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Single-shot laser-induced damage threshold of free-standing nanometer-thin diamond-like carbon foils

Dahui Wang[a,b], Wenjun Ma[a,*], Jianhui Bin[c], Klaus Alinger[c], Yinren Shou[a], Pengjie Wang[a], Jianbo Liu[a], Jungao Zhu[a], Zhengxuan Cao[a], Zhusong Mei[a], Hongyong Wang[e], Haiyang Lu[a], Chen Lin[a], Yanying Zhao[a], Joerg Schreiber[c], Xueqing Yan[a,d,*]

[a] State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100871, China
[b] State Key Laboratory of Laser Interaction with Matter, Northwest Institute of Nuclear Technology, Xi'an 710024, China
[c] Fakultät für Physik, Ludwig-Maximilians-University, Am Coulombwall 1, D-85748 Garching, Germany
[d] Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, China
[e] School of Environment and Chemical Engineering, Shanghai University, Shanghai 200444, China

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A B S T R A C T

Single-shot laser-induced damage threshold (LIDT) of free-standing nanometer-thin diamond-like carbon (DLC) foils was measured in vacuum environment for pulse durations from 50 fs to 200 ps. It is found that, due to higher surface defects density, the LIDT of free-standing ultrathin DLC foils is lower than that of bulk DLC by a factor of 3, and the damage fluence is almost a constant of about 0.1 J/cm^2 when the pulse duration is longer than 500 fs. Different from DLC films coated on silicon wafer, the damage fluence of free-standing DLC has a weak dependence on their thickness. Based on the measurement, the damage mechanism is illustrated by virtue of the carrier population analysis, and the requirement on the temporal laser contrast when DLC targets are used in relativistic laser-plasma experiment is discussed.

1. Introduction

Interaction of ultrahigh-intensity laser pulses with ultrathin foils recently becomes a very attractive topic in laser-plasma research. It has been demonstrated that energetic protons up to tens of MeV [1–3] and coherent extreme ultraviolet/X-ray [4–8] emission can be generated from such interaction at relativistic laser intensity ($I > 10^{18}$ W/cm^2), which may lead to table-top particle/radiation source for many applications [9,10]. For all of the experiments at such intensity, the laser-induced damage threshold (LIDT) of the targets is a crucial parameter that can significantly influence the results, as the targets must survive through the prepulse before interacting with the main pulse. Many kinds of free-standing ultrathin foils made of different materials have been exploited in experiments as targets but detailed studies on their LIDT were rarely seen. Usually it was assumed that the LIDTs of the thin foils are the same as the bulk materials.

However, there are several reasons disproving the reckless use of bulk material's LIDT for ultrathin foils in relativistic laser plasma experiments: (1) The thicknesses of the foils sometimes are only a few nanometers, which is already close to the skin depth of the materials and will impose significant influence on the laser absorption and heat dissipation process. (2) The optical properties of ultrathin free-standing foils in general are different from that of the corresponding bulk materials due to the existence of surface defects and internal stress. (3) Most LIDT measurements in literatures are performed in air with multiple shots, so defect incubation and fatigue effects are included. But the thin foil targets in relativistic laser plasma experiments are shot by single pulses in vacuum. Because of these reasons, the single-shot LIDT of free-standing ultrathin foils in vacuum environment needs to be specifically studied.

In this letter, we report on the experimental results of the single-shot LIDT of free-standing diamond-like carbon (DLC) foils measured in vacuum environment. The dependence of LIDT on the laser pulse duration and on the thickness of the foils is revealed. It is found that the LIDT of ultrathin DLC foil is lower than that of bulk DLC and weakly depends on the thickness. The damage fluence is almost a constant when the pulse duration is longer than 500 fs. Based on the measurements, the damage mechanism is illustrated by virtue of the carrier population analysis.

* Corresponding authors at: State Key Laboratory of Nuclear Physics and Technology, Peking University, Beijing 100871, China.
E-mail addresses: wenjun.ma@pku.edu.cn (W. Ma), x.yan@pku.edu.cn (X. Yan).

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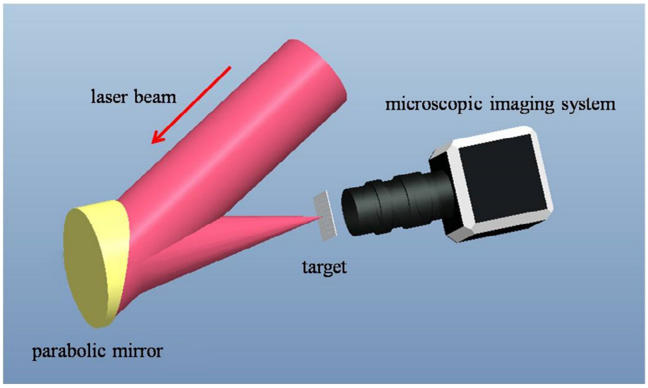

Fig. 1. Schematic configuration of experiments.

