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<tr>
<td>Citation</td>
<td>Applied Physics Letters, 108(1), p.011102_1·011102_4</td>
</tr>
<tr>
<td>Text Version</td>
<td>Publisher's Version</td>
</tr>
<tr>
<td>URL</td>
<td><a href="https://jopss.jaea.go.jp/search/servlet/search?5054510">https://jopss.jaea.go.jp/search/servlet/search?5054510</a></td>
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<tr>
<td>DOI</td>
<td><a href="https://doi.org/10.1063/1.4939231">https://doi.org/10.1063/1.4939231</a></td>
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<td>Right</td>
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Takayuki Kumada, Tomohito Otobe, Masaharu Nishikino, Noboru Hasegawa, and Terutake Hayashi

View online: http://dx.doi.org/10.1063/1.4939231
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/108/1?ver=pdfcov
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Dynamics of spallation during femtosecond laser ablation studied by time-resolved reflectivity with double pump pulses

Takayuki Kumada,1,a) Tomohito Otobe,1 Masaharu Nishikino,1 Noboru Hasegawa,1 and Terutake Hayashi2
1Kansai Photon Science Institute, Japan Atomic Energy Agency, 8-1-7 Umehidai, Kizugawa, Kyoto 619-0215, Japan
2Department of Mechanical Engineering, Faculty of Engineering, Kyushu University, 744 Motoooka, Nishi-ku, Fukuoka 819-0395, Japan

(Received 31 July 2015; accepted 18 December 2015; published online 4 January 2016)

The dynamics of photomechanical spallation during femtosecond laser ablation of fused silica was studied by time-resolved reflectivity with double pump pulses. Oscillation of reflectivity was caused by interference between the probe pulses reflected at the sample surface and the spallation layer, and was enhanced when the surface was irradiated with the second pump pulse within a time interval, $\Delta t$, of several picoseconds after the first pump pulse. However, as $\Delta t$ was increased, the oscillation amplitude decreased with an exponential decay time of 10 ps. The oscillation disappeared when $\Delta t$ exceeded 20 ps. This result suggests that the formation time of the spallation layer is approximately 10 ps. A second pump pulse with $\Delta t$ shorter than 10 ps excites the bulk sample. The spallation layer that is photo-excited by the first and second pump pulses is separated afterward. In contrast, a pulse with $\Delta t$ longer than the formation time excites and breaks up the spallation layer that has already been separated from the bulk. The formation time of the spallation layer, as determined in this experiment, is attributed to the characteristic time of the mechanical equilibration corresponding to the thickness divided by the sound velocity of the photo-excited layer. © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4939231]

When a solid sample is irradiated with an intense femtosecond laser pulse, the temperature and pressure increase in the photo-excited layer. The pressure propagates as compressive stress waves in the surface and depth directions. When one of the compressive waves is reflected from the sample surface and the spallation layer, and was enhanced when the surface was irradiated with the second pump pulse within a time interval, $\Delta t$, of several picoseconds after the first pump pulse. However, as $\Delta t$ was increased, the oscillation amplitude decreased with an exponential decay time of 10 ps. The oscillation disappeared when $\Delta t$ exceeded 20 ps. This result suggests that the formation time of the spallation layer is approximately 10 ps. A second pump pulse with $\Delta t$ shorter than 10 ps excites the bulk sample. The spallation layer that is photo-excited by the first and second pump pulses is separated afterward. In contrast, a pulse with $\Delta t$ longer than the formation time excites and breaks up the spallation layer that has already been separated from the bulk. The formation time of the spallation layer, as determined in this experiment, is attributed to the characteristic time of the mechanical equilibration corresponding to the thickness divided by the sound velocity of the photo-excited layer. © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4939231]

Therefore, non-thermal ablation would occur much faster than thermal ablation.

The aim of this study was to experimentally determine the formation time of the spallation layer. In our previous studies,6,7 we observed the separation behavior of the spallation layer from the sample surface by analyzing the oscillation of the time-resolved reflectivity, which is caused by repeated constructive and destructive interference between the probe pulses reflected from the layer and surface. However, the oscillation period (60–110 ps) was too long to determine the formation time of the spallation layer. Thus, in this work, we used another time-resolved reflectivity method with double pump pulses. The spallation layer produced by the first pump pulse is destroyed by the second pulse, which is confirmed by the oscillation of the time-resolved reflectivity disappearing. The formation time of the spallation layer, which is much less than the oscillation period, can be determined by measuring the destruction of the spallation layer.

