Self-formation of void array in Al_2O_3 crystal by femtosecond laser irradiation

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The femtosecond laser induced void array inside Al_2O_3 crystals was discussed. The void array was formed spontaneously under the irradiation of a single beam of infrared femtosecond laser which was focused at a fixed point inside the Al_2O_3 crystal sample. It was found that the regular voids only could be fabricated near the sample surface, which was different from the situation in CaF_2 single crystal reported before. The possible mechanism of the phenomena was also discussed.

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Femtosecond lasers, with extremely high electric field and ultrashort pulse duration, have become ideal tools to realize microscopic modifications for various materials[1]. Periodic microstructures induced by femtosecond laser in transparent materials were especially fascinated, because such structures have potential to be used as photonic elements such as photonic crystals and microgratings[2−6]. For a long time, such periodic structures can only be realized by multi-beam laser interference method[7]. Recently, Kanehira *et al.*[8] reported a one-step method of fabricating periodic nanovoid inside borosilicate glasses. In this method, the single beam of femtosecond laser was focused at one point inside the borosilicate glass, and quasi-periodical voids were then spontaneously formed along the propagation direction of the laser beam during the irradiation process.

Compared with glasses, crystals show more excellent optical properties in high luminescent efficiency and thermal stability etc. It is valuable to explore the similar void structures in various kinds of crystals by the one-step method. In this paper, we report the self-formation of voids in a kind of typical crystal using the same method. As a kind of important crystal, Al_2O_3 is widely used in the field of optical component and micromechanics. It owns many important mechanical, optical, and electrical properties, and has the potential for the fabrication of precise and well-defined micrometer-size structures[9]. It is found that regular voids can only be fabricated near the sample surface in Al_2O_3, which is different from the situation in CaF_2 single crystal which we reported before[10]. The possible mechanism of the observed phenomenon was discussed.

A regeneratively amplified Ti: sapphire laser with 800-nm wavelength, 120-fs pulse duration, and 1-kHz repetition was used in our study. The size of the polished Al_2O_3 crystal sample is 5 × 5 × 1 (mm). The laser pulse energy was adjusted from 1 to 30 μJ using a neutral density filter. The laser beam was focused at a fixed point inside the Al_2O_3 sample by a 100× objective lens with numerical aperture (NA) of 0.9. The number of laser pulses irradiated at the focusing point was controlled by an electronic shutter. The side view of the microstructure induced by the laser pulses in the Al_2O_3 sample was observed through an optical microscope. The experimental setup of the optical system which we used had been shown elsewhere[10].

Figure 1(a) shows a typical void array in Al_2O_3 generated by the irradiation of a femtosecond laser beam without moving the focal point. Quasi-periodic voids were spontaneously formed in Al_2O_3 below the focal point and lined up along the propagation direction of the laser beam. To fabricate this void array, the single pulse energy was 20 μJ and the laser beam was fixed at a point of 110 μm beneath the crystal surface during the irradiation process. The irradiation time was 0.064 s, corresponding to 64 pulses launched into the sample. The void array contains 16 voids, and the total length is 91 μm. The focal depth of the laser beam was measured in the side view optical microscope photograph of the sample.

At the same time, we compared the result in Al_2O_3 with that in CaF_2 when the laser pulse energy, irradiation time, and the focal depth were the same. Figure 1(b) is the laser induced microstructure in CaF_2 under the same experimental conditions, and there was no regular void, only a damaged line was produced below the focal point by irradiation. It seemed that regular voids could not be fabricated at that shallow position inside

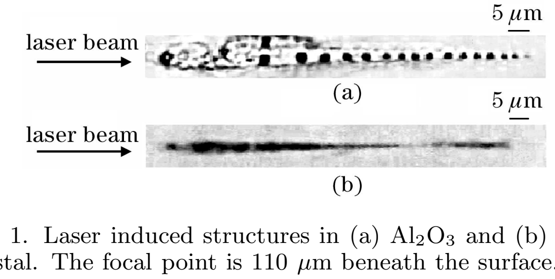

Fig. 1. Laser induced structures in (a) Al_2O_3 and (b) CaF_2 crystal. The focal point is 110 μm beneath the surface. The pulse energy is 20 μJ. 64 pulses are launched into the sample, and the length of the arrays both are 91 μm.

