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Standing electron plasma wave mechanism of void array formation inside glass by femtosecond laser irradiation

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ABSTRACT We investigate the mechanism of formation of periodic void arrays inside fused silica and BK7 glass irradiated by a tightly focused femtosecond (fs) laser beam. Our results show that the period of each void array is not uniform along the laser propagation direction, and the average period of the void array decreases with increasing pulse number and pulse energy. We propose a mechanism in which a standing electron plasma wave created by the interference of a fs-laser-driven electron wave and its reflected wave is responsible for the formation of the periodic void arrays.

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1 Introduction

The interaction of ultra-short laser pulses with transparent dielectrics has attracted much attention since the advent of the femtosecond (fs) laser [1–3], because it can lead to many interesting phenomena, such as filamentation [4, 5], birefringence [6], formation of a periodic void array [7–9], and so on. Among these phenomena, the formation of a periodic void array in the direction of laser propagation is one of the most puzzling phenomena because of the lack of a good physical understanding. For fs-laser-induced filamentation in glasses, it has been found that the dynamic competition between Kerr self-focusing and the plasma-induced defocusing effect can form multiple foci when focusing a fs laser pulse into the dielectrics. The distance between two adjunct foci is usually on the order of tens of microns [5]. In contrast, the period of the fs-laser-induced void array is only on the order of a few microns or even sub-micron [7–9]. In this paper, we systematically investigate the fs-laser-induced void arrays in fused silica and BK7 glass. In particular, we focus our study on the dependence of the period of the void array on the laser parameters, including pulse energy, pulse number, and focal depth. Based on the experimental results, we propose a physical mechanism by which the periodic void arrays can form owing to the interference of a fs-laser-driven electron plasma wave and its reflected wave.

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2 Experimental

Shown in Fig. 1 is a schematic diagram of the experimental setup. A regeneratively amplified Ti:sapphire laser at a central wavelength of 800 nm that emits 120-fs, 1-kHz, mode-locked pulses was used in our study. The glass samples were placed on a three-dimensional translation stage. The size of both the fused silica and BK7 glass is 10 mm × 5 mm × 3 mm with four sides optically polished with a roughness less than 10 nm. We focused the fs laser pulses into fused silica and BK7 glass to generate structural changes using a ×100 objective lens (NA = 0.9), and the measured waist radius of the laser beam in the focal region is approximately 1.0 μm. The laser pulse energy could be varied from 10 μJ to 40 μJ continuously by using a neutral filter. An electronic shutter was used to control the pulse number except for the single pulse, which was obtained by triggering the fs laser system itself. We captured in situ the top-view optical images of damage or voids with a transilluminated optical microscope and a charge-coupled device (CCD). After irradiation, the side view of the interior microstructure was obtained using the optical microscope.

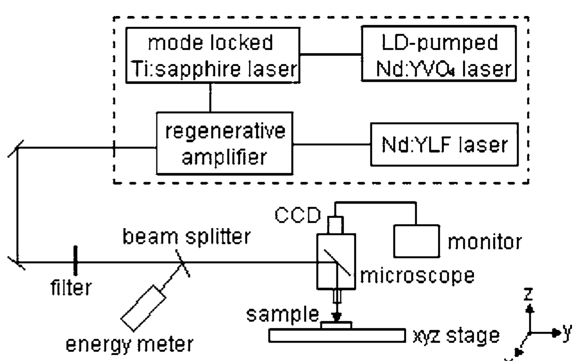

FIGURE 1 The schematic diagram of the experimental setup

3 Results and discussion

We first investigated the influences of the number of incident pulses and the focal depth on the generated void array. The geometrical depths chosen in the experiment were 200 μm, 400 μm, and 700 μm below the sample surface. Figure 2a shows typical void arrays generated by the irradiation

