Ultrafast UV laser induced dynamics in dielectric coating materials before laser damage

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ABSTRACT

The laser induced ultrafast dynamics in fused silica under few-cycle UV laser excitation is investigated in the present study. Using sub-10 fs UV laser pulses, we characterize the free carrier dynamics in the fused silica before laser damage. After laser excitation, free carrier in the conductive band is found to turn into self-trapped excitons within about 300fs. It is possible that the trapped exciton will result in the incubation effect under the condition of ultrafast high-frequency pulsed UV laser exposure.

Keywords: ultrafast laser spectroscopy, UV laser, self-trapped excitons, induced absorbance change

1. INTRODUCTION

Femtosecond laser-induced damage of dielectric coatings has always been a limiting factor for the stable operation of high-power laser system, because the development of lasers with ever higher average powers and peak intensities demands optical components like mirrors and gratings with high breakdown thresholds.[1-4] Especially, laser breakdown thresholds have been found to act as a function of pulse duration in the oxide dielectric films, which decrease when the laser pulse duration becomes shorter.[5] Also, The threshold fluence decreases with the number of pulses until it reaches the multiple pulse threshold, which is due to the accumulation of laser-induced defects during the pulse train together with the initial defects in the coating materials. Except the native defects, which is intrinsic to the material or stemmed from the material manufacturing coating process, the laser induced defects result from the self-trapped excitons forming states in the band-gap zone. Therefore, it is practically important to study the femtosecond pulse-matter interaction in the dielectric coating materials for the sake of enhancing the laser resistance of coatings. One the other hand, understanding the fundamental process of femtosecond pulse-matter interaction is desirable for fine controlled ablation for the laser micro-structuring. The structural modifications in dielectric materials will be induced by the femtosecond laser pulses, [6] which are originated characterized by multiphoton absorption and avalanche ionization.

Therefore, it is desirable to gain a thorough understanding of the physical processes involved in multiple-pulse interaction with the dielectric material, which is promising to reducing the laser induced material deterioration and helpful for the improvements of laser manufacturing methods. Even though there are already plenty of studies on femtosecond pulse-matter interaction to investigate the improvement of the laser-induced damage threshold of mirror coatings and/or laser micro-structuring, most of them are investigated by laser pulses excitation centered at 800 nm in the

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femtosecond regime. A lot of theoretical studies also have been made based on these experimental reulsts. The experimental study on the interaction between dielectric coating materials and few-cycle UV laser pulses has not yet been reported to the best of our knowledge. However, to what extend the laser induced dynamics under the excitation of laser pulse at 800 nm agree with the one using 400 nm excitation, it is still ambiguous.

As mentioned above, although many research studies have been carried out on carrier dynamics inside dielectric coating materials, for example inside the fused silica,[6,7] the basic ionization mechanisms of fused silica under direct excitation of ultrafast UV laser is not well understood, which points out the need to specify induced dynamics in dielectric coating materials under ultrafast UV laser excitation before laser damage. Furthermore, fused silica has attracted significant attention because of its technological importance both in photonics and in electronics. To gain a better understand of the multiphoton and avalanche ionization mechanisms, broadband ultrafast UV femtosecond laser pulses centered at 400nm with the pulse duration of 9 fs was used to excite the ultrafast carrier dynamics inside the fused silica, for the first time to the best of our knowledge. Laser induced difference absorbance changes of free carrier dynamics was measured with 1fs per step, and self-trap process from free carrier to self-trapped exciton has been observed.

2. EXPERIMENTS AND SAMPLE

In the present study, we used ultrashort UV femtosecond laser pulses centered at 400nm with pulse duration of 9fs in the inside the fused silica. Both pump and probe pulses were generated by a hollow fiber compressor system, and the laser system setup was shown in Fig. 1.[9,10] A commercial Ti:sapphire laser system (Coherent, Legend-USP) centered at 800nm operating at 1 kHz was utilized as the laser source, which provide 2.5 mJ pulse energy with the pulse duration of 35 fs. The fundamental laser pulse was reduced to about 900 μJ and then frequency doubled in a 200-μm-thick beta barium borate (BBO, type I, $\theta = 29.2°$) crystal. About 90 μJ pulses at 400 nm were focused into a hollow fiber filled with 0.8 atm argon gas, which has a 140 μm inner diameter and a 60 cm length. The output pulse energy after the hollow fiber was about 45 μJ. The output beam diameter was first reduced to about 2 mm by using two concave aluminum mirrors. Then, a pair of Brewster-angle-cut fused silica prisms and another dispersion compensating system composed of a grating and a deformable mirror was used for the dispersion compensation of the laser pulse. Clean sub-10 fs pulses were obtained with energy satellites smaller than 3%. The sub-10 fs pulse duration was characterized with a sample-cell window from a broken sample cell by using the self-diffraction frequency resolved optical gating (SD-FROG).

