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Ultrafast Breakdown of dielectrics: Energy absorption mechanisms investigated by double pulse experiments

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ABSTRACT

We investigate the mechanisms involved in the modification of dielectric materials by ultrashort laser pulses. We show that the use of a double pulse (fundamental and second harmonic of a Ti–Sa laser) excitation allows getting new insight in the fundamental processes that occur during the interaction. We first measure the optical breakdown (OB) threshold map (intensity of first pulse versus intensity of second pulse) in various materials (Al_2O_3, MgO, α-SiO_2). Using a simple model that includes multiphoton excitation followed by carrier heating in the conduction band, and assuming that OB occurs when a critical amount of energy is deposited in the material, we can satisfactorily reproduce this evolution of optical breakdown thresholds. The results demonstrate the dominant role of carrier heating in the energy transfer from the laser pulse to the solid. This important phenomenon is also highlighted by the kinetic energy distribution of photoelectrons observed in a photoemission experiment performed under similar conditions of double pulse excitation. Finally we show, in the case of α-SiO_2, that the initial electronic excitation plays a key role in the formation of surface ripples and that their characteristics are determined by the first pulse, even at intensities well below OB threshold.

1. Introduction

The use of short laser pulses for micromachining or modifying the optical properties, in the bulk or at the surface of dielectrics, is an extremely active field. Recently it was shown that temporal or spatial shaping of the pulse can lead to large improvement in the efficiency or the final quality of the laser manufacturing process [1,2]. Similarly, it was shown that the use of double pulse sequence can be a way to improve the ablation efficiency for instance or to achieve higher spatial resolution [3]. In order to understand the physics underlying the interaction of complex pulses or the sequences of pulses, and possibly for optimizing the laser parameters, investigation of the elementary processes involved is needed. In this work we report several experiments, in similar irradiation conditions, using a sequence of two short pulses. The main advantage of using such a sequence of pulses is to separate the non-linear excitation part, and the carrier heating part, as suggested in an early work with picosecond laser pulses [4]. Using such an excitation scheme we can control the interaction parameters and thus open new experimental windows and wider possibilities, by playing with the parameters of the two pulses: intensity, wavelength, polarization, duration, and time delay. We report three types of measurement: first the optical breakdown threshold mapping as a function of the intensity of the two pulses. A simple model allowing reproducing the measured thresholds is then presented. Then, photoelectron kinetic energy spectra show direct evidence of the carrier excitation-carrier heating sequence. Finally, a study of the morphology of the ablation craters demonstrates, in the case of quartz (α-SiO_2), that the initial electronic excitation determines the properties of the laser-induced periodic structures.

2. Double pulse Optical breakdown measurements

2.1. Experimental setup and method

The experiments have been made at the Saclay Laser-matter Interaction Center (SLIC: http://iramis.cea.fr/slic/index.php) facility. The laser is a chirped pulse amplification (CPA) Ti–Sa system, delivering up to 70 mJ at 800 nm, with a repetition rate of 20 Hz. The laser beam is initially separated in two beam lines each equipped

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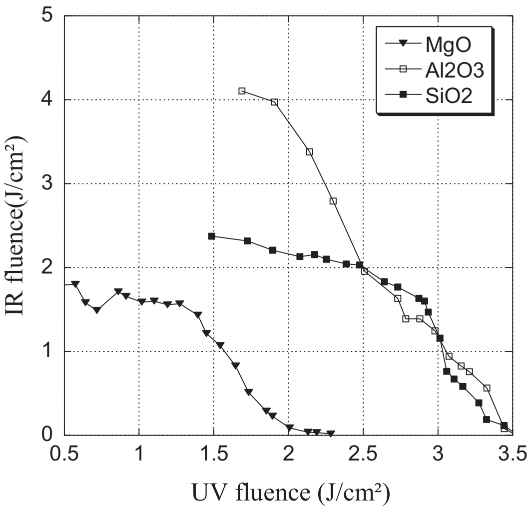

Fig. 1. Surface breakdown threshold for IR pulse at 800 nm (IR) as a function of the intensity of the pre-pulse at 400 nm (UV) for three materials: Al_2O_3 (empty squares), MgO (triangles) and α-SiO_2 (full squares). The delay between the two pulses is 1.5 ps.

with a compressor. We use one beam, frequency doubled, to generate the first pump pulse at 400 nm, and the other beam line for the second pump at 800 nm, with variable duration. The two beams are recombined with a dichroic mirror transparent for the fundamental and reflecting the second harmonic. They are then smoothly focused at normal incidence onto the surface of the sample with 300 mm focal length lens. The beam size has been measured to be 56 μm (at $1/e^2$) for the fundamental beam, and 26 μm (at $1/e^2$) for the second harmonic. To compensate the difference of foci for the fundamental and second harmonic, a long focal length is placed on the beam path of the fundamental before the dichroic mirror. For this set of experiments the duration of the pulse at 800 nm has been set at 300 fs (at FWHM), while the duration of the first pulse is kept at 50 fs (at FWHM). The delay between the two pulses is variable, and has been set to 1.5 ps (peak to peak) for this set of measurements.

