Chapter 4

Absorption of ultra-short laser pulses in \textit{in situ} formed clusters

Coupling of intense ultra-short laser pulse energy on interaction with matter is a very important aspect [71,91, 161], since the enhanced absorption leads to generation of energetic electrons, MeV ions, and x-rays. The x-rays produced by ultra-short ultra-intense laser have high peak brilliance, short pulse duration, and micron scale size. This makes it a potential source for various applications like time resolved x ray diffraction, imaging of live biological specimen etc. At moderate intensities ($\sim 10^{17} \text{W/cm}^2$) absorption of ultra-short laser pulses ($\sim 100 \text{fs}$) by planar solid targets is quite low, as described in Chapter 1 [39,59,64,65]. Various types of targets like gratings [162], structured targets [80], pre-deposited metal clusters [76], gas clusters [96], snow clusters [69] etc. have been used to enhance the coupling of the laser energy in matter. However, since the gratings are expensive, they are practically unusable on routine basis as targets for x-ray source. Next, high absorption (80-90\%) of ultra-short laser pulses has been observed in gas atom clusters [88,163]. The interest in the clusters stems from the fact that the electric field inside the cluster is highly enhanced at three times the critical density ($n_c$) [97,98] as mentioned in Chapter 1. As the clusters (at near solid density) absorb the laser energy, they get heated up and start expanding. During expansion, the density decreases. When the density approaches $3n_c$, the absorption increases rapidly due to the enhancement of the electric field inside the clusters [96]. Although gas clusters offer a debris-less source of x-rays, the keV x-ray conversion from gas clusters is rather small ($\sim 0.01\%$) and it is a low repetition rate source, with a further
limitation that there are few option of available gases. This limits the use of gas atom clusters as an x-ray source.

Metal clusters have also been used as targets for efficient laser energy absorption. They can be produced by high pressure dc sputtering on polished disks, or by magnetron gas aggregation. They can also be produced by atom pick-up method. All these methods involve two steps, first depositing the clusters on a planar solid substrate, and then using them as target for plasma formation [76]. As mentioned in Chapter 3, intense femtosecond laser irradiation of solid targets is a simple method of synthesis of clusters with few tens of nm diameter [134,160]. We also observed that a sub-ns pulse focused to high intensity can also do the same as observed in materials like Ag, In, Cu etc [147,164]. As mentioned in Chapter 3, the SEM pictures of the deposit taken at two pre-pulse intensities, one at a high intensity of $3 \times 10^{12}$ W/cm$^2$ and the other at a low intensity $10^{10}$ W/cm$^2$, clearly revealed a much higher cluster density corresponding to the deposition taken at the higher intensity of the sub-ns pulse. The particle density in this case ($I = 3 \times 10^{12}$ W/cm$^2$) was estimated to be $\sim$100 particles/µm$^2$ from the SEM pictures [147,164].

We have explored the possibility whether the in situ formed clusters/nano-particles can be used as targets for enhanced absorption of fs laser light and consequently higher x-ray generation. Silver clusters produced by a 30 mJ, 300 ps laser pulse were irradiated up to an intensity of $3 \times 10^{17}$ W/cm$^2$ by a 70 mJ, 45 fs compressed laser pulse from the same Ti:sapphire laser. Absorption of the laser light exceeding 70% was observed, resulting in an x-ray yield (>1 keV) of $\sim$60µJ/ pulse. This may constitute a much simpler means of intense x-ray generation using ultra-short laser pulses as compared to the irradiation of structured / pre-deposited cluster targets. Moreover, it offers higher x-ray conversion efficiency than that from gas clusters and planar solid targets.
4.1 Description of the experiment

The absorption measurements of intense femtosecond laser pulses by the in situ produced clusters/nano-particles, were carried out using the experimental setup shown in Fig. 4.1. The stretched 300 ps laser pulse, before the grating compressor, was split by a pellicle beam splitter and was used to produce silver clusters by focusing it on a planar solid target of silver, in normal incidence geometry. This pulse will be henceforth referred to as the cluster forming “pre-pulse”. The focused intensity of the pre-pulse beam was varied between $10^{10} - 4 \times 10^{12}$ W/cm$^2$. The transmitted part of the uncompressed beam (through a pellicle beam splitter) was time delayed through a delay setup and then compressed for getting a pulse of 45 fs duration (referred to as the “main pulse” or the “main beam”). This main beam (70 mJ, 45 fs) was focused very close to the target, at a distance of ~30 μm from the target surface (See Fig.4.1). This beam propagated parallel to the target surface and irradiated the clusters emanating from the pre-pulse.

