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Dynamics of spallation during femtosecond laser ablation studied by time-resolved reflectivity with double pump pulses

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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, $\Delta\tau$, of several picoseconds after the first pump pulse. However, as $\Delta\tau$ was increased, the oscillation amplitude decreased with an exponential decay time of 10 ps. The oscillation disappeared when $\Delta\tau$ exceeded 20 ps. This result suggests that the formation time of the spallation layer is approximately 10 ps. A second pump pulse with $\Delta\tau$ 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 $\Delta\tau$ 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, it is transformed into a tensile stress wave.[1–3] The tensile stress wave assists the separation of the upper part of the photo-excited layer as a thin layer. Linde *et al.*[4,5] observed interference fringes (Newton rings) arising from probe pulses reflected from the sample surface and the separated layer during time-resolved microscopy of metals and semiconductors. We recently observed this type of interference in fused silica and polymers in the form of oscillation of time-resolved reflectivity.[6,7] This non-thermal photomechanical effect, called spallation, has attracted intense interest in the development of high-precision micromachining techniques that suppress thermal damage.[8–11]

After excitation with a femtosecond laser pulse, the irradiated material undergoes energy transfer, diffusion, and relaxation over a wide range of timescales.[11] Thermal ablation takes place much later than the energy transfer from electrons to the lattice over a few picoseconds. Starting from nucleation sites in the photo-excited layer, liquid and gas gradually expand in the depth direction of the material. In contrast, non-thermal structural change can be driven directly by sufficiently dense photo-excited electrons, while the lattice remains vibrationally cold. 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, λ, 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, $\Delta\tau$, between the

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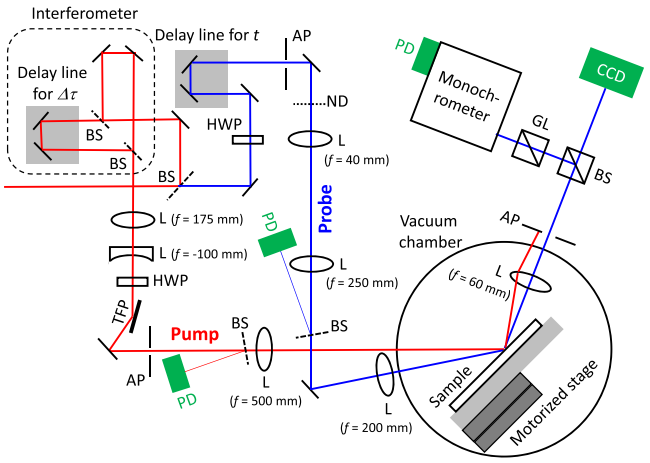

FIG. 1. Schematic layout for time-resolved reflectivity measurements. BS, beam splitter; L, lens; HWP, half-wave plate; TFP, thin-film polarizer; AP, aperture; ND, neutral density filter; PD, photodiode; and GL, Glan laser prism (for *p*-polarized probe pulse only).

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, θ_{probe}, of 55.5° ± 0.5°, 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 μm) were significantly smaller than those of the pump pulse (150 and 70 μm). 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 monochrometer 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).[6] The reflectivity increased steeply at $t = 0$ ps because of optical breakdown.[6,12–14] 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.[15] 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.[6]

The narrow lines in Figs. 2 and 3 show the reflectivity after irradiation of the first and second pump pulses with

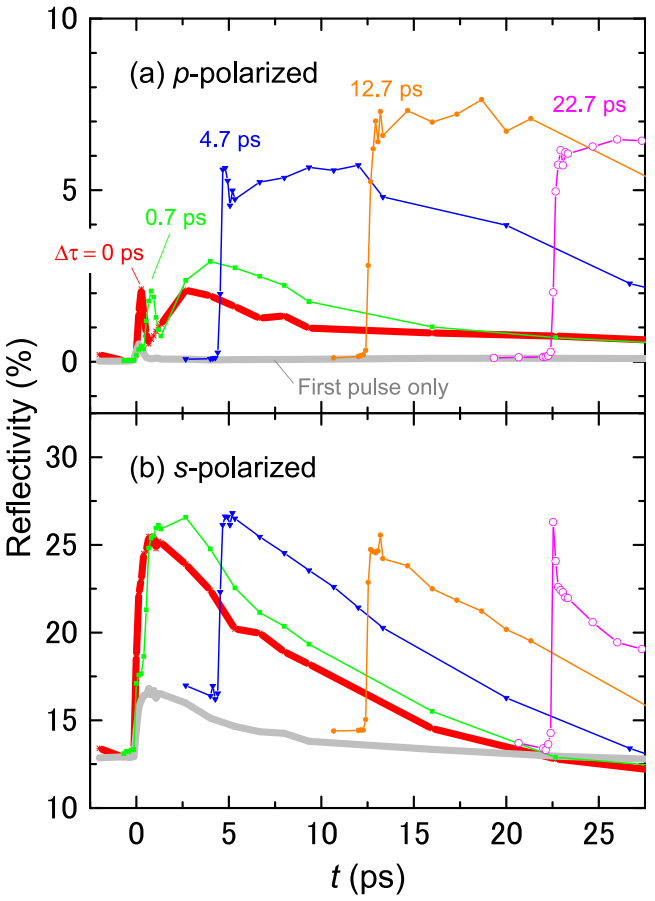

FIG. 2. Reflectivity of fused silica as a function of time for (a) *p*-polarized and (b) *s*-polarized pulses. Bold lines: reflectivity for the single pump pulse with the fluence of the first pump pulse (3.1 J/cm^2, gray), and sum of the first and second pump pulses (5.3 J/cm^2, red). Narrow lines: reflectivity for the double pump pulses (3.1 and 2.2 J/cm^2) with $\Delta\tau$ indicated above each peak.

