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Preparation of self-supporting diamond-like carbon nanofoils with thickness less than 5 nm for laser-driven ion acceleration

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ABSTRACT

Ultrathin (< 5 nm) self-supporting diamond-like carbon (DLC) foils are prepared by filtered cathodic vacuum arc (FCVA) deposition method as targets for laser-driven ion acceleration. The thickness and the morphology of these foils are characterized by atomic force microscope (AFM) and scanning electron microscope (SEM).

0. Introduction

Due to the limitation by the RF acceleration cavity and bending magnet, modern accelerators can be unexceptionally huge and costly. The acceleration gradient in conventional machines is typically less than 100 MeV/m due to sparks and breakdown of the metal wall. For more compact and cheaper accelerators, it is essential to scale up the acceleration gradient in a new way. In the past 15 years, with the invention of chirped pulse amplification (CPA) technique [1], terawatt to petawatt class laser systems have become available, which currently provide light intensity up to 10^{22} W/cm^2 [2]. By employing such high-intensity laser pulses as driving power, an acceleration gradient of over 1 TeV/m can be achieved in a plasma environment. The stable acceleration of quasi-monoenergetic electrons in the GeV energy range [3–7] is among the biggest successes of the field of high-intensity laser plasma interaction, including the generation of undulator radiation as a step towards next generation X-ray sources [8]. Ion accelerations with energy of several MeV/nucleon were also realized [9–11], with a maximum energy of around 60 MeV for protons [12]. Although still in the era of its infancy, table-top size and low energy consumption make laser-driven ion acceleration especially attractive and promising for the application of ion beam therapy against tumors [13].

Until recently, ion acceleration to the MeV/nucleon range relied on the interaction of laser pulses beyond 10^{18} W/cm^2 with micrometer thick targets. Relativistic electrons are generated and spread around the target foil and set up fields of TV/m at the plasma-vacuum boundaries, driving an expansion normal to the initial surface (TNSA, Target Normal Sheath Acceleration) [12,14–16]. Atoms at the rear surface in such fields are ionized and subsequently accelerated over micrometers to MeV energies. Due to the small acceleration time of the order of the pulse duration (< ps) and the small source size (10–100 μm), the quality of these beams in terms of emittance is exceptionally good [17] and the peak currents can be as high as 1 MA. However, it is inherent to this plasma expansion that the ion beams usually exhibit a continuous, exponentially decaying spectrum, where the portion of the useful high-energy ions is small.

In contrast to TNSA with thick targets, an alternative acceleration mechanism (Radiation Pressure Acceleration (RPA)) relies on the radiation pressure of lasers [18–21] and very thin targets. In this regime, the ultrathin target can be crudely viewed as a relativistic plasma mirror co-propagating with the laser pulse and accelerated as a whole. Compared to the TNSA regime, ion acceleration in the RPA regime has considerably higher efficiency (> 10%) [11] due to bunching of ions at higher energies, i.e. RPA can result in a quasi-monochromatic instead of continuous energy spectrum. Since the energy of ions is proportional to the laser intensity in the RPA regime, a 100 MeV/nucleon quasi-monochromatic ion beam for radiation therapy is expected with PW laser pulses of few tens of fs duration in the near future. Using ultrathin (< 5 nm) diamond-like carbon (DLC) foils and careful preparation of the laser pulses (removal of prepulses), this mechanism was recently realized experimentally [10].

Diamond-like carbon is a kind of amorphous carbon with a combination of sp^2 and sp^3 bonds [22]. DLC films can be very hard, strong, transparent, super-smooth, and chemically inert. Due to the combination of these properties, DLC is widely used as an ultrathin protective coating on magnetic hard disks and their

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reading head [23]. In the field of nuclear targets, Ivkova et al. [24] at the Kurchatov Institute fabricated ultrathin self-supporting DLC foils by sputter deposition and used them as long-lived stripper foils for high-energy heavy-ion tandem accelerators and effective secondary electron emitting foils for time-of-flight (TOF) spectrometers [25–28]. For laser acceleration experiments, DLC foils are ideal targets since they are very strong and transparent, which ensure their intactness under the radiation of pre-pulse before the arrival of main laser pulse. In this paper, we report our recent development in the preparation of ultrathin self-supporting DLC foils using the filtered cathodic vacuum arc (FCVA) technique that enabled a breakthrough in laser-driven ion acceleration.