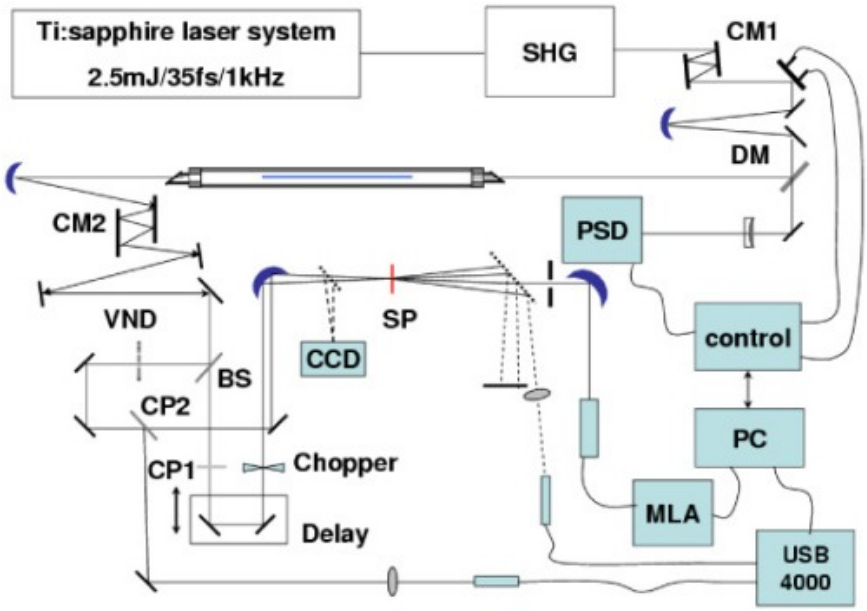

Figure 1. The 400nm hollow-fiber compressor and the pump-probe experimental setups; SHG: second-harmonic generation; CM1, CM2: chirped mirror; DM: dichroic mirror; VND: 0.1-mm-thick variable neutral-density filter; BS: 0.5-mm-thick beam splitter; CP1: compensate plate for VND; CP2: compensate plate for BS; SP: sample for pump-probe spectroscopy or BBO crystal for pulse characterization; MLA: 128-channel multi-lock-in amplifier; PSD: position sensing detector.; USB 4000: spectrometer. CCD: Charge Coupled Device (CCD) camera.

In the pump−probe experiment, the pump and probe configuration is the same as the SD-FROG measurement. The pump and probe beam diameters at $1/e^2$ on the dielectric coating material sample were measured to be about 110 and 105 μm, respectively, by a CCD camera. The pump and probe pulse energies can be tuned by using a step-variable 0.1-mm-thick neutral density (ND) filter. In the experiment, the pulse energies of the pump and probe beams were adjusted to be about 130 and 15 nJ, respectively. The pump-probe signal was collected using a bundle fiber, and then dispersed by a polychromator. And then the signal was guided to the photodetector by a 128-channel bundle fiber. The spectral resolution of the total system, which is composed of the monochromator, bundle fiber, and photodetector, was about 1.5 nm. All of the experiments were performed at a constant room temperature.

The SiO_2 films were deposited by electron beam evaporation with temperature 200 °C and a base pressure of 5×10^{-5} mpar. The substrates had been ultrasonically cleaned in petroleum ether before deposition. The film thickness were determined by the ellipsometer, and the optical thickness is $3\lambda/2$ (λ=400 nm). The deposition rate was approximately 0.3 nm/s.

3. RESULTS AND DISCUSSION

Since the high photon energy of the UV light, UV is more sufficient in generating seed electrons, which suggests that the multiphoton ionization is very easy to realize even using the pump energy of 130 nJ. Figure 2 shows the time dependence of the transient absorbance changes (ΔA) probed inside fused silica from 360nm to 440nm plotted in two dimensions. Using degenerate probe laser pulse with much weaker intensity than the pump, free carriers generated by femtosecond laser-multiphoton ionization have been observed. The results indicate that the observed ΔA signal are positive around 420 nm, which suggestion that what we observed is due to the laser-induced absorption, not bleaching or stimulated emission.

