Transient interference implications on the subpicosecond laser damage of multidielectrics

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Laser-induced damage in optical thin films with subpicosecond pulses is investigated. A model dedicated to optical interference coatings and based on the rate equation for free electron generation is introduced. It takes into account the transient interference effects induced by changes in the dielectric function during the laser pulse and its feedback effect on the electron density distribution in the multilayer stack. Simulations are compared to experiments on HfO_2 and Ta_2O_5 films with pulses ranging from 45 fs to 1 ps. It is shown that this approach can improve the interpretation of femtosecond and picosecond laser induced damage in thin films. © *2010 American Institute of Physics*. [doi:10.1063/1.3477961]

In the development of subpicosecond lasers, from table top lasers to tera/hexawatt class facilities, laser-induced damage to optical components is one of the main limitations. Such laser systems require specific coatings for controlling the light temporally (pulse compressors or stretchers), spatially (mirrors, beamsplitters, and polarizers), or spectrally (dispersive mirrors), and these coatings are often the weakest part when laser damage resistance is concerned. This issue is then one of the main concerns in the production of high quality coatings for short and ultrashort pulse lasers. In this field, it is of high interest for scientists and manufacturers to have available models for interpretation of damage phenomena and for describing the scaling of laser induced damage threshold (LIDT) as a function of operational parameters of the laser (pulse duration, wavelength, polarization, repetition rate, and number of pulses) and the multidielectric stack (dielectric functions, band gaps, thicknesses, and design of the system). The damage of dielectric materials in the femtoseconds (fs) up to few picoseconds (ps) range can be understood as a result of electronic processes. Free electrons will be generated during the laser pulse up to a level where the electron density in the conduction band reaches a critical density.[1] The free-electron generation in dielectrics can be described by the rate equation (RE) (Ref. 1)

$$\frac{dN}{dt} = W_{\mathrm{PI}}(E) + W_{\mathrm{AI}}(E,N) - W_{\mathrm{Loss}}(N). \quad (1)$$

The variation in the free electron density, N, in the material as a function of time and the electric field, E, depends on the rate of photoionization, (W_{PI}), the rate of avalanche ionization, (W_{AI}), and a relaxation rate, (W_{Loss}), that takes into account relaxation of electrons from the conduction band to lower electronic states. The relative role of photoionization and avalanche ionization in dielectrics excited by fs pulses is in debate in the literature,[2–6] particularly below 100 fs. The different parameters used in avalanche ionization (AI) or photo-ionization (PI) models are also not perfectly known (reduced mass used in the Keldysh expression, and the avalanche coefficient for instance). Furthermore, in the case of thin film materials, the material parameters can be quite different from their bulk counterparts and depend also on the deposition process. Then only the trends predicted by these models are usually used to interpret data. The RE has recently been used to scale the fs/ps-LIDT in dielectric thin films as a function of laser (pulse duration,[7] wavelength[8]) or material parameters (band gap).[7] It was prove to be a powerful approach on which we will base our interpretations in the following. In the case of photoionization, free carrier in the conduction band can arise either from tunneling (TI) or multiphoton (MPI) ionization. Using the Keldysh parameter,[9] it appears than in our experimental conditions, TI or MPI can occur. In this study, we then used the so called exact Keldysh theory for photoionization.[9] The corresponding reduced electron-hole mass for calculation of W_{PI} in the Keldysh expression is not known for the materials under study, especially in thin film form, and is usually used as a fit parameter in the literature. It has been set to the electron mass in our calculations. The Drude model of free carrier absorption has been used to get the avalanche coefficient. In this model another parameter, the Drude relaxation time, is unknown precisely for the materials under study. It has been set to 10^{-15} s (Ref. 1) in the calculations. One of the particularities of thin films is the presence of optical interference effects that induce an electric field distribution that has to be taken into account for interpretations. The introduction of a correction coefficient in the RE has been suggested to deal with interference effects in optical coatings.[10] Using the RE, an arbitrary damage criterion must also be set. It has been proposed and used in numerous studies that damage occurs when the plasma frequency reaches the laser frequency,[2] i.e., where linear absorption of the laser energy starts to increase strongly and a runaway process can take place. The free electron density in this case reaches values of $10^{21}-10^{22}$ cm^{-3}. Such electronic densities strongly affect the dielectric function of the material under irradiation.[11,12] The electronic density dependence of the refractive index can be described by the Drude model of free electron gas[13]

