Determination of Molybdenum Ablation Threshold for Ultrashort Laser Pulses in Atmosphere and Vacuum Using the Diagonal Scan Technique

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Abstract: we present the results of measuring metallic molybdenum ablation threshold in the 30 femtoseconds regime. The measurements were performed in atmosphere and vacuum, resulting in values equal to 1.6 TW/cm^2 3.8 TW/cm^2, respectively. This values difference can be credited to a plasma etching that helps the ablation in the atmosphere case.

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Introduction

Ultrashort pulse laser ablation of solids is due to an electron avalanche induced breakdown process[1, 2] which occurs when seed electrons are accelerated in the laser field, exponentially generating free electrons by collisions. The breakdown takes place when the plasma originated by the avalanche electrons reaches a critical density and transfers energy to lattice ions, which expand away from the surface after the pulse has passed. In metals, the seed electrons are always present (conduction band free electrons), and in dielectrics and semiconductors they are excited from the valence band to the conduction band by the pulse leading edge, either by multiphoton ionization[3, 4] or by tunneling induced by the laser field[5-7]. Although the seed electrons have dissimilar origins in different material types, a metallization occurs in dielectrics and semiconductor after they are produced, and the avalanche evolves deterministically in time[2, 8, 9] in the same way in all solid materials that now behave like metals[10, 11]. These mechanisms confer a nonselective characteristic to the ultrashort pulse ablation, and the intensity ablation threshold of a material, I_{th}, is the only parameter relevant to the etching process.

The established method[12] to determine the ablation threshold of a given material consists in ablating the material using a TEM$_{00}$ Gaussian beam at various intensities, and requires detailed knowledge of all the geometrical experimental parameters, the pulses energies and a series of measurements.

In previous works[13, 14] we introduced an alternative method to determine the ablation threshold in the ultrashort regime, based in the very precise definition of the etching region resulting from the nonlinear character of the ultrashort pulse ablation and in the almost inexistent lateral heat diffusion.

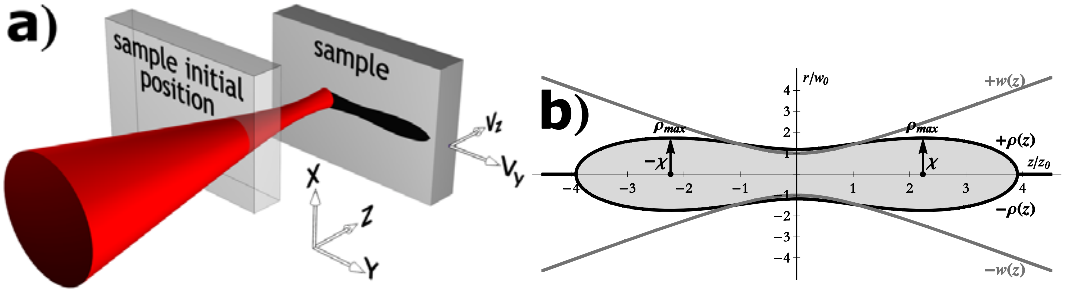

Fig. 1. Diagonal scan method. a) scheme of the experimental setup showing the sample movement. b) profile of the sample surface etching, ρ(z), by the laser beam, w(z) (the vertical and longitudinal axes normalized by the beamwaist and confocal parameter, respectively).

Our method, which is schematized in Fig. 1a, consists in scanning a solid sample diagonally across the beamwaist of a focused laser beam. The sample starts in a position where there is no ablation, then it is moved simultaneously in the *z* and *y* directions passing through the focus, up to the point where the ablation has ceased. This Diagonal Scan (D-Scan) etches a profile in the sample surface. If the pulse power is above the critical power[13] $P_{crit} = ½e\pi w_0^2 I_{th}$, the etched profile will be similar to the one shown in Fig. 1b, presenting two lobes, and in this case a simple equation relating the half maximum transversal dimension of the profile, ρ_{max}, the pulse power, P_0, and the sample ablation threshold intensity, I_{th}, is easily derived[13]:

$$I_{th} = \frac{P_0}{e\pi\rho_{max}^2} \cong 0.117\frac{P_0}{\rho_{max}^2}\,. \qquad (1)$$

The measurement of the ablation profile maximum half width, the pulse energy and time duration determine the ablation threshold intensity I_{th}; multiplying this value by the pulse duration gives the ablation threshold fluence, F_{th}.

In this work we present the results of measuring the ablation threshold for metallic molybdenum samples in atmosphere and vacuum using the D-Scan technique.

Experimental Setup

For the experiments we used metallic molybdenum samples irradiated by ultrashort pulses coming from a CPA Ti:Sapphire system composed by a Rainbow seeder and a Femtopower Compact Pro amplifier, both from FEMTOLASERS Produktions GmbH. This systems generates pulses shorter than 30 fs, at 4 kHz repetition rate, and energy up to 800 µJ. The beam was focused by a 7.5 cm focal distance converging lens, and the sample was placed in a one-axis translation stage controlled by computer, with its surface normal to the laser beam. The translation stage was aligned diagonally to the laser beam to ensure a movement through its waist. This setup was placed inside a vacuum chamber, and the measurements were performed at a vacuum of 10^{-6} mbar and atmosphere. The sample surface was photographed in an optical microscope, and the ablation profile was then measured.

