Ultrafast damage processes of laser mirrors and plasmonic nanostructures

Summary of the Ph.D. thesis

Benedek Nagy

Supervisor: Péter Dombi
Wigner Research Centre for Physics

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1 Introduction

For world-leading high-intensity laser systems, the damage threshold of various optical elements is a critical parameter. Failure due to one or more elements in a laser amplifier chain results in a significant loss not only in the performance and quality of the laser beam, but also in time and financial loss. Due to the finite intensity that can be used, amplifiar systems require the expansion of the beams, which requires optical elements with a diameter up to one meter. It is a difficult and expensive task to manufacture these elements in the right quality, which is a significant cost when designing such a large laser system. For all these reasons, it is important to know the damage threshold of the optical elements used and it becomes essential to perform systematic femtosecond damage threshold measurements, which contributes to the design of more intensity-resistant mirrors and other optical elements.

In parallel, we encounter an unexpected problem when the carefully prepared nanostructured samples used in our laser-matter interaction experiments do not withstand the intensity load imposed on them. A large percentage of today's experiments to study light-matter interactions use an extremely fragile sample made by nanofabrication process, the production of which requires not only time and financial expenditure, but also serious expertise. For this reason, it is particularly important to know the upper limits on the used samples, not to destroy them prematurely during experiments. These issues have proven to be particularly important in the use of nanoplasmonic samples, where the resulting nearfield can be as much as one hundred times the electric field of the illuminating laser pulse, and thus optical damage can very easily begin.

1.1 Damage Threshold of Laser Mirrors

In the case of large (>3 eV) band gap materials and in the range of picosecond pulse lengths physical damage occurs in the form of atomic layer ablation, conventional melting, boiling, color changes, and lattice defects [1]. In the femtosecond range, Coulomb repulsion occurs due to various ionization processes, as well as plasma formation and thermoelastic fracture [2].

An event can be called optical damage if an irreversible change occurs. In practice, this means that a visible change occurs that can be detected with modern, high-resolution imaging system or with another appropriate method on the surface of the material [1]. The damage threshold is the maximum power density that does not yet cause damage on the surface. The ablation parameter is a quantitatively measurable parameter of the damaged surface, such as the depth or diameter of the ablated crater. The ablation threshold is the maximum power density that does not yet cause ablation [1]. Below the damage threshold, a so-called incubation is observed, which is still reversible and not followed by ablation [3].

In recent years, significant progress has been made in the development of high power femtosecond lasers with a repetition rate in the MHz range. Among others, passive mode-locked ytterbium thin-disk lasers with relatively high pulse energy have emerged [4–6], and femtosecond optical fiber amplifiar systems [7]. Until the publication of my own results, little reliable data was available on the damage threshold measured at repetition rates in the MHz range. The damage threshold of a high-reflection mirror at 100 MHz was measured with a very strongly focused beam [8], and a so-called round-robin measurement is available, but there were differences in many important parameters between measurements at many different locations [9]. Angelov et al. performed comparative experiments with picosecond pulses at kHz and MHz repetition frequencies, but with an unknown number of pulses. They showed a difference of about a factor of two as a function of the band gap of the samples [10, 11]. However, due to the unknown number of pulses, the measurement cannot be considered as an accurate comparison of the damage thresholds caused by two repetition rates differing by orders of magnitude, because below a certain number of pulses the damage threshold has still a high dependency on the repetition rates. [12, 13].

In line with the above considerations, it became very important to perform measurements that meet the following three conditions: (i) they examine a femtosecond damage threshold, (ii) they use pulse trains arriving at repetition rates of kHz and MHz, and (iii) all other relevant pulse- and beam parameters are kept the same in a controlled manner for both the MHz and kHz repetition rates.

Based on the above considerations, I present in the thesis the results I obtained in the examination of the damage threshold of laser mirrors, first giving an exact comparison to the damage threshold measured at kHz and MHz repetition rates using a method developed by me, posessing several advantages. I performed these with a new measurement procedure I developed, which is also presented in the thesis.

1.2 Damage Threshold of Plasmonic Nanostructures

Metal nanoparticles play a significant role in many ultrafast plasmonic processes. The high laser intensity provided by femtosecond lasers is typically required to generate nonlinear effects. With a properly designed nanosystem high field-enhancement can be achieved by excitation of surface plasmons. For this purpose, both localized and propagating surface plasmons are suitable [14,15]. Surface plasmons (to name them accurately surface plasmon polaritons - SPP, and localized surface plasmons - LSP), are excitons on metal-dielectric or metal-vacuum boundaries which are caused by periodic charge rearrangement and charge density oscillations excited by an external excitation field. This can occur when the external field is an electromagnetic wave to which the charge density oscillations in the nanosystem couple [16].

For many applications, the extremely large local electric field generated by the field enhancement and femtosecond pulses even at low illuminance can cause rapid degradation of the nanoparticles. Therefore, in order to amplify plasmonic nonlinear effects as much as possible (e.g., maximizing the harmonic signal, photoelectron current, etc...), it is necessary to understand the laser-induced damage mechanisms of nanoparticles. It is necessary to know, for example, the damage threshold of nanoparticles produced by lithography. Measurements performed while taking into account the damage threshold increase the lifetime of the samples and maintain their stability during the experiments.

Contrary to typical damage threshold measurements on mirrors [T1-T4], real-time observation of plasmonic damage is hampered by the fact that nanostructures are too small for direct observation by optical methods (such as optical microscopy) and in addition many applications require a vacuum environment which makes it even more difficult to monitor the status of samples in real time. Despite the importance of the problem of plasmonic damage, there are only a few systematic studies on the damage of nanoparticles. So far, femtosecond laser-induced damage has been observed at high harmonic generation, after which the edges of the used nanorods become smoother and blunter [17] or the nanoparticles were destroyed by deformation and detachment [18] or simply melted [19, 20].