1. Preparation procedure

DLC films can be synthesized by several methods such as sputter deposition [29], pulsed laser ablation [30], ion beam deposition [31], and plasma enhanced chemical vapor deposition [32]. As an economic synthesis method, filtered cathodic vacuum arc (FCVA) has been applied for years in the protective coating of hard carbon films <5 nm [33]. Compared to other methods, the advantages below make FCVA especially suitable for the synthesis of self-supporting DLC foils: (1) its capability to provide hydrogen-free and highly transparent films with the best diamond-like properties (up to 85% of sp^3 bonding); (2) continuous and strong, ultrathin DLC films down to several atomic layers could be produced using pulsed arc at low deposition rates; (3) characteristics of deposited DLC foils could be controlled in a wide range of parameters using proper electrical bias, thus changing the energy of incoming carbon ions; and (4) FCVA is essentially a low-temperature deposition technique, which enables a variety of parting agents to be used to produce ultrathin self-supporting foils. A simplified illustration of our FCVA system is presented in Fig. 1. A highly ionized plasma is generated by a pulsed arc at the surface of a graphite cathode with average drift energy of 10–30 eV, then slightly accelerated by the anode. A 90° magnetic duct is set between substrate and anode to filter the macro-particles from the plasma. After that, most macro-particles are removed since they are not guided by the magnetic field. To control the energy of carbon ions, a bias is applied to the substrates. The deposition rate of DLC is controlled both by the current and by the pulse rate of the arc. To minimize the intrinsic stress of the DLC films, we adopted both a low bias voltage (20 V) and a low deposition rate (0.3 nm/min).

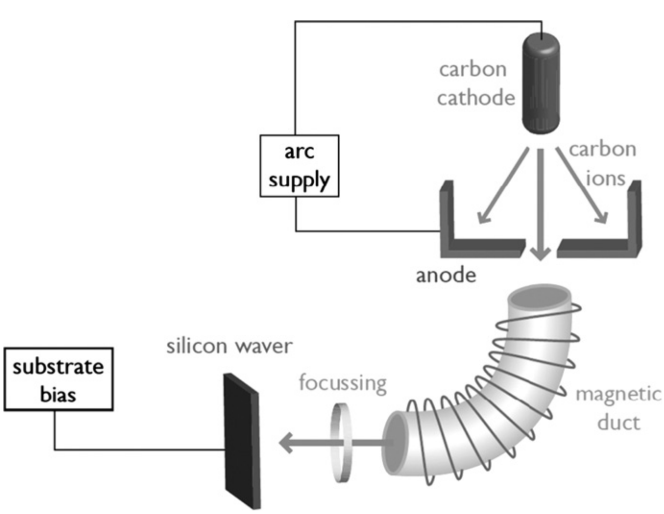

Fig. 1. Simplified illustration of our FCVA system for the deposition of DLC foils.

Polished silicon wafers were used as substrates for the deposition. Between the silicon wafer and the DLC, there is a parting agent layer for the release of the DLC film. It is well known that the parting agent substantially influences the major properties of the resulting foils. For ultrathin DLC foils, it was found that some extent of corrugation helps strengthen the foils. We adopted thermally deposited sodium chloride as parting agent, which has a corrugated morphology with a rms of about 10–20 nm. After the deposition process, DLC foils were floated in distilled water according to the standard procedure and mounted onto steel frames. The largest size of the self-supporting DLC foils mountable depends on their thickness and the parameters of the frames. Frames with round holes and smooth surface are the best according to our experiences. We succeeded to mount 3 nm thick DLC foils over circular apertures with a diameter of up to 1 mm.

