Time-Resolved Shadowgraphs of Material Ejection in Intense Femtosecond Laser Ablation of Aluminum

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The dynamic process of intense 50 fs laser ablation of aluminum is investigated by ultrafast time-resolved microscopy. A stripe pattern preceding phase explosion is clearly seen in the shadowgraph of 1 ns time delay. Intermittent material ejections are observed within the ejected plume after 2.5 and 7 ns time delay, respectively, which may be attributed to the material response to the generation of an extremely strong thermoelastic wave. Similar processes are also recorded in the ablation of silicon and glass samples, except for the glass samples, the intermittent material ejections are not found.

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The great development of ultrashort pulse laser technologies during the last decade has not only made the ultrafast laser a significant research tool in a broad area of scientific fields such as strong field quantum control [1], ultrafast diagnostics of chemical reactions [2], etc., but also has promoted many important applications in micro- and nanofabrications [3], sputtering [4], and even laser propulsion [5]. Although laser ablation with longer or nanosecond pulses has been extensively investigated over the past decades [6,7], picosecond and femtosecond laser ablation remains an attractive and relatively new phenomenon in the laser-matter interaction. The mechanisms of ultrashort pulse laser ablation are continuously under intense investigation for both technological and scientific interests.

Various theories such as phase explosion [8], Coulomb explosion [9], and the effect of a thermoelastic wave [10] are proposed to explain the fundamental mechanisms of ultrafast laser ablation. In particular, in the case of intense femtosecond laser ablation of solids, phase explosion occurs when the target surface is rapidly heated above the spinode or the critical temperature of the target and then adiabatically cools into the metastable region [8]. Coulomb explosion caused by the localized charge imbalance can take place when dielectric or semiconductor materials are irradiated by intense femtosecond laser pulses. Moreover, immediately after the target is heated by an intense ultrafast laser pulse, an extremely strong thermoelastic wave may be formed due to abrupt thermal expansion of the ablated region. Such a thermoelastic shock wave propagating into the target interior may cause spallation [11] or fragmentation [12] and eventually lead to material ejection. Theoretical calculations [12] also show that ultrafast laser ablation often does not involve only one kind of mechanism and is, de facto, a composite physical process depending on both laser parameters and target properties. Further studies are needed to identify what actually happens under different experimental conditions.

In this Letter, the novel dynamic process of femtosecond laser ablation of aluminum with energy fluence well above the ablation threshold is revealed. We choose aluminum as the primary sample of interest because of its well known properties. Shadowgraphs up to 10 ns time delay after a 50 fs laser pulse strikes the sample surface are recorded with a femtosecond time-resolved Mach-Zehnder interferometer. Two distinct dynamic phenomena, one characterized by the formation of the stripe pattern and the other best described as intermittent material ejections, are clearly identified and appear to be ubiquitous in the relatively intense ablation regime. The two dynamic phenomena are also found for silicon samples. But for glass samples, no obvious intermittent ejections are recorded. It is believed that the stripe pattern is actually the prelude of phase explosion, and the intermittent material ejections are the unique result of material response to an extremely intense thermoelastic wave. To the best of our knowledge, the shadowgraphs we recorded provide for the first time a direct dynamic picture of the interesting hybrid ablation process within the ps-to-ns time window associated with femtosecond laser–matter interaction in a much intuitional way. Discussions on the thermal and nonthermal characteristics of these recorded hybrid ablation processes due to the intense ultrafast laser heating will be also presented.

