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Generation of bubbles in glass by a femtosecond laser

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ABSTRACT

Permanent microscale bubbles with varied size and number density are induced in borosilicate glasses by adjusting the focusing depth (FD) of a tightly focused femtosecond laser. With continuously increasing of the focusing depth, the average size of generated bubbles experiences an increase–decrease process. However, the number density of generated bubbles experiences an opposite changing process compared to the change of the size. The possible mechanism for the bubble generation and changing with the focusing depth has been discussed.

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1. Introduction

The behavior of bubbles is not only ubiquitous but also complex in a multitude of fluid systems. It has been intensively studied on the experiment and theory and widely used in contemporary science and technology [1–5]. The discovery of single-bubble sonoluminescence gives bubble physics a boost [5]. Moreover, with the development of laser, arbitrarily shaped nonspherical bubbles can be generated and the trapping of bubbles in water is achieved [6,7]. The laser-induced bubbles in liquid can be utilized for the manipulation of carbon nanotubes, the generation of the shock wave etc. [8,9]. Obviously, the development of laser brings a promising prospect for the study and application of bubbles.

Recently, femtosecond lasers open up a new way to induce a microscale liquid region in solid materials, and subsequently investigate the behavior of bubbles in the liquid region under the extreme conditions. Long-lived bubbles in transparent materials have been induced by tightly focused femtosecond lasers [11,12]. Researchers can make these bubbles not only move upstream a few micrometers but also present anisotropic distribution controlled by laser writing direction [11,12]. However, the mechanism for the formation and evolution of bubbles induced by femtosecond lasers in solid materials is lacking and needs further investigation.

In this paper, we investigate the behavior of laser induced bubbles in glasses by adjusting the FD of a 250-kHz femtosecond laser. We

found that the average size of generated bubbles experiences an increase–decrease process, while the number density of the bubbles experiences an opposite changing process with the FD. We propose that the modulation of the laser fluence by the spherical aberration is responsible for the gradual change of the distribution, size, and number density of the generated bubbles with the FD.

2. Experimental

In our experiment, a commercial borosilicate glass (CAT No 7101. WHITE BRAND, Shanghai linglun Industrial Glass Co. Ltd.) with composition of 81SiO₂-13B₂O₃-4Na₂O-2Al₂O₃ (wt%) is used. A regeneratively amplified Ti:Sapphire laser emitting a train of 40 fs, 250 kHz mode-locked pulses is employed to micromachine the sample. The laser beam is tightly focused via a $100 \times$ oil-immersion objective (NA = 1.25) into the sample. The sample is moved at a constant speed of 20 µm/s along the x-axis, which is perpendicular to the laser propagation axis (z axis). The schematic graph of the laser writing has been presented in our previous work [13]. The beam waist diameter is estimated to be ~0.8 µm and the pulse energy is 2.4 µJ measured before the objective. The oil used in the experiment is 1-bromonaphthalene with the refractive index n = 1.658. After laser micromachining, the top-view of the induced bubbles in the laser-writing track is imaged by a transilluminated optical microscope. To investigate the location and size of bubbles in the modified zone, we do a side polish until the bubbles is exposed at the surface. An atomic force microscope (VEECO, Dimension 3100) and a field-emission microprobe analyzer (JEOL, JXA-8100) are used to determine the size of these voxels.

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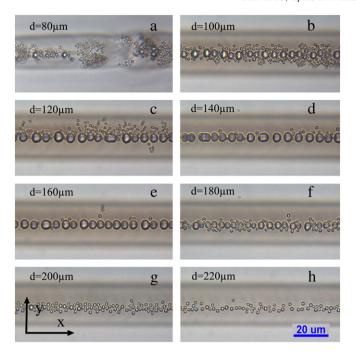


Fig. 1. The bubbles induced at different FDs. The pulse energy is fixed at 2.4 μ J and the scanning speed along x axis is 20 μ m/s. An 100× oil-immersion objective (NA = 1.25) is employed.

3. Results and discussion

Fig. 1 shows the optical images of bubbles in xy plane at varied FD (d), adjusted from 80 to 220 µm. Here, the FD is defined as the depth of the focal spot beneath the sample surface if the glass would have the same refractive index as the used oil. When $d = 80 \mu m$, numerous small bubbles appear irregularly within the laser-writing track. However, when d is in the range of 100 to 180 µm, it is observed that the induced bubbles basically consist of two parts: well aligned bigger bubbles at the center of the laser-writing track and random smaller bubbles around the bigger bubbles. It is can be seen that in Fig. 1(c), almost all the bubbles locate in one side of the bigger bubbles, leaving the other side with few smaller bubbles. We think that it is caused by the local impurity of the glass sample. When the FDs are 200 and 220 µm (the maximum working distance of the used objective), smaller bubbles with a homogeneous size are formed. We notice that the size of the bigger bubbles experiences an increasedecrease process with the FD, while the number density of the smaller bubbles has an opposite changing tendency. The results indicate that the distribution, size, and number density of the generated bubbles strongly depend on the FD. In the experiment, a linearly polarized femtosecond pulse with the direction of polarization along the x-axis is used. When a $\lambda/2$ wave plate is employed to change the laser polarization, we did not find difference in the distribution, size, and number density of the generated bubbles.