2. Experimental setup and methods

In order to obtain the single-shot LDT of the foils under the environment as in laser plasma experiments, we developed a method to perform the measurement at the shooting position in the vacuum chamber. The experiments were carried out by using the ATLAS Ti: Sapphire laser facility at the Max-Planck-Institut für Quantum Optik. The laser beam, target chamber, and target imaging system were the same as in relativistic laser plasma experiments (Fig. 1). The targets were precisely positioned at the focus spot with high-precision linear stages and shot by the laser pulses with different intensity and durations. After each shot, the target was front-illuminated by a collimated light source, and imaged by a 12 bit charged coupled device (CCD) behind a long-working-distance microscope objective lens to check the damage caused by the pulse. After saving the image of the shot target, a fresh target was moved in for the next shot to avoid multi-pulse effects. The full width of half maximum (FWHM) diameter of the focus spot was 15 µm, which was measured by the same microscopic imaging system as that for target imaging. The pulse duration was adjusted from 50 fs to 200 ps by changing the group delay dispersion (GDD) of the pulses. A FROG system (Swamp Optics) and a second-order auto correlator were used to measure the pulse duration. The pulse energy was varied by using different neutral density optical filters before the compressor. All the filters were thinner than 2 mm to avoid the self-phase modulation and pulse front distortion. A pyroelectric detector PE50 (OPHIR) was used to measure the pulse energy and calibrate the filters. The measured rms of the energy variation was less than 2%.

Free-standing DLC foils with thickness of 5 nm, 20 nm and 40 nm were used as targets. They were fabricated by filtered cathodic vacuum arc deposition (FCVAD) on NaCl layer and floated on a holder with 500 µm pinhole [11] as free-standing targets. The ratio of sp^3 bonds in such DLC foils was about 85%. Because of the high sp^3 bonds level, they have wide band gap up to 3.5 eV [12], high strength [13] and inert chemical properties. In previous studies, it has been found that they normally outperform many other targets like amorphous graphite and plastic foils in relativistic laser plasma experiments. Fig. 2 shows the morphology of 40 nm DLC targets after shooting by 50 fs laser pulses at different intensity, and the intensity distribution around the focus spot. At the peak intensity of

1.2×10^{12} W/cm^2 (Fig. 2(a)), the laser pulse didn't induce distinguishable damage on the foil. The foil was intact. At the peak intensity of 4.2×10^{12} W/cm^2 (Fig. 2(b)), a small pinhole with diameter of 18 µm was found on the foil. So the foil was already damaged at that intensity. The edge of the pinhole turned to dark due to the graphitization of DLC. With the increase of laser intensity, the damaged area increased in size as well. At the intensity of 1.3×10^{13} W/cm^2(Fig. 2(c)), the average diameter of the hole had increased to 36.5 µm. It should be noticed that the damaged area of the targets had the shape highly similar to the intensity distributions of the laser pulses. This indicates that the damage was intensity-determined, and lateral thermal damage was neglectable.