Our experimental setup is shown in Fig. 1. A Ti:sapphire femtosecond laser amplifier (HP-Spitfire, Spectra-Physics Inc.) is used to generate 50 fs laser pulses with a central wavelength of 800 nm. The pump and probe pulses are generated using a 50:50 beam splitter so that each pulse has a pulse energy of ~0.2 mJ. The probe beam is frequency doubled with a 2 mm thick BBO crystal with Type I phase matching. The aluminum target (also a silicon or a glass target) is mounted on a five-axis combo translation stage. In order to avoid the unwanted air ionization, the sample is placed slightly before the focal point as illustrated in the dashed block of Fig. 1. Two optical delay lines are used for achieving the specific time delays between the pump and probe pulses. The estimated energy fluence of the focused pump beam at the sample surface is about 40 J/cm^2. A 10-bit digital CCD camera (Dalsa 4M15) synchronized with

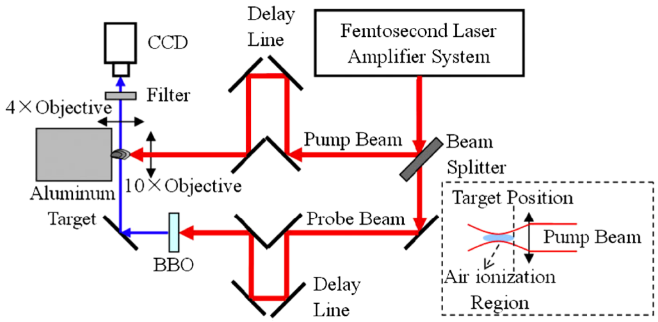

FIG. 1 (color online). Experimental setup.

the femtosecond laser system is used to record the time-resolved shadowgraphs of material ejection after the target is ablated with a single 50 fs laser pulse. Each time after a shadowgraph is recorded at a specific time delay, the target is shifted to a new position with a fresh area before the next shadowgraph is taken. A 400 nm bandpass filter together with some neutral density filters is used to block the residual 800 nm light in the probe beam and the fluorescence generated during the ablation from entering the CCD camera.

Figure 2 shows a sequence of time-resolved shadowgraphs of material ejection after a single 50 fs laser pulse of 40 J/cm^2 impinges on the aluminum target. Shadow of the ejected material can be clearly seen in Fig. 2 for the time delay of 100 ps. It is interesting to notice that starting from the shadowgraph of 500 ps, there are more and more bright regions in the shadow of the ejected material, and as evidenced in the shadowgraph of 700 ps or larger, these

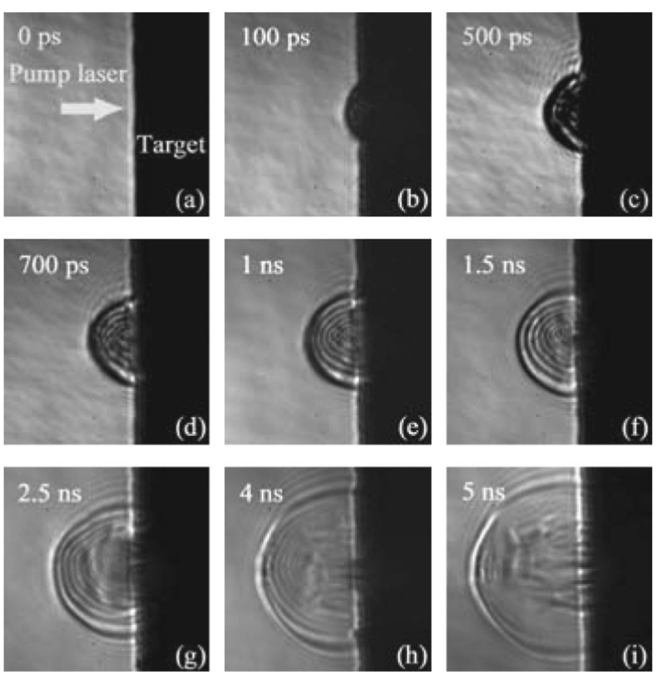

FIG. 2. Time-resolved shadowgraphs of material ejection at the indicated time delays after an aluminum target is ablated by a 50 fs laser pulse of 40 J/cm^2. Frame size: 170×170 μm.

bright regions evolve into a series of nearly concentric and semicircular stripes. This stripe pattern becomes clearer for time delays of 1 ns and 1.5 ns. For larger time delays, the contrast of the stripe pattern gradually decreases, and the material ejection near the target surface can be seen in the shadowgraph of 2.5 ns or larger.