2. Characterization

2.1. Thickness measurement

For highly precise measurements of the DLC thickness as well as surface morphology analysis, a commercial profilometer equipped with an atomic force microscope (AFM) sensor (MicroProf, FRT Fries Research & Technology) was used. Depending on the sample, the vertical resolution of our AFM can usually be as high as 0.2 nm, which is good enough for the thickness determination of DLC nanofoils. Characterization of the thickness was done on a reference silicon wafer rather than on the parting-agent-coated actual substrate. Before placing the reference wafer next to the actual substrate and starting the deposition, a mark line was drawn by a marker pen on its surface. After the deposition, the mark line was erased from the reference wafer with acetone, resulting in a clean, uncoated silicon surface separated from the DLC-coated area by a sharp step, which was used to define the thickness of the DLC. Fig. 2 shows a typical AFM topography for thickness determination of DLC foils. From the profile at the step, we can determine the thickness of that film to be about 3 nm.

2.2. Morphology of the DLC nanofoils

One unique feature of DLC films is their sub-nanometer smoothness [34]. Their surface morphologies are mainly determined by the substrates they are deposited on. In our case, the surface morphology of DLC nanofoils is determined by that of the sodium chloride layer. Fig. 3 shows the morphology of a DLC foil attached to the parting agent layer. The roughness of about 20 nm comes from the distinct grains of the polycrystalline salt. Besides AFM, we adopted scanning electron microscope (SEM) to characterize the self-supporting DLC nanofoil after the floating process. Fig. 4 shows the SEM images of a self-supporting DLC nanofoil. At the $10\times10\ \mu m^2$ scale, the film has satisfactory smoothness, which is essential for laser acceleration. At the $1\times1\ \mu m^2$ scale, structures with a size of 100 nm can be seen, which is consistent with the AFM result shown in Fig. 3.

3. Conclusion

By taking advantage of a filtered cathodic vacuum arc (FCVA) deposition method, we produced self-supporting ultrathin diamond-like carbon (DLC) nanofoils down to 3 nm. These nanofoils are extremely robust for handling and transportation. In combination with their optical transparence at moderate laser intensity, they are ideal targets for laser-driven ion acceleration in the radiation pressure acceleration regime.

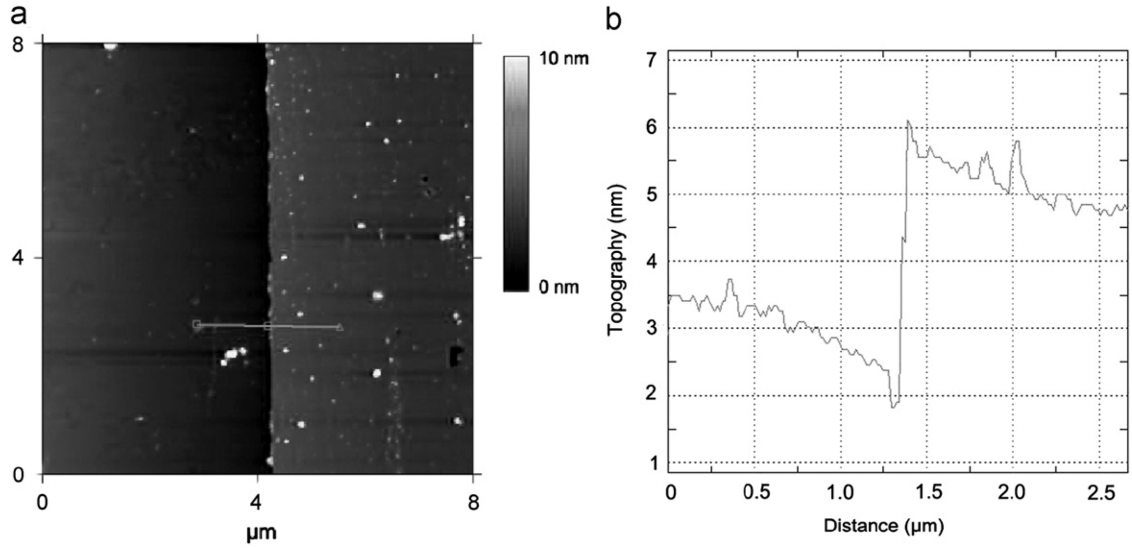

Fig. 2. (a) Topography of a reference sample used for the thickness measurement. The red line marks the region of the linear profile in (b). (b) Linear profile near the boundary of DLC-coated area and uncoated area.

