Pump-probe imaging of the fs-ps-ns dynamics during femtosecond laser Bessel beam drilling in PMMA

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Abstract: A pump-probe shadowgraph imaging technique was used to reveal the femtosecond-picosecond-nanosecond multitimescale fundamentals of high-quality, high-aspect-ratio (up to 287:1) microhole drilling in poly-methyl-meth-acrylate (PMMA) by a single-shot femtosecond laser Bessel beam. The propagation of Bessel beam in PMMA (at 1.98 × 10^8 m/s) and it induced cylindrical pressure wave expansion (at 3000-3950 m/s in radius) were observed during drilling processes. Also, it was unexpectedly found that the expansion of the cylindrical pressure wave in PMMA showed a linear relation with time and was insensitive to the laser energy fluctuation, quite different from the case in air. It was assumed that the energy insensitivity was due to the anisotropy of wave expansion in PMMA and the ambient air.

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1. Introduction

The femtosecond (fs) laser has been considered to be a promising tool for drilling microholes in almost any kind of material [1–4] due to its extreme peak intensities and short interaction timescales. However, debris redeposition and tapering effects preclude high-quality, high-aspect-ratio microhole fabrication using Gaussian beams [2, 4], especially for microholes with a diameter below 5 μm and an aspect ratio exceeding 100.

Bessel beams provide a good solution for drilling high aspect ratio microholes by spatial beam shaping [5–8]. In our previous work, we reported that a fs laser with a transverse intensity profile of Bessel beam can generate small diameter, high-aspect-ratio microholes without taper in poly-methyl-meth-acrylate (PMMA) [9]. By optimizing the pulse energy and focal depth position, long and repeatable microholes with diameters of 1.4–2.1 μm and aspect ratios exceeding 460 were fabricated. This is a benefit of the near constant intensity distribution along the propagation axis of the Bessel beam, whose maximum intensity is strongly localized in the beam center over a distance that exceeds the Rayleigh range of Gaussian beams by orders of magnitude.

Generally, drilling with a conventional Gaussian beam is usually an accumulation of multiple pulse ablation due to the limited ablation depth of a single laser pulse [1–4], while for Bessel beam, only one single fs laser pulse is capable of drilling a deep microhole in PMMA [5, 9]. The difference mainly reflects in the energy deposition and transfer dynamics that occurs within femtosecond-to-nanosecond time scale [10]. Recent years, by utilizing various ultrafast imaging techniques, the time-resolved dynamics of plasma generation, shockwave propagation, and material ejection during fs laser ablation have been studied extensively [11–13]. For example, Zeng et al. [14] investigated the propagation of laser-induced shockwaves from ablation inside of cavities to simulate the situation of multi-pulse drilling. Papazoglou et al. [15] studied the laser-induced ultrafast electron dynamics and subsequent material changes in PMMA using inline holographic microscopy, and they found that the sub-ps photochemical processes play an important role in the generation of long void structure in PMMA. These time-resolved studies gave an insight into the mechanism of fs laser drilling with traditional Gaussian beams. But to date, the fundamentals of fs laser Bessel beam drilling in PMMA have not yet been revealed.

In this paper, we report on an experimental investigation into the energy transfer and diffusion dynamics at the early 30 ns during single-shot fs laser Bessel beam drilling in PMMA. An axicon combined with a 4-F system was used to generate a confined Bessel beam to drill high-quality, high-aspect-ratio microholes in PMMA; and time-resolved images of the drilling process were recorded by the pump-probe shadowgraph imaging technique from the side view. The propagation of a Bessel beam in PMMA and it induced cylindrical pressure...
wave expansion were observed during drilling processes. The influence of laser energy and focal depth on the pressure wave expansion in PMMA and in air was also studied.

2. Experiment

The experimental setup is shown in Fig. 1, which is a typical two-wavelength pump-probe shadowgraph imaging system. The pump pulse and the probe pulse were divided from a single 50 fs, 800 nm laser pulse, which was generated by a Ti:sapphire fs laser amplifying system (Spitfire, Spectral Physics Inc.). The pump pulse was converted from a Gaussian beam profile to a Bessel beam by passing through an axicon lens with a base angle of 2°. The diameter of the pump laser beam was confined to 6 mm by an aperture located before the axicon to confine the Bessel beam to a proper length (180 mm). A 4F system composed of a convex lens (f = 200 mm) and a 20 × objective lens (NA = 0.42, Olympus) was employed to focus the Bessel beam into the PMMA sample (10 × 20 × 1 mm, Hefei Kejing) at a normal incidence with respect to the sample surface. The probe pulse was frequency doubled by a beta barium borate (BBO) crystal with Type I phase matching, and then directed transversely through the Bessel beam focus area in PMMA. The sample was vertically fixed on a translation stage with the biggest surface faced orthogonally to the probe beam. The top surface of the sample was polished with a polisher. The transmitted shadowgraph image of the fabricated area was collected by a tube lens and recorded by a charge-coupled device (CCD). A 400 nm bandpass filter was used to block the residual 800 nm wavelength of the probe pulse and the fluorescence radiation generated during laser ablation from entering the CCD.