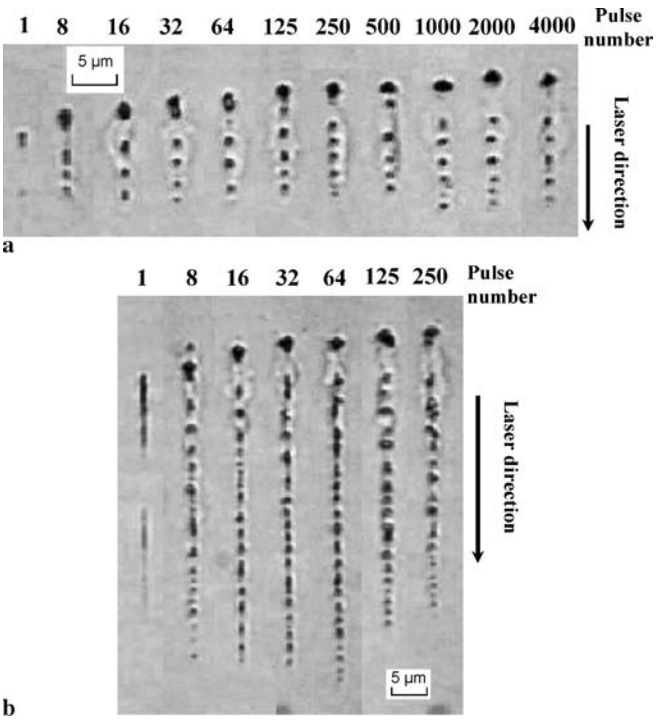

FIGURE 2 Side-view micrographs of void arrays in fused silica when irradiated by fs laser beam focused by a ×100 objective lens. (**a**) 13.9 µJ/pulse, focal depth: 400 µm; (**b**) 29.6 µJ/pulse, focal depth: 700 µm

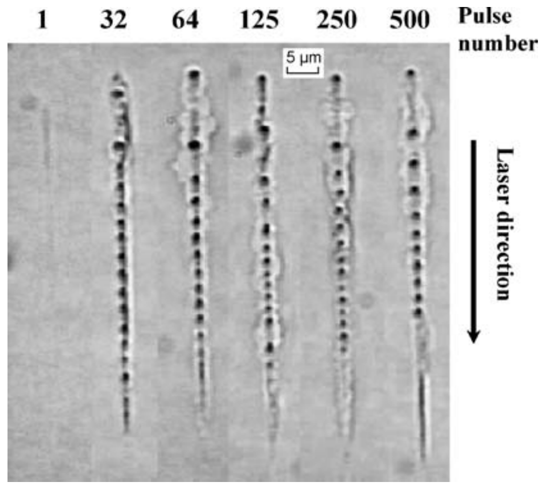

FIGURE 3 Side-view micrographs of void arrays in BK7 glass when irradiated by fs laser beam focused by a ×100 objective lens. 26.1 µJ/pulse, focal depth: 400 µm

