied by processing parameters. This work has potential applications in 3D optical storage, photonic crystal and integrated optics, and provides novel insight into the interaction between the single femtosecond pulse and the transparent materials.

### 1 Introduction

Femtosecond lasers have become a promising tool for fabricating microstructures inside transparent materials, primar-

ily due to their ultra-short pulse duration and ultra-high peak power, which have distinct advantages over the continuous-wave and long-pulse lasers [1–5]. Among these applications, the fabrication of void arrays in the bulk of dielectrics has attracted special attention for its potential applications in three-dimensional (3D) optical storage, photonic crystal and integrated optics [5–8].

Since the early observation of the femtosecond-laser induced periodically aligned nano-voids inside conventional borosilicate glasses by S. Kanehira et al. in 2005 [9], many groups have been devoted to exploring the applications and formation mechanisms of the self-organized void arrays. For example, Xiao Hu et al. [10] reported the self-formation of quasi-periodic void structure with the length of several hundred micrometers inside CaF<sub>2</sub> crystal. The quasi-periodical voids along the propagation direction of the laser beam were formed spontaneously after the irradiation of a single femtosecond laser beam which was focused at a fixed point inside the crystal sample. They also found another void array grown vertically to the laser propagation direction due to the light passing through the side facet of the sample [11]. Eiji Toratani et al. [12] fabricated a 90°-bend waveguide in fused silica by serving voids as an optical mirror. Juan Song et al. [13] suggested that spherical aberration, which resulted from the refractive index mismatch between air and fused silica glass, was the main reason for the formation of the self-organized void array.

However, most 3D void array structures are fabricated using the point-by-point in situ irradiations of several femtosecond laser pulses. This method is inefficient and limits its industrial applications. For example, consider one-point irradiation spends 2 seconds: to fabricate a void structure of dimension 1 × 1 mm<sup>2</sup> and period 5 μm will spend over 20 hours. If the quasi-periodic void structure can be formed by single femtosecond laser pulse, the 3D micro-void struc-

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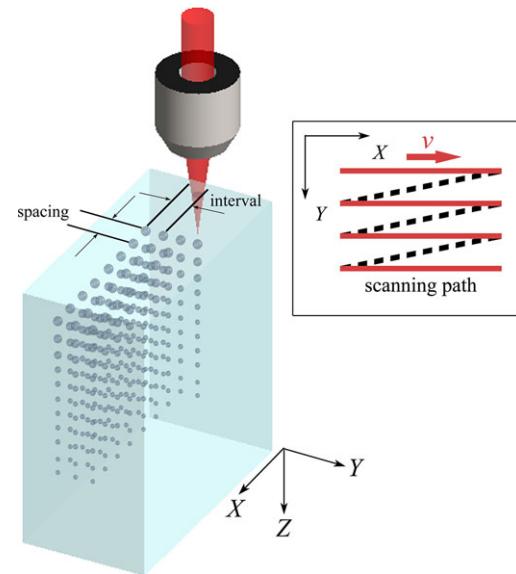
tures will be efficiently fabricated by fast-scanning the laser pulses (at a repetition rate of several kilohertz) line-by-line inside the sample. Compared to the classic multiple *in situ* irradiations process, the whole process using this method will be shortened by hundreds or even thousands times. Furthermore, the formation mechanism of quasi-periodic void structure was still controversial; the quasi-periodic void structure formed by single femtosecond laser pulse will provide novel insight into the interaction between the single femtosecond pulse and the transparent materials.

In this paper, we demonstrated that quasi-periodic void structure can be self-formed in transparent materials by single femtosecond laser pulse, and the dependence of the 3D morphologies on the pulse energy is experimentally investigated. The formation mechanisms have been discussed comparatively in detail. Subsequently, 3D void arrays with areas of  $1 \times 1 \text{ mm}^2$  are successfully manufactured in fused silica bulks within a few minutes. The experimental results show the quasi-periodic void structures have stable reproducibility and the arrangement of micro-void structures can be easily varied by the processing parameters. The diffractive patterns of micro-void arrays show a good performance as diffractive beam splitters.