Both the laser source and the CCD were operated in external gating mode, and a homemade time controller was used to synchronize the laser pulse output and the CCD exposure. For each experiment at a specific time delay, the sample was shifted to a fresh point by the translation stage; and then three images were taken before, during and after the pump pulse irradiation, respectively; recording the background, the transient signal and the final result, respectively. Both the background and the final result images were taken with the pump pulse blocked. Only one pump pulse was applied when taking the signal image. In order to get a better signal-to-noise ratio, the background image was subtracted from the signal and result images (unless otherwise noted, all the images in this paper were background corrected). The time delay between the pump and the probe pulse was controlled by an optical delay line in the probe light path. The zero time delay point was defined as the time when the front of the pump pulse impacted the surface of the PMMA sample.
3. Results and Discussion

Figure 2 shows a series of time-resolved shadowgraph images of the fs laser Bessel beam drilling process in PMMA, with a pulse energy of 23 µJ. The left part of the dashed line indicates the air, while the right part is the PMMA target. The interface is not so clear due to the background subtraction. The Bessel beam (pump laser) impinges on the target from the left.

It can be seen from the images that as soon as the fs laser Bessel beam entered into the PMMA target [Fig. 2(a)], a small dark strip appeared close to the PMMA surface that resulted from electron excitation by the rising edge of the fs laser pulse. At greater delay, the plasma channel grew longer [Figs. 2(b)-2(g)], reflecting the propagation of the laser field in the PMMA. In theory, the propagation velocity of the Bessel beam focus in PMMA can be derived from geometrical optics [16] as:

\[ v = \frac{c}{n \cos \beta} \]  

where \( c \) is the speed of light in vacuum, \( n \) is the refractive index of PMMA, and \( \beta \) is the cross-angle of the Bessel beam in PMMA. By assigning \( n = 1.54 \) and \( \beta = 14.4^\circ \), the propagation velocity of the Bessel beam focus in PMMA was theoretically calculated to be \( 2.01 \times 10^8 \) m/s. Propagation of the plasma front inside the PMMA at different laser energies is plotted in Fig. 3. The extension of the plasma channel showed a linear relation with time and little dependence on laser energy. The extension speed of the plasma channel was measured to be approximately \( 1.98 \times 10^8 \) m/s, which agreed well with the theoretically calculated propagation speed of the Bessel focus in PMMA. This further confirmed that the extension of the plasma channel was resulted from the successive ionization of PMMA due to the propagation of Bessel beam focus in the material. Note that this is a superluminal propagation process (\( 1/\cos \beta \) times the light speed in PMMA) due to the special focus mechanism of the Bessel beam [16].
Fig. 2. Time-resolved images during fs laser Bessel beam drilling in PMMA (see also in Visualization 1). The left part of the dashed line indicates the air, while the right part is the PMMA target. A single 23 μJ laser pulse with a diameter of 6 mm was converted into a Bessel beam profile by an axicon with a base angle of 2° and then demagnified by a factor of 22 through a 4-F system for drilling the hole.

At 1.6 ps, the energy transition from the laser pulse into the PMMA material was completed; meanwhile, the plasma channel reached a maximum length of ~318 μm and stopped growing longer thereafter. This length was shorter than the full Bessel beam focus length in PMMA (~460 μm) because the Bessel beam was not fully focused into the material. By comparison, it was observed that the diameter (1.6 μm) and length (323 μm) of the final fabricated microhole [Fig. 2(q)] coincided well with the plasma channel, which indicated that the heat diffusion effect of fs laser ablation was negligible.

The plasma channel showed little change from 2 ps to 100 ps [Figs. 2(i)-2(j)]. But from 500 ps, a cylindrical wave separated from the plasma channel and gradually expanded outward along the radial direction [Figs. 2(k)-2(p)]. Meanwhile, a hemispherical wave with a tip was also generated in air around the entrance of the microhole [Figs. 2(m)-2(p)]. The pressure waves in air and in PMMA were not in the same focal plane of the imaging system due to the different refractive index of PMMA and ambient air. Thus, there were some fringes around the edge of the pressure wave in air. The tip in the pressure wave in air was due to the air ionization which changed the geometry of the hot center [13]. The “silent period” from 2 ps to 100 ps was considered to be the electron-ion energy transfer time [17]. When the ions obtained enough energy from the energetic electrons, the trapped hot plasma induced an extreme high pressure and temperature in the focal area, which then actuated an explosive expansion of the material from the hot center into the surroundings with a speed exceeding the speed of sound. The pressure wave produced density variations in the PMMA that translated to refractive index variations, providing the contrast seen in the shadowgraph image [18].
Fig. 3. The extension of the plasma channel as a function of time. The experimental data were measured with five different pulse energies. The straight line is the theoretical propagation of the Bessel beam focus in PMMA.