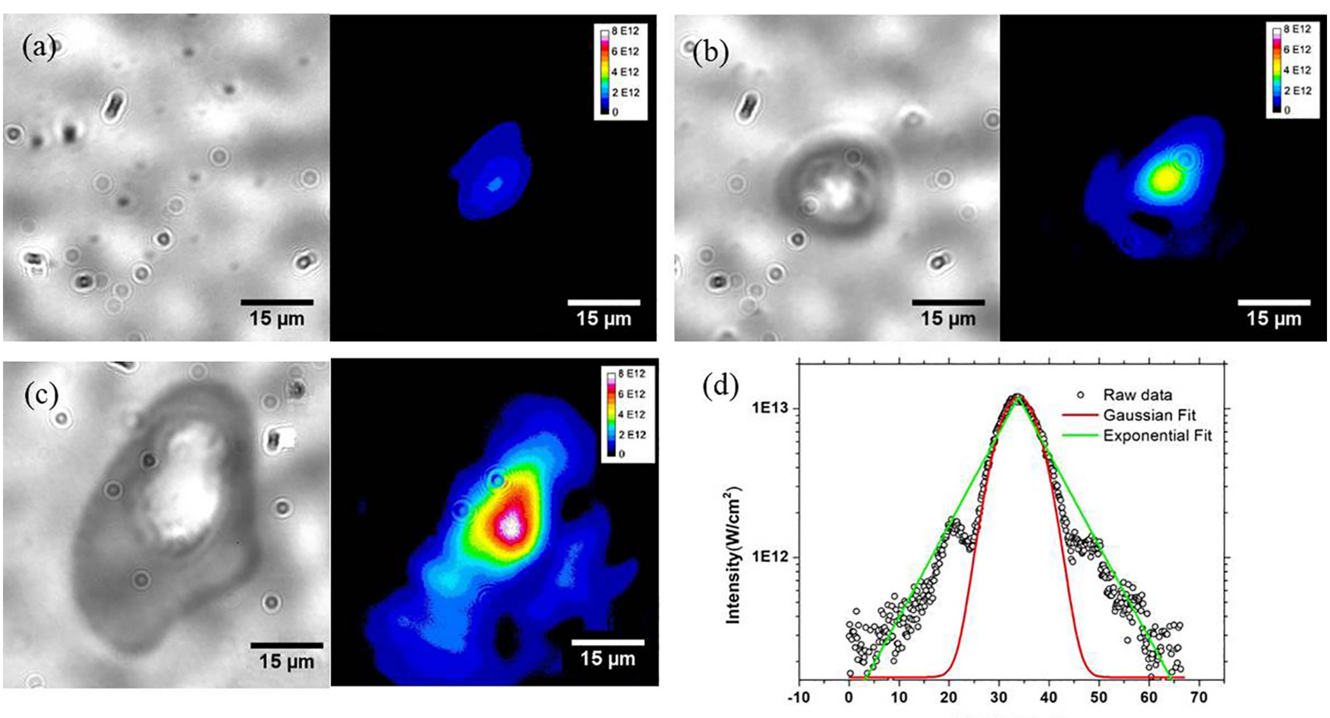

Fig. 2. Damage morphology of DLC targets (left panel) and corresponding intensity distribution of the laser pulses (right panel) at the intensity of 1.2×10^{12} W/cm^2 (a), 4.2×10^{12} W/cm^2 (b) and 1.1×10^{13} W/cm^2 (c). (d) A line-out of the intensity distribution (scattered black circle) around the focus spot. The distribution is Gaussian with $\sigma_{FWHM} = 14.6\ \mu$m nearby the focus spot. Far away from the focus spot, the intensity distribution can be fitted by $I = I_0 \exp(-x/7.5\ \mu\text{m})$.

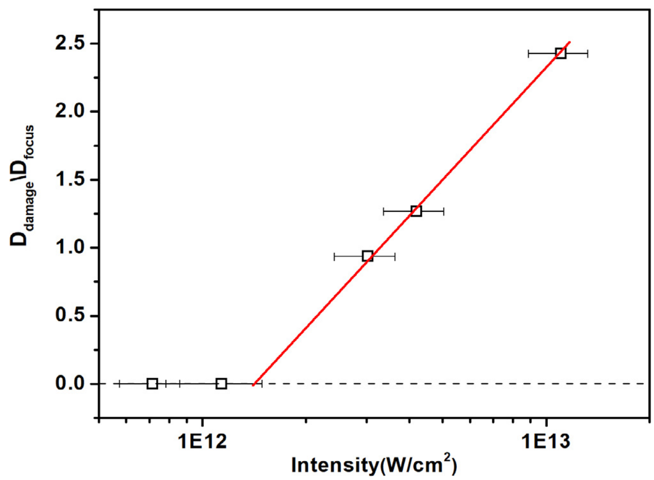

Fig. 3. The normalized diameter of the damaged area with respect to the laser intensity.