## 2 Experimental setup

In our experiments, the laser source is a Ti:sapphire oscillator-amplifier system (FEMTOPOWER Compact Pro, FEMTOLASERS), which delivered 800 nm, 30 fs Gaussian laser pulses at a repetition rate of 1 kHz. The pulse energy could be continuously varied by a variable neutral density filter, and the laser pulses are focused via a  $100\times$  microscope objective (Nikon, N. A. = 0.80). The sample used in our experiments is fused silica of dimensions  $10 \times 10 \times 5 \text{ mm}^3$ , all the surfaces of which have been polished. Before laser irradiation, the sample is cleaned by ultrasonic bath for 15 min in acetone and rinsed in de-ionized water.

Figure 1 shows the schematic illustration for the 3D void array using single-femtosecond-pulse formation method, and the insert presents the moving path of the sample. The femtosecond laser pulses are tightly focused by a high numerical aperture (N. A.) objective into the fused silica, the sample is located on a 3D stage controlled by a computer. Suppose the moving speed of the sample is set as  $v$  in mm/s, the repetition rate of laser pulses is 1 kHz, then the spacing between two adjacent voids in one irradiation line is  $v$  in  $\mu\text{m}$ . If the diameter of the void,  $d$ , is smaller than  $v$  in  $\mu\text{m}$ , that is  $d < v$  in  $\mu\text{m}$ , then we can obtain a separated void array rather than a continuous line. In order to improve the uniformity of a void array, we keep the direction of scanning irradiation unchanged; the dotted lines in scanning path represent the lines with laser irradiation, and the dashed lines

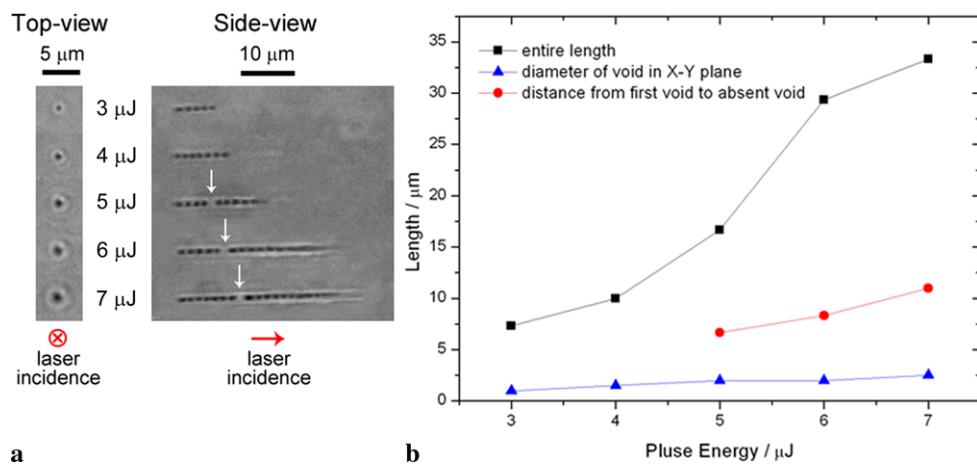


**Fig. 1** Schematic illustration for fabrication of 3D quasi-periodic void structure array. The *insert* shows the scanning path: the *dotted lines* in scanning path represent the lines with laser irradiation and the *dashed lines* represent the lines without laser irradiation

represent the lines without laser irradiation. In our experiments, we set the interval between two adjacent scanning lines equal to the spacing between two adjacent voids in one scanning line, so we can obtain a void array the periods of which in  $X$  and  $Y$  directions are equal.