Based on the number of nanoparticles displaced from the substrate, the damage of plasmonic nanorods [21] was also quantitatively analyzed, where a damage threshold of 1.2 $\frac{\mathrm{mJ}}{\mathrm{cm}^2}$ was determined. In addition, others have created nanoholes at power well above the damage threshold [22]. Laser-induced deformation of nanoparticles does not help to understand the damage thresholds, as these experiments take place at laser intensities where nanoparticles are sure to undergo significant deformation [23–26].

Based on all of this, I aimed to determine the maximum focused laser intensity at which nanoparticles produced by electron beam lithography can still be safely used for nonlinear applications. In the scope of this work, I also experienced other interesting phenomena, which I also thoroughly examined.

2 Results

2.1 New method for measuring the damage threshold of laser mirrors

I developed a new measurement method to measure laser induced damage threshold (LIDT). Compared to known and frequently used methods (ISO 1-on-1, ISO S-on-1, and R-on-1), this method requires a significantly smaller sample area and can be performed significantly faster. I have shown that the new method results in lower femtosecond damage thresholds compared to measurements similar to the ISO standard. However, since the measurement procedure exposes the mirror to a load similar to the real exposure, the value I have determined can be used more realistically and safely in sizing modern femtosecond laser systems. Another advantage of the measurement method is that it can be used for very strongly focused beams with a Rayleigh lengths of few μm-s.

This result is presented in the following journal articles where I am the first author: T1,T2. This result is supported by further publications: T3,T4.

2.2 Femtosecond damage threshold at a MHz repetition rate

I have experimentally demonstrated that laser pulses arriving at a repetition rate of the order of MHz significantly reduce the value of LIDT compared to those arriving at a repetition rate of the order of kHz. I realized that at repetition rate of MHz complete thermal relaxation does not occur in the focalspot, so thermal effects contribute significantly to femtosecond damage at this repetition frequency. All this leads to a decrease in the damage threshold. I compared the experimental results with numerically modeled results and those showed high agreement.

This result is presented in the following journal article where I am the first author: T1.

2.3 Measurement of the damage threshold of plasmonic nanoparticles

I also extended my research on mirrors to nanooptic systems. I experimentally defined the damage threshold of plasmonic nanoparticles prepared by electron beam lithography. I validated the damage with an electron microscope and defined the damage threshold of such special samples, which play an important role in nonlinear optics, by evaluating the microscopic images and using an extrapolation method. From the position that the result is significantly lower than the damage threshold of either the metal or the substrate material, I have shown that the damage is induced by the optical nearfield.

This result is presented in the following journal article where I am the first author: T5.

2.4 Intensity dependent nearfield induced damage and nearfield mapping

I investigated in detail the morphological changes of plasmonic nanoparticles written on the surface in the intensity range critical for the damage processes. In doing so, I observed as a new phenomenon the nearfield induced partial separation of nanoparticles from the surface, as well as the local inversion and stochastic removal of the particles. I have shown, using higher intensities, that there is an intensity at which nanoablation on the substrate underneath the nanoparticle can be performed such that the ablated pattern accurately maps the nearfield distribution.

This result is presented in the following journal article where I am the first author: T5.

Thesis related publications

T1 B.J. Nagy, L. Gallais, L. Vámos, D. Oszetzky, P. Rácz, P. Dombi, „Direct comparison of kilohertz- and megahertz-repetition- rate femtosecond damage threshold," Optics Letters **40** (11), 2525-2528, (2015).

T2 B.J. Nagy, L. Vámos, D. Oszetzky, P. Rácz, P. Dombi, „Femtosecond damage threshold at kHz and MHz pulse repetition rates," Proc. SPIE **9237**, 923711, (2014).

T3 V. Csajbók, L. Szikszai, B.J. Nagy, P. Dombi, „Femtosecond damage resistance of femtosecond multilayer and hybrid mirrors," Optics Letters **41** (15), 3527-3530, (2016).

T4 V. Csajbók, Z. Bedőházi, B.J. Nagy, P. Dombi, „Ultrafast multipulse damage threshold of femtosecond high reflectors," Applied Optics **57** (2), 340-343, (2018).

T5 B.J. Nagy, Zs. Pápa, L. Péter, C. Prietl, J. R. Krenn, P. Dombi, „Near-field-induced Femtosecond Breakdown of Plasmonic Nanoparticles," Plasmonics **15** (2), 335-340, (2020).

Additional publications

S6 M.Zs. Kiss, B.J. Nagy, P. Lakatos, Z. Göröcs, Sz. Tőkés, B. Wittner, L. Orzó, „Special multicolor illumination and numerical tilt correction in volumetric digital holographic microscopy," Optics Express **22** (7), 7559-7573, (2014).

S7 P. Rácz, B.J. Nagy, K. Ferencz, P. Dombi, „Intracavity Herriott-cell testbed for large-aperture femtosecond optics" Laser Phys. Lett. **11** (12), 125805, (2014).

S8 J. Vogelsang, J. Robin, B.J. Nagy, P. Dombi, D. Rosenkranz, M. Schiek, P. Groß, C. Lienau, „Ultrafast electron emission from a sharp metal nanotaper driven by adiabatic nanofocusing of surface plasmons", Nano letters **15** (7), 4685-4691, (2015).

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