Figure 1 shows the experimental setup, which is slightly modified from the one used for the time-resolved reflectivity measurements in Ref. 6. A laser pulse from a Ti:sapphire chirped pulse amplification system with a wavelength, $\lambda$, of 795 nm, pulse duration of 60 fs, and a repetition rate of 10 Hz was split into pump and probe pulses. The pump pulse was split further into double-pulse trains by an interferometer, narrowed by a pair of convex and concave lenses, and passed through a set of a waveplate and a thin-film polarizer to control the fluence. The split pulse trains were focused on the surface of a 1.5-mm-thick fused silica sample plate (Helalux) in a vacuum chamber at an incidence angle of 45° with s-polarization. The time interval, $\Delta t$, between the

a)Author to whom correspondence should be addressed. Electronic mail: kumada.takayuki@jaea.go.jp. Present address: Nuclear Science and Research Institute, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan.
Double pump pulses was controlled by using an optical delay line in the interferometer, and $\Delta \tau$ at the origin was determined by the interference spectrum of the double pump pulses. To avoid the effects of interference on the ablation, we obtained the reflectivity at $\Delta \tau = 0$ ps by using a single pump pulse with a fluence equal to the sum of the fluences of the double pump pulses.

A $p$- or $s$-polarized probe pulse was passed through an optical delay line, the beam diameter of which was expanded by using a pair of convex lenses, and then, the pulse was tightly focused at the center of the irradiation spot of the pump pulse at an angle, $\theta_{\text{probe}}$, of $55.5^\circ \pm 0.5^\circ$, which is very close to the Brewster incident angle. The time, $t$, at which the reflectivity was increased by irradiation of the first pump pulse was defined as the origin. We ensured that the diameters of the major and minor axes of the ellipsoidal spot of the probe pulse (30 and 15 $\mu$m) were significantly smaller than those of the pump pulse (150 and 70 $\mu$m). The fused silica sample plate was moved along its surface plane during the measurement so that a fresh spot was exposed to every laser shot. The reflected probe pulse was spatially filtered by an aperture, polarization-filtered by a Glan laser prism, and spectrally filtered by a monochromator to remove the scattered pump pulse and breakdown emission before measuring its fluence with a photodiode.

The bold lines in Figs. 2 and 3 show the reflectivity after irradiation of the single pump pulse with fluences of 3.1 and 5.3 J/cm$^2$, which were higher than the ablation threshold (3 J/cm$^2$). The reflectivity increased steeply at $t = 0$ ps because of optical breakdown. The reflectivity for the $s$-polarized probe pulse monotonically decreased with a decay time of approximately 10 ps, whereas that for the $p$-polarized probe pulse showed a dip at $t \approx 1$ ps owing to resonance absorption by gas plasma blown out from sample surface. The reflectivity oscillated with a period of approximately 60 ps, which was caused by repeated constructive and destructive interference between the probe pulses reflected at the bulk sample surface and the spallation layer.

The narrow lines in Figs. 2 and 3 show the reflectivity after irradiation of the first and second pump pulses with...
fluences of 3.1 and 2.2 J/cm², respectively. Although the fluence of the second pump pulse was lower than that of the first pulse, the reflectivity at the timing of the second pump pulse \( t = \Delta t \) increased more than that at \( t = 0 \) ps. This result shows that, while the first pump pulse generated initial seed carriers via multiphoton excitation, the second pulse multiplied the preexisting carriers via impact ionization.\(^6\)\(^7\) The peak reflectivity for the \( s \)-polarized probe pulse at \( t = \Delta t \) was independent of \( \Delta t \), indicating that the carrier density produced by the double pump pulses was independent of \( \Delta t \), at least up to 20 ps. In contrast, the peak reflectivity for the \( p \)-polarized probe pulse increased with increasing \( \Delta t \) up to approximately 10 ps. This result suggests that the resonance absorption of the \( p \)-polarized probe pulse became less significant because of the diffusion and dissipation of the gas plasma produced by the first pump pulse.