## 3 Results and discussion

The 3D morphologies of the quasi-periodic void structures induced by different laser energies are shown in Fig. 2. The pulse energies are 7, 6, 5, 4 and 3  $\mu\text{J}$ , respectively. The focal spot is 200  $\mu\text{m}$  beneath the sample surface, and the scanning speed  $v = 10 \text{ mm/s}$ . In the top-view images, there are concentric circles around the void, which agrees well with the top view of laser fluence distribution in Ref. [13]. From the side-view image, a string of voids formed beneath the focal spot along the laser propagation direction, which is the direction of  $Z$ -axis. The first void starts from about 300  $\mu\text{m}$  beneath the sample surface, which is about 100  $\mu\text{m}$  beneath the focal spot. This is also in agreement with the simulated fluence distribution around focal spot in Ref. [13]. Surprisingly, there is an absent void present when the pulse energy increased higher than 5  $\mu\text{J}$ , as shown in side-view image (as white arrows indicate). And the locations of absent voids are varied by the incident pulse energy. At the end of the structures, the voids degenerated to a long cavity. Figure 2(b) shows the entire lengths of the quasi-periodic void structures, average diameters of the voids in  $X-Y$  plane and the distances from the first void to the absent void as functions of incident laser energy.



**Fig. 2** The 3D morphologies of quasi-periodic void structures induced by different pulse energies in our experiments. (a) The top-view and side-view images of fabricated void strings. The pulse energies are: (from top to bottom) 3, 4, 5, 6 and 7  $\mu\text{J}$ . The focal spot was 200  $\mu\text{m}$  beneath the sample surface, and the scanning speed  $v = 10 \text{ mm/s}$ . The

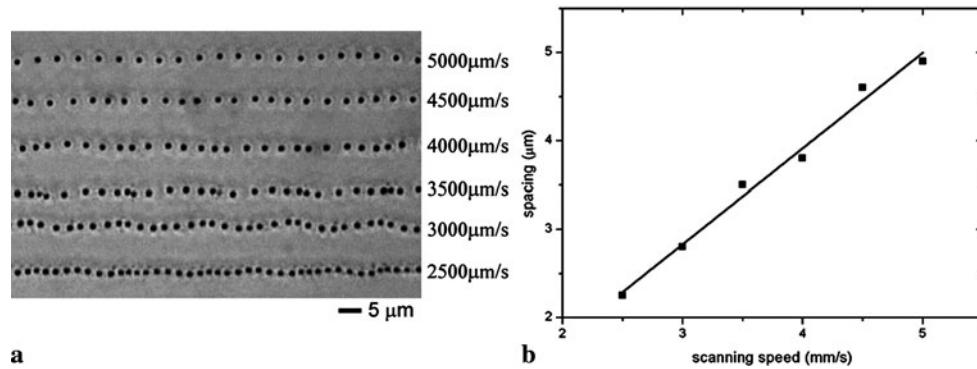
white arrows in side-view image indicate the locations of absent voids. The directions of laser incidence are also shown in the figure. (b) The entire length of the quasi-periodic void structures, the average diameters of the voids in  $X-Y$  plane and the distances from the first void to the absent void as functions of incident laser energy

The entire length of quasi-periodic void structures is elongated with the increase of laser energy, but less than  $\sim 100 \mu\text{m}$  reported in former papers [9–11]. We attribute the reduction of the entire length to the decrease of incident pulse energy and pulse number. The incident laser energies employed in our experiments are several microjoules, and in the former papers they are about tens of microjoules. The quasi-periodic void structure is induced by single laser pulse in our experiments; in the former papers, the structure is induced by tens, or even hundreds of laser pulses. The transparent sample absorbs lower incident laser energy, which is determined by the product of pulse energy and number, rapidly; so the entire length of quasi-periodic void structures decreased. We also fabricated quasi-periodic void structure by 500 pulses with the pulse energy of 5  $\mu\text{J}$ , and found that the entire length was about 50  $\mu\text{m}$ , which is close to  $\sim 60 \mu\text{m}$  reported in Ref. [13]. Furthermore, with propagating and consuming of laser energy for a certain distance, the void structure fails to occur, and a long cavity is formed at the tail of the structure, which also agrees with the earlier reports [13, 14].