The expansion of the cylindrical wave at different laser energies is plotted in Fig. 4 as a function of time. Unexpectedly, we found that the experimental data measured at different laser energies superposed with each other. When the laser energy increased from 10 μJ to 60 μJ, there was only a slight increase of less than 10% in diameter for time delays shorter than 5 ns. For larger time delays, the diameter of the cylindrical wave was almost uninfluenced by the laser energy, reflecting an energy insensitivity of the wave expansion process. Furthermore, the cylindrical wave was found to propagate linearly with time at all energies. The expansion speed in radius was measured to be 3000-3950 m/s, varied from sample to sample due to the property difference. This speed was comparable to the acoustic velocity in PMMA (2630 m/s) [19], and also in good agreement with the measured result of Papazoglou et al. (2930 m/s) [16]. It has been widely reported that the expansion of laser-induced shockwave in air [12, 14, 20, 21] usually conforms to the Sedov’s point source blast wave theory [22], i.e. the expansion speed not only depends on the input energy but also exponentially decays with time. However, in our experiment, both the energy insensitivity and linear expansion character make the cylindrical wave quite different from the widely reported shockwave in air. This is mainly due to that the cylindrical pressure wave in our experiment is induced by a bulk ablation, which is quite different from the widely reported situation of surface ablation.

Fig. 4. The diameter expansion of the pressure wave in material as a function of time at different laser energies.

Figure 5 shows a series of shadowgraph images taken at different laser energies at a fixed time delay of 10 ns. Although the diameter of the cylindrical wave doesn’t change with the laser energy, there is an evident increase in diameter of the central plasma channel when at
higher laser energy, indicating an increased laser energy absorption at higher laser energy. This in some extent precludes the probability of saturation effect in laser energy absorption [23] that may lead to the energy insensitivity of the cylindrical wave. In addition, in contrast to the cylindrical wave in PMMA, the expansion of the pressure wave in air depends greatly on the laser energy, it gets larger when under higher pulse energies. It may be assumed that the energy is easier to release through the axis direction towards the ambient air than the lateral direction in bulk PMMA, which explains the energy insensitivity of the cylindrical wave in PMMA.

![Fig. 5. The influence of laser energy on the expansion of pressure wave in air and in PMMA. The femtosecond laser Bessel beam irradiated the PMMA from the top. All of the images were taken at the same time delay of 10 ns with the same focal depth of F = 370 μm (the value of F indicates a relative distance between the last focusing lens and the surface of the PMMA sample).](image)

Although pressure waves appeared in both the lateral and the entrance of the hole, there was no pressure wave in the other end of the hole, i.e., the bottom of the hole. This indicated that the pressure wave expansion in air was easier than in PMMA. In order to verify whether a pressure wave would appear in another end of the hole that was located in PMMA, the shadowgraph images taken at different focal depths are compared in Fig. 6, at a fixed time delay of 30 ns. The results showed that as long as one end of the hole was in contact with the air, the pressure wave only appeared in the air end; and when the Bessel beam was fully focused into the interior of the PMMA [Fig. 6(a)], no pressure waves were observed at both ends of the hole. This indicates that both the expansion in air and the lateral expansion in PMMA were easier than the longitudinal expansion in PMMA and that the expansion of the pressure wave in PMMA was a two-dimensional cylindrical expansion process. In addition, as the focal depth into the PMMA decreased, the tip in the pressure wave in air grew longer due to a longer ionized area in air. The diameter of the pressure wave in PMMA remained almost unchanged due to the near constant intensity distribution along the propagation axis of the Bessel beam focus. However, a slight increase in the width of pressure wave in air was observed. This indicated that the pressure wave in air tends to be an isotropic three-dimensional spherical expansion rather than a two-dimensional cylindrical expansion like that of the pressure wave in PMMA.
4. Conclusion

In summary, time-resolved images during drilling high-quality, high-aspect-ratio (up to 287:1) microholes in PMMA by a single-shot femtosecond laser Bessel beam were obtained over the early 30 ns by using the pump-probe shadowgraph imaging technique. The propagation of Bessel beam in PMMA and it induced cylindrical pressure wave expansion were observed during drilling processes. It was unexpectedly found that the expansion of the pressure wave in PMMA showed a linear relation with time and was insensitive to the fluctuation of laser energy. This is quite different from the expansion of shockwave in air, reflecting an energy relaxation difference in surface and bulk material. It was assumed that the energy insensitivity of the pressure wave in PMMA was due to the anisotropy of wave expansion in PMMA and in air.

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