Investigation of ultrashort pulse laser ablation of solid targets by measuring the ablation-generated momentum using a torsion pendulum

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Abstract: 50 fs - 12 ps laser pulses are employed to ablate aluminum, copper, iron, and graphite targets. The ablation-generated momentum is measured with a torsion pendulum. Corresponding time-resolved shadowgraphic measurements show that the ablation process at the optimal laser fluence achieving the maximal momentum is primarily dominated by the photomechanical mechanism. When laser pulses with specific laser fluence are used and the pulse duration is tuned from 50 fs to 12 ps, the generated momentum firstly increases and then remains almost constant, which could be attributed to the change of the ablation mechanism involved from atomization to phase explosion. The investigation of the ablation-generated momentum also reveals a nonlinear momentum-energy conversion scaling law, namely, as the pulse energy increases, the momentum obtained by the target increases nonlinearly. This may be caused by the effective reduction of the dissipated energy into the surrounding of the ablation zone as the pulse energy increases, which indicates that for femtosecond laser the dissipated energy into the surrounding target is still significant.

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1. Introduction

The dynamic process of ultrashort laser ablation has been a subject of great interest to a wide range of researchers for over two decades [1,2] due to its great significance both theoretically [3] and practically [4]. However, a full understanding of the ultrafast laser ablation has not yet been achieved. The ejected plume generated during the ultrashort laser ablation of solid targets can exert propulsive momentum on the ablated target [5]. The knowledge of the ablation-generated momentum has great application potentials in the forthcoming technologies, such as laser induced inertial confinement fusion [6] and ablative laser propulsion [7]. The magnitude of this momentum also contains useful information about the laser ablation process and its underlying mechanisms. Unfortunately, only few measurements of ultrashort laser ablation-generated momentum have been reported [5,8,9], and furthermore most of them are for the investigation of ablative laser propulsion not for the study of the mechanisms of ultrashort laser ablation.

In this paper, the momentum induced in the ablation of solid targets by 50 fs - 12 ps laser pulses is experimentally investigated. A torsion pendulum is employed to measure the ablation-generated momentum. The dependence of the momentum on the laser fluence, laser pulse width, and laser pulse energy is determined for aluminum, copper, iron, and graphite targets, and the underlying ablation mechanisms are studied. The time-resolved shadowgraphs of 50 fs laser pulses ablating the copper target with different laser fluences are recorded using the pump-probe technique, which shows that the transition from photothermal to photomechanical mechanism results in the dependence of the ablation-generated momentum on the laser fluence. When the laser pulses with specific laser fluence are employed, the generated momentum firstly increases rapidly and then remains almost constant as the laser pulse width increases from 50 fs to 12 ps. This might be attributed to the change of the ablation mechanism from atomization [10] to phase explosion [11]. A nonlinear momentum-energy conversion scaling law is found that larger laser pulse energy can lead to higher momentum-energy conversion efficiency due to the decrease of the energy dissipated into the surrounding volume of the ablation spot.

2. Experimental setup

In our experiments, two femtosecond laser amplifier systems are employed to generate ultrashort laser pulses. One is a commercial Ti: Sapphire femtosecond laser amplifier system (HP-Spitfire, Spectra-Physics Inc.) that generates laser pulses with pulse energy up to 2 mJ and a central wavelength of 800 nm. Its laser pulse width is adjustable from 50 fs to 12 ps by changing the effective grating spacing. Another laser system used in the experiment is a TW home-built femtosecond laser amplifier system capable of generating 50 fs laser pulses with
pulse energy up to 150 mJ and a central wavelength of 800 nm. The two laser systems both output Gaussian laser beams with a beam quality factor of about 1.5.

The experimental setup measuring the ablation-generated momentum is shown in Fig. 1(a). Two pieces of identical targets are symmetrically mounted at both ends of the pendulum. The suspension thread of the torsion pendulum is made of fused silica fiber. A beam of He-Ne laser is reflected by a small square mirror attached to the center of the pendulum bar, and is then focused by Lens B onto a CCD camera (LU135M, Lumenera Inc.) to record the rotational angle of the pendulum. Before measuring the ablation-generated momentum, the torsion pendulum is calibrated by recording its free oscillation period and amplitude decay. With the measured maximal rotational angle of the pendulum under the strike of each single laser pulse, the ablation-generated momentum can be derived from an oscillator model [12]. To eliminate the air disturbance, the torsion pendulum is placed inside an airproof box.

