Nonlinear Behavior and Damage of Dispersive Multilayer Optical Coatings Induced by Two-Photon Absorption

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ABSTRACT

We have observed and studied a nonlinear response of dispersive dielectric multilayer mirrors (DM). It was found that the structure of the mirror itself causes strong enhancement of the electric field inside the multilayer stack consequently triggering strong two-photon absorption (2PA). We have developed a mathematical model, that allows estimation of the coefficient of the 2PA, β, subsequent prediction and to some extent tuning of the strength of the nonlinear response of any multilayer coating.

Keywords: dispersive mirrors, thin film coatings, ultrafast optics, two-photon absorption

1. INTRODUCTION

Implementation of dispersive coatings, namely DM[1–3] significantly advanced ultrafast science and technology in the last two decades allowing routine generation of few-cycle[4–6] and later one-cycle[7] or even sub-cycle[8] light pulses. As the oscillating field of the few-cycle pulse is comprised to a very short period of time, the peak intensity of the pulse might reach PW orders of magnitude thus allowing us to enter strong-field regime and explore whole family of new optical phenomena.[4] Emerged generation of traces and isolated soft x-ray attosecond pulses[9,10]started the era of attosecond science.[11] However, the performance of the optics until now had yet been constrained to the linear domain only. Here we show, that while the ultrafast systems are yielding PW level intensities their specialized optics, namely dispersive mirrors, driven in the multi TW regime, starts to be prone to nonlinearities.

As Fourier-transform limited (FTL) pulse propagates through any medium, it gets stretched many times above FTL due to the accumulation of the wavelength dependent group delay (GD). In order to restore the original pulse width, one needs to introduce the equal amount of dispersion of the opposite sign. This procedure is called "compression". Historically, there are several techniques for performing the re-compression.[12,13] One that gained the most recognition is the implementation of DMs. The term DM if referred to the multilayer thin film coating capable of introducing wavelength dependent GD or group delay dispersion (GDD), the first derivative of the GD in frequency domain. Functionality of DM relies on the penetration effect[1] and implementation of resonant, fabry-perot like, structures.[14] Combined action of both can be described as if different wavelengths penetrate and get reflected in different depths inside the multilayer stuck, therefore introducing wavelength dependent GD. Addition of the resonant cavities creates light traps inside the structure, that assist in creating extra long time delays for narrow parts of the spectrum. Modern DM to some extent exploit both basic techniques.

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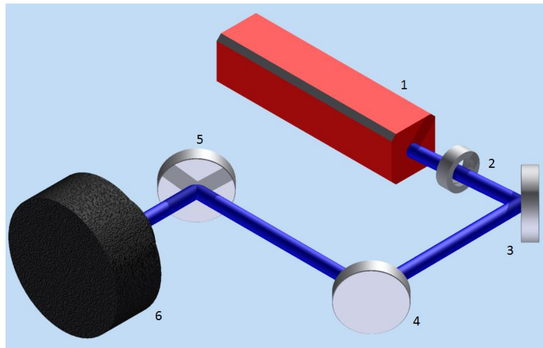

Figure 1. Test set up. 1 - source, 2 - iris, 3- folding mirror, 4-focusing mirror, f=2m, 5 - tested mirror, 6 - pyroelectric power head

Table 1. Design parameters of the MX-series

Design name	Material pair	Introduced GDD (fs^2)
M1	Ta_2O_5/SiO_2	-150
M2	Ta_2O_5/SiO_2	-180
M3	Ta_2O_5/SiO_2	-50
M4	HfO_2/SiO_2	-150
M5	HfO_2/SiO_2	-50

2. EXPERIMENT

We have tested a series of DM in the set up depicted in Figure 1. Laser source 1, is the frequency doubled output of the Ti:Sph amplifier system yielding $\sim$ 40fs up-chirped pulses centered at 400nm. Exploitation of the programmable dispersive filter-DAZZLER (not pictured) -allows step-wise attenuation of the transmitted power without compromising temporal and spectral profile of the pulse. With implementation of gentle focusing scheme (focusing mirror 4, focal length f= 2m), we are able gradually very incident intensity and multiply the irradiance by order of magnitude. The investigated mirror is slided along the focusing beam while the spectrally and temporally averaged power is being measured with pyroelectric power head. Thus our measurement procedure mimics to some extent the z-scan technique.[15]