The single-shot LIDT can be deduced from the images of the target after shooting. A rough estimation can be made by observing the laser intensity at the edge of the damaged area, which is about $(2 \pm 1) \times 10^{12}$ W/cm^2 as shown in Fig. 2A more accurate way to determine the LIDT is by extrapolating the laser intensity to the value at which the damage of the foil just happens. Fig. 3 depicts normalized diameter of the damaged area with respect to the peak intensity of 50 fs laser pulses. The horizontal axis represents peak laser intensity on the targets, and the vertical axis is D/D_{focus}, where D is the mean diameter of damaged area andD_{focus} is the FWHM of the focus spot. In our case, D is obtained according to the following procedure. First, we got the skeleton of the damaged regions of the targets whose margins show the drastic change of the gray value. Then we calculate the area of the skeleton region and then obtain the diameter of the damaged area by normalizng it to a circular region. It is reported in previous studies that D and the laser fluence have the relationship of $(D/D_{focus})^2 \propto lnF$ [14,15] when the laser intensity has a Gaussian distribution, where F is the maximum pulse fluence. In our case, instead, we observed the relation as $D/D_{focus} \propto lnI$ as shown in Fig. 3, where I is the peak intensity of the laser. This is because in our case the laser intensity outside the $3D_{focus}$ of the focus spot has an exponential distribution instead of Gaussian distribution as shown in Fig. 2(d) due to the wavefront distortion of the high-power laser. When the peak laser intensity is much higher than the damage threshold, the diameter of the damaged area is determined by the intensity far away from the focus spot.

By extrapolating the data to $D/D_{focus} = 0$ in Fig. 3, we obtain$I_{th} = 1.4 \times 10^{12}$ W/cm^2, which is the single-shot damage threshold of laser intensity (DTI) for 40 nm DLC at pulse duration of 50 fs. Then, the corresponding single-shot damage threshold of laser fluence (DTF) F_{th} can be derived as $F_{th} = I_{th} \times \tau = 0.07$J/cm^2, where $\tau = 50$ fs is the pulse duration of the laser pulses.

3. Results and discussion

3.1. The dependence of LIDT on the pulse duration

Using the method described above, we performed series damage threshold measurements by changing the duration of the laser pulses. Fig. 4 shows the measured DTI and DTF of40 nm DLC targets for pulse duration τ50 fs to 200 ps. The DTI varies in a very wide range depending on the duration of the pulse with a minimum value 4×10^8 W/cm^2 @200 ps and maximum value1.4×10^{12} W/cm^2 @50 fs. However, the derived DTF varies little in the range of 0.07–0.1 J/cm^2. The highest DTF is 0.1 J/cm^2 at 500 fs. Compared to the DTF of micrometer-thick DLC foils, the DTF for 40 nm DLC is smaller by a factor of 3 [16,17].

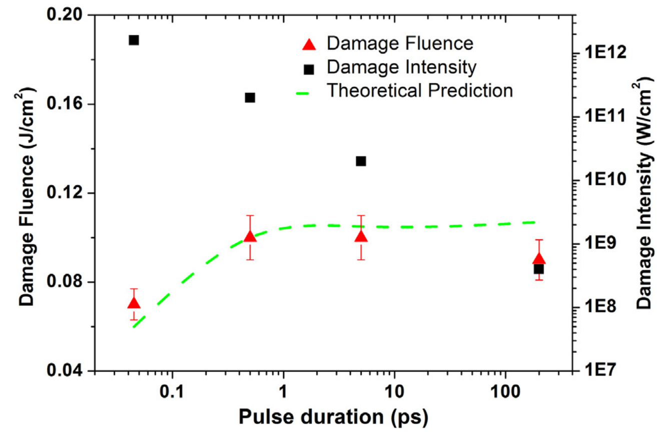

Fig. 4. Dependence of the damage threshold of fluence (left panel) and intensity (right panel) on the laser pulse duration.

The optical damage mechanism of ultrathin DLC targets can be deduced from the duration dependence of DTF. For transparent dielectric materials, optical breakdown happens when the density of free carriers (electrons, holes) equals to the critical density [18]. The major ionization process of the carriers is multiphoton ionization and avalanche ionization [19]. Multiphoton ionization is important for the generation of seed electrons in the conduction band. These free electrons absorb laser energy by inverse bremsstrahlung. When an electron achieves the energy equal to the band gap, subsequent impact ionization promotes another valence electron into the conduction band, which in-turn results in a stronger absorption, leading to the avalanche ionization process. The defects in the material can result in seed carriers as well, which will significantly reduce the LIDT. We can use the rate equation of free electrons [20,21] to analyze the evolution of the free electron density n(t) in DLC as

$$\frac{dn(t)}{dt} = \delta I(t)n(t) + \sigma_k I^k \tag{1}$$

where δ is the avalanche coefficient, k is the smallest multiphoton order, σ_k is the k_{th} absorption cross section and $n(0) = n_0$. For our case, the photon energy $\hbar\omega = 1.55$ eV and band gap $\Delta = 3.5$ eV. The corresponding k is 3.