Figure 1(b) illustrates the optical layout (side view) for laser ablation of the solid targets mounted on the pendulum. To determine the relative position between the ablated target and Lens A, laser pulses with intensity well below the target ablation threshold are used to irradiate the target and the back reflected light is recorded by a CCD camera (MTV188EX, Mintron Inc.). Lens A is mounted on a one-dimensional translation stage. By changing the distance between Lens A and the target, laser fluence from 0.1 J/cm$^2$ to 100 J/cm$^2$ on the target surface can be achieved. Lens A and Mirror 2 are both mounted on a vertical translation stage by which the laser beam can be moved up or down after each pulse strikes the target, insuring that each laser pulse always strikes a fresh target area.

3 Results and discussions

In order to facilitate the following analyses and discussions, we will first briefly review a key parameter, the momentum coupling coefficient [13], which is often used in the propulsion technology. The momentum coupling coefficient describes the momentum-energy conversion efficiency and is expressed as $C_m = p/E$, where $p$ is the momentum generated by a single laser pulse and $E$ is the laser pulse energy in this case. In the following part of this paper, all the data presented are obtained for single laser pulse.

3.1 Laser fluence effect on the ablation-generated momentum

The dependence of the momentum coupling coefficient on the laser fluence is shown in Fig. 2 for a copper target ablated by laser pulses with pulse energy of 1.1 mJ and pulse duration of 50 fs focused by a 100 mm focal length lens. The focal lens has a diameter of 25.4 mm larger than the incident laser beam diameter of 9 mm ($1/e^2$). In Fig. 2, the optimal laser fluence that can generate the maximal momentum is 4 J/cm$^2$ for the copper target. Similar relations between the momentum coupling coefficient and the laser fluence also exist for other kinds of targets, such
as aluminum and iron whose optimal laser fluences are measured to be 9 J/cm$^2$ and 1.7 J/cm$^2$ respectively under the same experimental conditions.

![Graph showing the dependence of the momentum coupling coefficient on the laser fluence for a copper target ablated by 1.1 mJ, 50 fs laser pulses focused by a 100 mm focal length lens. 1 dyne = $10^{-5}$ N.]

**Fig. 2.** Dependence of the momentum coupling coefficient on the laser fluence for a copper target ablated by 1.1 mJ, 50 fs laser pulses focused by a 100 mm focal length lens. 1 dyne = $10^{-5}$ N.

In the following, the underlying physical mechanism associated with the data shown in Fig. 2 will be revealed by the pump-probe technique. The time-resolved shadowgraphs are recorded of the ultrafast dynamic process of femtosecond laser ablating the copper target. The ablation laser pulse and the focal condition are the same as those used in Fig. 2. The output femtosecond laser pulse is divided into two pulses by an 8:2 beam splitter. One pulse is used as the pump pulse to ablate the target; the other pulse after frequency doubling is used as the probe pulse to record the ablation process at selected time delays. The detailed description about the experimental setup for recording time-resolved shadowgraphs can be found in Ref [14].

The time-resolved shadowgraphs of ablating the copper target are presented in Figs. 3 and 4 which are respectively recorded when the copper target is placed at the focal plane of the focal lens and at the position associated with the optimal laser fluence. The indicated time in each shadowgraph represents the delay time when the picture is taken after the laser pulse strikes the target. For each shadowgraph, a single laser pulse ablates a fresh spot on the target surface. The long and narrow channel in Fig. 3 is the air shock wave associated with direct air ionization. In Fig. 4, however, no such channel is present because the laser fluence in these cases is not high enough to ionize the ambient air. In Figs. 3 and 4 the ejected target material is all surrounded by the shock wave caused by the target material ablation.

![Time-resolved shadowgraphs of material ejection recorded at the indicated time delays after a copper target is ablated by 1.1 mJ, 50 fs laser pulses. For each time delay, the target that is placed at the geometrical focus of the 100 mm focal length lens is moved to a fresh spot and only one pulse is fired. The horizontal solid arrow in the shadowgraph of 1 ns time delay indicates the laser pulse propagation direction. The narrow channel in front of the target is generated due to the laser-induced air ionization. Frame size: 320 $\mu$m × 240 $\mu$m.]