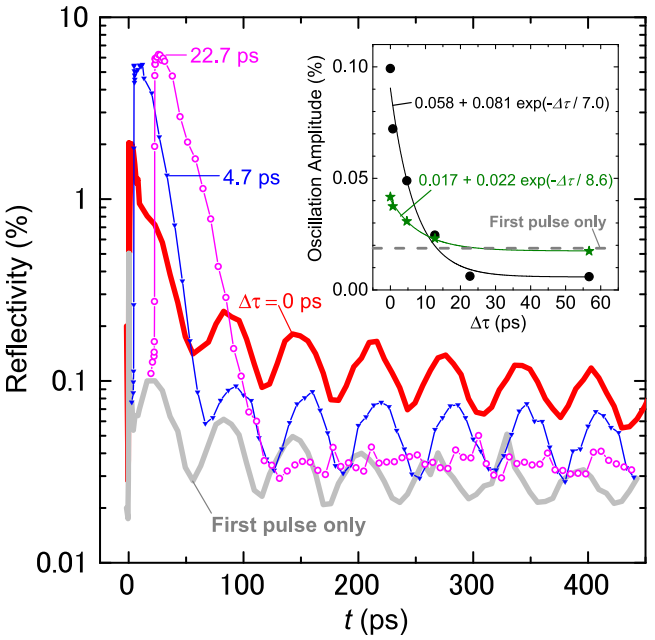

FIG. 3. Reflectivity of fused silica in a logarithmic representation for the *p*-polarized probe pulse. The notation of each line is same as that in Fig. 2. The inset shows the oscillation amplitude at the fourth cycle (t = 200–250 ps) that was after irradiation of the second pump pulse with the fluence of 2.2 J/cm^2 (black circles) and 0.5 J/cm^2 (green stars), respectively. The curves show the exponential decay function fit to the data. The gray dashed line shows the results for the first single pump pulse.

fluences of 3.1 and 2.2 J/cm^2, 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 ($t = \Delta\tau$) 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.[16,17] The peak reflectivity for the *s*-polarized probe pulse at $t = \Delta\tau$ was independent of $\Delta\tau$, indicating that the carrier density produced by the double pump pulses was independent of $\Delta\tau$, at least up to 20 ps. In contrast, the peak reflectivity for the *p*-polarized probe pulse increased with increasing $\Delta\tau$ 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 of the reflectivity for the *p*-polarized probe pulse was increased by the irradiation of the second pump pulse at $\Delta\tau = 0$ or 4.7 ps, but decreased at $\Delta\tau = 22.7$ 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 $\Delta\tau$, the decrease in the oscillation amplitude with increasing $\Delta\tau$ can be accounted for by only the decrease in thickness.

Next, we discuss how the thickness decreased with increasing $\Delta\tau$. 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 $\Delta\tau$ 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/cm^2 that is much less than the ablation threshold was irradiated at $\Delta\tau \gg 10$ 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

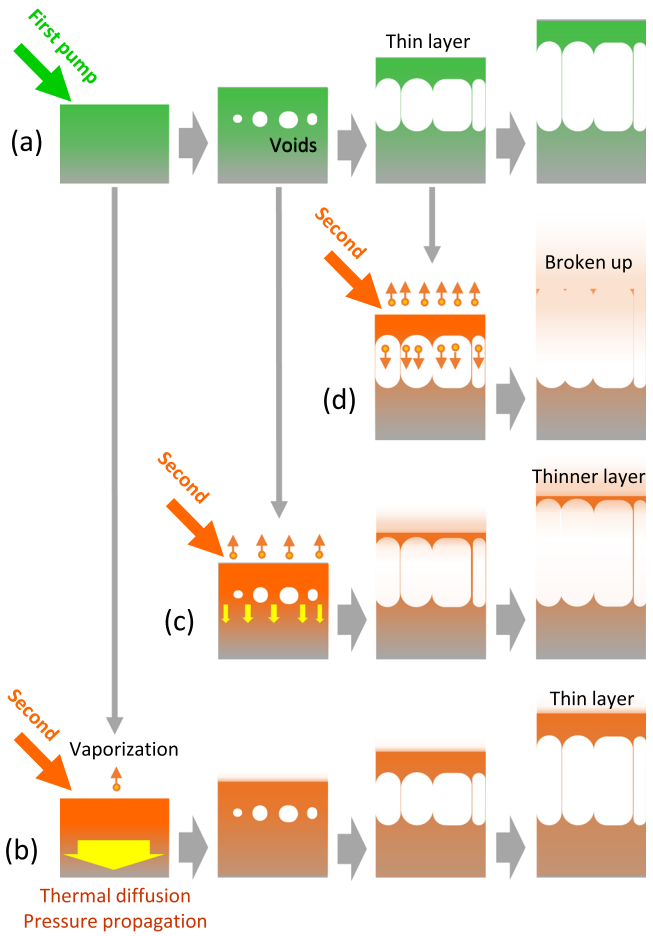

FIG. 4. (a) Schematic of spallation after irradiation with the first pump pulse. (b)–(d) Schematic of spallation after irradiation of the second pump pulse before, during, and after the formation and growth of voids, respectively.

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, τ_s, of mechanical equilibration of the photo-excited layer, which is given by l and sound velocity, c_0, as[2,3,18]

$$\tau_s \approx l/c_0. \quad (1)$$

By substituting $l \approx 100$ nm from the reflectivity and crater depth measurements[13] and $c_0 = 5720$ m/s (Ref. 19) into Eq. (1), τ_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$10 ns, 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.

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