Chapter 9

Near complete absorption in carbon nano-tubes.

The plasma produced from solid targets using an ultra-short laser pulse is a point like, pulsed source of x-rays of ultra-short duration [72, 208]. Such a source can be very useful for applications like mammography [170, 209], phase contrast imaging [210, 211], and radiography [212]. The $K_{\alpha}$ x-ray line emission from the ultra-short pulse laser produced plasma can provide greatly improved contrast, spatial resolution as well as better dose utilization for mammography applications [213]. Among various targets, molybdenum has an edge over others since its characteristic x-ray emission at 17.5 keV falls in the 17.3-28.5 keV range, which is most optimal wavelength range for mammography [214]. For realizing a compact table top x-ray source, it is necessary to enhance the conversion efficiency of laser into the $K_{\alpha}$ photons. Among the various processes involved in the absorption of the incident intense laser pulse in the plasma, processes like resonance absorption, $J \times B$ heating, and vacuum heating lead to the generation of hot electrons [31, 52, 59]. These hot electrons penetrate the surface layer of cold plasma and interact with the solid core of the target to produce its characteristic x-rays [109, 210]. Increasing the laser intensity is not always useful to increase the $K_{\alpha}$ flux as the more-energetic electrons produced at higher intensity penetrate deeper into the target and produce x-ray photons deep inside the target, resulting in their re-absorption [109, 210], and increases the size of the source resulting in poor spatial coherence. Moreover, in this case, as the x-rays come from different depths, the pulse duration of the x-rays also increases. Therefore, there is an optimal hot-electron temperature for efficient $K_{\alpha}$ x-ray conversion, which is roughly 5-6 times the K-shell ionization energy [109, 210]. In view of this, the best way to enhance the $K_{\alpha}$ emission is to generate more
hot electrons with the desired optimal temperature [109, 210]. One way to achieve this is to utilize the electric field enhancement through resonances in nano-structures, leading to the generation of an increased fraction of hot electrons [72, 76, 87, 177, 202, 215-218]. For this purpose, carbon nano-tubes (CNTs) can be good choice since they have a hollow structure which supports two surface plasmon resonance modes, resulting from the hybridization of the cylindrical cavity plasmon and nano-rod plasmon [219]. It is therefore not very surprising that, as explained earlier in Chapter 8, the CNT plasma supports resonance at two electron densities, and depending on the degree of hollowness, the resonance can be tuned to even close to solid density [220]. A resonance at high density is desirable since if the field enhancement takes place at an instant when the electron density is also high, a larger number of electrons can gain energy from the enhanced field. Unfortunately, this is not the case with planar solid, nano-rod, or cluster targets, where the resonance density is of the order of the critical density (For Ti:sapphire laser, \( n_c \approx 1.7 \times 10^{21}/\text{cm}^3 \)). For solids, the resonance takes place at \( n_c \) [31], at \( 2n_c \) for nano-rods [177,202], and at \( 3n_c \) for clusters [96]. Hence, it may be interesting to use CNT coated on planar Mo target to study laser energy absorption and x-ray emission.

In the Chapter 8, the theoretical aspects of laser nano-tubes interaction and its merits were discussed in detail. We performed experiments with planar target coated with carbon nano-tubes to study the effect of hollow structure in laser energy absorption, x-ray emission, and electric field enhancement. We describe here a comparative study of the laser energy absorption and the hard x-ray emission in the 2-20 keV range from uncoated planar molybdenum, and planar molybdenum coated with CNTs. Near complete energy absorption of intense ultra-short laser pulses is observed in the case of CNT deposited on planar Mo. The enhancement in K\(_\alpha\) emission in CNT coated Mo target over planar Mo target is observed to be very sensitive to the
laser intensity. It is also observed that for nano-tubes of a certain degree of hollowness, there exists an optimum laser intensity for maximum $K_\alpha$ x-ray enhancement compared to the uncoated planar target. The results are explained considering the hollow structure of the nano-tube plasma which facilitates resonant electric field enhancement at two plasma electron densities during plasma evolution in time as discussed in Chapter 8. This resonantly enhanced localized field at a density much larger than the critical density $n_c$ leads to highly efficient hot electron generation, which results in enhanced $K_\alpha$ emission.