The dynamic process revealed in the shadowgraphs in Fig. 2 may be best explained with phase explosion [8]. The formation of the stripe pattern at the time delay of near or above 700 ps in fact implies that phase explosion is just about to occur. According to our numerical estimation based on the two-temperature model [13], when the intense 50 fs laser pulse with the same laser parameters as those used in our experiment hits the aluminum sample, the target surface will be overheated up to $\sim 10^5$ K, which is more than an order of magnitude higher than the thermodynamic critical temperature of aluminum (5720 K). This inevitably leads to an extremely high pressure in the ablated region [8], and such a great pressure will be released through the adiabatic expansion. Shadowgraphs of shorter than 2.5 ns in Fig. 2 only show the outward expansion of the laser heated area. The corresponding inward expansion compresses the inner part of the target and causes the transformation of thermal energy to mechanical energy, which is responsible for inducing the thermoelastic wave and causing material ejections at larger time delays. The thermal phase explosion and nonthermal thermoelastic wave mechanisms are combined at this point. In fact, the existence of a thermoelastic wave directly underneath the surface layer at which phase explosion takes place has been predicted by molecular dynamics simulations [12].

The formation of the stripe pattern in Fig. 2 actually represents the cooling process of the ejected material. As the material expands, it cools down close to the critical temperature where anomalies happen [14]. Around the critical temperature, the material will undergo a transition from a superheated metal liquid to a dielectric liquid and becomes nearly transparent [14]. In such cases, the outer edge of the ejected material appears opaque because the incident angle of the probe beam is close to 90° at these locations. In other words, the stripe pattern is in fact the result of diffraction of the ejected plume. In the shadowgraph of 500 ps, no stripe pattern can be seen except some bright regions inside the shadow of the ejected material. This is because at 500 ps, the temperature of the plume is still much higher than the critical value, and a very large portion of it remains opaque. We have also used an 800 nm probe beam to record the shadowgraphs and found the stripe pattern becomes sparser compared with that of using 400 nm probe beam. So the diffractional origin of the stripe pattern is further confirmed.

When the plume further cools to reach the metastable state, homogeneous nucleation occurs, and phase explosion takes place [8]. In this case, the density and refractive index of the plume decrease due to the transition from a liquid to a mixture of vapor and liquid, which leads to the blurring of the interface between the ambient air and the ejected material and consequently blurs the stripe pattern. This process is evident in the shadowgraphs with time delays from 2.5 to 5 ns in Fig. 2. The same type of stripe pattern is also observed in the shadowgraphs recorded at ~1 ns after a silicon or a glass sample is ablated with a 50 fs laser pulse of 40 $\mathrm{J/cm^2}$. The generation and the vanishing processes of the stripe pattern for these two different materials are very similar to those observed for aluminum, which convinces us that the origin of the stripe pattern described above must be valid in a rather broader sense.

Three shadowgraphs at larger time delays in Fig. 3 show the developments of a shock wave and a contact front after the event of phase explosion. The shock wave is formed once the energetic ablated material ejects into the ambient air and compresses the air at its front. However, in the early stage of laser ablation, only shadows of the ejected material are observed, and no shock waves are present in those shadowgraphs of shorter than 2.5 ns. Shadowgraphs from 2.5 to 5 ns (in Fig. 2) actually record the decay of the stripe pattern and the emergence of the shock wave. In the shadowgraph of 7 ns, the shock wave can be clearly seen, and the stripe pattern is almost vanished.

