

Strong optical nonlinearities in dispersive dielectric chirped mirrors below the damage threshold

Guan Gui^{*}, Amitava Adak, Manika Dandapat, Daniel Carlson, Drew Morrill, Henry Kapteyn[#], Margaret Murnane, and Chen-Ting Liao[§]

JILA and Department of Physics, University of Colorado and NIST, 440 UCB, Boulder, Colorado 80309, USA

[#] KMLabs Inc., 4775 Walnut Street, Suite 102, Boulder, CO 80301

^{*}guan.gui@colorado.edu; [§]ChenTing.Liao@colorado.edu

Abstract: We experimentally characterized an unexpectedly-strong nonlinear response in dispersive dielectric multilayer mirrors below the known damage threshold. We observed a strong reflectivity decrease, local heating, transient spectral modifications, and time-dependent absorption of the incident pulse. © 2020 The Author(s)

OCIS codes: (190.1900) Diagnostic applications of nonlinear optics, (230.2035) Dispersion compensation devices, (320.1590) Chirping, (310.4165) Multilayer design

1. Introduction

Dispersive dielectric multilayer mirrors, also called chirped mirrors (CMs), are widely used as dispersive optical components in ultrafast optics to manage dispersion of broadband and ultrashort laser pulses. One obvious advantage of using a CM is its high energy handling capability. As a reflective optical component with a nominal laser-induced damage threshold (LIDT) in the range of ~ 200 - 350 mJ/cm² [1], they appear to be an excellent choice for dispersion compensation for high-energy pulses in the few-cycle range. In this work, we surprisingly identified nonlinear effects in CMs even when using near infrared lasers with moderate laser fluence well below the LIDT. These nonlinear responses, e.g., reflectivity decrease, local heating, transient spectral changes, and time-dependent absorption of the incident pulse, relate to the incident laser peak intensity, not the fluence, and do not result in damage to the mirror even in the long term.

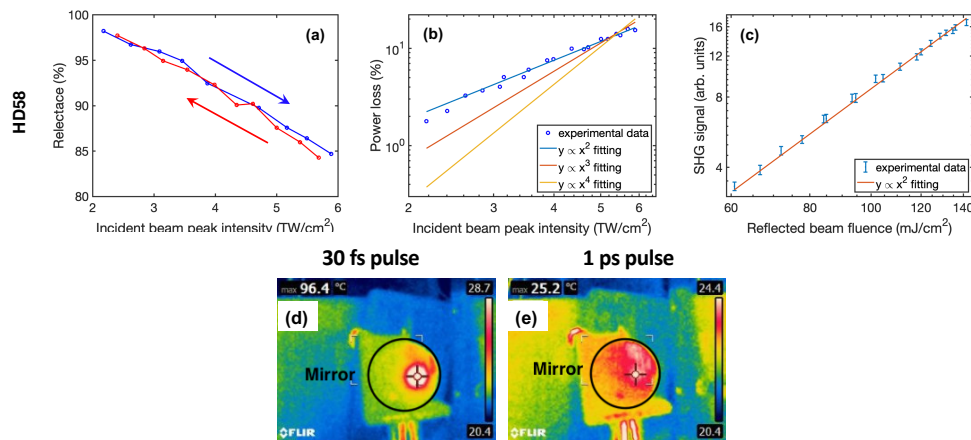


Fig.1 Intensity-dependent performance of HD58 chirped mirror. (a) Measured reflectance as a function of incident laser peak intensity. Blue (red) data points and the arrow indicates the increasing (decreasing) peak intensities when taking the measurements, verifying a the reproducible and fully reversible effect. (b) Power loss on reflection as a function of peak intensity on a log-log scale, along with power-law fits showing that the absorption fits a quadratic power law. (c) second harmonic generation (SHG) signal generated with the reflected laser beam, as a function of the reflected laser fluence on a log-log plot. These SHG signal fits a quadratic power law, indicating the duration of the reflected pulses was not significantly changed by the strong nonlinear absorption. (d) and (e) are thermal images of the mirror when illuminated by the same laser fluence with short (~ 30 fs) and long (~ 1 ps) pulses.

2. Experiment

As an example, we experimentally investigated highly dispersive HD58 CMs from UltraFast Innovations GmbH. Our Ti:sapphire laser amplifier (Red Dragon, KMLabs Inc) delivers up to 16 mJ at 1 kHz repetition rate with a nominal pulse duration of ~ 30 fs at ~ 790 nm. The mirrors we studied consist of Ta₂O₅ ($n = 2.09$ at 800 nm) and SiO₂ ($n = 1.45$ at 800 nm) multilayer dielectric stacks. Figure 1(a) shows the power loss from the mirror on reflection, as a function of incident laser peak intensity. A power loss of up to $\sim 16\%$ (reflectance down to $\sim 84\%$) was observed at ~ 5.89

