Chapter 2

Experimental setup and diagnostic system

The laser used in the present experimental study is a 10 TW, 45 fs, 10 Hz Ti:sapphire laser system. In this chapter, we present a brief description of this laser and the characterization of various parameters of the laser pulse. This is followed by the description of experimental arrangement for high order harmonic generation, their detection and measurement.

2.1. Ti:sapphire laser system

The laser used in the experimental study is a chirped pulse amplification (CPA) technique based commercial Ti:sapphire laser system. The oscillator generates nanojoule pulses of ~20 fs duration at 75 MHz. A block diagram of the laser system is shown in Fig. 2.1. Generation of such small duration pulses is made possible by a mode-locking technique known as Kerr-lens mode-locking (KLM) [2]. This technique exploits the self-focussing of the laser pulses inside the lasing medium due to Kerr effect for the generation of femtosecond pulses. In such oscillators, initially the laser is in continuous free running mode. Then the laser cavity is disturbed externally. The disturbance of laser cavity results in the generation of very short duration pulse known as ‘coherent spike’. Coherent spike is a noise pulse for which all the lasing modes are inherently locked. Due to its small duration, the coherent spike has highest intensity and hence it undergoes largest self-focussing (due to Kerr effect) inside the laser crystal, and it gets amplified inside the lasing medium. The pulse duration of coherent spike would then be increased due to dispersion, by the laser crystal and air path. These factors introduce positive
dispersion in the laser pulse and to compensate these dispersions, passive components that provide negative dispersion are required. Prism pairs [3] and chirp mirrors [4] are generally employed for such applications. In this way one gets a stable femtosecond laser pulse having repetition rate corresponding to the optical path of the laser oscillator.

These pulses are then stretched using a pulse stretcher. The pulse stretcher uses a grating pair and a cylindrical mirror (known as Öffner type stretcher [5]), introduces positive linear chirp in the laser pulse and increases its duration from ~20 fs to ~200 ps. In a positive linearly chirped pulse, the wavelength of the pulse decreases linearly in time i.e. the initial portion of the pulse has longer wavelengths and the later portion of the pulse has shorter wavelengths.

The stretched pulse is injected into a regenerative (regen.) amplifier which is a low gain multipass amplifier that works in small signal amplification regime. In our case, the single pass gain of the regenerative amplifier is ~2, whereas the overall gain of the amplifier is ~10^5. Its high gain is because it is basically a seeded oscillator. The energy of the seed pulse gets amplified to few micro joules in the regenerative amplifier [6]. The regen. amplifier is cavity dumped every 100 msec (i.e. at 10 Hz or at lower rep-rate, as desired).

In addition to the main pulse, there are laser pre-pulses present before the main pulse. The pre-pulses present in the regen. output pulse are mainly of two types, 1) amplified spontaneous emission (ASE), generated by the amplification of spontaneous emission in regenerative amplifier and 2) pre-pulse generated by the leakage of the pulse
To reduce the pre-pulse level, a pulse cleaner is used. The pulse cleaner consists of a fast Pockels cell which rotates the polarization of the laser pulse by $90^\circ$ for a small time window of about 5 ns. This pulse is then passed through a polarizer. The polarizer rejects the pre-pulses and passes the main laser pulse (which is rotated by $90^\circ$ by the Pockels cell), resulting in reduction of pre-pulse level. The rejection ratio of the pre-pulse depends on the extinction of the polarizers and switching of the Pockels cell. This pulse is then amplified in two bow-tie multipass amplifiers (one pre-amplifier and the other main amplifier) to achieve its final energy of $\sim 600$ mJ.

The amplified stretched pulse is then allowed to pass through a pulse compressor resulting in a laser pulse of 45 fs duration, and energy $\sim 450$ mJ. The pulse compressor employs a parallel grating pair along with a retro mirror to compensate the positive chirp of the laser pulse [7]. The negative chirp introduced by the pulse compressor depends on the separation between the two gratings. As one increases the separation the negative chirp is increased and at optimum value of the grating separation, the negative chirp introduced by the compressor compensates the positive chirp of the stretched laser pulse. Increasing/decreasing the grating separation from this optimum value results in negatively/positively chirped pulses. It may be noted that the pulse is not compressed to its original duration at oscillator. This is due to the reduction of pulse spectrum through gain narrowing during the amplification of the laser pulse [8].