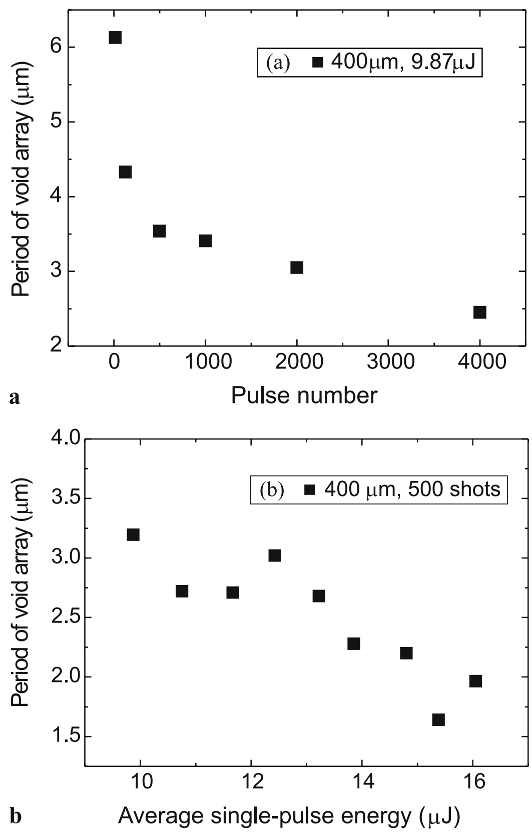

FIGURE 4 Dependence of the period of void array generated in fused silica on (**a**) the laser pulse number with a fixed pulse energy of 9.87 µJ, and on (**b**) the laser pulse energy with a fixed pulse number of 500 shots. The focal depth is 400 µm in both cases (**a**) and (**b**)

of the focused fs laser beam in fused silica at a geometrical focal depth of 400 µm. One can see that when a single laser pulse was used, a void array could not be formed. Instead, single fs laser pulses tend to induce dashed-line-shaped filamentation along the laser propagation direction. When multiple fs laser pulses were used for irradiation, void arrays were readily formed inside the glasses, whereas the periods of the arrays were not uniform within the entire void array structures. Namely, the period of each void array at the two ends is longer than that in the central area. Figure 2b presents the void arrays formed by focusing fs laser pulses 700 µm beneath the surface of the fused silica. In this case, both the size and the period of the void arrays were reduced. The non-uniform periods of the void arrays were also observed in BK7 glass, as shown in Fig. 3. In fact, this non-uniform feature of the void arrays has also been reported in [7].

In order to understand the mechanism of the formation of the void array, we then made a statistical analysis of the period of the void array using Fourier transformation (FT). We defined that the average period of each void array is determined by the position of the highest peak in its Fourier spectrum. We then plotted the period of the void array as functions of the pulse numbers and pulse energies. As can be seen in Fig. 4a and b, the period of the void array decreases with increasing pulse number and pulse energy. Although the relationship shown in Fig. 4 was obtained in fused silica for the focal depth of 400 µm, a similar relationship was also found with other focal depths and in BK7 glass. Additionally, we also found that the shortest period (~ 2 µm) observed in these experiments was almost independent of the focal depth.

Since the ultra-short characteristic of the fs pulses makes their intensity easily beyond 10^{14} W/cm^2, the propagation of the fs pulses in the dielectric is highly nonlinear, and several nonlinear effects begin to take effect, such as the self-focusing effect resulting from Kerr nonlinearity and the defocusing effect caused by the generated plasma. Some researchers indeed observed the multiple refocusing phenomena of the laser pulses by capturing the plasma fluorescence and have already shown convincing numerical simulation results [5]. However, the periods of the multi-foci in those works are usually significantly longer than the periods of the void arrays. The periods of the void arrays mentioned here are very short with the minimum ones approaching $\sim 2\,\mu$m; thus, it is hard to be understood in the physical picture of repeated self-focusing and defocusing.