TW/cm². Note that the maximal loss is beam size averaged, so the local maximal reflectance reduction is > 20%. The loss is plotted on a log-log scale in Fig. 1 (b) along with power-law fits that clearly indicate an absorption quadratic with incident power, indicating a two-photon absorption process. Although neither the bandgap of Ta₂O₅ (~4.2 eV) nor of SiO₂ (~7.5 eV) supports two-photon absorption at ~790 nm (~1.57 eV), potential defect states in the forbidden bands of Ta₂O₅ could allow for two-photon absorption [2]. We also measured a second harmonic signal generated by the reflected near infrared beam as shown in Fig. 1(c). This second harmonic signal, as a function of fluence for a reflected beam, can be fit well by a quadratic power-law function, which implies that the duration of the reflected pulses is not significantly altered by the nonlinear absorption. To distinguish a peak intensity effects from fluence-related effects, we further varied the incoming chirp of the pulse, stretching the pulse to ~1 ps to reduce the peak intensity. Figure 1 (d) and (e) shows thermal images of the mirrors as the pulse chirp is changed at constant fluence. The temperature can easily reach ~100 °C when illuminated by short (~30 fs) pulses, with the temperature profile following the Gaussian profile of the incident laser beam. With long (~1 ps) pulses, the temperature remains close to the room temperature. This behavior clearly indicates a peak intensity-dependent effect.

To study the mechanism behind the nonlinear response, we also conducted time-resolved pump-probe measurements. Here we used a pump pulse at a moderately strong peak intensity of ~2.5 TW/cm², and a probe pulse at a low peak intensity < 10 GW/cm² overlapped with the central part of the pump beam. The time-resolved power loss of the probe beam from HD58, as well as another CM design HD1631, are shown in Fig. 2 (a). The CMs show a transient and large power loss within ~1 ps time scale, followed by a slow ~10's of ps recovery. Figure 2(b) shows a time-resolved spectral change, i.e., transient absorption spectrogram, of the probe beam in the presence of a pump beam. The strongest spectral absorption is around ~810 nm, which is red-shifted by ~20 nm with respect to the peak spectrum of the incident laser. The absorption at ~810 nm can go as high as ~35%. The same red shifting is also observed on the side panels of Fig. 2(b), which shows the probe spectrum at representative pump-probe delays of 0 ps, 4 ps, and 24 ps. The transient power loss can be understood by multiphoton absorption and free electron, and the slow recovery is probably related to the formation of long-lived self-trapped excitons [3, 4].

To better understand the observed nonlinear effects, we also simulated the electric field intensity in the dispersive multilayer stacks. The simulation shows order-of-magnitude scale field enhancements, especially in high-index Ta₂O₅ layers, which has lower bandgap and dominant contribution to nonlinear effects. The field enhancement leads to a wavelength-dependent nonlinear absorption, despite that it shows no signs of cumulative damage before catastrophic failure.

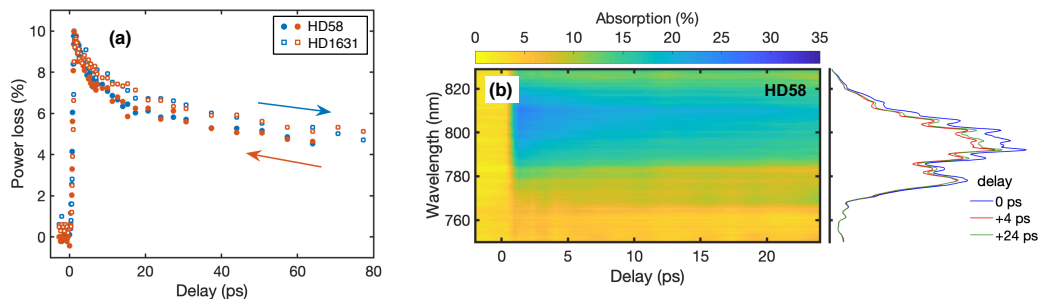


Fig.2 (a) Time-resolved power loss curve of HD58 and another HD1631 mirror. (b) Time-resolved spectral change, namely, transient absorption spectrogram, of the probe beam in the presence of a pump beam on HD58 mirror. The side panel shows the probe beam spectrum at representative pump-probe delays.

3. References

- [1] I. B. Angelov, S. A. Trushin, V. Pervak, S. Karsch, Z. Major, A. von Conta, and F. Krausz, "Investigation of the laser-induced damage of dispersive coatings," *Laser-Induced Damage Opt. Mater.* 2011 8190, 81900B (2011).
- [2] O. Razskazovskaya, T. T. Luu, M. Trubetskov, E. Goulielmakis, and V. Pervak, "Nonlinear absorbance in dielectric multilayers," *Optica* 2, 803 (2015).
- [3] D. Grojo, M. Gertsvolf, S. Lei, T. Barillot, D. M. Rayner, and P. B. Corkum, "Exciton-seeded multiphoton ionization in bulk SiO₂," *Phys. Rev. B* 3–6 (2010).
- [4] Z. j Rudolph W, Mero M, Sabbah A J, "Femtosecond dynamics of dielectric films in the pre-ablation regime," *Appl. Phys. A Mater. Sci. Process.* 324, 317–324 (2005).