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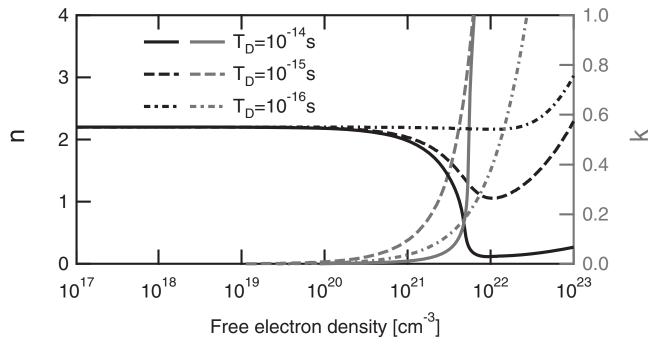

FIG. 1. Variation in the real (n) and imaginary (k) part of the complex index as a function of the free electron density, considering Eq. (2).

$$\tilde{n}(N) = \sqrt{n_0^2 - \frac{Ne^2}{m^* \epsilon_0} \frac{1}{\omega^2 + i\omega/\tau_D}}, \tag{2}$$

where n_0 is the refractive index of the unexcited material, m^* is the effective mass of electrons, ϵ_0 is the free space dielectric permittivity, ω is the laser pulsation, and τ_D is the Drude relaxation time. For illustration the variation in the complex refractive index of a coating material (hafnia) with electronic density is given in Fig. 1.

These changes in complex index will lead to modifications of the electric field repartition during the laser pulse. At the location of the peaks of standing electric-field, the optical response of the dielectric material initially due to the bound electrons will gradually acquire a metal-like behavior with free carrier effects. This can totally change the electric-field repartition during the pulse and the prediction obtained with models when applied to optical interference coatings. The ultrafast evolution of reflection and transmission properties of thin films during subpicosecond irradiation has experimentally been shown and attributed to the electric film modulation across the film.[11] In order to deal with these phenomena, the dependence of the electric field repartition on the complex index variations has to be taken into account. It can be done by adding the spatial and temporal variations in the electric field in the RE

$$\frac{dN^i(z,t)}{dt} = W^i_{\mathrm{PI}}[E(z,t,N^i)] + W^i_{\mathrm{AV}}[E(z,t,N^i),N^i] - W_{\mathrm{Loss}}[N^i,t], \tag{3}$$

with the superscript "i" referring to the layer, i, of the stack. Practically, to solve this equation each film "i" of the stack can be divided in slices located at z_j, each slice being characterized by its electronic density, N_j, and refractive index, $n_j(N_j)$. The problem is then equivalent to a multilayer stack in which the electric field has to be recalculated for each time iteration when solving Eq. (3). This approach involves coupled differential equations that are solved numerically. The electric field distribution is calculated with the matrix method, which offers the ability to change the polarization and the incidence angle of the incoming wave.[14] A pulse laser with a Gaussian temporal shape is considered but arbitrary shapes can easily be implemented (addition of a pedestal for instance). The pulse however is considered to have a negligible spectral dispersion which can be a limitation of our approach in some multilayer systems. The spatial pulse length needs also to be large compared to the film thickness.

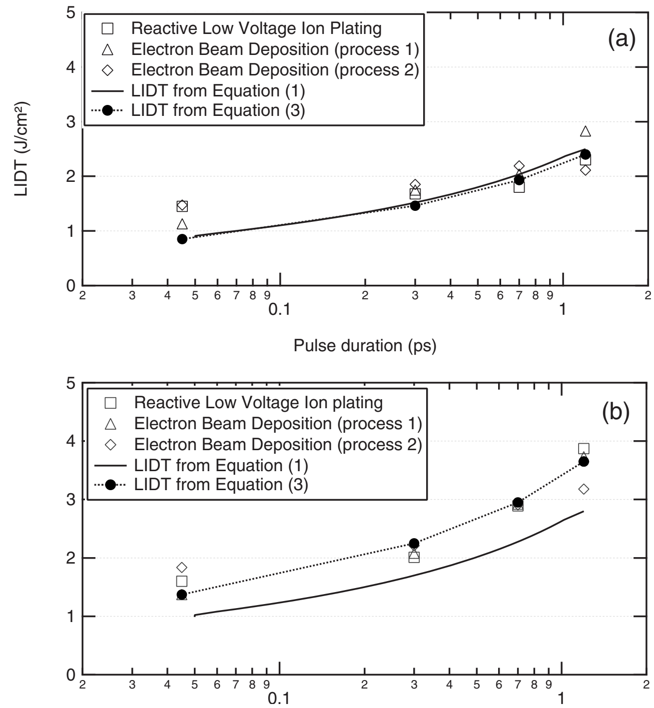

FIG. 2. Comparison of experimental measurements to theoretical prediction. (a) 2H hafnia single layers; (b) 2H/3 hafnia single layers.

