Photomechanical effects on femtosecond-laser ablation of fused silica studied using time-resolved reflectivity

Takayuki Kumada, Hiroshi Akagi, Ryuji Itakura, Tomohito Otobe, and Atsushi Yokoyama
Kansai Photon Science Institute, Japan Atomic Energy Agency, Kizugawa, Kyoto 619-0215, Japan
kumada.takayuki@jaea.go.jp

Abstract: We observed oscillation of time-resolved reflectivity of femtosecond-laser-ablated fused silica. We ascribe the oscillation to the interference between probe pulses reflected from a sample surface and a thin layer produced by the photomechanical effect.
OCIS codes: (350.3850) Materials processing; (350.3390) Laser materials processing

1. Introductions

In femtosecond laser ablation, materials are ablated not only by the photothermal effects, but also by photomechanical effects driven by the relaxation of laser-induced stresses [1-3]. Linde et al. [4] observed interference fringes (Newton rings) in time-resolved microscopy of metals and semiconductors irradiated with a femtosecond laser pulse with the fluence F_{pump} slightly above the ablation threshold, and attributed the fringes to the interference between probe pulses reflected from sample surface and a thin layer, which is separated from the sample by the tensile stress (spallation). The thin layer is produced only when materials are ablated by the photomechanical effects. Therefore, observation of the thin layer is important on the interrogation of the photomechanical effects on femtosecond laser ablation.

The interference fringes have been observed only on metals and semiconductors, but not on transparent dielectrics. It has been concluded from this result that transparent dielectrics are directly vaporized or fragmented. Since bandgaps of transparent dielectrics are much wider than the photon energies of infrared and visible pump laser pulses, electronic excitation is initiated by multiphoton absorption, and then accelerated by avalanche ionization processes. The latter process leads to an abrupt increase in density of photo-excited electrons (carriers) above the ablation threshold, and cause optical breakdown. It has been believed that thin layer cannot be produced at such heavily excited conditions.

However, we suspected that, even if thin layer is generated on transparent dielectrics, it could not be observed by the time-resolved microscopy. Reflectivities of transparent dielectrics are much less than those of metals and semiconductors, whereas clear contrast in reflectivity ($\Delta R \approx 10\%$) between constructive and destructive interferences is needed to recognize the fringe in the time-resolved microscopy.

In this study, we succeeded in observing oscillation with an amplitude $\Delta R \approx 0.1$ % in the time-resolved reflectivity of fused silica for the first time. Similar to the interference fringes in the microscopy, this oscillation is attributed to the interference between sample surface and the thin layer produced by the spallation mechanism.

2. Experiments

A femtosecond laser pulse from Ti:Sapphire regenerative and multi-pass amplifiers (Thales α 10US-A, Δt = 60 fs, 10 Hz) was separated into pump and probe pulses. The *s*-polarized pump pulse (λ_{pump} = 795 nm) was focused onto a sample surface with an incident angle of 47 ± 2° and a diameter of 50–170 μm. The *p*-polarized probe pulse (λ_{probe} = 795 and 398 nm) was focused with the angle $\theta_0 = 56 \pm 2°$, which is close to the Brewster angle of transparent dielectrics, and a diameter of 10–30 μm. In contrast to the time-resolved microscopy whose probe pulse measured an entire image of the focal spot of the pump pulse, our probe pulse was focused specifically at the center of the focal spot of the pump pulse. The sample was moved along its surface plane during the measurement so that a fresh spot could be exposed at every laser shot.

3. Results

Figure 1(a) shows the time-resolved reflectivity of silicon. After the pump pulse is irradiated at t = 0 ps, the reflectivity oscillates with a period $t_{\mathrm{period}} \approx 500$ ps. This oscillation is attributed to the repetition of constructive and destructive interferences between probe pulses reflected at the sample surface and thin layer produced by the spallation mechanism as a function of the distance between them. If one assumes that the thin layer is separated at a constant velocity v, the increment of the distance d_{period} during t_{period} is related to v and λ_{probe} using the Bragg's law,

$$d_{\mathrm{period}} = v\, t_{\mathrm{period}} = \lambda_{\mathrm{probe}} / (2 \cos \theta_0). \quad (1)$$