In past years, the periodic patterns on the surfaces of many materials produced by laser irradiation have been intensively investigated and frequently reported [10]. The periods of the surface ripples are usually on the order of the incident laser wavelengths. Moreover, it is found that the formation of the ripples can be easier when the surfaces are irradiated using multi-pulses instead of using a single pulse. The laser-excited surface-plasma-wave scattering was one of the mechanisms used to explain this phenomenon [11]. In this paper, we suggest that the laser-excited plasma wave can generate not only the periodic microstructures on the surface, but also inside the materials such as periodic void arrays in silica glass or BK7 glass. When a 120-fs laser pulse of $10\,\mu$J (which is the minimal pulse energy used in our experiments) is focused into a focal spot size of $\sim 1\,\mu$m inside glasses, the peak intensity reaches the 10^{16} W/cm^2 level. Under such high peak intensity, free electrons are easily generated inside glasses by a series of nonlinear optical effects including multi-photon absorption, tunnel ionization, and avalanche ionization [12]. The free electrons generated by the leading part of the fs laser pulse then interact with the following part of the fs laser pulse, forming an electron plasma wave (EPW). The physics of EPW excitation has been intensively investigated in the research field of laser wake field acceleration, in which the huge electric field existing in an EPW is used for accelerating electrons to high energy [13]. Three main schemes of EPW production have been proposed, including laser beating wave, laser wake field, and laser self-resonant wake field. In the former two schemes, resonant excitation conditions are required so that the fs laser pulses used for EPW production must be either of the pulse duration close to the EPW wavelength or of the temporal profile periodically modulated with a period close to the EPW wavelength. Since our fs laser pulse duration is approximately 120 fs, such resonant excitation conditions cannot be satisfied. The third scheme (the laser self-resonant wake field scheme) does not require a specific temporal profile of the fs driven pulse. Instead, a relativistic intensity beyond 10^{15} W/cm^2 is needed to excite a large-amplitude EPW [14]. Compared with our experimental conditions, the relativistic intensity required for a self-resonant wake field can readily be satisfied. In this manner, the relativistic self-focusing and the Raman instability modulate the laser pulse temporal envelope into a train of pulses verifying the resonant condition $\omega_{\mathrm{pl}}\tau \approx 1$, where ω_{pl} and τ are the electron plasma frequency and the pulse duration, respectively. These sub-pulses excite an EPW to large amplitude [13].

It should be noted that the EPW is a traveling wave; thus, it alone cannot form a stationary and periodic electron density modulation. However, when the laser pulses propagate deeply into the glasses, the losses of the pulse energies combined with the diffraction will reduce the laser intensity, leading to the termination of EPW production. Therefore, after a certain propagation distance, the free electron density generated by the fs laser pulse will abruptly drop, forming a large density gradient. It is well known that when a plasma wave propagates into a region with a large plasma density gradient, it will be reflected and then interfere with the forward-traveling wave, creating a standing wave pattern [15]. This standing EPW will lead to the stationary density distribution [16]. Since the optical breakdown in glass is sensitive to the free electron density, the standing EPW would induce periodic microexplosions, which result in the periodic void array structure. This physical picture is also supported by the observation in [7] that aligned void structures can more easily be generated when the filamentation path reaches the bottom surface of the glass sample where an abrupt decrease of electron density naturally occurs. However, in our experiments, we found that void arrays could also be formed inside glasses without the filamentation path approaching the bottom of the glass samples.

The above-mentioned mechanism allows us to evaluate the electron densities generated by fs laser pulses because the free electron density determines the wavelength of the EPW and, consequently, the period of the generated void array. In this scenario, the halved wavelength of the EPW should be equal to the period of the void array. The wavelength of the EPW can be expressed as $\lambda = 2\pi\nu/\omega_{\mathrm{pl}}$, where $\omega_{\mathrm{pl}} = \sqrt{q_{\mathrm{e}}^2 n_0/\varepsilon_0 m_{\mathrm{e}}}$, ν is the light velocity in the dielectric, m_{e} is the mass of an electron, n_0 is the intensity-dependent plasma density, q_{e} is the electric charge of an electron, and ε_0 is the vacuum dielectric constant [17]. It is apparent that the wavelength of the EPW is inversely proportional to the electron density; thus, it can be understood that the intervals of the adjunct voids at the two ends of the void arrays are always longer than the period of the array in its central area, because the laser peak intensity is the highest in the central area. The high peak intensity in turn generates high free electron density to form a short-period void array. Based on the analysis, the electron density required for forming an array of a period of $2\,\mu$m can be estimated as $\sim 10^{19}$ cm^3. This electron density level is consistent with the previously reported results in fs laser irradiation of dielectrics [2, 12].

The results in Fig. 4 can now be understood straightforwardly. Since the periods of the void arrays are determined by the electron densities generated by fs laser pulses, the higher the electron density, the shorter the array period. This feature is well reproduced in Fig. 4a and b. It is known that for glasses repeatedly exposed to multiple fs laser pulses, defects can be created by the early pulses which lower the ionization threshold for the later pulses. Thus, with the increased pulse numbers, the ionization process in glass can be enhanced, leading to increased electron density which leads to a decreased array period. On the other hand, increasing pulse energy can also raise the electron density. Thus, in both cases, the period of the void array can be shortened following the trends displayed in Fig. 4a and b.