As an experimental illustration of this effect, we chose two simple cases involving in each case only one material. One set of samples consist of hafnia single layers deposited on superpolished fused silica substrates. Two thicknesses were chosen (2H and 2H/3, where H represents one-quarter wave layer at 1030 nm), leading to different intensity repartitions and values in the stack. In the case of 2H thickness the intensity has two maxima: at the Air/film and film/substrate interface. In the second case (2H/3), it is maximum at the layer/substrate interface. The thin film properties depend on the deposition process and their characteristics can change with the deposited thickness. This can increase the complexity for interpretations. In order to bypass this difficulty, different set of samples were manufactured with three deposition process involving different materials and methods (see Ref. 15 and other references therein for the detailed deposition processes and characteristics). To avoid any experimental artifact resulting from the damage tests, three different independent facilities were used for this study (complete details of the measurements will be presented in another paper). The samples were tested with 1030 nm pulses of 45 fs, 300 fs, 670 fs, and 1.2 ps. The focusing element on each facility has been chosen in order to obtain laser spot sizes in the same range, i.e., between 30 and 50 μm. Single shots were used in order to avoid any incubation or fatigue effects known to occur in thin film materials. A statistical approach involving 50 tested sites at each fluence was used to test the sample. Damage in our study is defined as an irreversible modification detected by a Nomarski microscope at higher magnification. The results of the measurements are given in Figs. 2(a) and 2(b).

Considering Figs. 2(a) and 2(b) independently, variations in damage threshold between the samples made with different deposition techniques can be observed. The analysis of this difference is not the subject of this paper but it can be linked to small differences in properties between the samples (band gap, dielectric function). However, the same tendency is observed on all samples when comparing Figs. 2(a) and 2(b): the LIDT of 2H/3 layers is higher than the 2H layers. Depending on the samples, the increase in LIDT is between 10% and 25% at 45 fs, 20% and 25% at 300 fs, 30% and 60% at 600 fs, and 30% and 70% at 1.2 ps. The prediction based on Eq. (1) has been calculated and plotted on the figures. PI and AI rates discussed before have been taken for the calculation, with bulk material parameters, as well as a correction factor that takes into account interference effects in the coatings.[10] It should be noted that the values used for the calculations were not adjusted to fit the data since relative comparison were of interest. It can be seen from these theoretical results that the differences in LIDT between thick and thin films cannot be explained by the "classical" treatment. If we take into account the transient effects, as described in Eq. (3), a relatively good agreement is obtained between experiments and results to describe the LIDT dependence as a function of pulse duration. In the case of 2H layer, the E-field peaks are located at the air/film and film/substrate interface. The electronic density will then strongly increase during the pulse at these two locations. Calculations with Eq. (3) show that the E-field peak at the film/substrate interface is strongly reduced but the air/film interface value is weakly affected. Equations (1) and (3) provide then the same results and damage is located at the air/film interface. In the case of 2H/3 layer, the E-field peak is located at the film/substrate interface, where damage should occur according to Eq. (1). Calculations with Eq. (3) show that during the pulse, the E-field peak value is decreased which lead to higher damage thresholds from the prediction of Eq. (1).

Another set of samples, Ta_2O_5 single layers deposited on superpolished fused silica substrates, were made. Four thicknesses were chosen: 4H, 2H, H, and H/2. The samples were tested at 1030 nm, 530 fs. The results and the comparison to the model described above are shown on Fig. 3.

Again, a relatively good agreement is obtained between experiments and results on the description of LIDT as a function of the film thickness. The description of fs/ps LIDT is then clearly improved with this approach.

Analysis of experiments in thin films requires precise knowledge of the temporal and spatial modulation of the electric field due to interference effects. A specific approach was developed and presented in this paper that allows one to take into account the changes in the dielectric function during the laser pulse. Several experimental examples have been given to illustrate the interest of our model. It can improve the interpretation of fs and ps laser induced damage in thin films, and can be taken into account when designing damage resistant coatings.

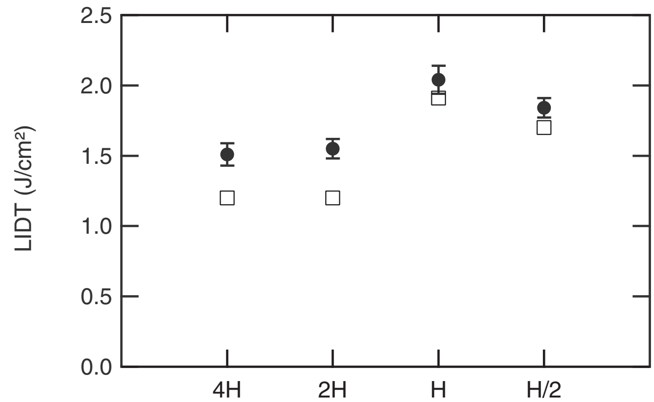

FIG. 3. Comparison of experimental measurements to theoretical prediction made on Ta_2O_5 single layers of different thicknesses. H represents one-quarter wave layer at 1030 nm.

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