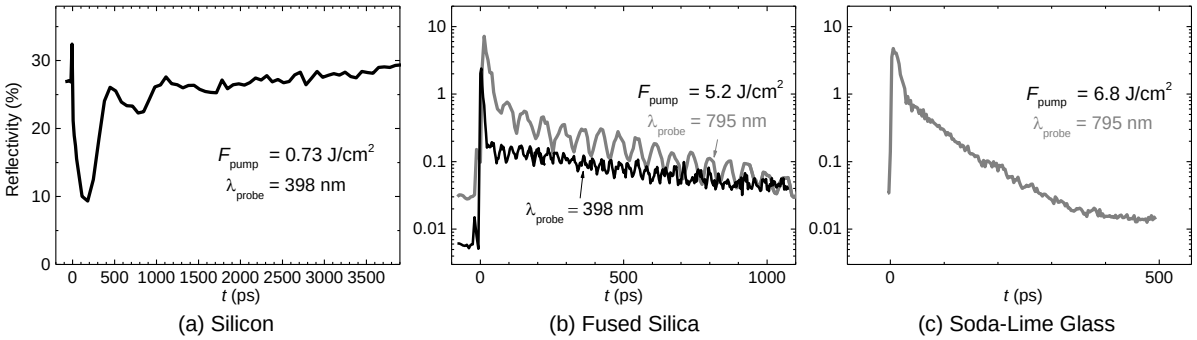

Fig. 1. Time-resolved reflectivities of silicon, fused silica, and soda-lime glass.

The velocity v is determined from Eq. (1) to be approximately 700 m/s, which is consistent with the results of the time-resolved microscopy [4].

Figure 1(b) shows the reflectivity of fused silica. After the reflectivity drastically increases due to optical breakdown at t = 0 ps, it oscillates with t_{period} of 64 ps for λ_{probe} = 795 nm and 32 ps for λ_{probe} = 398 nm. Since t_{period} is proportional to λ_{probe}, the oscillation is attributed to the interference. Similar to the result of silicon, we concluded that this oscillation is due to the interference between probe pulses reflected at the sample surface and thin layer. The oscillation amplitude is comparable to the reflectivity at each t, indicating that the reflection coefficient at one surface is comparable to that at the other. This is the strong evidence that photo-excited layer is separated into two parts, one of which is left on the sample surface and the other separated from the surface by the spallation mechanism. It is interesting to note that the oscillation is observed even at F_{pump} = 15 J/cm^2, which is five times higher than the ablation threshold (not shown), indicating that the thin layer is produced even at such high F_{pump} .

Figure 1(c) shows the reflectivity of soda-lime glass. Although the reflectivity increases at t = 0 ps in a similar manner to that of fused silica, it decreases much faster than that of fused silica without showing any oscillation. This result suggests that the thin layer is not produced. The remarkable decrease in the reflectivity is probably due to absorption of the probe pulse by fragments.

4. Discussion

Our result of fused silica opposes the prevailing idea that transparent dielectrics, which are excited intensely through the avalanche process, would be directly vaporized or fragmented. The separation velocity of the fused silica layer (11000 m/s) determined by substituting t_{period} into Eq. (1) is an order of magnitude higher than that of metals and semiconductors [4]. This result is consistent with the idea that transparent dielectrics should be excited much more than metals and semiconductors. We consider that fused silica forms thin layer because of its exceptionally high viscosity at high temperatures. Fused silica does not have a specific melting point at which first-order phase transition to liquid occurs. Instead, as temperature increases, the viscosity of fused silica continuously decreases. For example, at the softening point of fused silica (1660°C), viscosity of fused silica is ten orders of magnitude higher than that of silicon, and sixth orders than that of soda-lime glass. It is tough to decompose such high viscous layer of fused silica into fragments, whereas soda-lime glass can be fragmented due to such relatively low viscosity.

5. Summary

In this study, we found the experimental evidence of the spallation in fused silica. In contrast to conventional theories, our results suggest that the photomechanical effects play an important role on femtosecond laser ablation of transparent dielectrics. We speculate that the photomechanical effects are not minor effect but generally play an important role on femtosecond laser ablation of a variety of materials at various laser conditions.

[1] S. I. Anisimov, N. A. Inogamov, A. M. Oparin, B. Rethfeld, T. Yabe, M. Ogawa, and V. E. Fortov, "Pulsed laser evaporation: equation-of-state effects," Appl. Phys. A **69**, 617-620 (1999).

[2] P. Lorazo, L. J. Lewis, and M. Meunier, "Thermodynamic pathways to melting, ablation, and solidification in absorbing solids under pulsed laser irradiation," Phys. Rev. B **73**, 134108 (2006).

[3] C. Wu and L. V. Zhigilei, "Microscopic mechanisms of laser spallation and ablation of metal targets from large-scale molecular dynamic simulations," Appl. Phys. A **114**, 11-32 (2014).

[4] D. von der Linde and K. Sokolowski-Tinten, "The physical mechanisms of short-pulse laser ablation," Appl. Surf. Sci. **154-155**, 1-10 (2000).