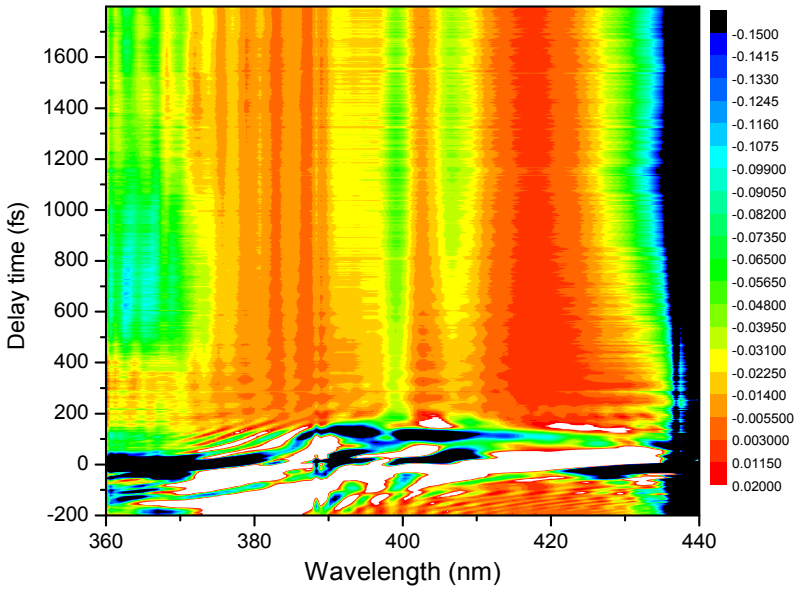

Figure 2. Two-dimensional pseudo-color display of the time dependence of the absorbance changes (probe photon energy versus probe delay time).

Fig.3 shows a typical time-resolved transient absorption signal observed from -200 fs to 1800 fs at 420 nm, which is picked out from the two dimensional plot shown in Fig. 2. Laser induced absorption signal of free electrons is observed to undergo an ultrafast decay. Because we are applying a probe pulse with rather weaker laser energy than the pump pulse, the observed dynamics is purely due to the pump induced material characteristics in fused silica, which means the probe is just used for detection and will not influence the pump induced dynamics anymore. Using this method, we can study the pure dynamics of free electron, for example the self-trapped exciton formation. The lifetime of the free carriers is calculated to be 300±5 fs using biexponential fitting method. It indicates that the free carriers generated by femtosecond laser-multiphoton ionization in the conductive band are trapped into self-trapped excitons after an ultrafast decay within about 300fs.

Even though we using weak-energy probe pulse, it seems we can still observe the effect of self-trapped exciton on multiphoton ionization. As shown in figure 3, we notice that the real-time trace does not go to zero after the trapping, but it decays very slowly and the final lifetime is beyond our measure time. As reported in previous studies in fused silica, the self-trapped excitons have a lifetime ranging from 30 to 300 ps.[11] Reasonable explanation is that these trapped excitons generated by pump laser can be excited and ionized by the probe pulse and turned into the free electrons again after trapping, which is considered to be the contributors of the long-time term in the real-time traces. It is well known that the damage in the fused silica occurs when the free electron density reaches a critical density value. Therefore, the trapped excitons with much longer lifetime than the initial generated free carriers are probably also one of the reasons for the incubation effect occurred in the laser damage experiments under the condition of ultrafast high-frequency pulsed UV laser exposure.[5]

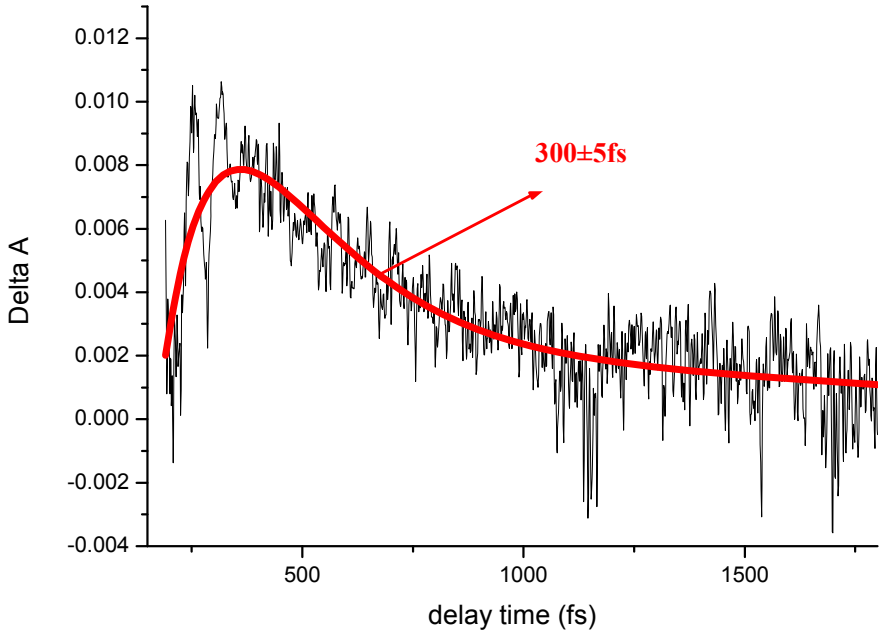

Figure 3. Real-time difference absorption trace at 420nm.

4. CONCLUSION

Using ultrafast UV laser, we have observed induced dynamics in dielectric coating materials before laser damage. Furthermore, we have shown that due to the laser-induced absorption the observed ΔA signal are positive around 420 nm. Meanwhile, within about 300fs, the free carrier in the conductive band will trap into self-trapped excitons. It is possible that these trapped excitons are excited and ionized after trapping.

5. ACKNOWLEDGEMENT

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