Femtosecond laser-induced color change and filamentation in Ag^+-doped silicate glass

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We investigate the influence of multiple filamentation (MF) on the micromachining in Ag^+-doped silicate glass irradiated by a 1-kHz femtosecond laser. The thresholds of MF and color change (CC) are measured for both linearly and circularly polarized laser beams. The results demonstrate that the thresholds of MF and CC are very close. The thresholds of CC and MF for circular polarization increase by ∼1.4 times compared with linear polarization. Circular polarization can suppress the number of filaments to some extent compared with linear polarization. However, it is difficult to obtain CC without any filamentation if circular polarization technique is used alone.

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In recent years, femtosecond (fs) laser microfabrication has received intensive attention because of its powerful microprocessing abilities. Various microstructures, such as ripples, gratings, and different patterns have been fabricated by direct writing or multiple beam interference[1−4]. However, it is worthy to note that most fabricated structures exist either on or slightly beneath the surface of material. We can ascribe these results to the limit of work distance of objective lens and low resolution. The latter is caused by some unavoidable nonlinear effects, such as self-focusing and filamentation. When we want to obtain a large area of pattern by photolithography deep in material with a long focal length convex lens, the low resolution effect is more pronounced. In this case, the laser power used is more powerful than focusing with an objective lens. Then self-focusing and filamentation are more easily produced. Up to now, the mechanism of self-focusing and filamentation of a powerful fs laser beam propagating in an optical medium is well known[5] . When the incident laser power is more than the critical power for self-focusing P_{cr}, filamentation occurs. At higher incident power, a single input beam can also break into several long and narrow filaments, which is known as multiple filamentation (MF). The direct visualization of MF has been presented in many media using a charge-coupled device (CCD) camera[6] . The general explanation for MF in the literature is that it is caused by noise in the input beam. Since noise is random, this implies that the MF pattern is unpredictable and tends to be different from shot to shot[7]. Therefore, MF is very difficult to control and usually harmful to many practical applications.

In this letter, we investigate the influence of MF on the micromachining in Ag^+-doped silicate glass induced by fs laser when focusing with a convex lens with 1-m focal length, and measure the thresholds of MF and color change (CC) irradiated by linearly and circularly polarized laser beams. The purpose is to provide some useful information for micromachining. Ag^+-doped silicate glass is chosen, since it exhibits large third-order nonlinear susceptibility and ultrafast nonlinear response, and has been used extensively for microfabrication[8]. Additionally, the irradiated area in this material by fs laser can turn to yellow color after subsequent heat treatment, which is an attractive phenomenon for microfabrication. Based on this color change, the designed pattern is possible to be recorded if an appropriate laser power is chosen. Under this laser power, only CC occurs, while MF does not. The mechanism of CC is well known[8]. Similar to the developed photosensitive glasses, after the irradiation by fs laser, free electrons are released from the substrate (from a sensitizer for photosensitive glasses), while Ag^+ captures the electron and becomes an Ag atom. These Ag atoms will precipitate in the fs laser irradiated area after subsequent heat treatment (around 500 °C), and then show yellow color.

The composition of the Ag^+-doped silicate glass sample used in this study was $0.1Ag_2O{\cdot}70SiO_2{\cdot}10CaO{\cdot}20Na_2O$ (mol%). Reagent grade SiO_2, $CaCO_3$, Na_2CO_3, and AgO_2 were used as starting materials. The detailed glass preparation procedure has been described elsewhere[9]. Before the experiment, the glass samples were cut into small sizes of 10×5×2 (mm) and then six-side polished.

A schematic diagram of the experimental setup is shown in Fig. 1. A regenerative amplified Ti:sapphire laser (Legend USP, Coherent Inc.) was used in our study. The repetition rate, wavelength, and pulse duration of the fs laser were 1 kHz, 800 nm, and 40 fs, respectively. The samples were placed on a three-dimensional (3D) translation stage controlled by a computer. We focused the pulses inside the Ag^+-doped silicate glass to generate structural changes using a convex lens with 1-m focal length. The geometrical focal depth was fixed at 800 μm below the sample surface,

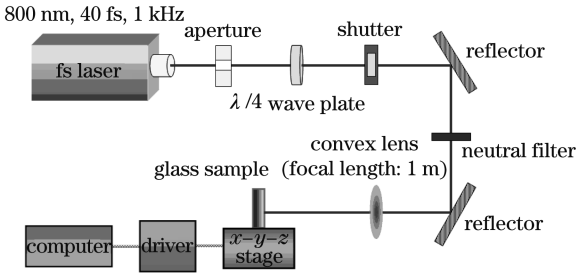

Fig. 1. Schematic diagram of experimental setup.

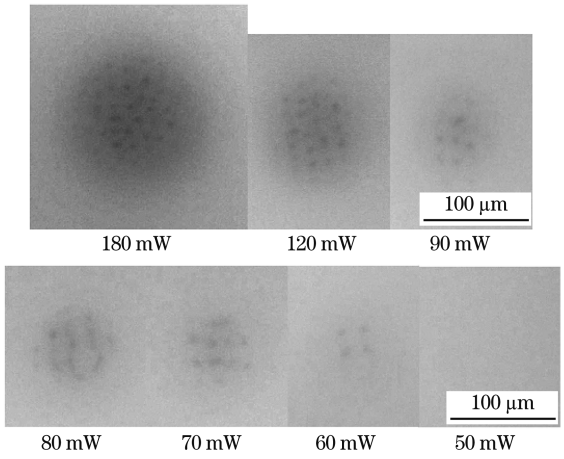

Fig. 2. Top views of MF when irradiated by linearly polarized fs laser.

and the estimated diameter of focal spot was approximately 100 μm (1/e). For each irradiation, the laser exposure time was 1 min, and after that, the stage was moved to a new area. The same experiments were repeated for three times. The laser power could be varied from 1 to 400 mW continuously by using a neutral filter. After irradiation, the samples were further annealed. The temperature was firstly ramped from room temperature to 500 °C at a speed of 5 °C/min, and then held at 500 °C for 30 min. Finally, the samples were naturally cooled down to room temperature. We then observed the top and side views using an optical microscope (Olympus BX51).