Moreover, there is an absent void in the laser propagation direction when the pulse energy increases up to 5  $\mu\text{J}$ . The simulated laser fluence distribution in Ref. [13] also shows a notable minimum at 125  $\mu\text{m}$  beneath focal spot when both spherical aberration and nonlinear effects take place, which gives a validation for our experimental results. However, we have not found any absent voids in the quasi-periodic void structures induced by multiple laser pulses in former papers. We also fabricated quasi-periodic void structure by multiple pulses, and did not observe the absent void. This difference may result from the energy accumulation of successive pulses; after tens or hundreds of pulses inci-

dence, the minimum of incident laser fluence distribution is higher than the formation threshold of the void, and the absent voids are vanished. The location of the absent void will move close to the first void when the incident laser energy decreased, as shown in Fig. 2(a). We think the formation and variation of the absent void may be the following: The periodical distribution of laser fluence induced by the spherical aberration and the nonlinear effects is depended on the incident laser energy; the spherical aberration is beneficial to the formation of periodical distribution whereas the nonlinear effects go against it. When the incident laser energy gets lower, the spherical aberration occupies the leading position, the nonlinear effects can be neglected, and a string of voids is formed. With the increase of laser energy, the spherical aberration and the nonlinear effects are intensified, and the nonlinear effects cannot be neglected anymore; consequently, the absent void is presented. A further increase of laser energy would further intensify the spherical aberration and nonlinear effects, but the accession rate of nonlinear effects is lower than the spherical aberration; as a result, the minimum of laser fluence moves to the tail of decayed periodical laser fluence distribution, and the absent void moves far away from the first void.

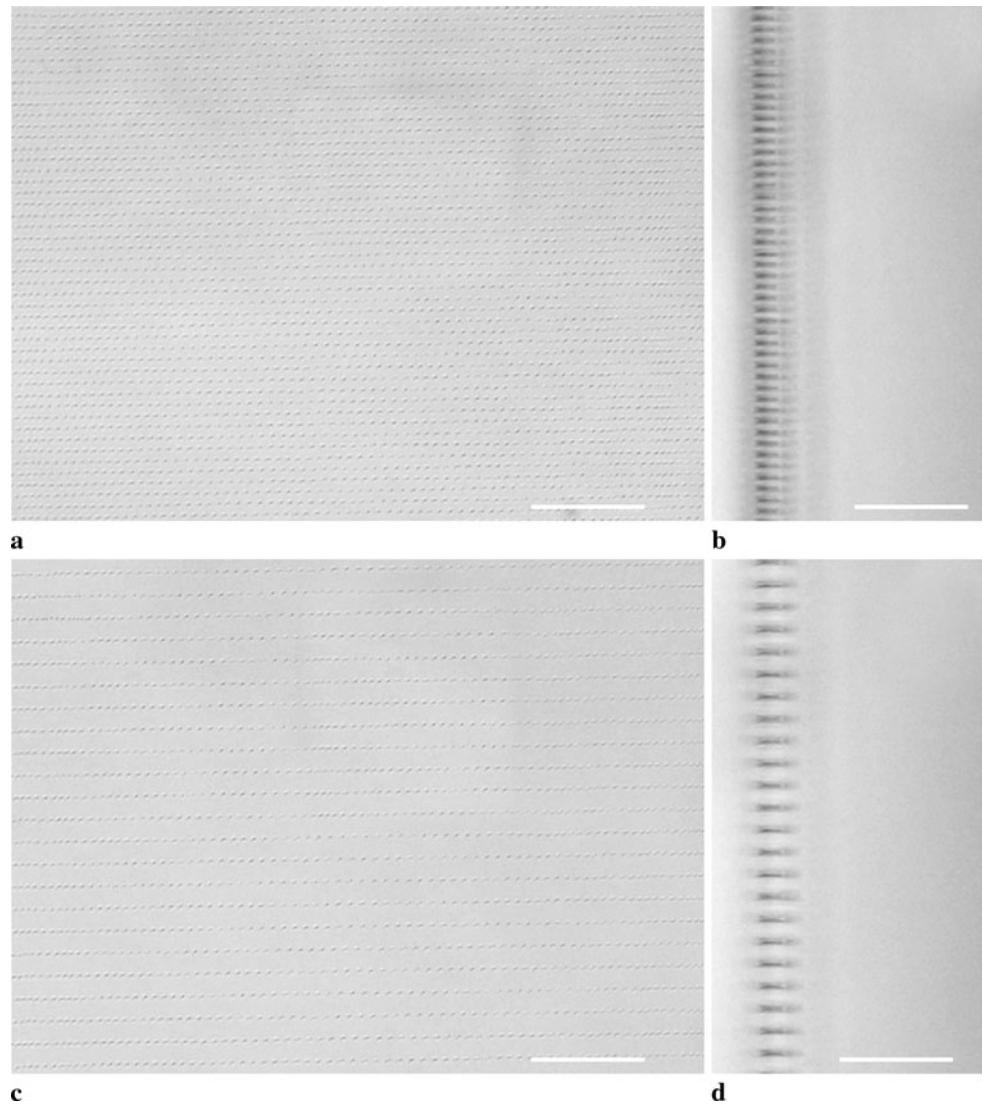
Figure 3(a) shows the top-view image of the void ranks formed by the femtosecond laser irradiation with different scanning speeds, and the average spacing between two adjacent voids in one rank is plotted in Fig. 3(b) against the scanning speed. The single-pulse energy was 5  $\mu\text{J}$ , the focal spot was 200  $\mu\text{m}$  beneath the sample surface. Clearly, the spacing between two adjacent voids in one rank is proportional to the scanning speed, which indicates the period of void array can be changed by scanning speed. In practice, this spacing is determined by the incident laser energy