The pulse duration was measured with an interferometric autocorrelator (Femtometer, from FEMTOLASERS), and the pulse energy was measured with an Ophir PE-50 energy meter.

Results

In Fig. 2 we present the pulses interferometric autocorrelation measurement. From the pulse spectrum the central wavelength was determined by calculating the spectrum center of gravity, and this wavelength was used to calculate the fringe period on the autocorrelation trace. From it we determined a pulse duration (FWHM) of 28.5 fs. For the experiments presented here the pulse energy was fixed at 100 µJ, resulting in a peak power P_0 equal to 3.5 GW.

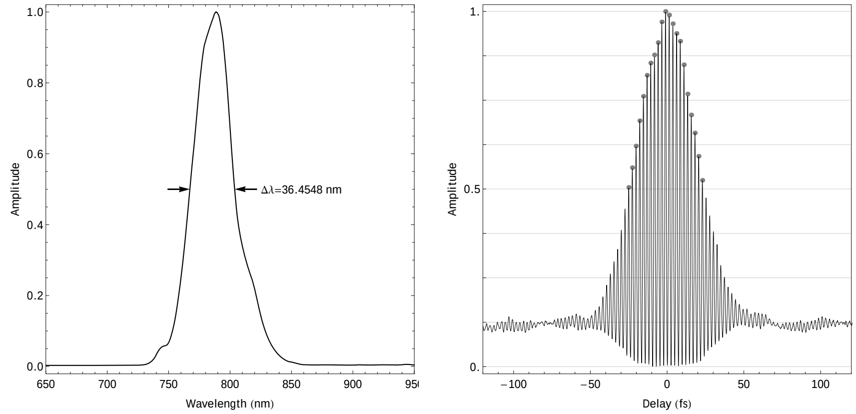

Fig. 2. Ultrashort pulses spectrum (left) and interferometric autocorrelation (right). The spectrum central wavelength at λ_0=778 nm and the 20.5 peaks on the autocorrelation trace above the 50% level results in a pulse duration (FWHM) below 28.5 fs assuming a 1.897 deconvolution factor (sech^2 pulse).

In Fig. 3 photographs of the ablation profiles obtained in atmosphere and vacuum are shown. Fig. 3a presents the sample with the two profiles (upper one created in atmosphere, lower one in vacuum), Fig. 3b shows the profile obtained in atmosphere along with two marks done 1 mm apart to be used as scale, and Fig. 3c exhibits the profile obtained in vacuum with the same scale as in micrograph 3b. From these images, the maximum transversal dimension measure for the sample in atmosphere was $2\rho_{max}$=322 µm, and for the vacuum one was $2\rho_{max}$=207 µm. Using these values in equation (1), we determined the ablation threshold in atmosphere to be $I_{th}=1.6\times10^{12}$ W/cm^2, and in vacuum to be $I_{th}=3.8\times10^{12}$ W/cm^2, corresponding to ablation fluences equal to 0.04 J/cm^2 and 0.11 J/cm^2, respectively.

Comparing Figs.3b and 3c it can be seen that a larger heat discoloration area occurs around the ablation performed in vacuum, indicating that the atmosphere contributes to remove excess heat deposited by the laser pulses.

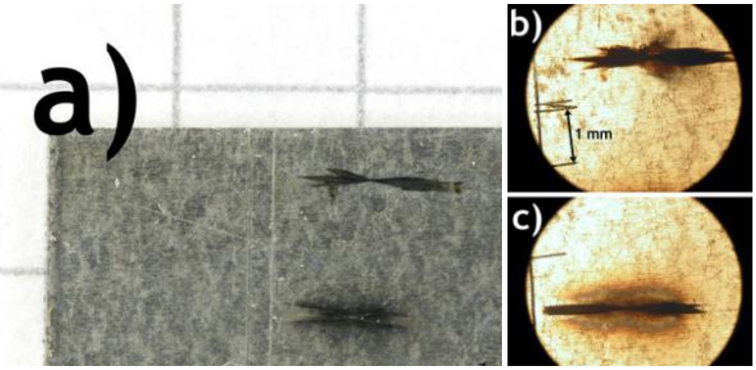

Fig. 3. a) molybdenum sample showing the ablation profile obtained in atmosphere (upper) and vacuum (lower). b) ablation profile in atmosphere, along with the scale used to measure the maximum ablation transversal length. c) ablation profile in vacuum, with the same scale as the one in atmosphere.

Conclusions

We have measured the molybdenum ablation threshold in the 30 femtoseconds regime in atmosphere and vacuum using the D-Scan technique. We have found that the ablation threshold in vacuum that is more than 2 times greater than in air. One possibility for explaining this is to assume that in atmosphere a fraction of the pulses energy is used to ionize the air and this somehow contributes to remove molybdenum atoms from the surface, or the plasma formed during the ablation etches the surface. On the vacuum case, the ejected material quickly leaves the surface, and the plasma cannot contribute to the etching. However, the dimensions were measured in the photographs, and a better method to determined them should be used to ensure the values are correct, since the ρ_{max} is squared in the denominator in eq. (1) and fluctuations on it lead to big variations in the ablation threshold values.

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