Tailored Femtosecond Pulses for Nanoscale Laser Processing of Dielectrics

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Abstract

We present an experimental demonstration of laser control of two basic ionization processes in dielectrics on intrinsic time and intensity scales. To that end we generate temporally asymmetric pulse trains and double pulse sequences on the femtosecond time scale and show, that it is the timing of an intense photoionizing sub-pulse that can turn on or off electron-electron ionization. This is observed via different thresholds for surface material modification. The thresholds are determined with the help of Scanning Electron Microscopy. We create robust structures one order below the diffraction limit. Additionally we discuss simulations on the free electron density. These simulations support the control statement. Our approach opens the route to develop tailored pulse shapes for controlled nanoscale material processing of dielectrics.

Keywords: femtosecond pulse shaping , nanoscale processing of dielectrics, control of ionization processes in dielectrics

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] [3] [4].

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 that in the end lead to phase transitions or the creation of voids. A large number of experiments devoted to the study of the microscopic ionization processes 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 [5] [6] [7] and recent pulse-train experiments [8] 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 [9] [10] up to well above the breakdown threshold [11] [12]. These studies show that below the breakdown threshold multi photon ionization can be invoked as a main ionization mechanism and well above the threshold the plasma reflectivity prevents further energy deposition into the material.

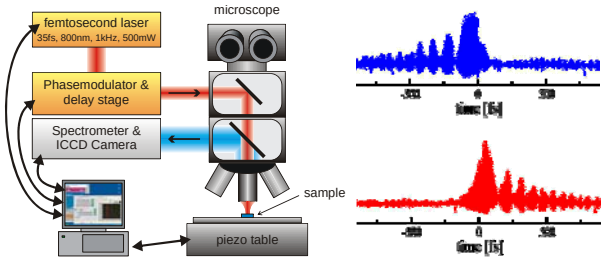

Fig. 1:Left: Schematic of experimental set up. Right: Crosscorrelation of asymmetric shaped pulses (a cubic phase was applied in the frequency domain)

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 [13] [14] [15] [16]: While photoionization (usually multi-photon ionization) generates electrons with low kinetic energy in the conduction band, impact ionization requires electrons of high kinetic energy exceeding the band gap energy. The absorption of this additional energy from the laser light by intraband absorption proceeds on a timescale in the femtosecond range as the process of

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Drude absorption involves microscopic collisions on a femtosecond timescale. Its probability grows with laser intensity.

In our work we make use of temporally asymmetric femtosecond pulses of identical fluence and identical statistical pulse duration in order to control photoionization and electron-electron impact ionization. Control leads to different final electron densities 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 [17] with a microscope setup for material processing (see Fig. 1) [18,19]. 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. The prism compressor in the amplifier is adjusted to precompensate the dispersion in the subsequent beam path. The pulses are split into two beams using a Mach-Zehnder interferometer. One arm includes a calibrated home built spectral phase modulator [4] while the other arm contains a stepper motor driven delay stage. After the interferometer the pulses are focused via a 50x 0.5 NA objective to a spot diameter of 1.4 µm ($1/e^2$ value of intensity profile). For pulse diagnostics a two-photon photodiode is placed in the interaction region. Shaped pulses are characterized via their implemented phase functions occasionally checked via direct crosscorrelation measurements (see Fig. 1).

For material processing phase shaped femtosecond pulses or double pulse sequences are focused onto the samples. 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 chemically cleaned and coated with platinum for post-mortem scanning electron microscopy (SEM) analysis.

Results and Discussion

In certain parameter ranges we obtain reproducible lateral substructures that are an order of magnitude below the diffraction limit. By timing an asymmetric double pulse sequence, these substructures can be switched on and off (see Fig. 2).

Systematic studies with phase shaped laser pulses based on third order spectral phase modulations (cubic phase) leading to asymmetric temporally shaped laser pulses (see Fig. 1) [20] [21] revealed a change in the threshold depending on whether the direct pulse shape or the time inverted profile was used (see Fig. 3).

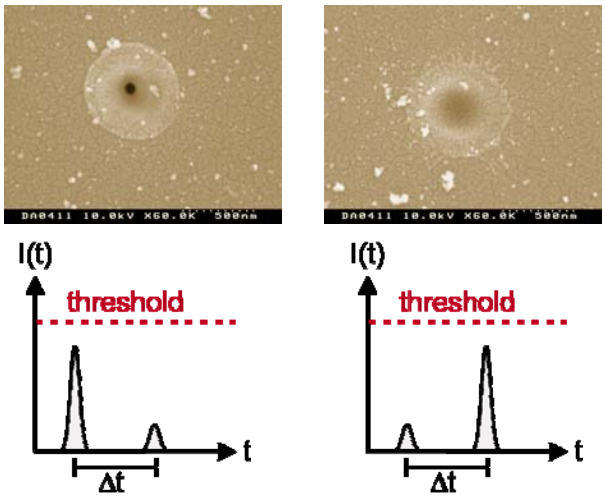

Fig. 2: SEM images of ablation structures in fused silica (top) for double pulse interaction (Δt = 5ps)(below). Left: the observed substructure has a diameter of 70 nm and is 20 times below the diffraction limit

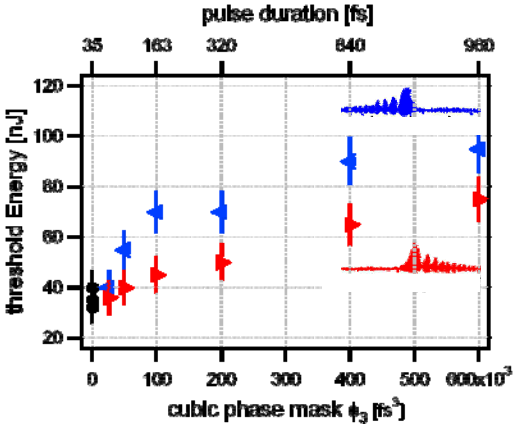

Fig. 3: Threshold for surface damage in fused silica obtained with different cubic phase parameters

Preliminary theoretical simulations based on a model described in [14] show, that the two different temporal profiles address the two ionization mechanisms in a different fashion: it is the timing of an intense photoionizing sub-pulse that can turn on or off electron-electron ionization [22]. The observed nanoscale structures are attributed to propagation effects – primarily filamentation – and are remarkably stable with respect to fluence. We will address this aspect in our future work.

We conclude that control of ionization processes with tailored femtosecond pulses is suitable for robust control of laser processing of high band gap materials.

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