According to Sedov's blast wave theory [15], for a hemispherical shock wave, the relation between the radius r of the shock wave front and the time t may be expressed as $r = Ct^{2/5}$, where C is a constant that depends on the energy to drive the shock wave and the density of undisturbed air. A plot of this equation with $C = 0.21\ \mathrm{s^{-2/5}\ m}$ is given as the solid line in Fig. 4 together with the radii of the plume fronts deduced from the shadowgraphs of shorter than 5 ns (the open circles) and the radii of the shock wave fronts for larger time delays (the solid squares).

In Fig. 4, both the circles and the squares are all in the close vicinity of the solid line, which means they all can be approximated well by Sedov's equation. So the opaque outer edge of the ejected material actually overlaps with the shock wave front, and thus there is no shock wave in the shadowgraphs of shorter than 2.5 ns. As time elapses, phase explosion happens, the opaque outer edge of the plume blurs and finally disappears, and therefore the shock wave front becomes more and more visible from 2.5 to 5 ns in Fig. 2, and eventually becomes clearly observable at larger time delays in Fig. 3.

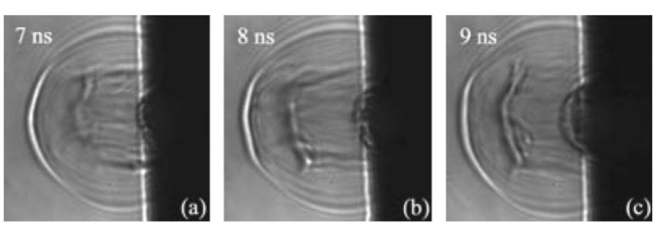

FIG. 3. Time-resolved shadowgraphs of material ejection at larger time delays after the same aluminum sample is ablated by a 50 fs laser pulse of 40 $\mathrm{J/cm^2}$. Frame size: $200 \times 200\ \mu\mathrm{m}$.

From the shadowgraph of 2.5 ns or larger, it can be seen that further material eruption takes place in the interior of the ejected plume or the shock wave, which forms the contact front. As mentioned above, when a 50 fs laser pulse ablates aluminum, the induced pressure at the target surface leads to the adiabatic expansion whose inward part will produce a thermoelastic wave, which could be the origin of the material ejection forming the contact front. Because this ejection appears only at 2.5 ns or later, there is a time interval between this ejection and the initial one forming the stripe pattern. If the ejection that forms the contact front is generated by the outward expansion of the laser heated area, such a time interval would be zero.

The propagation of the stress generated by the thermoelastic wave can be calculated based on an analytic solution given in Ref. [10] without considering any material removal and the corresponding structure change. Figure 5 shows our calculated results, where a negative stress represents a compressed stress [10]. When the thermoelastic wave propagates inside the target, the heated material will first undergo a compressed stress and thus move towards the interior of the target as shown in Fig. 5.

The 50 fs laser pulse of 40 $\mathrm{J/cm^2}$ will induce a peak negative stress in an order of magnitude of 1000 GPa [16], according to the calculation given in Fig. 5. Such an extremely high stress is much larger than the maximum endurable stress of aluminum, which is typically less than 0.6 GPa. So when the compressed stress exceeds the maximum endurable stress, the lattice of aluminum will be destroyed, but the compressed material will continue to move towards the target interior due to the compression force imposed on it. The distance between the adjacent atoms thus decreases further, leading to the sharp and huge increase of the repelling force between the adjacent atoms which eventually becomes so strong that it overcomes the compression force and ejects the material away from the target, causing a further material eruption.

From the generation of the compressed stress at the target surface to the repelling force becoming strong enough to overcome it, there is a developing time corresponding to the time interval between the material ejection

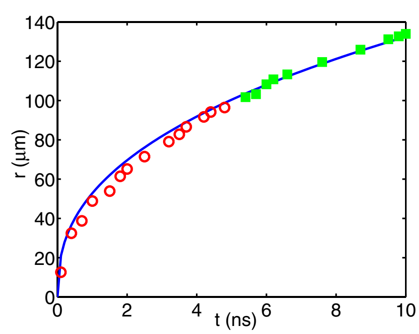

FIG. 4 (color online). Measured radii of the plume fronts (open circles) and the shock wave fronts (solid squares) as a function of time delay. The solid line represents the radius of the shock wave front calculated according to Sedov's theory.

