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Surface roughness-aided hard X-ray emission from carbon nanotubes

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Abstract. Efficient low debris hard X-ray source based on multiwalled carbon nanotubes (MWNT) irradiated by intense, femtosecond laser over an intensity range of 10^{15}–10^{17} W cm^{-2} μm^2 is reported. The MWNT targets yield two orders of magnitude higher X-rays (indicating significant enhancement of laser coupling) and three orders of magnitude lower debris compared to conventional metallic targets under identical experimental conditions. The simple analytical model explains the basic experimental observations and also serves as a guide to design efficient targets to achieve low-debris laser plasma-based hard X-ray sources at low laser intensities suitable for multi-kHz operation.

Keywords. Carbon nanotubes; intense femtosecond lasers; X-rays; local field enhancement.

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1. Introduction

The advent of intense, ultrashort solid-state chirped pulse amplification lasers (USUIL) have come up with possibilities of realizing ultrashort high peak brightness sources of photons and particles on a table top [1]. These sources provide unique opportunities to study the matter under extreme conditions, challenging in terms of proper scientific understanding and promising for their potential technological applications [2]. When a USUIL pulse interacts with matter, it instantaneously ionizes it generating free electron which further couples more laser energy by collisional as well as collisionless absorption processes such as resonance absorption, vacuum heating [3] etc. leading to an avalanche breakdown of the material resulting in the formation of hot dense plasma. The electrons oscillating under the laser electric field collide with the surrounding ions giving rise to transfer of electromagnetic energy to the plasma which in turn produces energetic photons while recombining back.

As the electrons play the essential role in producing bremsstrahlung radiation, it is of primary interest to make the electron generation process more efficient.

The generation of hard X-rays from the laser produced near solid density plasmas essentially depends on the initial interaction of the laser pulse with the plasma and the subsequent hydrodynamic evolution of the plasma together with the energy redistribution. So far efficient laser coupling to plasma has been achieved by employing intentional pre-plasmas at the cost of increase in X-ray pulse duration [4,5]. Bagchi *et al* [6] suggested use of nanostructured surfaces as a route to efficient laser coupling in plasma. A clear enhancement in the low to moderately high-energy X-rays has been noticed.

In this report, we present a highly efficient, low-debris hard X-ray source based on multiwalled carbon nanotubes (MWNT) irradiated by intense, femtosecond laser. In the process we also demonstrate the critical role of surface roughness which couples the laser energy to plasma very efficiently. The characteristics of the plasma produced by MWNT is compared with copper surface (Cu, used as substrate) to see the relative efficiency of the structured surface.

2. Experimental arrangement

Experimental studies are performed with p-polarized, 50 fs, 800 nm, 10 Hz Ti:sapphire laser (THALES, ALPHA 10) pulses focussed in $f/4$ geometry by an off-axis parabolic mirror having a high reflectivity in 800 nm region, at an angle of 23° with respect to the target normal direction. The focal spot radius is found to be 10 μm ($1/e^2$) using an equivalent imaging technique. The targets are placed side by side to attain identical experimental conditions. A motorized stage assembly ensures a fresh target region for every laser shot. The base pressure of the chamber is kept at 10^{-6} Torr.

The hard X-ray measurements in the 20–300 keV range are performed with a calibrated NaI(Tl) detector gated in time with respect to the incident laser pulse to ensure almost background-free data acquisition. The detector is covered by thick lead walls to reduce the pile-up events. The detector has almost 100% efficiency in the detection range considered [7]. The signal from the detector is collected and analysed by a multichannel analyser (MCA) attached to a computer.

The energy of the ions emitted from the plasma normal to the target surface is measured by a channel electron multiplier (CEM) used in proportional mode, placed 97 cm away from the target subtending an angle of 26 msr on the focal spot, using conventional ion arrival time measurement technique. Four large annular Faraday cups (AnFC) [8] are placed at different angles (θ) of 5, 8, 12 and 17° with respect to the axis of the time-of-flight spectrometer to measure the total flux as well as the angular divergence of the ions emitted from the plasma.