We have measured the evolution of the optical breakdown threshold for the second pulse as a function of the intensity of the first pulse. The measurements are performed as follows: the intensity of the first pulse is set at a given value, and the intensity of the second pulse is gradually increased (with the help of a half-wave plate placed before a polarizer) until a permanent modification of the surface can be observed (constrast change). This modification is monitored with a diode laser illuminating the front surface, which is imaged on a CCD camera with a large magnification. The sample is moved between two laser shots (a shot corresponding to a sequence of two pulses), such that we measure the single shot breakdown threshold. The term of damaging or breakdown being subjective different thresholds can be measured depending on the technique used. To check the sensitivity of our measurements, we have also observed the surface of the samples with a SEM microscope (see paragraph below for details). Thus we could validate our optical in situ observation, which proved to be sensitive enough to distinguish the first threshold as defined by any permanent modification of the surface [5], to be distinguished from the ablation threshold, which is defined by material removal. In our investigations, OB threshold has been synonym to damage threshold.

The results of these measurements are reported for three materials, namely Al_2O_3, MgO and α-SiO_2 – with the bandgap 8.7, 7.7 and 9.2 eV respectively – in Fig. 1.

The shapes of the curves are the same for the three types of crystals. This shape is expected and indicates the evolution of breakdown from infrared (IR) alone to ultraviolet (UV) alone, while the UV pulse intensity is increased. This is why we observe a plateau at low UV energies. It should be noted that we have stopped the measurements when the UV prepulse seemed to have no influence on the IR breakdown threshold. Thus the first point of each curve can be considered as the breakdown threshold for the IR pulse alone, within the error bars of our measurements, which is below 5%. Then this plateau is followed by a fast decrease of the IR energy needed to reach the breakdown threshold. This means that the damage threshold is rapidly modified as soon as the UV intensity is high enough to promote a noticeable density of carriers in the conduction band (corresponding to the UV beam absorption threshold).

As can be seen, MgO exhibits the weakest threshold whereas, it is similar, at least at high UV intensities – above 2.5 J/cm^2 – for Al_2O_3 and SiO_2. The only difference between the last two materials is seen at low UV intensities, Al_2O_3 being able to sustain a higher IR intensity.

2.2. Modeling the double pulse breakdown thresholds

In order to explain quantitatively these experimental observations, we have adapted a simple model, which has been developed and successfully used to calculate the ablation craters depth and its evolution with pulse intensity [6], to the double pulse excitation. This model takes into account the nonlinear excitation from the valence band to the conduction band, and the heating of carriers by the laser pulse. This heating can be due to the first exciting pulse itself, or due to the second pulse in the pair of pulses if the carrier lifetime is larger than the delay between the two pulses: this is the case for Al_2O_3 and MgO, having "free" carriers lifetime of several tens of picosecond. We have seen that this heating in the last case is essentially due to sequences of resonantly enhanced single or multiphoton processes within the conduction band [7]. For the seek of simplicity and to avoid a too large number of parameters, we will assume here that this carrier heating process occurs by sequence of single photon processes. Thus the equations describing the interaction are written as follows:

$$\frac{\partial N_1}{\partial t} = N_\nu \sigma_{n_1} F_1^{n_1} + N_\nu \sigma_{n_2} F_2^{n_2} - \beta_1 N_1 F_1 - \beta_2 N_1 F_2$$

$$\frac{\partial N_2}{\partial t} = \beta_1 N_1 F_1 + \beta_2 N_1 F_2$$

$$N_\nu = N_0 - N_1 - N_2$$

$$\frac{\partial F_1}{\partial z} = -n_1 \hbar\omega_1 N_\nu \sigma_{n_1} F_1^{n_1} - \beta_1 N_1 F_1$$

$$\frac{\partial F_2}{\partial z} = -n_2 \hbar\omega_2 N_\nu \sigma_{n_2} F_1^{n_2} - \beta_2 N_1 F_2$$

where β_1 and β_2 are the single-photon absorption cross sections (per photon) for electrons in the conduction band, n_1 and n_2 are the number of photons necessary for multiphoton absorption, N_ν, N_1 and N_2 represent the population density of carriers in the valance band, and in two fictive states of the conduction band separated by one photon energy and σ_{n1} and σ_{n2} are the reduced multiphoton absorption cross sections. F_1 and F_2 are the photon fluxes for the first (UV) pulse of frequency ω_1 and the second (IR) pulse of frequency ω_2 respectively. We assume propagation over extremely short distance – the crater's depth at breakdown threshold does not exceed 100 or 200 nm [6] – and changes in intensity for UV and IR pulses are only due to depletion effect from multiple or single photon processes. The change of reflectivity, which was found to be weak under the present experimental conditions, also contributes to the decrease of the pulse intensity and has been taken into account. The sample is initially at rest, thus N_1 and N_2 are set