**Fig. 3.** Time-resolved shadowgraphs of material ejection recorded at the indicated time delays after a copper target is ablated by 1.1 mJ, 50 fs laser pulses. For each time delay, the target that is placed at the geometrical focus of the 100 mm focal length lens is moved to a fresh spot and only one pulse is fired. The horizontal solid arrow in the shadowgraph of 1 ns time delay indicates the laser pulse propagation direction. The narrow channel in front of the target is generated due to the laser-induced air ionization. Frame size: 320 $\mu$m × 240 $\mu$m.
Fig. 4. Time-resolved shadowgraphs of material ejection recorded at the indicated time delays after a copper target is ablated by 1.1 mJ, 50 fs laser pulses. The target is placed at the position where the optimal laser fluence exists. The solid arrow in the shadowgraph of 0 ns time delay indicates the laser pulse propagation direction. Frame size: 620 μm × 460 μm.

By comparing Fig. 3 and Fig. 4, one distinct characteristic can be found: when the laser fluence of the pump beam on the target surface is relatively high (over 100 J/cm²), an opaque plume can be seen near the target surface in the shadowgraph of 1 ns time delay in Fig. 3; however no such opaque plume appears when the laser fluence becomes relatively low (see Fig. 4). In Fig. 3, as the laser fluence of the pump beam is high enough to ionize the ambient air, the material on the target surface is commonly considered to be removed through the atomization process [10] and so the opaque plume is composed mainly of the ionized atomic target material. It is inferred that the plume is opaque because its plasma frequency is higher than the frequency of the probe beam at the early stage of the ablation. As time elapses and plume expanses, the opaque plume becomes more and more transparent due to the decrease of its density and thus plasma frequency.

When the laser fluence of the pump beam on the target surface is only ~4 J/cm², no such opaque plume is observed and instead there are many stripes normal to the target surface within the shock wave shown in Fig. 4. The material particles ejected from the target will vent the air in their path, leading to the non-uniform distribution of the residual air within the shock wave, which is considered to be the reason for the appearance of such stripes when irradiated by the probe beam. By observing the stripe spacing in Fig. 4, it can be deduced that the ejected particles have a relatively large scale even in the micrometer range. As mentioned above, all these stripes are perpendicular to the target surface, indicating that these large particles have a preferential ejection direction normal to the target surface. As the ultrafast laser heating can induce extremely high temperature and stress gradients inside the target along the laser beam propagation direction, i.e., normal to the target surface, these large-scale particles may be originated from the release of the non-uniform stress, which is in nature a consequence of the photomechanical effect [10]. Note that photothermal mechanism, such as phase explosion, always needs a period of time of 1 ns - 10 ns to form droplets [15] after laser pulses strike the target and thus is not likely to be the mechanism that generates the large-scale particles in the case of Fig. 4.

Similar ejected material normal to the target surface can also be observed in Fig. 3 at a larger time delay (such as 6 ns time delay). In Fig. 3 the laser fluence at the target surface is very high, but at a deeper layer inside the target the laser fluence of the pump beam can decrease to a value close to the laser fluence at the target surface in Fig. 4 because the laser fluence decreases sharply towards the inside of the target. But the ablated target material ejected perpendicularly from the target surface in Fig. 3 cannot be simply attributed to the photomechanical effect because a relatively long time lag exists between the strike of the laser pulse and the ejection of the ablated target material.
As the air shock wave is generated due to the target material ablation, the shock wave front can be approximately taken as the plume front of the ejected target material. Therefore, based on the time-resolved shadowgraphs in Figs. 3 and 4, the time dependent relation of the ejected plume velocity along the normal direction of the target surface can be readily obtained and presented in Fig. 5. It should be noticed that in Fig. 5 the velocity of the ionized atomic target material (hollow squares) is apparently greater than that of the ablated target material (solid circles) due to the photomechanical effect. This may help us to obtain a physical explanation to the curve shown in Fig. 2, namely, the ablation process without fast ejected atomic material can generate more momentum. It can be inferred that the ultrafast ablation process dominated by the photomechanical process has a larger material removal rate. Ablation induced by femtosecond laser with high laser fluence such as the case in Fig. 3 can generate atomic ejected material with high velocity, but on the whole it only removes lesser target material. On the other hand, smaller momentum is generated due to the weak ablation induced by laser pulses with low laser fluences just as shown in Fig. 2. Therefore, the optimal laser fluence often has a mediate value. The above analyses can also apply to the case of aluminum and iron targets.