4 Conclusions

To conclude, we have experimentally investigated how the period of the fs-laser-induced void array inside glass depends on the irradiation conditions. A new model based on the standing plasma wave scenario was proposed to explain the formation of the void array. Reasonable agreement between the experimental results and the theoretical analysis was obtained. Currently, plasma physics has not yet been frequently employed to explore the interaction of dielectrics with fs laser pulses, in which the electron density can easily reach a level as high as $\sim 10^{20}\,\mathrm{cm}^3$ and the laser peak intensity can exceed $10^{16}\,\mathrm{W/cm}^2$. Our research shows that the microstructures left in the glasses exposed to fs laser pulses can provide an easy and effective means for diagnosing plasma parameters, shedding a new light on the high-temperature, high-density plasma physics.

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REFERENCES

1 S. Juodkazis, K. Nishimura, S. Tanaka, H. Misawa, E.G. Gamaly, B. Luther-Davies, L. Hallo, P. Nicolai, V.T. Tikhonchuk, Phys. Rev. Lett. **96**, 166 101 (2006)
2 N.T. Nguyen, A. Saliminia, W. Liu, S.L. Chin, R. Vallée, Opt. Lett. **28**, 1591 (2003)
3 K. Miura, J. Qiu, H. Inouye, T. Mitsuyu, Appl. Phys. Lett. **71**, 3329 (1997)
4 L. Sudrie, A. Couairon, M. Franco, B. Lamouroux, B. Prade, S. Tzortzakis, A. Mysyrowicz, Phys. Rev. Lett. **89**, 186 601 (2002)
5 Z. Wu, H. Jiang, L. Luo, H. Guo, H. Yang, Q. Gong, Opt. Lett. **27**, 448 (2002)
6 E. Bricchi, B.G. Klappauf, P.G. Kazansky, Opt. Lett. **29**, 119 (2004)
7 S. Kanehira, J. Si, J. Qiu, K. Fujita, K. Hirao, Nano Lett. **5**, 1591 (2005)
8 E. Toratani, M. Kamata, M. Obara, Appl. Phys. Lett. **87**, 171 103 (2005)
9 D. Ashkenasi, H. Varel, A. Rosenfeld, S. Henz, J. Herrmann, E.E.B. Campbell, Appl. Phys. Lett. **72**, 1442 (1998)
10 D. Ashkenasi, A. Rosenfeld, H. Varel, M. Wähmer, E.E.B. Campbell, Appl. Surf. Sci. **120**, 65 (1997)
11 S.R.J. Brueck, D.J. Ehrlich, Phys. Rev. Lett. **48**, 1678 (1982)
12 C.B. Schaffer, A. Brodeur, E. Mazur, Meas. Sci. Technol. **12**, 1784 (2001)
13 J.R. Marquès, J.P. Geindre, F. Amiranoff, P. Audebert, J.C. Gauthier, A. Antonetti, G. Grillon, Phys. Rev. Lett. **76**, 3566 (1996)
14 S. Tzortzakis, G. Méchain, G. Patalano, Y.-B. André, B. Prade, M. Franco, A. Mysyrowicz, J.-M. Munier, M. Gheudin, G. Beaudin, P. Encrenaz, Opt. Lett. **27**, 1944 (2002)
15 M.D. McFarland, A.Y. Wong, Phys. Rev. Lett. **80**, 5540 (1998)
16 P. Michelsen, H.L. Pécseli, J. Juul Rasmussen, N. Sato, Phys. Fluids **20**, 1094 (1977)
17 W. Hoyer, J.V. Moloney, E.M. Wright, M. Kira, S.W. Koch, J. Phys. **11**, 153 (2005)