By fitting the measured DTF values at different pulse duration using linear regression method based on Eq. (1), we obtained the parameters for the DLC foils as $\delta = 60$ cm^2/J, $\sigma_3 = 10^{20}$ cm^{-3}ps^{-1}(cm^2/TW)^3, and $n_0 = 5 \times 10^{19}$ cm^{-3}. Here, n_0 is the intrinsic seed electron density (ISED) due to the defects in the material. The black dash line in Fig. 4 depicts the calculated DTF using the obtained parameters. Compared to other results obtained from bulk DLC as $\delta = 80$ cm^2/J, $\sigma_3 = 10^{20}$ cm^{-3}ps^{-1}(cm^2/TW)^3, and$n_0 = 3.6 \times 10^{17}$cm^{-3}, the values of δ and $\sigma_3$are similar, but the n_0 is larger by 2 order of magnitude [22,23]. The experimental results show that the DTF of ultrathin DLC is approximately a constant value of about 0.1 J/cm^2 for $\tau > 500$ fs. This trend is the result of high ISED. Because of the high ISED, avalanche ionization in ultrathin DLC can be easily triggered even without enough contribution from multiphoton ionization. The free electron rate equation can be simplified as $\frac{dn}{dt} = \delta I(t)n_0$. As a result, the DTF isa duration-independent value determined by $F_{th} = \frac{2}{\delta}\ln\left(\frac{n_{cr}}{n_0}\right)$ [24].

For an ultrashort pulse, tunneling effect could contribute to the ionization and becomes significantly important for LIDT. It could be signified by the Keldysh parameter of $\gamma = \omega\sqrt{2m\Delta}/(eE)$ [25], where m is the mass of the electron, e is the charge of the electron, and E is the peak laser field. When $\gamma < 1$, the tunneling effect is the dominate ionization mechanism. For the parameters $\lambda = 800$ nm, $\Delta = 3.5$ eV, and $I_{th} = 2.0 \times 10^{12}$ W/cm2, $\gamma \approx 3.8$ is yielded. At this time, multiphoton ionization used in Eq. (1) still dominates laser absorption [26].

3.2. The dependence of LIDT on the thickness of DLC foils

In previous studies, the LIDT of laser-deposited DLC film coated on Si wafer were studied [27,28]. It's found that the DTF linearly scales up with the thickness of DLC if the films are thinner than the optical penetration depth l_{pe} $\alpha_{eff}{}^{-1}$, here α_{eff} is the effective absorption coefficient taking into account of the influence defects. We measured the LIDT of 5 nm and 20 nm DLC at 50 fs pulse duration as well. It turned out that the DFT of 5 nm and 20 nm DLC is 0.068 J/cm^2 and 0.07 J/cm^2, respectively. By comparing to the DFT of 0.07 J/cm^{2}of 40 nm DLC, we can draw to a conclusion that the dependence of LIDT on the thickness of nanometer-thin DLC foils is weak. It implies that the foils we investigated synthesized through FCVAD were close to or thicker than the optical penetration depth. This is possible because the effective absorption coefficient of our free-standing DLC foils should be large considering their high intrinsic carrier density.

4. Conclusions

In conclusion, we have performed experimental study on the single-shot optical damage threshold of nanometer-thin free-standing DLC foils for pulse durations from 50 fs to 200 ps. Compared to bulk DLC or DLC film coated on wafers, free-stand ultrathin DLC foils have lower damage threshold. More important for the application of the foils as targets in laser-plasma experiment, is that their damage fluence is almost a constant value of about 0.1 J/cm^2 for pulse durations from 500 fs to 200 ps. Our results can be used as important criterion to judge whether the DLC foils can survive through the prepulse or ASE pedestal for a certain high-power laser. For the state-of-art high-power laser facilities nowadays, the peak laser intensity is in the order of 10^{20} W/cm^2. Based on the obtained LIDTs, if one wants to use free-standing ultrathin DLC as targets in relativistic laser-plasma experiments, the temporal contrast ratio of the laser should be higher than 10^{11}-10^{12}:1 at nanosecond scale, 10^{10}-10^{11}:1 at tens of picosecond scale, and 10^{9}-10^{10}:1 at a few picoseconds scale accordingly. This requirement is highly challenging for current laser technology. Even if cross-polarized wave generation (XPW) or optical parametric chirped pulse amplification (OPCPA) techniques are used, single plasma mirror or double plasma mirrors system is still needed. Thus, for applications that requires high operation rate like laser-driven hadron therapy, new techniques that can improve the contrast by 10^2-10^4 in a wide temporal range at high operation rate are highly demanded.

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