*Fig.4.1 : A schematic diagram of the experimental setup*
beam irradiating target. For absorption measurements, the main beam energy was measured with and without pre-pulse produced plasma in its path, to get normalized transmission through the plasma plume. After the main beam propagated through the plasma, the transmitted light was collected using a large diameter lens (75 mm diameter, 100 mm focal length) and made incident on a pyro-electric detector (Gentec, sensitivity 3V/J) for energy measurement. Another more sensitive calorimeter (Gentec, sensitivity 164 V/J) was placed outside the chamber to sample the scattered light collected by another lens of focal length 50 mm placed on the plasma chamber wall window in a direction of 45° w.r.t. the target. The scattering signal was very low and the angle integrated scattered laser light intensity, normalized to the incident beam, was estimated to be ≤1%. Thus, the reduction in transmission of the main pulse beam was predominantly due to its absorption by clusters. Next, an x-ray p-i-n diode (Quantrad) filtered with two aluminized polycarbonate filters having cut-off (1/e transmission) at 0.9 keV, was used to measure the x-ray radiation emitted by the laser irradiated clusters. The detector subtended a solid angle of 410 μsr at the source and was kept at 45° angle from the target.

4.2 Optimization of absorption in the in-situ formed clusters

Once confirmed that efficient production of cluster is taking place then conditions of maximum absorption and x-ray emission from in-situ formed clusters has to be found. For this intensity of pre-pulse beam and main pulse beam has to be varied but first the delay must be optimized. It is expected that the pre-pulse intensity must be high such that efficient cluster production take place.

a) Effect of delay and cluster forming laser pulse intensity: Absorption measurements of the main laser beam were carried out at the maximum main beam intensity of
3x10^{17} \text{ W/cm}^2, for different intensities of the pre-pulse beam. First, a small delay of 10 ns was kept between the cluster forming pre-pulse and the main pulse. A low absorption was observed as shown in Fig.4.2.

![Absorption vs Pre-pulse Intensity](image)

**Fig.4.2**: The variation of the main pulse absorption as a function of the pre-pulse laser intensity at a main beam intensity of 3x10^{17} \text{ W/cm}^2. The absorption is studied at two delays of 10 ns and 75 ns between the pre-pulse and the main pulse.

Despite the increase in absorption with pre-pulse energy, the absorption did not increase much (<10%) even when the pre-pulse was focused at higher intensities >10^{12} \text{ W/cm}^2, where a larger amount of cluster formation is expected. The delay was then increased to 75 ns. From Fig.4.2 it is seen that at this delay, there is a low absorption of ~10% recorded for intensity around 10^{10} \text{ W/cm}^2. As the pre-pulse intensity is increased, the absorption increases rapidly up to 10^{12} \text{ W/cm}^2, and thereafter, it shows saturation in the range (1-4)x10^{12} \text{ W/cm}^2, corresponding to a high absorption of ~70%.

The above observations can be understood as follows. At the delay of 10 ns, even at the highest pre-pulse intensity of 4x10^{12} \text{ W/cm}^2, the large clusters of size ~10 - 40 \text{ nm (radius)}
having a typical velocity $\sim 10^2$-$10^3$ m/s [135,152, 155] will travel a distance of only $\sim$ 1-10 $\mu$m from the target surface and hence will not reach the interaction volume of the main pulse, passing 30 $\mu$m away from the surface. Thus, the main beam will interact with only the plasma produced by the pre-pulse beam (and perhaps some very small clusters traveling with a higher velocity), resulting in the observed low absorption. Increasing the delay results in the arrival of more and more clusters in the region of main beam. The observed increase in absorption with increasing pre-pulse intensity is then simply understood from the increase in number as well as the velocity of clusters produced at higher pre-pulse intensity [147,164].