9.1 Description of the Experiment

![FIG. 9.1: A schematic diagram of the experimental setup](image)

Figure 9.1 shows a schematic diagram of the experimental setup. The plasma was produced by focusing 90 mJ, 45 fs (FWHM) Ti:sapphire laser pulses ($\lambda = 790$ nm) on a planar molybdenum with / without pre-deposited 25 μm thick layer of CNTs. Synthesis of carbon nano-tubes was done by cracking of acetylene in presence Fe as catalyst in quartz tube. The substrate material was placed in the central part of the tube, where a temperature of ~ 900° C was maintained. The p-polarized laser beam was incident on the target at an angle of 45° with respect to the target normal in the intensity range of ~1.6x10^{16} W/cm^2 to 2.5 x10^{17} W/cm^2. The laser energy absorption was estimated by collecting the reflected laser light using a large aperture
collection lens (to collect specularly reflected as well as diffusely reflected laser light). Diffused scattering of the laser light in other directions was measured to be insignificant. The x-ray spectrum in the 2-20 keV region was recorded using a dispersion-less spectrograph with a 25 μm thick Ni foil (cut-off at 7.9 keV).

9.2 Results

Figure 9.2 shows the transmission electron microscope (TEM, Model : Philips CM200) image of the CNTs. The nano-tubes are clearly seen to be hollow, having an average outer diameter of 30 nm and a degree of hollowness of ~ 0.9 (the degree of hollowness is defined as the ratio of the inner to outer radii of a nano-tube).

![TEM image of the CNTs showing the hollow structure](image)

**FIG. 9.2: TEM image of the CNTs showing the hollow structure**

Figure 9.3a shows the variation of the absorption of the 45 fs laser pulses in the planar Mo target and CNT coated Mo target, in the laser intensity range ~1.6 x10^{16} W/cm^2 to 2.5x10^{17} W/cm^2. A near complete absorption is observed in CNT coated Mo target indicating efficient coupling as compared to the planar Mo. Figure 9.3b shows the hard x-ray emission spectrum from planar Mo and Mo coated with CNTs at a laser pulse intensity of 8 x10^{16} W/cm^2. The
spectrum shows a large enhancement of K$_\alpha$ and bremsstrahlung emission from CNT coated Mo as compared to planar target.

![Graph showing absorption and x-ray emission spectra](image)

**FIG. 9.3**: a) Percentage absorption of 45 fs ultra-short laser pulse in planar Mo (solid triangles) and Mo coated with CNTs (solid circles) as a function of the laser intensity; b) Hard x-ray emission spectrum recorded from planar Mo and Mo coated with CNTs, at a laser intensity of $\sim 8 \times 10^{16}$ W/cm$^2$.

Figure 9.4a shows the intensity of the K$_\alpha$ emission from planar Mo and Mo coated with CNTs, as a function of the laser intensity. It is observed that the K$_\alpha$ emission from CNT coated Mo target is always higher as compared to that from planar Mo. It is important to note here that with increasing laser intensity, the K$_\alpha$ yield from planar Mo keeps on increasing monotonically, with no sign of saturation, unlike the peaking seen in lower and medium Z targets like Al and Ti respectively [221, 222]. This observation eliminates any possibility of re-absorption of the K$_\alpha$ photons generated by the hot electrons penetrating inside the target, even at the highest laser intensity ($2.5 \times 10^{17}$ W/cm$^2$) used in our experiments.
FIG. 9.4: a) $K_{\alpha}$ generation from planar Mo (hollow triangles) and Mo coated with CNTs (solid triangles) as a function of the laser intensity, b) The enhancement factor of $K_{\alpha}$ yield from Mo coated with CNT w.r.to planar Mo.

Figure 9.4b shows the enhancement factor of the $K_{\alpha}$ yield from Mo coated with CNT w.r.to planar Mo. Interestingly, although the absorption was close to 100% in the intensity range used in our experiments, the $K_{\alpha}$ enhancement from CNT coated target (w.r.to uncoated Mo target) varies drastically with the intensity. The enhancement was only 3.5 times at lower intensity of irradiation of $1.6 \times 10^{16}$ W/cm$^2$. As the laser intensity is increased, a sharp increase of x-ray enhancement is observed. The $K_{\alpha}$ x-ray emission enhancement peaks at an intensity of $8 \times 10^{16}$ W/cm$^2$, where the enhancement is ~ 75 times of that obtained from planar Mo. As the laser intensity is increased further, one observes a sharp drop in the enhancement of $K_{\alpha}$, and the enhancement decreases to just 3 times at the highest irradiation intensity of $2.5 \times 10^{17}$ W/cm$^2$. 
Such a sharp enhancement at a particular intensity is indicative of a very efficient hot electron generation process. Such a peak behavior in x-ray enhancement has been also observed recently by Bagchi et al using CNT coated Cu target [84]. They studied the hard x-ray bremsstrahlung emission and observed enhancement utilizing the CNTs, but did not discuss the reason for the peak behavior.