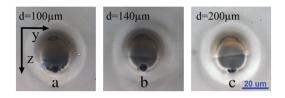


Fig. 2. The side view of bubbles at three different FDs (100, 140, 200 $\mu m)$ induced with the same laser parameters.

To investigate the size change of bubbles with the FD as well as the location of bubbles in the modified region, the observation from the side (yz plane) is essential. Fig. 2 shows the side view of bubbles taken by an optical microscope at d = 100, 140 and 200 μ m. It is interesting to observe that all of bubbles are at the bottom of the laser modified region, instead of the central region, suffered the highest intensity from the focused laser beam. In addition, it is also observed that the biggest one is at the lowest point. Fig. 3(a) shows the AFM images of 3-dimensional view of a hollow voxel, which originate from a polished bubble at $d = 160 \, \mu m$ in Fig. 1e. Sectional analysis yields a hollow voxel width of about 4.45 µm and a depth of about 2.18 µm, as shown in Fig. 3(b). The AFM image and data present a near hemispherical feature. The width of hollow voxels as a function of the FD is shown in Fig. 3(c). When the laser focal point moves from 80 to 220 µm, the width of these hollow voxels experiences a gradual rise and fall, and the maximum width is at the depth of 120 µm. Previous studies pointed out that micro-explosion was responsible for the formation of bubbles or voids in transparent materials when they were induced by tightly focused femtosecond lasers [10,14]. Those hollow voxels are submicrometer in diameter and located in the center of the laser irradiation area. In addition, exposure for one time can induce only one voxel in those studies. Obviously, the features in previous studies [10,14] are different from our results, which mean that the theory of micro-explosion is not suitable to the origin of bubbles in our experiment. So, we need to further clarify the formation process of the hollow voxels in our experiment.

The formation of bubbles in glasses is attributed to two typical properties of a 250-kHz femtosecond laser, higher peak power and repetition rate. Juodkazis et al. demonstrate that a single pulse with a few hundred nJ can induce a temperature increase of 0.5 MK and extreme pressures of 10 TPa at the focused volume [14]. Moreover, the heat accumulation effect will be present in dielectrics due to their relatively low thermal conductivity if the interval between the pulses is lower than about 10 µs [14,15]. In our case, when the sample is scanned by a 250-kHz femtosecond laser with energy of 2.4 µJ at a speed of 20 µm/s, each exposed region experiences tens of thousands of shots and the temperature around the focal volume can reach a few thousands of Kelvins by the accumulated heat, which results in localized melting. These unique conditions can induce intensive physical and chemical changes in the melted region, such as boiling, evaporation, the occurrence of plasma, bond breaking and reforming etc. Under such conditions, the nucleation of bubbles can be induced from several possible reasons. First, during the laser exposure in glasses, a phase explosion occurs in the focal region and results in a rapid transition from solid or liquid to a mixture of vapor, which is contributed to the homogeneous nucleation of bubbles [16]. Second, since the laser intensity exceeds 10¹⁴ W/cm² in our experiment, the plasma is induced in the focal region, which plays a role as condensation nucleus for the heterogeneous nucleation [10,14]. Third, as we use borosilicate glass samples, the liquid B₂O₃ is easily vaporized to undissolved gas in the superheated liquid, helping the heterogeneous nucleation [17]. Although the reason for the bubble nucleation is complex in this experiment, the three mechanisms may play a major role in the nucleation process.

Next, we discuss the dynamics of the formation of bubbles. During the bubbles generation, they suffer the optical radiation pressure (ORP) from the incident laser, and gain the initial momentum along the laser propagation axis [18,19]. Then, the radial shock wave drives the bubbles away from the laser-irradiated region [14]. On the other hand, with the increase of laser shots, the laser-induced temperature difference results in a radial pressure gradient driving bubbles away from the center of the melted region [15]. As a result, the abovementioned three actions collectively overcome buoyancy and the viscous force, and drive the bubbles to move from the focal center to the bottom of the melted region. It is clear that this moving is from high-temperature region to low-temperature region. So, during the motion, the pressure inside the bubbles is bigger than outside the bubbles, which makes the bubbles

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