Fig. 5. Dependence of the ablation plume velocity normal to the target surface on the delay time. Hollow squares present the case that the copper target is placed at the focus of the focal lens, and solid circles present the case that the copper target locates at the position where the optimal laser fluence exists.

3.2 Laser pulse width effect on the ablation-generated momentum

The effect of the laser pulse width on the ablation-generated momentum is investigated through using laser pulses with pulse energy of 1.1 mJ and pulse duration ranging from 50 fs to 12 ps. The experimental results are shown in Fig. 6(a). Four kinds of targets, including aluminum, copper, iron, and graphite are used which are all placed at the geometrical focus of the 100 mm focal length lens; (b) enlarged view of the data points for iron target with laser pulse duration less than 2 ps.

Fig. 6. (a) Dependence of the momentum coupling coefficient on the laser pulse width when 1.1 mJ laser pulses are used to ablate aluminum (solid squares), graphite (solid circles), copper (solid triangles), and iron (solid inverse triangles) targets. All targets are placed at the geometrical focus of the 100 mm focal length lens; (b) enlarged view of the data points for iron target with laser pulse duration less than 2 ps.
The trend presented by the data points in Fig. 6(a) is obtained when laser pulses with specific laser fluence are employed and it is expected that as the laser fluence changes, the dependence of the momentum coupling coefficient on the laser pulse width may change correspondingly.

The specific relation shown in Fig. 6(a) is believed to be originated from the change of the ablation mechanism with the increase of the laser pulse width. When the laser pulse width is short, the laser pulse energy is deposited mainly within a thin surface layer characterized by the optical absorption depth due to less effectiveness of thermal diffusivity. This will result in a high deposited energy density inside the target which makes a large part of the absorbed laser energy be used to remove the target material through instantaneous atomization [10]. The target material ablated through the atomization process has a high ejection velocity as it is shown in Fig. 5. However, as the laser pulse only heats a thin surface layer, the amount of the removed target material is small and hence a smaller momentum coupling coefficient is obtained in this case just as shown in Fig. 6 for 50 fs laser pulses. (In this case, there are still other mechanisms causing the target material to be ablated off in the later stage of the ablation, but the amount of the consumed laser energy and thus the generated momentum in the later ablation stage is believed to be relatively small. This is because a large part of the laser pulse energy is used to ablate target material through atomization process just as described above.)

As the laser pulse width increases, the laser-heated volume extends due to the thermal relaxation and the deposited laser energy density inside the target decreases accordingly. The target material near the surface may be ablated off still through the atomization process, but due to the decrease of the laser energy density in the target, more absorbed laser energy is employed to ablate the target material through photomechanical mechanism or phase explosion with a smaller ejection velocity. Therefore, due to the increase of the heat affected zone, more material will be ejected with a smaller mean velocity as a result of the reduced mean energy density deposited in the target. The rapid increase of the momentum coupling coefficient in Fig. 6(a) is understandable when the relation between the kinetic energy $E_k$ and the momentum $p$ [$E_k = (1/2)mv^2 = (1/2)pv$] is analyzed. From the simple relation between $E_k$ and $p$, also assuming that the energy conversion ratio from the laser energy to the kinetic energy of the ablated target material does not decrease with the increase of the laser pulse width, it can be readily deduced that larger momentum will be generated when more target material is removed with slower ejected velocity. Examination of the ablation craters with an atomic force microscope (Dimension 3100, Veeco Inc.) indeed shows that the total volume of the removed aluminum target material ablated by a single laser pulse with the same parameters as that used in Fig. 6(a) increases as the laser pulse width increases from 50 fs to 2 ps [16]. Therefore the mean velocity of the ejected target material calculated based on the corresponding data in Fig. 6(a) decreases, which presents an argument for the above analyses.

When the laser pulse is longer than 2 ps, the volume of the removed aluminum target material measured by AFM keeps nearly constant as the pulse width increases [16]. Because phase explosion is always considered as a process determined by the amount of the deposited energy rather than the pulse width [17,18], the dominating physical mechanism of the ablation process with laser pulse widths longer than 1 - 2 ps shown in Fig. 6(a) is expected to be phase explosion [11]. Further experiments we performed also show that the momentum coupling coefficients respectively generated by 200 ps and 10 ns laser pulses ablation of the copper target are nearly of the same value as that generated by 12 ps laser pulses in Fig. 6(a). This further demonstrates that phase explosion is the dominating ablation mechanism for laser pulse durations longer than 1 - 2 ps because for high fluence nanosecond laser pulses phase explosion is the only primary material-removal mechanism [11]. Finally it should be noted that the momentum coupling for shorter pulses can be either smaller or larger than that for longer pulses, depending on the laser fluence.