CaF_2 by laser beam.

On the contrary, we found that regular voids could not be fabricated at deep positions inside Al_2O_3. Figure 2(a) is the damaged line in Al_2O_3 crystal induced by laser beam with focal depth of 300 μm. The single pulse energy is 30 μJ and the irradiation time is 0.5 s. There is no regular void formed under such irradiation condition, just some damaged marks left. However, in our previous study on CaF_2, the void array can be produced even in much deeper positions. Figure 2(b) shows the regular voids generated by irradiation of laser beam focused at 950 μm beneath the CaF_2 surface, and the single pulse energy and the irradiation time were the same as in Al_2O_3, which is shown in Fig. 2(a).

We also studied the effect of the focal depth beneath the sample surface on the length of void array in Al_2O_3. The focal depth variation was made by moving up the crystal sample. Laser pulses with energy of 20 μJ were focused by a 100× objective lens, the irradiation time was fixed as 0.5 s. Figure 3 shows the relationship between the focal depth and the void array length. As the focal depth increases, the length of the void array formed by irradiation of these laser pulses increases firstly. However, when the focal point is deeper than 70 μm beneath the surface, the length of the void array decreases inversely, and the void structure becomes not regular gradually. The rule of the phenomena in Al_2O_3 is similar to that in CaF_2.

From the experimental results shown above, we can conclude that the formation of regular voids strongly depends on the depth of the laser focal point, when the pulse energy is fixed, the voids only appear in a fixed depth range, and the depth range is different for different materials. If the position of the focal point is out of the fixed range, no regular voids will appear after the laser

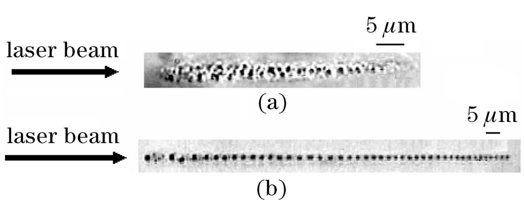

Fig. 2. (a) Damage line in Al_2O_3 crystal, and the focal point is 300 μm beneath the surface. (b) Voids structure in CaF_2, and the focal point is 950 μm beneath CaF_2 surface. The pulse energy is 30 μJ. 500 pulses are launched into the samples.

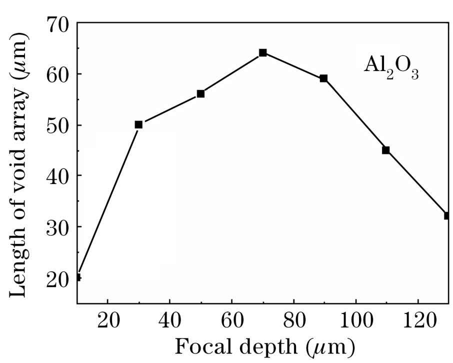

Fig. 3. Length of the void array versus the laser focal depth beneath the entrance surface. Laser pulse energy is 20 μJ and the irradiated time is 0.5 s.

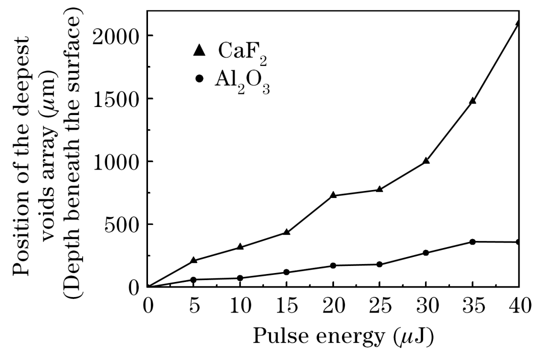

Fig. 4. Depth of the deepest regular void structure that can be fabricated by the laser pulses in samples versus the pulse energy.

irradiation, and only a damage line will be left. We compared the position of the deepest void array induced by laser beam in CaF_2 with that in Al_2O_3 when changing the laser pulse energy, which is shown in Fig. 4. In this figure, the depth is determined by measuring the focal depth of each void array. It also shows that under the irradiation with the same pulse energy, the voids can be induced in a much deeper position in CaF_2 than in Al_2O_3. For example, when the pulse energy is 30 μJ, void array can be induced only when the laser beam was focused shallower than 270 μm beneath the surface in Al_2O_3, but with the same energy, the deepest void array would appear at about 1000 μm beneath the sample surface in CaF_2.