The energy and repetition rate of the laser can be varied by changing the gain of multi-pass amplifiers and the ejection frequency of the pulse ejector, respectively. In the Table 2.1, we summarize the important parameters of the laser system.
2.2. Characterization of the laser pulse

Prior to any laser-plasma interaction experiment, it is essential to characterize the laser pulse for its various parameters viz. the pulse energy, spectrum, duration, beam divergence, and the pre-pulse contrast ratio, as these parameters influence the interaction process. The laser energy can be measured by standard pyro-electric energy meters. In the following, a brief account of the techniques for measuring the remaining parameters, and actual measurements are presented.

2.2.1. Laser spectrum

The spectrum of the final laser pulse was measured using an optical spectrograph with wavelength range 570 nm - 1100 nm. A typical spectrum of the laser pulse is shown in Fig. 2.2. The measured laser spectrum has a peak wavelength at 790 nm and bandwidth (FWHM) of about 20 nm (Δλ). The temporal profile of the laser pulse is expected to be hyperbolic secant squared (sech²) type. This profile is generated because such pulses self-maintain their envelope (known as soliton pulses) during propagation in a chirp compensating system [9]. Applying uncertainty principle to the hyperbolic secant squared (sech²) pulses, the product of frequency bandwidth and pulse duration should be

\[ Δντ \geq 0.315 \]  [10], (or \( Δλ(nm)τ(fs) \geq 660 \) for 790 nm laser). As per this relation the

<table>
<thead>
<tr>
<th>Laser Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse duration</td>
<td>45 fs (minimum)</td>
</tr>
<tr>
<td>Wavelength</td>
<td>790 nm ± 10 nm (central)</td>
</tr>
<tr>
<td>Bandwidth</td>
<td>20 nm</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>1, 2, 3, 5, 10 Hz, or single</td>
</tr>
<tr>
<td>Energy per pulse</td>
<td>Upto 450 mJ</td>
</tr>
<tr>
<td>Power</td>
<td>Upto 10 TW</td>
</tr>
<tr>
<td>Maximum intensity</td>
<td>Upto ( 4\times10^{18} ) W/cm²</td>
</tr>
</tbody>
</table>

Table 2.1 Parameters of the Ti:sapphire laser
expected smallest pulse duration comes out to be ~34 fs, which is smaller than the observed pulse duration of ~45 fs. The larger duration of observed pulse may be due to the nonlinear chirp in the laser pulse which cannot be compensated by the pulse compressor.

2.2.2. Pulse duration measurement

Measurement of pulse duration is an important aspect of the ultrashort laser pulse - plasma interaction. Intensity autocorrelation method [11] is used to measure the duration of ultrashort laser pulses. In the second order autocorrelator shown in Fig. 2.3a, a beam splitter splits the incoming laser beam into two, which are then passed through a nonlinear crystal for second harmonic generation. If the two beams overlap both in space and time, a second harmonic beam is generated in the direction mid-way between the two beams (non-collinear phase matching). The temporal overlap of the two beams is achieved by the adjustment of the delay line of one arm of the autocorrelator. The angle between two beams and the laser pulse duration together determine the spatial width of the second harmonic correlated beam. A ray diagram of the intensity autocorrelator is shown in Fig. 2.3.
The detector shown in Fig. 2.3 is a space resolved detector like a CCD camera. A ray diagram for the generation of autocorrelation trace from the overlap of the two ultrashort pulses is shown in Fig. 2.4. Only in the case of spatial and temporal overlap of two beams, the central second harmonic pulse is generated, known as autocorrelator trace. As mentioned earlier, the width of the central trace depends on the duration of the laser pulse and the temporal profile of the laser pulse. In a way, the measurement of the autocorrelation trace width does not give full information about the ultrashort pulse, as one has to know (or assume) the shape of the laser pulse in order to know the exact duration of the pulse.

The width of the autocorrelator trace \( W \) is related to the pulse width \( \tau \) (FWHM) of the laser pulse and the cross-over angle \( \theta \) between the two overlapping beams (see Fig. 2.4) by the relation [11]

\[
\tau = \frac{2}{K} \frac{W \sin(\theta/2)}{c}
\]  

……… (2.1)
where $K$ is a constant which depends on the exact temporal shape of the laser pulse, and $c$ is the velocity of light. For a hyperbolic secant squared ($\text{sech}^2$) pulse, $K = 1.55$. Fig. 2.5 shows a typical autocorrelator signal and its trace obtained using Promise software.