Tested mirrors, denoted as MX-series, differ by two key parameters: the value of introduced GDD and material pair. For coating production we have used Ta_2O_5/SiO_2 and HfO_2/SiO_2 as the material pairs that are most commonly used for visible (VIS) region of the spectrum. HfO_2/SiO_2 is also used for ultraviolet (UV) region. The mirrors of the series were designed to introduce different amounts of GDD, ranging from -50fs^2 to -180fs^2. The relevant mirror data are presented in Table. 1

We observed a nonlinear response of the MX-series mirrors in the form of intensity dependent reflectance (Fig. 2), i.e. the reflectance was decreasing with increasing of the incident intensity. The plots of the Fig. 2 show clear dependence of the observed nonlinearity from particular mirror parameters. First, the used material pair. Mirrors M1 and M4 introducing same amount of GDD yet produced out of different material pairs also behave noticeably different. Second, the strength of the nonlinear response is correlated with the introduced GDD, the less the value of the introduced GDD is, the weaker the response is. M3 produced out of the same material as M1 and M2, but introducing the least dispersion has significantly weaker drop of the reflectance. We have additionally tested quarter-wave high reflector (QWHR) that introduces virtually zero GDD and did not observe any sufficient evidences of its nonlinear behavior, Fig. 2.

It is important to mention, that until the point of an optical breakdown the effect appears to be reversible (Fig. 3), i.e. if the incident intensity is decreased, the reflectance instantly rolls back to higher values.

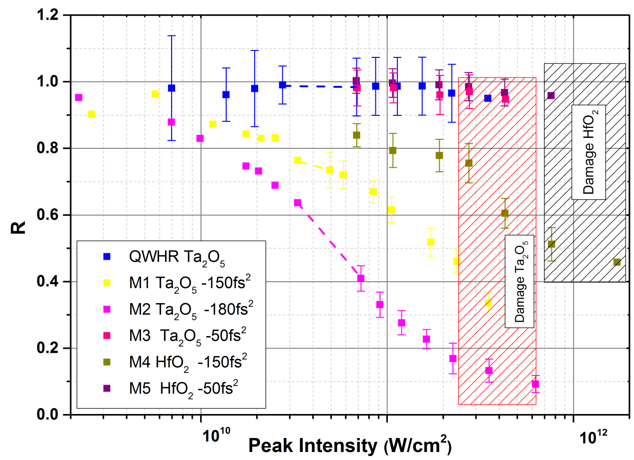

Figure 2. The intensity dependent reflectance of the MX-series mirrors.

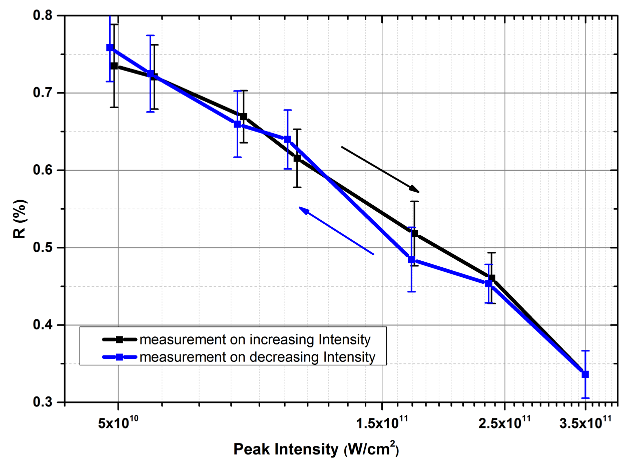

Figure 3. The reversibility of observed nonlinearity; if the intensity is decreased, the reflectance rolls back to higher values.

Observed effect might be interpreted and understood if one takes the structure of the DM into consideration. As the operational principle of most DMs is the penetration effect, the particular minimal optical thickness needs to be reached in order to introduce desired GDD. The more GDD one wants to introduce, the thicker the coating gets, the deeper the field penetrates into the multilayer stack involving more and more material into interaction. Addition of the resonant cavities creates "hot spots" inside the stack, where electric field gets drastically enhanced. Both effects acting together create distribution of the electric field very different to those observed in single layers or QWHR (Fig. 4). From the plots of Fig. 4 it is seen, that the electric field inside the DM stack gets significantly enhanced. In "hot spots" regions the electric field amplitude is several times higher, than it is in single layers or QWHR. Achieved enhancement appears to be enough to trigger peculiar nonlinearities.