Figure 2 shows the typical top views generated by the irradiation of linearly polarized focused fs laser beam in Ag^+-doped silicate glass. One can see that the spot size increases, and yellow color becomes much deeper with increasing laser power. It is worthy to note that there are some small dots embedded in the yellow spot patterns, which are caused by MF as mentioned above. The typical side view of MF is shown in Fig. 3, and many narrow filaments are observed, which propagate across the whole side. With increasing the incident laser power, the filament number increases much more. There are honeycomb-shaped dots on the laser irradiation spot. The surface damage occurs when we increase the laser power to 220 mW. In order to show this trend clearly, we have made statistics on the number of filaments as a function of incident laser power, as the diamonds shown in Fig. 4, where the data is the average over three experimental results. It is found that the filament number increases almost linearly with laser power. Fs laser-induced CC now has performed promising applications in optical storage, colored sculpture[8], and so on, but MF tends to follow CC due to many nonlinear effects when laser propagates in material. In order to avoid this phenomenon, reducing laser power is an optional way. But when reducing the laser power to a low value, CC becomes too weak to be identified, as shown in the case of 50-mW irradiation in Fig. 2. With increasing laser power slightly to 60 mW, CC along with the undesirable MF induced dots appears again. This indicates that the thresholds of CC and MF are very close for Ag^+-doped silicate glass, which is in the range of 50−60 mW. As a result, it is difficult to obtain CC alone under a larger laser power by normal machining process. The critical peak power of self-focusing for silica glass P_{cr} is about 2.0 MW[10]. When a 40-fs laser pulse of 50 μJ (which is the threshold of CC and MF) is focused inside glasses, the laser peak power reaches 1.5 GW level. Under such a high peak power, MF occurs easily.

It has been reported that circularly polarized laser can suppress MF compared with linearly polarized laser[7]. So we investigate the CC and filamentation inside Ag^+-Top doped silicate glass by the irradiation of circularly polarized laser. In our experiments, a $\lambda/4$ wave plate was used to achieve the circular polarization. The experimental results are shown in Fig. 5, which indicate that the thresholds of CC and MF increase by ∼1.4 times compared with the case of linear polarization, in the range of 70−80 mW, whereas two thresholds are still very close. The dots in Fig. 4 show the dependence of number of filaments on incident laser power, from which we can find that filamentation is suppressed to some extent compared with linear polarization. The surface damage threshold is also found increasing to 250 mW. However, it is difficult to obtain CC pattern without any filamentation if circularly polarized laser is used alone because of their close threshold values. The laser-field amplitude for circular polarization is only

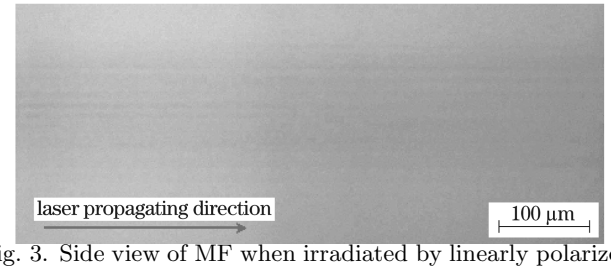

Fig. 3. Side view of MF when irradiated by linearly polarized fs laser.

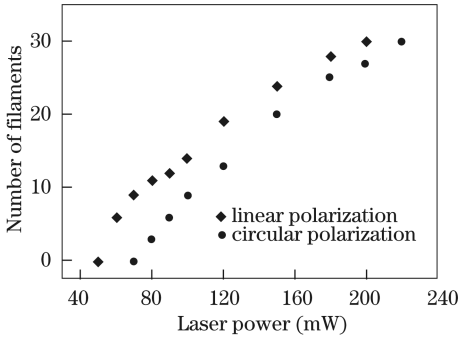

Fig. 4. Dependence of the number of filaments on incident laser power.

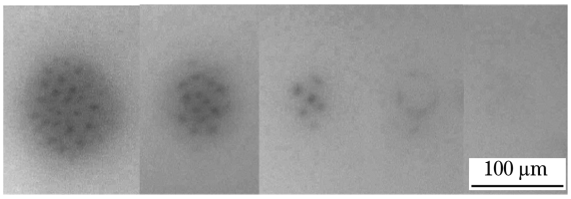

Fig. 5. Top views of MF captured by optical microscope when irradiated by circularly polarized fs laser.

$1/\sqrt{2}$ of that for linear polarization under the same laser intensity. Consequently, the nonlinear coefficient is lower for circularly polarized laser beam than linearly polarized laser beam[11], which leads to a much higher self-focusing threshold for circular polarization. Therefore, compared with linearly polarized laser, the number of filamentation induced by circularly polarized laser is less.

In conclusion, we investigate CC and MF inside Ag^+-doped silicate glass by using an 800-nm, 1-kHz fs laser. The thresholds of CC and MF irradiated by linearly and circularly polarized laser beams were measured. The results indicate that circular polarization can suppress the number of filaments to some extent, and the thresholds of CC and MF for circular polarization increase by ∼1.4 times compared with linear polarization, which is in the range of 70-80 mW, whereas it is difficult to obtain CC pattern without any filamentation if circular polarization technique is used alone because of the close threshold values of CC and MF. Our study provides some information for structuring patterns inside material, and how to achieve CC only is another question we will investigate in the future.

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