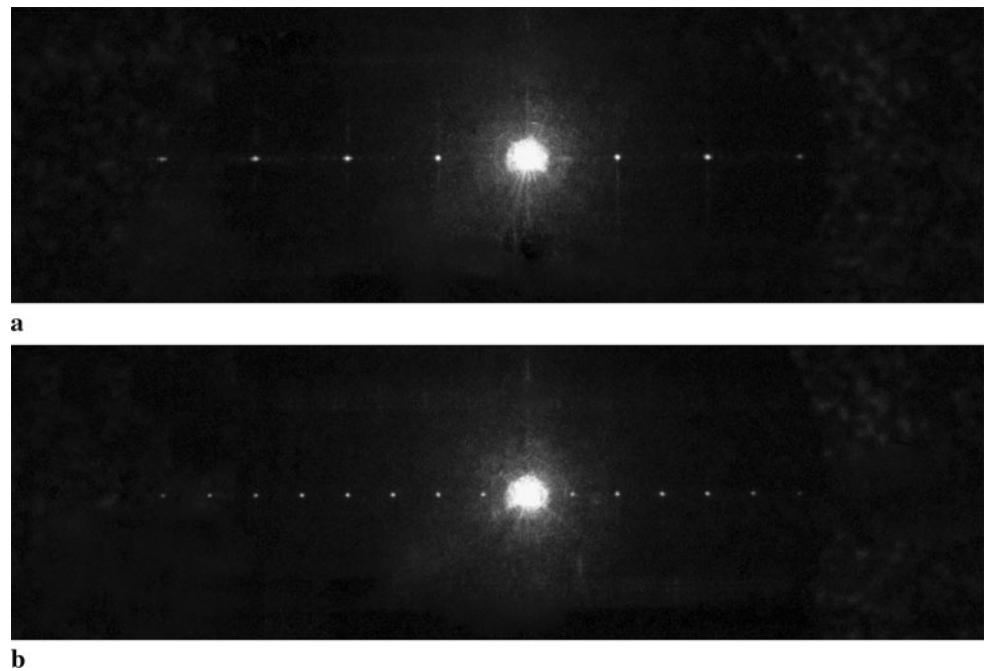
**Fig. 3** The dependence of spacing between two adjacent voids on the scanning speed. The single-pulse energy was 5  $\mu\text{J}$ , the focal spot was 200  $\mu\text{m}$  beneath the sample surface. **(a)** The top-view image of the void ranks formed by the femtosecond laser irradiation with scanning

speed of (from top to bottom) 5, 4.5, 4, 3.5, 3 and 2.5 mm/s. **(b)** The average spacing between two adjacent voids in one rank plotted against the scanning speed