Figure 3 shows that the oscillation amplitude for the \( p \)-polarized probe pulse was increased by the irradiation of the second pump pulse at \( \Delta t = 0 \) or 4.7 ps, but decreased at \( \Delta t = 22.7 \) ps. As shown in the inset, the oscillation amplitude decreased with an exponential decay time of approximately 10 ps. Theoretically, the oscillation amplitude increased with the increasing areal density of carriers, namely, the product of the carrier density and thickness, in the spallation layer.\(^6\) Because the carrier density is independent of \( \Delta t \), the decrease in the oscillation amplitude with increasing \( \Delta t \) can be accounted for by only the decrease in thickness.

Next, we discuss how the thickness decreased with increasing \( \Delta t \). Figure 4 shows a schematic of spallation caused by the first and second pump pulses. When a solid sample is irradiated with a single pump pulse with a fluence slightly higher than the ablation threshold, voids are formed and grow at a depth of approximately half the thickness of the photo-excited layer assisted by tensile stress.\(^1\)\(^3\) The upper part of the photo-excited layer is separated as the spallation layer when these voids merge together [Fig. 4(a)].\(^2\)\(^3\) When the second pump pulse is irradiated before these voids are formed, it multiplies carriers in the photo-excited layer on the bulk sample. The upper part of the layer, in which carriers are produced by the first and second pump pulses, is separated [Fig. 4(b)]. As \( \Delta t \) increases, the voids hinder the propagation of the photo-excitation energy from the second pump pulse into the bulk sample. Thus, larger numbers of atoms evaporate from the photo-excited layer [Fig. 4(c)]. When the second pump pulse excites the spallation layer that has already been separated from the bulk sample, the photo-excitation energy provided to the layer cannot propagate into the bulk because of the lack of mechanical contact. Therefore, the spallation layer breaks up as a result of intense evaporation, and the oscillation of the reflectivity disappears accordingly [Fig. 4(d)]. This model was supported by the result that the oscillation was not destroyed when the second pump pulse with the fluence of 0.5 J/cm² that is much less than the ablation threshold was irradiated at \( \Delta t \approx 10 \) ps [Inset of Fig. 3]. The fluence would be too low to destroy the spallation layer.

In contrast to the oscillation amplitude, the exponential decay time of the oscillation amplitude was independent of the fluence of the second pump pulse within a measurement error of 2 ps. This result supports the model that the decay time is determined by the formation time of the spallation layer. Theoretically, the spallation occurs when the compressive stress wave generated at a depth of approximately half the thickness, \( l \), of the photo-excited layer propagates toward the surface, and the tensile stress wave propagates from the surface to the depth at which the voids are formed.\(^1\)\(^2\) Thus, the formation time is roughly estimated to be the characteristic time, \( \tau_s \), of mechanical equilibration of the photo-excited layer, which is given by \( l \) and sound velocity, \( c_0 \), as \(^2\)\(^3\)\(^18\)

\[
\tau_s \approx \frac{l}{c_0}. \quad (1)
\]

By substituting \( l \approx 100 \) nm from the reflectivity and crater depth measurements\(^1\)\(^3\) and \( c_0 = 5720 \) m/s (Ref. 19) into Eq. (1), \( \tau_s \) is estimated to be 20 ps, which is comparable to the formation time of the spallation layer determined in this experiment. The formation time of the spallation layer was longer than that of plasma, which was estimated to be less than 1 ps from the dip in the reflectivity for the \( p \)-polarized probe pulse owing to resonance absorption in Fig. 2(a), but shorter than that of thermal ablation and evaporation (\( \leq 100 \) nm, Ref. 11).

In conclusion, the measurement of time-resolved reflectivity with double pump pulses revealed that spallation
occurs after mechanical equilibration of the photo-excited layer. This result shows that spallation is a collective motion of atoms in the photo-excited layer. The time of the spallation is determined by the thickness and sound velocity, which are irrelevant to the thermal effect.

We acknowledge Professor Y. Kayanuma of the Tokyo Institute of Technology and Dr. N. Inogamov of the Landau Institute for fruitful discussion. We also thank Dr. Y. Kurosaki of JAEA for reading this manuscript. This study was supported in part by the CPhoST program funded by the Special Coordination Funds for Promoting Science and Technology commissioned by MEXT of Japan, and a Grant-in-Aid for Scientific Research C from JSPS (Grant No. 15K04706).