The mechanism of the phenomena can be studied here. We suggested that the voids were formed during the laser propagation process which is usually considered to be the competition between self-focusing by the Kerr effect and defocusing by the plasma[11−17]. The process was usually expressed by Schrödinger equations involving self-focusing effect, group velocity dispersion (GVD), impact ionization, and multiphoton ionization (MI)[14]. This theory is usually used in condensed transparent nonlinear mediums, like glasses and crystals[14,15]. The details of the voids formation mechanism were discussed elsewhere[10].

In this paper, our experimental results indicate that the depth of the focal point strongly influences the formation of voids in different crystals. Under the irradiation with the same pulse energy, the voids can be induced in deep positions in CaF_2 while in Al_2O_3 voids only can be formed in shallow positions. Our previous experimental results also showed that in CaF_2, if high pulse energy was launched in shallow positions, voids could not be formed and only damage line left. We suggested that under this situation, the material was damaged badly, therefore, regular voids were destroyed and could not be recognized. But if we reduced the laser power to a proper value, the voids also could be formed in a shallow position in CaF_2.

From these phenomena, we conclude that the pulse energy at the focal point is important for the formation of the voids in crystals, and we suggest that the power of the laser beam decreases when it propagates through the crystal before it reaches the focal point. For different crystals, the change of the power through propagation is different. Al_2O_3 and CaF_2 crystals are transparent at

800 nm, so the absorbance of the femtosecond laser would be small during the laser propagation process. Therefore, we think that when the original pulse energy is high enough, the femtosecond laser pulses would propagate and refocus in crystals[14]. As we know, GVD plays a key role in the change of the pulses energy in dispersive nonlinear mediums. Because of the GVD[18], the width of the pulse will increase as it propagates in samples, thus the peak power of the pulses will decrease at the same time. Usually we use the GVD parameter k_2 to describe the effect of GVD on the pulse width in the propagation process. For Al_2O_3, its k_2 is 400 fs^2/cm at 800 nm[19], which is much bigger than that of CaF_2 (-5 fs^2/cm)[20]. Therefore, in Al_2O_3, the effect of GVD on pulse shape is more obvious than that in CaF_2 when the propagation distance is the same. When laser beam focused at a deep position, the pulse will be broadened intensively in Al_2O_3, so peak power of the pulse will decrease at the same time.

Except the effect of GVD on the voids formation, the spherical aberration of the lens in different crystals also might influence the laser power. Because of the large refractive index mismatch at the interface between the air and the crystal, the blurring of the focal spot caused by the serious spherical aberration may also lower the peak power of the pulse[21]. The refractive index at 800 nm for Al_2O_3 is 1.7, which is bigger than that of CaF_2 (1.4), so the serious spherical aberration in Al_2O_3 is larger than that in CaF_2, and this leads to lower pulse peak power in Al_2O_3 compared with CaF_2 when the original laser power and the propagation distance are the same.

Voids formation requires high peak power to induce micro-explosive[16]. For Al_2O_3, pulse peak power will decrease intensively along the propagate direction, it cannot induce regular voids in deep positions, as shown in Fig. 3(a). However, for CaF_2, the effect of GVD and spherical aberration on the decrease of pulse peak power are not so obvious in deep position as that in Al_2O_3, and the peak energy will still be enough to induce voids. In addition, the damage threshold of CaF_2 is smaller than that of Al_2O_3[22]. So, in deeper position inside the crystal, voids still can be induced when pulse energy launched into the crystal is the same.

In summary, regular void array was self-formed by the irradiation of femtosecond laser pulses in Al_2O_3 crystal. It was found that compared with CaF_2 crystal, regular voids only could be fabricated when laser focused at a shallow position in Al_2O_3, and the formation of regular voids strongly depended on the depth of the laser focal point in different materials. The mechanism of these phenomena was also discussed.

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