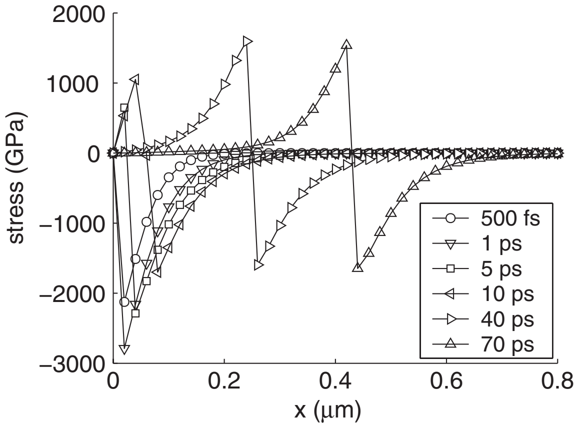

FIG. 5. Calculated time-dependent stress distribution in the interior of the aluminum target as a result of thermoelastic effect. The coordinate x originates from the surface and points towards the target interior.

forming the contact front and the one forming the stripe pattern. Unfortunately, the specific time interval cannot be derived from the calculations shown in Fig. 5.

Also, we notice that in the shadowgraphs from 7 to 9 ns, a growing dark area appears near the target surface [17]. We consider that the dark area represents another material ejection after the one forming the contact front. As this ejection appears at 7 ns when phase explosion has ended for a relatively long time, it can only be attributed to the effect of thermoelastic wave. As mentioned above, the outward release of the repelling force leads to the material ejection forming the contact front; its inward release will compress the inner part of the target, and after a specific developing time, another release of the repelling force will induce another ejection, which is the one appearing at 7 ns. Further bursts of material ejection may appear at even larger time delays, which unfortunately cannot be observed in our experimental setup due to the limited maximum delay line. When the energy fluence of the pump pulse changes from 40 to 10 J/cm^2, the dynamic process of femtosecond laser ablation of aluminum changes mainly in spatial scale, the stripe pattern and the intermittent material ejections can still be clearly observed in the shadowgraphs. So the phenomena we observed are rather ubiquitous in the relatively intense ablation regime.

The intermittent ejections are also seen in the shadowgraphs of femtosecond laser ablation of a silicon sample with the same energy fluence of 40 J/cm^2, but the ejection forming the contact front is noticeably weaker compared with that of aluminum. For the glass sample, however, when ablated with the laser pulse of the same parameters, no obvious intermittent ejections are observed. The fact that the electron mobility gets weaker and weaker from silicon to glass favors the generation of Coulomb explosion, which as another mechanism of causing damages to the material structure could hamper the effect of thermoelastic wave through decreasing its strength, and finally avoid the formation of the intermittent ejections.

In summary, the distinctive dynamic characteristics of intense femtosecond laser ablation of aluminum are revealed through time-resolved shadowgraphs. It is found that in the intense ablation regime associated with a typical laser energy fluence of 40 J/cm^2 at the sample surface, ultrafast laser ablation can be characterized as a hybrid material removal process composed of both thermal and nonthermal ablation mechanisms. For the metallic material of aluminum and the semiconductor material of silicon, the ablative dynamic process is primarily governed by the photothermal mechanism of phase explosion and the photomechanical mechanism of thermoelastic wave, and the latter is responsible for the recorded interesting phenomenon of intermittent bursts of material ejection. For the dielectric material of glass, no intermittent material ejections are observed, which could be due to the emergence of Coulomb explosion that hampers the formation of a strong thermoelastic wave. It is believed that the intermittent ejections could be better predicted with molecular dynamics calculations [12] based on an improved microscopic modeling at atomic and molecular levels, which will be the focus of our future work.

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