9.3 Discussion of the results

To qualitatively understand these observations in Figs 9.3 and 9.4, we determine the electric field and hence the effective laser intensity inside a laser irradiated nano-tubes based on the steps shown in Chapter 8. The laser electric field inside the nano-structure determines the absorption and ionization processes, and hence governs the hot electron generation and the x-ray emission [72, 76, 87, 177, 202]. The electric field enhancement leads to the increase of effective intensity of the laser pulse interacting with the nano-tubes and hence leads to enhancement of the hot electron temperature [72, 76, 87, 177, 202]. It is quite valid to assume that, for an ultra-short pulse of 45 fs duration, the hydrodynamic motion is almost frozen and the nano-tubes geometry is intact during its interaction with the laser. Let the applied instantaneous laser field strength be $E_o$ (the applied intensity $I_o$), oriented perpendicular to the axis of the nano-tubes of “hollowness” $a_o/b_o$ (where $a_o$ and $b_o$ are the inner and outer radii), and the instantaneous dielectric constant be $\varepsilon = (1 - n_e/n_c)$, where $n_e$ is the electron density). The electric field applied at any arbitrary angle with respect to the tube axis can always be resolved in to a parallel and perpendicular component along the axis. As shown in Chapter 8 the field applied parallel to the tube axis does not get modified. The electric field inside the hollow nano-tube (perpendicular to its axis) is calculated using the Laplace equation in cylindrical coordinates neglecting the z variation and under quasi-
static approximation i.e. \( a_o, b_o << \lambda \). Applying appropriate boundary conditions, the magnitude of the electric field inside the nano-tube region between \( a_o \) and \( b_o \) can be calculated to be

\[
E_{in} = \sqrt{A_z^2 + \frac{B_z^2}{r^2} - \frac{2A_z B_z}{r^2} \cos 2\theta}, \quad \text{where} \quad r \text{ and } \theta \text{ are the radial and azimuthal cylindrical coordinates,}
\]

\[
A_z = -\frac{2E_0(\varepsilon + 1)}{\Delta}, \quad B_z = -\frac{2E_0a_o^2(\varepsilon - 1)}{\Delta}, \quad \Delta = (\varepsilon + 1)^2 - \left(\frac{a_o}{b_o}\right)^2(\varepsilon - 1)^2. \]

The root mean square (r.m.s.) electric field enhancement \( \left\langle \frac{E_{in}}{E_0} \right\rangle \) can be calculated by taking the spatial average of field as

\[
\frac{1}{\pi(b_o^2 - a_o^2)} \int_0^{2\pi} \int_{a_o}^{b_o} \left(\frac{E_{in}}{E_0}\right)^2 r dr d\theta. \]

From this, the r.m.s. intensity enhancement ratio is calculated as \( \left\langle \frac{I_{in}}{I_0} \right\rangle = \frac{1}{I_0} \left( A_z^2 + \frac{B_z^2}{a_o^2 b_o^2} \right) \). The electric field is resonantly enhanced when \( \Delta = 0 \), which happens for two value of \( a_o/b_o \) i.e. \( \frac{a_o}{b_o} = \pm \left(\frac{\varepsilon + 1}{\varepsilon - 1}\right) \). By using the value of \( \varepsilon = 1 - n_e/n_c \), the electric field or the effective intensity in the nano-tubes plasma is resonantly enhanced at two densities \( n_H = \frac{2n_c}{1 - \left(\frac{a_o}{b_o}\right)} \), \( n_L = \frac{2n_c}{1 + \left(\frac{a_o}{b_o}\right)} \). Figure 9.5a shows the variation of the high and low resonance density with degree of hollowness, where the dashed vertical line is for the CNTs used in our experiments with \( a_o/b_o = 0.9 \), for which \( n_H = 20 n_c \) and \( n_L = 1.05 n_c \). It follows from Fig. 9.5a that higher resonance density \( (n_H) \) can be increased by choosing a nano-tube of smaller wall thickness. This is in contrast to the case of solid clusters and nano-rods \( (a_o = 0) \), where the field enhancement occurs only in the vicinity of \( 3n_c \), and \( 2n_c \) respectively [96-100, 177, 202]. Figure 9.5b shows the variation of the r.m.s. intensity
enhancement factor of the incident laser intensity inside a CNT (having a degree of hollowness of 0.9) as a function of the electron density. The effective intensity inside nano-tubes shows two peaks at densities mentioned above, but it is the peak corresponding to the high density resonance that governs the hot electron generation more effectively. This is