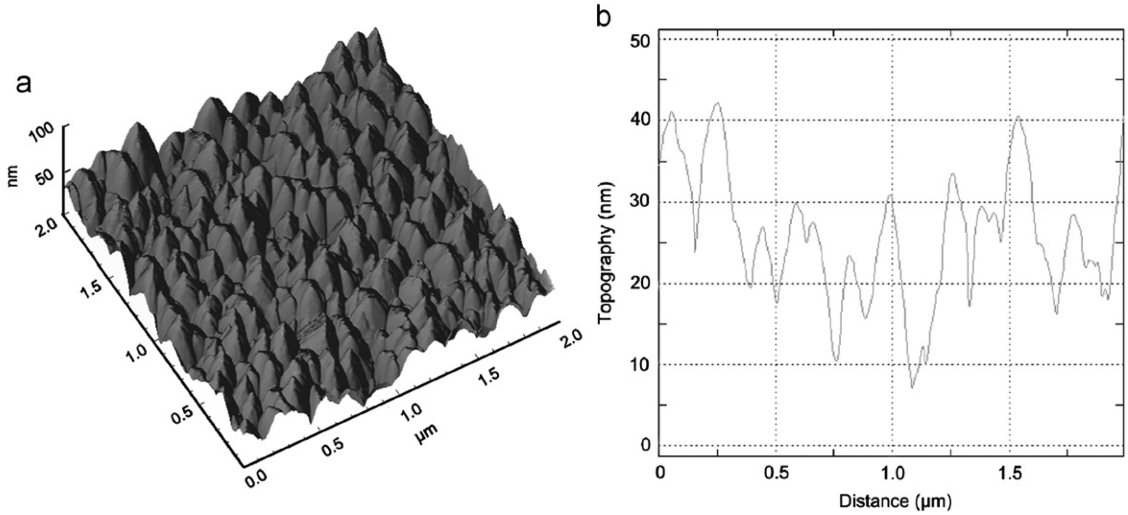

Fig. 3. Topography of a DLC foil on NaCl sacrificial layer. (a) AFM image of $2 \times 2\ \mu m^2$ region. (b) Linear profile of the DLC foil.

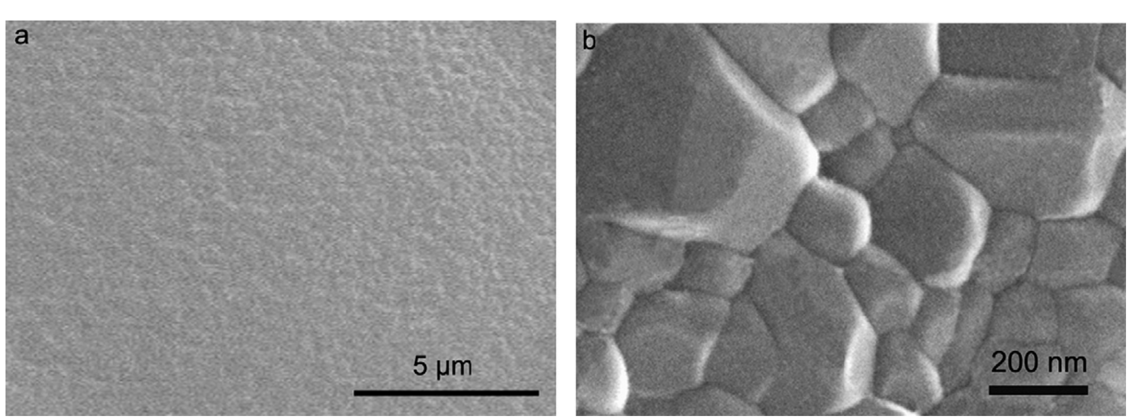

Fig. 4. SEM images of a self-supporting DLC nanofoil.

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