Precise measurement of the cross-over angle $\theta$ is a difficult task and some error is always involved with such measurement. We have used another simple approach for the calibration of time axis with the CCD pixels. In this method, a small delay is introduced in one of the laser pulses. The peak of autocorrelation trace is shifted by the amount of the delay. The difference between the two autocorrelation peaks directly gives the time delay in terms of the CCD pixel. For example, if the delay between two pulses is changed by $10 \, \mu m$, the peak of overlapping pulse is shifted by $\sim 33.3$ fs. Thus the pixel shift of autocorrelator trace is equal to $33.3$ fs. This method gives an accurate means of the measurement of the pulse duration. In this geometry, the formula for pulse duration measurement gets modified as

$$
\tau = \left( \frac{W \, \text{pixel}}{K} \right) \times \text{calibration factor} \left( \frac{\text{fs}}{\text{pixel}} \right) \quad \text{…….. (2.2)}
$$

The measured pulse duration was $45$ fs.
2.2.3. Beam divergence measurement

In laser-plasma experiments, peak intensity of the focussed laser beam on the target is an important parameter that governs the interaction processes. It is therefore necessary to measure the focal spot size, which is governed by the laser beam divergence and the focal length of the focussing optics. The peak laser intensity is given by

\[ I_0 = \frac{E}{\tau \left( \pi \omega^2 \right)} \]  \hspace{1cm} \text{......... (2.3)}

where \( E \) is the pulse energy, \( \tau \) is the pulse duration, and \( \omega \) is focal spot radius. The schematic of the laser beam divergence measurement is shown in the Fig. 2.6. The laser focal spot on the target is magnified and imaged on a CCD camera using a microscope lens. To avoid any damage to the CCD sensor, the pulse energy prior to focussing the laser beam was reduced by reflecting the beam from multiple glass surfaces. The FWHM diameter \( (2\omega_0) \) of the focal spot was measured to be \( \approx 18 \mu \text{m} \) for a lens of focal length 500 mm. Since the laser focal spot diameter is equal to the product of the laser beam divergence \( (0) \) and the focal length \( (f) \) of lens as \( f \times 0 \), one gets the full divergence of the final laser beam to be 36 micro-radians. Fig. 2.7 shows the measured focal spot of the laser. One can see that the spot is nearly circular and the intensity profile of the laser spot is close to Gaussian. It should be noted here that insertion of any filter in the path of the laser distorts the shape of the focal spot. Hence we have used multiple reflections from
anti-reflection windows (surface quality ~ $\lambda/10$) to reduce the laser intensity to desired levels.

![Image](image.png)

**Fig. 2.7** a) Magnified image of laser focal spot on CCD, b) intensity profile of the laser spot closely matching the Gaussian fit.

After measurement of these parameters (i.e. laser pulse duration, beam divergence, and laser energy), we can now estimate peak intensity of the laser pulse (with a lens of focal length 500 mm) to be ~ $10^{18}$ W/cm².

### 2.3. Experimental setup

A schematic of the experimental layout is shown in Fig.2.8. In this experiment the laser was operated at lower power of ~3 TW corresponding to a laser energy of ~150 mJ per pulse. To generate the plasma plume, a part of the uncompressed laser beam

![Diagram](diagram.png)

**Fig. 2.8** Schematic of the experimental setup
(pulse energy of ~ 20 mJ, pulse duration of 200 ps, central wavelength at 790 nm) was split from the main beam by a beam splitter. This beam was focussed at normal incidence by a f/10 spherical lens of 500 mm focal length on the surface of target, which was kept in a vacuum chamber evacuated to 10⁻⁵ mbar. The focal spot of the pulse at target surface was ~ 600 μm. The intensity of this beam (referred to as the “pre-pulse” beam) at the target surface was ~10⁹–10¹⁰ W/cm². The interaction of the laser pulse with the target surface at these low intensities generates the plasma plume mainly consisting of neutrals and singly charged ions. The plasma plume serves as the medium for harmonic generation [12]. In one of our experiments the details of which will be discussed in Chapter 7, we have generated the plasma plumes by focussing the prepulse at intensity ~10¹³ W/cm².