The MWNTs are synthesized on copper substrate through the catalytic pyrolysis of a ferrocene–xylene mixture at around 675°C fed into a tubular two-stage quartz reactor [9]. Scanning electron microscope (SEM) images reveal that MWNT are grown perpendicular to the surface of the copper substrate with a thickness of about 1–10 μm depending on reaction time. In the present experimental study we have used MWCNTs having a length of 1.1 μm and a width of 100 nm as shown in

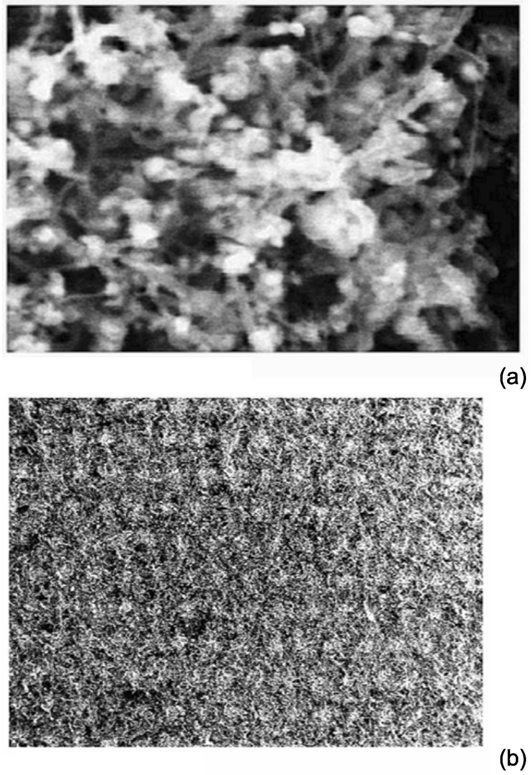

Figure 1. Scanning electron microscope (SEM) image of (**a**) multiwalled carbon nanotubes and (**b**) copper substrates.

figure 1a. The characteristic of the plasma generated on this sample is compared with polished copper surface as shown in figure 1b.

3. Experimental results

The first measurement performed on these samples was the relative estimates of the bremsstrahlung yield for a fixed number of incident laser shots. Figure 2a shows the ratio of bremsstrahlung yield from MWNT and Cu samples. The two orders of magnitude X-ray yield from MWNT is evident. Notably the ratio increases with increasing laser intensity up to an intensity level of 4×10^{16} W cm^{-2} clearly indicating a large absorption of laser energy. After this, the ratio goes down because of the damage of the MWNT coating, a behaviour similar to the previous experimental observations [6]. Examining the sample under optical microscope reconfirms the fact that the coating is completely ripped off the surface of the substrate. The bremsstrahlung measurement, acquired over 4000 independent laser exposures at count rates less than 0.1 counts/s ensuring no pile-up events, at a laser intensity of 2.1×10^{16} W cm^{-2}, reveals 'hot' electron temperature distribution of 6.5 ±

1.5 keV and 14.9 ± 2.4 keV from MWNT compared to 9.3 ± 1.6 keV. During these measurements the count rates were kept less than 0.1 counts/s to minimize the occurrence of any pile-up events.

It is to be noted that in the case of intense, short pulse laser produced plasmas, two distinct kinds of electrons are produced. The generation of electrons though starts with collisional process but soon taken over by more efficient collisionless processes such as resonance absorption (RA), vacuum heating [3] etc. In RA, it is the damping of the electron oscillations set up by the incident p-polarized laser pulses giving rise to higher component of the bremsstrahlung radiation. This process can be conveniently expressed in the form of scaling law [10], T_{hot} (keV) $= 14(T_{\mathrm{c}}I\lambda^2)^{0.33}$ where T_{c} is the bulk plasma electron temperature in keV at the critical density, I is the intensity of the laser in units of 10^{16} W cm^{-2} and λ is the wavelength of the laser in μm. From the experimentally determined bremsstrahlung temperatures, the enhanced localized electric field near the tip of the nanostructures comes out to be 19.5 times the incident laser electric field.

Now, looking from the perspective of localized field enhancement near a sharp structure, the electric field enhancement by a nanotube can be modelled [11] as $E_{\mathrm{local}} = 1.2(2.15 + L/r)^{0.9}$, where r is the diameter and L is the length of the nanotubes. In this case, the field enhancement factor comes out as 21 times the incident laser intensity, a remarkable agreement with experimental observation.

