From Carrier Dynamics inside Fused Silica to Control of Multiphoton-Avalanche Ionization for Laser Machining

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Abstract: Using pump-probe measurements, we characterize carrier decay time inside fused silica and measure deeply bound self-trapped excitons. With pump-probe delay, we also control free carrier injection and the subsequent avalanche process for laser machining applications.

OCIS codes: 190.4180 Multiphoton processes, 320.7130 Ultrafast processes in condensed matter, including semiconductors

1. Introduction

The electron plasma induced by tightly focused femtosecond laser pulses in fused silica can cause structural modifications that form the basis for many interesting applications, for example, the formation of microscale three-dimensional optical elements [1-3]. The fundamental process is originated through a nonlinear ionization process characterized by multiphoton absorption and avalanche ionization. Although considerable amount of knowledge has been gained in the past years on carrier dynamics during nonlinear ionization inside fused silica [4-7], the relationship between the basic ionization mechanisms and the permanent modifications in the bulk of fused silica is not well understood, which hinders our ability to control the process with ultimate precision in materials modification. In this study, we use pump-probe technique with varied relative laser intensities to measure time-evolution of free carrier dynamics. The objective is to understand multiphoton and avalanche ionization mechanisms so that we can control them for laser machining applications.

In the experiments, we use 800 nm femtosecond laser pulses with three different pulse durations (48, 85, 140 fs) and peak energy up to few hundred nanojoules. Each laser pulse is split into a pump and a probe and later recombined into a collinear beam with variable delays from 0 to about 2 ns between them. The beams are focused inside a fused silica sample with a microscope objective (NA=0.25) approximately 150 μm from the incident surface. Free electrons are injected by the linearly polarized pump pulse. The probe pulse is cross polarized and follows the pump with a preset time delay. The transmitted light is collected and collimated using another microscope objective with a higher numerical aperture (NA=0.5). The sample is continuously moved during the measurements at a speed high enough for the laser repetition rate so that each laser shot is fired on a fresh spot.

2. Free Carrier Trapping Dynamics

Figure 1 shows a time-resolved measurement of the transmission of a weak probe (E_{probe}=7 nJ) as a function of delay between pump and probe inside fused silica. The pulse duration is 48 fs. The pump energy of 64 nJ is above the ionization threshold and it produces an underdense microplasma estimated at a density of $n_e \sim 5\times10^{19}$ cm^{-3}. The black transmission curve indicates an ultrafast decay of free electrons, and the trapping time is estimated by the blue line as 162±8 fs. Using longer pump pulses (85 fs and 140 fs), we change the free carrier density and repeat the measurements. The results suggest similar ultrafast trapping of free carriers with exponential decay.

3. Lifetime of Deeply Bound Self-Trapped Excitons

Free carriers injected by a pump pulse are trapped in deep self-trapped excitons after an ultrafast decay within about few hundred fs. These trapped excitons are readily ionized with a relatively weak probe pulse. Using a probe of 49 nJ which is slightly below the ionization threshold for the conditions used, Figure 2 shows that the lifetime of the self-trapped electrons extends beyond 2 ns as indicated through time-resolved multiphoton absorption of the probe pulse.

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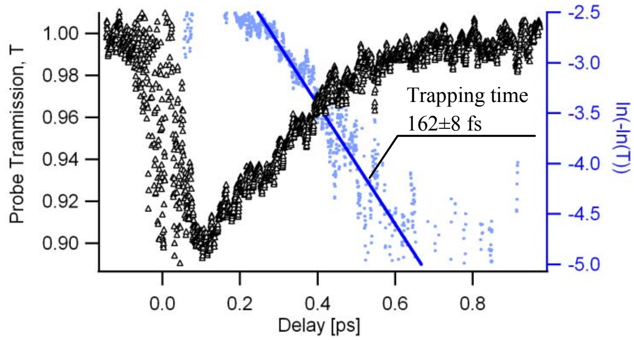

Fig. 1: Variation of the linear transmission of a focused weak probe (E_{probe}=7 nJ) with delay time. The pump energy is 64 nJ which produces an underdense microplasma (n_e~5×10^{19}cm^{-3}). The blue line fit to the data points indicates an exponential decay of free carriers.

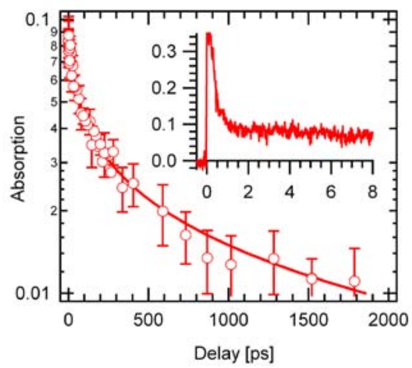

Fig. 2: Variation of the absorption of a focused probe pulse with delay time. The probe energy is E_{probe}=49 nJ. The long lasting absorption curve indicates a long lifetime with a non-exponential decay. The inset shows the initial fast decay process.

4. Control of Multiphoton and Avalanche Ionization for Laser Machining Applications

Our measurements show that, once they are freed, the bound or self-trapped exciton electrons can be used as seed electrons for avalanche absorption by the probe pulse. We will show that the seed electrons allow us to show that avalanche ionization plays a role in femtosecond multiphoton breakdown. The test is independent of any model assumptions.

5. Conclusions

Using pump-probe methods we have measured ultrafast free carrier dynamics in fused silica and the room temperature life time of trapped excitions. Furthermore, we have shown that a pump-probe pulse pair can also be used for controlling avalanche ionization. If the pump pulse is 3rd or higher harmonic of the fundamental 800 nm laser light, then it should be possible to extend controlled laser machining to the nano-scale.

6. References

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