**b) Effect of laser pulse intensity on absorption :**

![Graph](image)

*Fig. 4.3: Variation of the main beam absorption as a function of its intensity. The pre-pulse beam intensity is fixed at 4x10^{12} W/cm^2.*

Next, the absorption of the main beam was studied as a function of its intensity, at a fixed pre-pulse intensity of 4x10^{12} W/cm^2. For this purpose, calibrated neutral density filters were used to attenuate the main beam energy, starting from the highest value. The neutral density filters were put before the compressor in the path of the uncompressed beam thereby eliminating the pulse
spreading and additional B integral issues. The absorption variation for silver target is shown in Fig.4.3, where the intensity of the main beam was varied in the range of $3 \times 10^{16} - 3 \times 10^{17}$ W/cm$^2$. When the intensity was increased by one order of magnitude (from $\sim 3 \times 10^{16}$ W/cm$^2$ to $3 \times 10^{17}$ W/cm$^2$), the absorption increased from $\sim 25\%$ to $70\%$. The increase in absorption was initially fast and at higher intensities the increase in absorption was slow.

At a fixed pre-pulse beam intensity of $4 \times 10^{12}$ W/cm$^2$, the increase in absorption with increase in the main pulse intensity can also be understood in terms of the $3n_c$ resonance absorption in clusters [88,97,98]. As a cluster absorbs the laser energy and gets heated up, it starts expanding and the density decreases from near solid density and approaches $3n_c$. The absorption increases rapidly due to the rapid enhancement of the laser electric field inside the cluster at this density, when the dielectric constant of the cluster approaches a value of $-2$ at $3n_c$ [97-99]. The clusters irradiated at higher laser intensity will be at higher temperature and will expand faster. Hence they will reach comparatively closer to $3n_c$ during the laser pulse (resulting in higher absorption) than those irradiated at lower intensity [88,98]. As a result, one gets lower absorption at lower intensities as seen in Fig.3. Moreover, the rate of change of electron density in a cluster after irradiation is expected to be fast initially for smaller radius (as $dn_e/dR \propto R^{-4}$), hence with increasing intensity of main pulse, a slower change in electron density is expected as most of the expansion occurs during the initial part of the pulse. This explains qualitatively the fast rise in absorption followed by a slower increase of absorption when the main pulse intensity is increased.

c) Effect of multiple shots: It may be relevant to point out that a high absorption was observed only when fresh surface of the target was irradiated. When the pre-pulse laser beam was incident on a previously irradiated spot on the target, the absorption decreased. Fig.4.4
shows the main pulse absorption after a number of pre-pulse shots were fired at the same place. It is seen that the absorption decreases with the number of pre-pulse shots.

![Graph showing absorption decrease with pre-pulse shots](image)

*Fig. 4.4: Variation of the absorption of the main beam with nᵗʰ pre-pulse shot fired at the same place in silver target. The fitted curve is only guide to the eyes*

The above behavior may be understood as follows. When the pre-pulse irradiates a solid surface it forms a crater. On subsequent laser irradiation at the same spot, there will be a recession of the crater surface as more and more material ablates out. Thus the clusters produced from irradiation of crater region will take a longer time to reach the interaction region and would increasingly miss interaction with the main beam. This inference is also supported from the observation that when the target, after few shot irradiation, was brought slightly closer (~10 µm) to the main beam, the absorption showed a slight increase compared to that in the unshifted position. Next, as stated above, the absorption did not reduce to zero even after a number of shots were fired at the same place. This may be due to arrival of some smaller clusters in the interaction region even after crater formation.
4.3 KeV x-ray emission measurements

Now, we present the results of the x-ray emission measurements from the *in situ* produced clusters by irradiation of 45 fs laser pulse heating. Fig.4.5 shows the intensity of x-ray emission as a function of the pre-pulse beam intensity at the 45 fs heating pulse of maximum intensity of $3 \times 10^{17}$ W/cm$^2$. It is seen that the x-ray intensity increases with pre-pulse beam intensity. This behavior is consistent with our measurements of a higher absorption of the main beam at high pre-pulse intensity (shown in Fig.4.2). With increase in pre-pulse intensity, the number of clusters interacting with the main pulse increases. As a result, the absorption increases and the x-ray yield also increases, showing a variation somewhat similar to that of the absorption.