Figure 2b shows the comparison of maximum ion energy emitted from both the samples. It is evident that except at the lowest incident laser intensity used in this experiment, Cu emits higher energetic ions compared to MWNT-coated layers. The maximum ion energy emitted from Cu with increasing laser intensity reasonably follows the scaling law [12] $(I\lambda^2)^{0.4}$ whereas for MWNT, the maximum ion energy steadily goes down. At the lowest laser intensity, the MWNT is seen to be emitting higher energetic ions because of plasma formation whereas for Cu, the plasma might have barely formed. These observations clearly indicate that the MWNT-coated layer is a very efficient laser energy absorber and the plasma formation threshold intensity is lower for MWNT due to its structural property.

The ion accelerating electric field can be written as [13] $E_{\mathrm{accl}} = T_{\mathrm{e}}/\max(L_{\mathrm{n}}, \lambda_{\mathrm{D}})$, where T_{e} (eV) is the 'hot' electron temperature, λ_{D} is the Debye length and L_{n} is the local plasma scale length. The localized enhancement of the electric field near the tip of the nanotube facilitates early plasma formation thereby coupling more laser energy. But this localized excitation deforms the plasma sheath layer leading to a large localized plasma scale length and non-planar plasma expansion. As a consequence, the ion accelerating electric field magnitude is considerably reduced. Also, the non-planar plasma expansion leads to a divergent ion emission reported in the next paragraph.

The angular divergence of the ions measured by the annular Faraday cups (AnFC) at two different laser intensities used in the experiment is presented in figure 2c. It is clearly seen that the ion yield from the MWNT surface is two orders of magnitude less than that from Cu. Notably, the ion emission from MWNT is extremely divergent in nature compared to Cu where the ion emission is mostly in the forward direction. This presents a great practical advantage of using nanotubes as moderately hard X-ray source compared to any other laser produced plasma-based X-ray sources reported in the literature. This reconfirms the previous conclusion

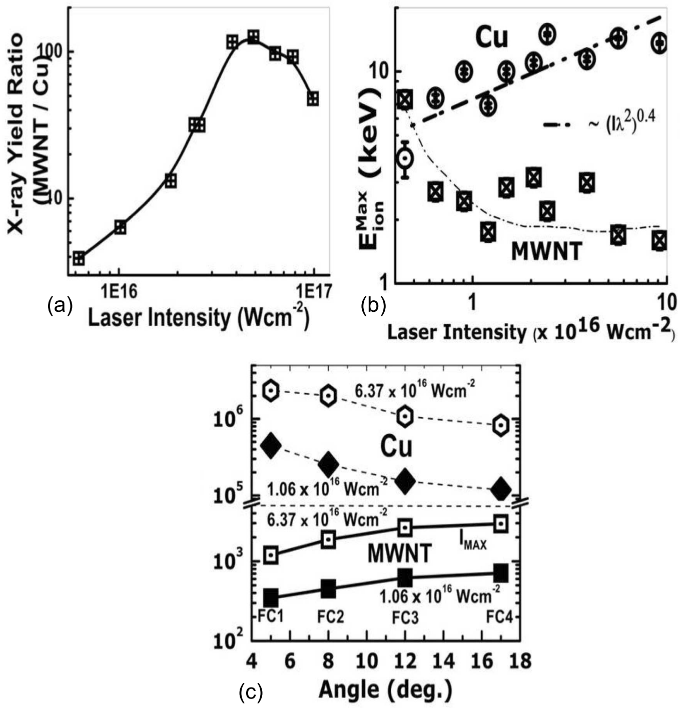

Figure 2. (**a**) Ratio (MWNT/Cu) of bremsstrahlung yield (crossed square) with increasing laser intensity. (**b**) Maximum ion energies from MWNT (square) and Cu (circle). The dash–dot line (Cu) represents the experimentally obtained scaling law behaviour $(I\lambda^2)^{0.4}$ of maximum ion energy with increasing laser intensity. The dotted line (MWNT) is drawn for visual aiding. (**c**) Spatial divergence of the ejected ion flux at two different laser intensities. Notice the clear change in pattern for MWNT.

of MWNT as an efficient laser absorber but a poor ion emitter and supports the occurrence of non-planar plasma expansion as predicted earlier.

In conclusion we report a highly efficient, inexpensive, low debris and high repetition rate moderately hard X-ray point source based on laser-produced plasma for a plethora of applications such as diffraction imaging, lithography, material processing, to name a few.

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