**Fig. 4** **(a), (c)** top-view and **(b), (d)** side-view images of the fabricated void arrays of dimensions  $1 \times 1 \text{ mm}^2$ . The incident single-pulse energy was 5  $\mu\text{J}$ . The scanning speeds were **(a), (b)** 10 mm/s and **(c), (d)** 20 mm/s, so the periods of the two void arrays are **(a), (b)** 10  $\mu\text{m}$  and **(c), (d)** 20  $\mu\text{m}$ , respectively. All of the scale bars in four images are 100  $\mu\text{m}$



**Fig. 5** The diffractive patterns of the fabricated void arrays presented in Fig. 4 with normal incidence of He–Ne laser ( $\lambda = 632.8$  nm). (a) The diffractive pattern corresponding to the fabricated void array shown in Fig. 4(a). (b) The diffractive pattern corresponding to the fabricated void array shown in Fig. 4(c)



and specialized requirements. However, lowering scanning speed may not allow to separate the void from each other, so there will form a continuous line rather than a series of separated voids.

Thus, we fabricated void arrays of dimensions  $1 \times 1 \text{ mm}^2$  to identify the efficiency of processing. Figure 4 shows the top-view and side-view images of the void arrays. The incident single-pulse energy was  $5 \mu\text{J}$ . The scanning speeds were (a), (b)  $10 \text{ mm/s}$  and (c), (d)  $20 \text{ mm/s}$ , so the periods of voids in  $X-Y$  plane were  $10$  and  $20 \mu\text{m}$ , respectively. Both these void arrays were fabricated within 5 minutes, and the 3D morphologies have a nice uniformity.

The corresponding diffractive patterns of micro-void arrays presented in Fig. 4 are shown in Fig. 5. The incident light is He–Ne laser ( $\lambda = 632.8$  nm) with normal incidence. From the figures we can see that the diffractive patterns are similar to the diffractive patterns of one-dimensional grating, rather than two-dimensional grating. According to the Babinet principle, the void array with normal incidence has similar diffractive pattern to two-dimensional grating, but the results are different; we attribute this difference to that the arrangement of fabricated void arrays in  $X-Y$  plane is not strictly aligned. From the top-view images in Fig. 5, the void arrangement has slight horizontal deviation due to the moving and positioning precision of moving stages, and in vertical direction the voids are distributed comparatively uniformly. So, because of the destructive superposition in deviated direction, the diffractive patterns are similar to the diffractive patterns of one-dimensional grating.

#### 4 Conclusions

In summary, a method for high-efficiency fabricating of large-area periodic micro-void arrays in transparent materials by femtosecond laser pulses has been proposed. We experimentally demonstrated that quasi-periodical void structures can be induced by single femtosecond laser pulse in fused silica, and a  $1 \times 1 \text{ mm}^2$  void array can be fabricated within several minutes. Using this method, the whole process was shortened by hundreds or even thousands times. The discussion of formation mechanism showed that the interface spherical aberration and the nonlinear effects have notable influence on the formation of the quasi-periodical void structures. The diffractive performance shows that the fabricated micro-void arrays have a good performance as diffractive beam splitters, and the diffractive patterns are similar to the diffractive patterns of one-dimensional grating. Our scanning method is favorable to the large-area array fabrication, and provides novel insight into the interaction between the single femtosecond pulse and the transparent materials, which has potential applications in optical storage, photonic crystal and integrated optics.

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