3. THEORY

There are two possible nonlinearities to be triggered. 2PA and/or optical Kerr-effect. Involvement of one of these two is able to explain the observed differences in nonlinear behavior between coatings produced out of different materials. As it is expected that change in the nonlinear refractive index,n_2,(and so respectively the coefficient of the 2PA) for dielectric/semiconductor materials is inversely proportional to the electronic band gap,[16] HfO_2 with its band gap of ~ 5eV[17] should have smaller nonlinear coefficients than Ta_2O_5 with its band gap of ~ 4eV[18] and therefore weaker nonlinear response.

If the Kerr-effect had taken place, the transmission of the coating would have changed. Simultaneously the

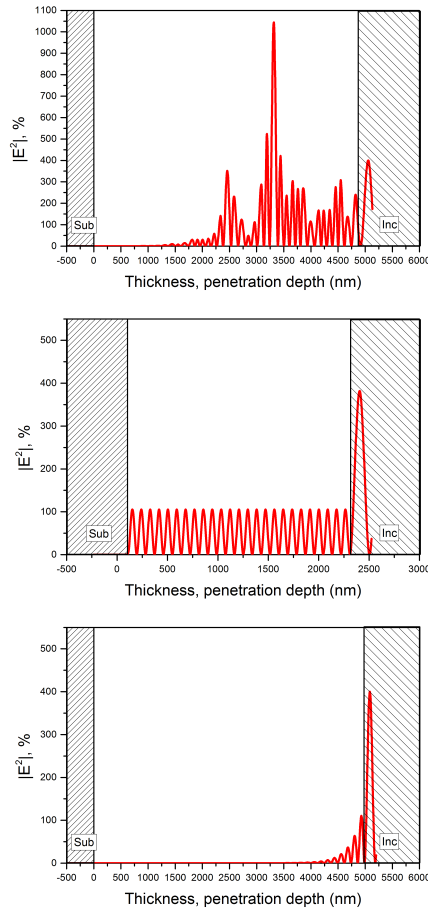

Figure 4. Distribution of the electric field from top to bottom in DM, single layer, QWHR.

GDD would have been affected. However, we have monitored the transmission and did not observe any change in all the intensity range of interest. TG FROG[19] measurement also confirmed, that dispersion properties of the mirrors were not affected.

At the same time, appearance of 2PA only causes increase of the losses, since part of the energy is converted to heat, without any noticeable impact on dispersion. In order to confirm this hypothesis we performed thermal measurement where we directly measured surface temperature of the irradiated mirrors. We used commercial FLIR SC305 Infrared Camera (FLIR systems Inc.). Each measurement last 240 sec, with camera frame rate of 9Hz. We have registered that the surface of MX mirrors warms up drastically more in comparison to the surface of the QWHR (Fig. 5). This was considered as the sufficient argument in support of the 2PA suggestion. Following 2PA direction, we have developed a mathematical model that allows prediction of the nonlinear behavior of any requested multilayer coating.

We simulated 2PA as induced extinction coefficient proportional to the intensity of the electric field inside the multilayer stack: $\chi(z) = \beta * |E(z)|^2$, where z is the coordinate along mirror's cross section. We have considered the extinction coefficients of the high-idex materials, in our case Ta_2O_5 and HfO_2 only, as their band gaps are narrower that the band gap of SiO_2 ($\sim$ 7eV[20]). The model allows to not only simulate the monochromatic cases (Fig. 6.) but, with implementation of an averaging procedure, emulate the intensity dependent behavior of the optics supporting broad spectra.

