Tailored Femtosecond Pulses for Nanoscale Laser Processing of Dielectrics

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Abstract. Laser control of two basic ionization processes in dielectrics on intrinsic time and intensity scales with temporally asymmetric pulse trains is investigated. We create robust structures one order below the diffraction limit.

Introduction

Lasers delivering ultrashort pulses have emerged as a promising tool for processing wide band gap materials for a variety of applications ranging from precision micromachining on and below the wavelength of light to medical surgery [1] [2]. Within this context it is the transient free-electron density in the conduction band of the dielectric that plays a fundamental role in addition to various propagation and relaxation mechanisms. A large number of experiments makes use of the threshold of observed damage as experimental evidence for exceeding a certain critical electron density after the laser interaction. These involve pulse duration measurements [3] [4] [5] and recent pulse-train experiments [6] all showing a strong dependence of the damage threshold on pulse duration and on pulse separation. Direct studies of transient electron densities range from intensities below [7] [8] up to well above the breakdown threshold [9] [10]. The temporal evolution of the free-electron density and the role of the fundamental ionization processes are strongly depending on time and intensity as well as on the instantaneous frequency [11] [12] [13]. Two main processes for generating free electrons are multi photon ionization (MPI) and Avalanche-Ionization (AI). MPI requires no free initial free electrons and has highest efficiency for shortest pulses AI on the other hand needs initial free electrons and needs time to establish. In our work we make use of temporally asymmetric femtosecond pulses in order to control MPI and AI. Control leads to different final electron densities (and energies) as the direct temporal profile and the time inverted profile address the two ionization processes in a different fashion. This results in observed different thresholds for material modification in fused silica as well as in reproducible lateral structures being an order of magnitude below the diffraction limit.

Experiment

In our experiment we combine femtosecond pulse shaping techniques [14] with a microscope setup for material processing [15] [16]. Linear polarized laser pulses with 35 fs full width at half maximum (FWHM) pulse duration and a central wavelength of 790 nm are provided by an amplified Ti:Sapphire laser system. After

passing a calibrated home built spectral phase modulator [17] the pulses are focused via a 50 x 0.5 NA objective to a spot diameter of 1.4 µm ($1/e^2$ value of intensity profile). The pulse shaper is operated in a parameter regime far away from severe space time coupling effects [18] as can be seen for ex. from quantum optical measurements with the same set up [19]. Shaped pulses are characterized in the interaction region. The sample is translated by a 3-axis piezo table to a new position for each shot. A typical measurement pattern consists of an array of points where we vary the pulse shapes, energy and focal z-position. After laser processing the samples are analyzed via scanning electron microscopy (SEM).

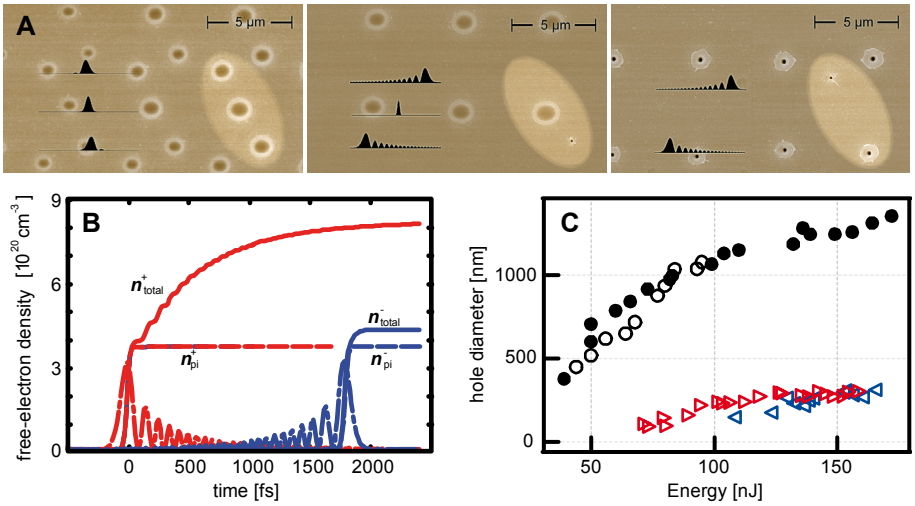

Fig. 1: A) SEM micrographs of a measurement pattern on fused silica: For an applied energy E and focal position a triplet of applied laser pulses is highlighted by the ellipse. Negative, zero and positive TOD were used. Normalized temporal intensity profiles are sketched for comparison between different TODs. *Left:* low TOD (statistic pulse duration of 2 σ = 50 fs; E = 77 nJ) results in negligible differences between created structures. *Middle:* high positive TOD (statistic pulse duration of 2 σ = 960 fs; E = 71 nJ) results in a change of structure size and threshold energy. *Right:* the threshold energy for ablation with high negative TOD is reached with E = 110 nJ. Here the unshaped pulse is suppressed in order not to mask structures with TOD. **B)** Transient free-electron density n_{total} (solid lines) as calculated with help of the MRE, together with the density of electrons provided by photoionization n_{pi} (dashed lines) and the corresponding transient intensities (dashed-dotted lines) of the pulse with positive TOD (index +) and negative TOD (index -), respectively. **C)** Diameters of ablation structures as a function of pulse energy for unshaped pulses (circles), for (+) shaped pulses (triangles pointing right) and for (-) shaped pulses (triangles pointing left). Without changing the focus spot diameter structures below 300 nm are obtained over a large energy range thus providing a large process window for creation of nanostructures. The smallest structures are about 100 nm in diameter.

Results and Discussion

Systematic studies with phase shaped laser pulses based on third order dispersion (TOD) leading to asymmetric temporally shaped laser pulses revealed a change in the threshold depending on whether the direct pulse shape or the time inverted profile

was used (see Fig. 1A). Theoretical simulations based on a multiple rate equation (MRE) model described in [12] show, that it is the timing of an intense photoionizing sub-pulse that can turn on or off AI [16] (see Fig. 1B). The observed nanoscale structures are an order of magnitude below the diffraction limit and remarkably stable with respect to variations in laser fluence (see Fig. 1C) [20]. We propose that it is the interplay of MPI creating free electrons in a spatially very confined region followed by AI that restricts the area of reaching the critical electron density (and energy) that may eventually lead to the nanoscale structures seen for positive and negative TOD pulses.

Conclusions

We conclude that control of ionization processes with tailored femtosecond pulses is an important prerequisite for robust control of laser processing of high band gap materials and that our strategy opens the route to develop tailored pulse shapes for controlled nanoscale material processing of dielectrics.

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