FIG. 9.5: a) The variation of high and low resonance density with degree of hollowness, where the dashed vertical line is for the class of CNTs used in our experiments with \(a_r/b_0=0.9\); b) The r.m.s. intensity enhancement factor inside the hollow nano-tube, compared to the incident intensity of the laser (the inset shows a top view of CNT with a hollowness factor of 0.9).

because, when an intense laser pulse irradiates the CNTs, the plasma is created in the foot of the pulse when the laser intensity exceeds the plasma formation threshold (~\(10^{14}\) W/cm\(^2\) for 100 fs pulse) and the plasma density becomes super-critical (> \(n_c\)) [194, 200, 201]. Subsequently, as the intensity rises sharply, the electron density also rises because of the intensity dependent
ionization processes like tunnel ionization, optical field ionization, and electron impact ionization [223]. During the ionization phase, the electron density reaches the resonance condition that is determined by the chosen target geometry. It is desirable that for most efficient generation of hot electrons, the high resonance density $n_H$ is achieved at the peak of the laser pulse [96, 97, 177]. If the laser intensity is too low, the electron density generated at the peak falls short of that required for resonance. If the laser intensity is too high, the electron density generated at the peak overshoots the value required for resonance. In both the cases, although the effective intensity is enhanced w.r.t the applied laser intensity, the enhancement is not as strong as when the resonance condition is met, as can be also be seen from Fig. 9.5b. The sharp peaked $K_{\alpha}$ x-ray emission enhancement at an intensity of $8 \times 10^{16}$ W/cm$^2$ is probably because in this case, the resonant electron density is reached at the peak of laser pulse, close to the high resonance density $n_H = 20 \ n_c$ for the CNTs used in our experiment. In general, it follows that for nano-tubes of a certain degree of hollowness there exists an optimum laser intensity for maximum x-ray enhancement.

Coating CNTs on planar targets is a versatile method which, in principle, should be applicable to any material to enhance the x-ray emission, since the role of the CNTs is to facilitate high absorption and efficient hot electron generation. Taking into consideration the hot electron generation and transport in a material, there exits an optimum intensity and hot electron temperature [109,210] for maximum $K_{\alpha}$ yield. For materials with higher atomic number (Z), the optimum intensity is higher, and for lower Z materials, the optimum intensity is lower [109]. Consequently, one needs to coat CNTs of greater degree of hollowness on higher Z materials. Since such CNTs have a higher resonance density, at higher intensity of irradiation, the high density resonance may occur closer to the peak of laser pulse, leading to its better absorption.
Similarly, for lower Z materials, lower intensity of irradiation is needed to obtain the optimal Kα generation. Therefore, one has to optimize the laser intensity and the CNT geometry (degree of hollowness) depending on the target whose Kα one is interested in.

Next, it is important to consider the role of the thickness of the CNT layer in this process. A very thick layer of CNTs will inhibit the propagation of the hot electrons generated in the CNTs into the base material and will thereby reduce the x-ray conversion in the base material. Moreover, a thick CNT layer will also attenuate the x-rays produced in the base material. On the other hand, a too thin layer of CNTs will not give rise to sufficient hot electron generation. So one needs to optimize the laser intensity, CNT geometry (degree of hollowness) and CNT layer thickness for different target materials to achieve high x-ray conversion, and this will be studied in future.

To summarize, it is observed that near complete energy absorption of intense ultra-short laser pulses is recorded in CNTs deposited on planar molybdenum. The hollow structure of the nano-tube plasma facilitates resonant electric field enhancement at high electron density during the ionization phase of the nano-tubes. This resonance enhancement at a density much larger than the critical density (n_c) leads to highly efficient hot electron generation, which results in enhanced Kα emission. In general, hollow tube targets facilitate more efficient hot electron generation and consequent x-ray emission, and this may be useful as a compact, low cost, table top, ultra-short duration x-ray source.