While the dependence of the momentum coupling coefficient on the laser pulse duration seems similar for the four different types of targets as shown in Fig. 6(a), a zoom-in view of the data points for the iron target (see Fig. 6(b)) reveals a unique feature for laser pulse duration less

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than 300 fs. In Fig. 6(b), it is evident that for the iron target, as the laser pulse duration increases, the momentum coupling coefficient first decreases and reaches a minimal value at about 300 fs, then gradually climbs up and reaches a relatively constant value. Such a special behavior associated with the iron target has also been observed in our previous experiment [5] but remains not well understood.

3.3 Laser pulse energy effect on the ablation-generated momentum

In this section, femtosecond laser pulses with pulse energy up to 90 mJ are generated by the terawatt femtosecond laser system and employed to ablate the aluminum target attached to the torsion pendulum. As the output laser pulses have a beam diameter of ~50 mm ($1/e^2$), a focal lens ($f = 500$ mm) with a diameter of 100 mm is used so that the laser beam can be completely accommodated.

Figure 7(a) presents the dependence of the ablation-generated momentum on the laser fluence for 400 fs laser pulse ablation of the aluminum target with four different pulse energies. By averaging three adjacent data points, a trend line is added to every group of data in Fig. 7(a). The vertical solid lines in Fig. 7(a) mark the laser fluences of 0.2 J/cm$^2$, 0.6 J/cm$^2$, 2 J/cm$^2$, and 4 J/cm$^2$ respectively. The values of the ablation-generated momentum at these laser fluences on the trend lines in Fig. 7(a) are replotted in Fig. 7(b) to display the relations between the ablation-generated momentum and the laser pulse energy at the given laser fluences. It can be seen from Fig. 7(b) that the increasing rate of the momentum with the laser pulse energy is apparently much faster than the linear increasing rate shown by the linear scaling line. Note that, in order to achieve the same fluence for different pulse energies, the laser spot size on the target surface must be adjusted. The nonlinear trend indicates that the ablation-generated momentum does not scale linearly with the ablation area. Therefore a nonlinear scaling law exists between the ablation-generated momentum and the laser pulse energy, and the momentum coupling coefficient grows with the increase of the laser pulse energy.

The physical origin of the nonlinear momentum-energy conversion scaling law may be attributed to the fact shown as follows. When the pulse energy is raised by $\beta$ times, the laser spot area on the target surface must be increased also by $\beta$ times to keep the laser fluence unchanged. However, the perimeter of the ablation zone is only increased by $\beta^{1/2}$ times in this case, which inevitably reduces the energy diffused into the surrounding area. Thus, more laser energy is used for ablation and transferred to the momentum of the ejected material.

4. Conclusions

The dependence of the momentum coupling coefficient on the laser fluence is investigated using the torsion pendulum and the optimal laser fluence is obtained for femtosecond laser
ablation of aluminum, copper, and iron targets. Time-resolved shadowgraphs of laser ablation of the copper target indicate that the ablation process generates the maximal momentum is dominated by the photomechanical mechanism. It in turn demonstrates that the photomechanical mechanism is a highly efficient way to remove the target material, which is in agreement with the description in Ref [19].

As the laser pulse duration increases from 50 fs to 12 ps, the momentum coupling coefficient first increases rapidly and then remains nearly constant when laser pulses with specific laser fluence are used. The characteristic pulse width that divides the above two regimes is about 1 - 2 ps, only varying slightly for different target materials. The change of the ablation mechanism from atomization to phase explosion with the increase of the laser pulse width is responsible for such an observation.

It is also found that under the same experimental conditions the ablation-generated momentum increases nonlinearly as the increase of the laser pulse energy and the laser spot area on the target. Such a nonlinear momentum-energy conversion scaling law demonstrates that even for femtosecond laser pulses, the dissipation of the energy into the surrounding area of the ablation site is still significant and laser pulses with larger pulse energy and spot area while keeping the laser fluence unchanged can lead to higher energy utilization efficiency.

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