![Graph](image)

*Fig.4.5: X-rays emission intensity from silver clusters as a function of the pre-pulse intensity, for main beam intensity of $3 \times 10^{17}$ W/cm$^2$."

The spectrally and temporally integrated x-ray yield (for $h\nu \geq 1$ keV and up to 10 keV beyond which the response of p-i-n diode drops due to finite detector layer thickness) at the main beam intensity of $3 \times 10^{17}$ W/cm$^2$ and the pre-pulse intensity of $4 \times 10^{12}$ W/cm$^2$, was measured to
be \( \approx 60 \ \mu J \), assuming isotropic emission. For the above condition of the pre-pulse and the main pulse, the recorded absorption of the high intensity 45 fs pulses was \( \approx 70\% \). Thus, the percentage conversion efficiency of the laser energy into x-rays is \( \approx 8.5 \times 10^{-2} \% \).

The above high yield of the x-rays achieved from silver clusters can be due to several reasons. Irradiation of the metal cluster with intense femtosecond pulses leads to enhanced absorption. Due to the enhanced field inside the clusters, highly charged ion species are produced [165-167]. Therefore, the clusters give a high x-ray output due to increased collisionality, and higher ionization and excitation inside the clusters caused by laser heating [163,166,167]. Due to the above reasons, a high percentage conversion efficiency of laser energy into x-rays is observed. In conventional Xe gas cluster targets, the percentage conversion efficiency of laser energy into x-rays (\( h\nu > 1 \) keV) is reported to be between 1.7-4.2 \( \times 10^{-3} \% \) [163]. Nickel target in various forms, when irradiated with 1 ps pulse of \( 10^{17} \) W/cm\(^2\), produces efficient x-rays above 900 eV [70]. The reported percentage conversion efficiency of 70 mJ laser energy in to x-rays (\( h\nu > 900 \) eV) from flat nickel target is 2.8 \( \times 10^{-3} \% \), for Ni nano-groove target it is 2.8 \( \times 10^{-2} \% \), for Ni “smoke” targets it is 1.3 \( \times 10^{-1} \% \), and for Ni black target it is 1.4 \( \times 10^{-1} \% \) [70]. This suggests that the silver clusters produced \textit{in situ} is a better source of x-rays than the gas clusters and solids. Moreover, it offers a simple single step method for keV x-ray generation than the structured targets which are difficult to fabricate and are expensive.

Next, the x-ray emission in directions making large angle with target normal is nearly debris free. This is because, while the x-ray emission from the \textit{in situ} produced clusters is expected to be isotropic, the debris expand predominantly in a direction normal to the target, making it practically a debris-free source in a direction perpendicular to the target normal. The present scheme of cluster formation has also the advantage that the interaction of the generated
clusters with the main beam is slightly away from the solid target. This prevents heat conduction from clusters to the cold target substrate, leading to enhanced plasma temperature and increased x-ray emission, than in the case of metal clusters pre-deposited on a planar target.

Thus we have observed a high absorption of ~ 70% of high intensity ultra-short laser pulses in silver clusters in situ produced by a sub-ns pre-pulse. Effect of temporal delay (between the pre-pulse and the main pulse) and the pre-pulse intensity was studied to achieve high absorption of the fs pulses in these clusters. The high laser light absorption resulted in an efficient keV x-ray generation. A high conversion efficiency of ~8.5x10^{-2} % of the laser energy into x-rays (hν ≥0.9 keV) is observed. Moreover, the x-ray source is practically debris-free.