By numerical fitting of the data measured for M2 (Fig. 6), we were able to estimate 2PA coefficient, β, to be $\sim 4.1 * 10^{-21}$ [m^2/V^2], which corresponds to more commonly used $\alpha_2 \sim 4.3 * 10^{-9}$ [cm/W]. Due to the lack of data, we were not able to compare the estimated value of β or α_2 to the similar research. Only from the comparison to

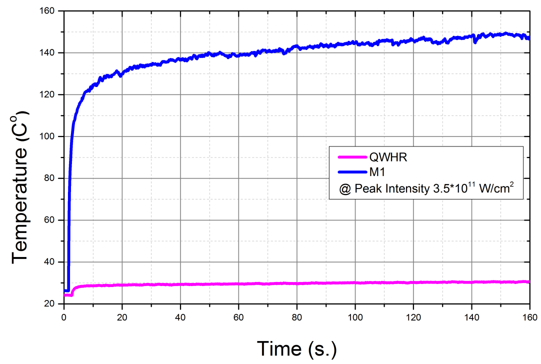

Figure 5. Surface temperature of the irradiated MX-series mirror and QWHR.

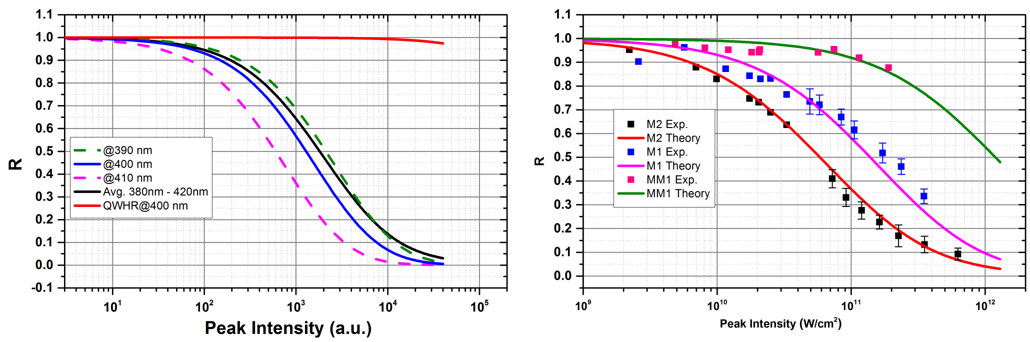

Figure 6. Implementation of the developed model. On the left - simulated reflectance for monochromatic and averaged cases; On the right - fitting of measured data to developed model.

other oxides, like Al_2O_3~ 0.09*10^-9 [cm/W][21] and TiO_2 ~ 17*10^-9 [cm/W][22] we concluded that the suggested value is rather adequate. After plugging the estimated value of β to the design of the mirror M1, we were able to obtain convincing correspondence between predicted and measured data, thus confirming consistency of the developed model.

4. RESULTS

It is feasible to implement the developed model to the already adopted thin film design techniques, therefore opening possibilities for the actual tuning and to some extent tailoring the intensity dependent behavior of the coatings. However, the algorithm requires multi-dimensional optimizations and several solution of nonlinear problems and thus is rather time-consuming. It is also possible to use simpler approach. In particular case we were interested in suppressing the influence of the 2PA. Then, having the approximate value of β, we estimated the value of the induced extinction coefficient at the maximum of the incident intensity and added it to the linear absorption of the high-index material. Following this approach, we have developed a new series of the 2PA-optmized designs, denoted as MMX. There specification is presented in Table 2.

Table 2. Design parameters of the MMX-series.

Design name	Material pair	Introduced GDD (fs^2)
MM1	Ta_2O_5/SiO_2	-150
MM2	Ta_2O_5/SiO_2	-100
MM4	HfO_2/SiO_2	-150

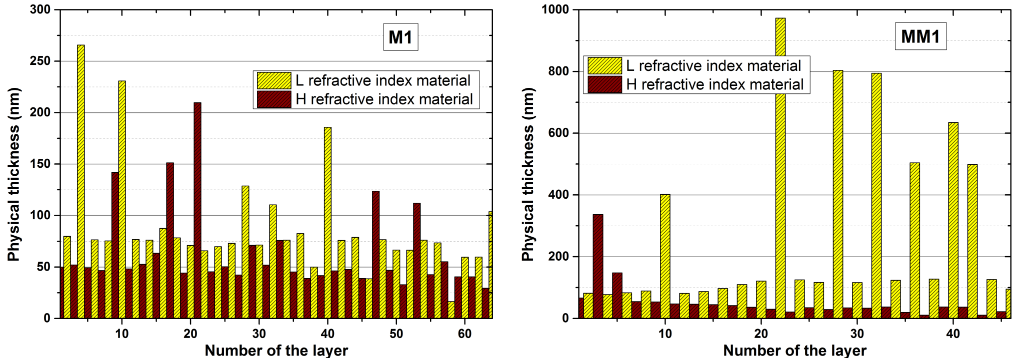

Figure 7. Designed multilayer stack, M1 - on the left, MM1 - on the right

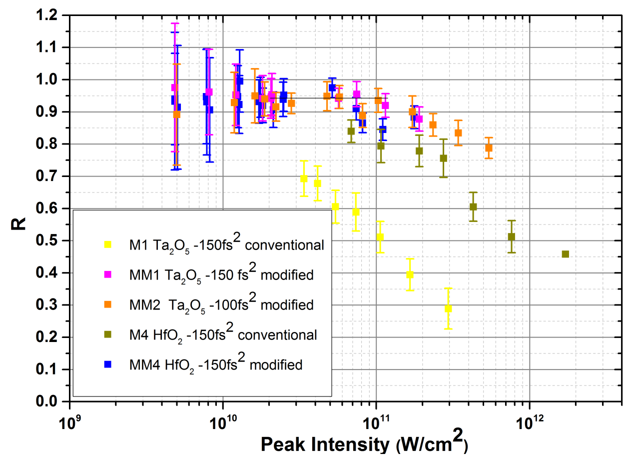

Figure 8. Comparison of the reflectance of the mirrors MX and MMX sreies.

The designed stacks of the mirrors M1 and MM1 are depicted in Fig. 7. One can immediately recognize several features of the MMX series. While in the case of M1, stack is containing approximately the same amount o the high- and low- index materials and materials are distributes homogeneously, in case of MMX stack, the presence of the low-index material is increased in favor of high-index material, and high-index layers are concentrated near substrate, where electric field is should be weaker (see Fig. 4).

The reflectance data of the 2PA-modified designs of MMX series are presented in Fig. 8. The plots of the Fig. 8 reveal significant improvement of the performance of the MMX series. The mirrors of the series have higher average reflectance and appearance of the nonlinearity is postponed to higher intensities. It is also remarkable, that the performance is not longer strongly dependent neither on the introduced GDD, nor on the used high-index material. That might signalize that, due to the modified electric field distribution, now the 2PA in SiO_2 starts to be pronounceable. Therefor it would be rational, in order to create optimal design, to estimate and take into account the 2PA coefficient of SiO_2 as well.

In order to find out, if the improvement of thermal performance also takes place, we have repeated the thermal tests, see Fig. 9. As it can be see, the thermal performance of the MMX mirrors has improved as well. The 2PA-optimized mirrors warm up significantly less that the non-optimized designs.

5. CONCLUSIONS AND OUTLOOK

For first time to the best of our knowledge we have observed nonlinear response of the dispersive dielectric multilayer coatings. We have found out, that the structure of the multilayer stack causes strong enhancement of the internal electric field, which in turn stimulates significant 2PA absorption, that in case of no enhancement would not have had an influence. We have developed the mathematical model, that allows quantitative and

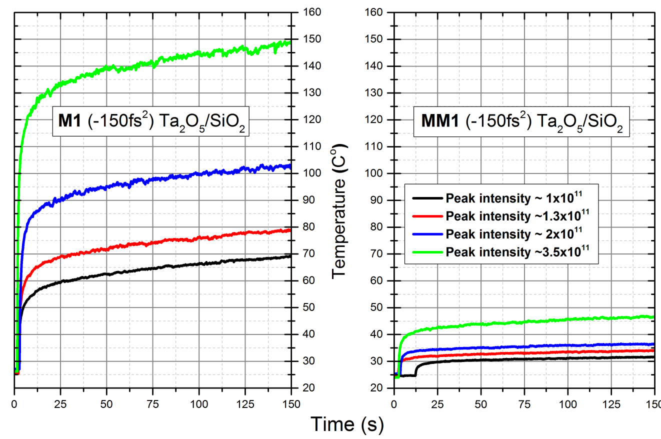

Figure 9. Comparison of the thermal performance of the mirrors MX and MMX sreies.

qualitative simulation of observed effect. Implementation of the developed model into design routine will permit creation of the multilayer structures with, to some extent, tailored intensity behavior, what is applicable to the development of vast range of future photonic devices.

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