The laser pulse transmitted from the beam splitter was passed through a delay line and then passed through the vacuum compressor to generate the 45 fs pulse (will be referred as main pulse). This pulse has energy ~130 mJ. The delay between pre-pulse and the main pulse could be adjusted in the range of 6 ns to 130 ns by changing the length of the delay line. The main laser pulse was focussed in the preformed plasma plume using a spherical lens of focal length 500 mm, and its intensity in the plume was kept at 10¹⁵–10¹⁶ W/cm². The intensity was adjusted by changing the position of the focussing lens. At the best focus of the lens the intensity of the laser can reach to ~10¹⁸ W/cm². However increasing laser intensity beyond the optimum range of 10¹⁵–10¹⁶ W/cm² reduces the intensity of harmonics. This will be further discussed in Chapter 5. Positive or negative chirp could be introduced in the laser pulse by changing the separation between the two gratings in the compressor. In this way, we could generate up to 15 ps long negatively chirped pulse and upto 5 ps long positively chirped pulse.
The interaction of the focussed laser beam with the preformed plasma plumes results in high order harmonic generation. The harmonic radiation was detected by a flat field XUV spectrograph of variable spectral range. The XUV grating disperses the HHG spectrum in the horizontal direction after its passage through a slit. The spectrograph went through many improvements during the course of the work. More details about the spectrograph are given in the next sub-section.

Various targets were used to generate the plasma plumes. These targets can be broadly divided into two parts: a) bulk targets (such as Ag, In, Cr, Mn etc.) and b) nanostructured targets (such as C_{60}, carbon nanotubes, nanoparticles of Ag, Au, etc.). The plasma plume generated from the interaction of the pre-pulse and target surface mainly consists of neutral atoms or singly charged ions (in the case of bulk materials) or mildly heated nanoparticles (in the case of nanostructured targets). One can place bulk surface / nanostructured target surface on a target holder of 4 mm width and 50 mm length. The targets of approximately 4 mm × 4 mm size were used. Multiple targets were placed on the holder so that the properties of HHG from these targets could be studied without breaking the vacuum. The emission from plasma plume was imaged through a port to record the optical spectrum of the plasma plumes.

To study the HHG from an elongated plasma plume, the pre-pulse beam was focussed on the target surface using an assembly of two crossed cylindrical lenses of focal lengths 550 mm and 450 mm. This combination gave a focal spot of 2 mm × 300 μm (FWHM). The intensity of pre-pulse at target surface was ~10^9 W/cm². The length of plasma plume was varied using a variable width slit placed before the lens assembly. The length of plasma plume was varied in the range of 0.8 mm to 2 mm in step size of 0.4 mm.
The laser parameters used in experimental study are summarized in Table 2.2.

<table>
<thead>
<tr>
<th>Laser Parameter</th>
<th>Value in Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Repetition rate</td>
<td>1 Hz, 10 Hz</td>
</tr>
<tr>
<td>Delay between pre-pulse and main pulse</td>
<td>6 ns to 120 ns</td>
</tr>
<tr>
<td>Energy (main pulse)</td>
<td>130 mJ</td>
</tr>
<tr>
<td>Energy (pre-pulse)</td>
<td>20 mJ</td>
</tr>
<tr>
<td>Intensity (main pulse)</td>
<td>$10^{15} - 10^{18}$ W/cm²</td>
</tr>
<tr>
<td>Intensity (pre-pulse)</td>
<td>$10^9 - 10^{13}$ W/cm²</td>
</tr>
<tr>
<td>Main pulse duration</td>
<td>45 fs to 15 ps</td>
</tr>
<tr>
<td>Pre-pulse duration</td>
<td>~ 200 ps</td>
</tr>
</tbody>
</table>

Table 2.2 Laser parameters during experiment

2.4. XUV spectrograph for harmonic detection

The generated harmonics were detected by an in-house developed XUV spectrograph. In the initial configuration the spectrograph comprised of a dispersing element and an imaging system. A schematic of the XUV spectrograph is shown in Fig 2.9. The dispersing element of the spectrograph was a variable line-spacing flat field grating which focussed the dispersed spectrum at a flat surface normal to its plane. To image the dispersed spectrum, an assembly of micro-channel plate (MCP) and optical CCD camera was used. The MCP, placed at the focal plane of the grating, converted the XUV signal to blue-green fluorescent signal, which was then imaged onto a CCD camera [12]. The MCP-CCD assembly was mounted on a bellow to facilitate translation of the detector assembly in the direction of dispersion. This allows selection of a suitable

![Fig. 2.9 A Schematic of XUV spectrograph](image-url)
spectral range required for a particular experimental study. A typical spectrum of high order harmonic generation from the silver plasma plume using the above spectrograph is shown in Fig. 2.10.

![Image of a spectrum with exposure time 20 sec]

**Fig. 2.10: Typical spectrum from the XUV spectrograph**

The above spectrograph was later on modified to achieve higher sensitivity of detection through focusing of the line spectrum [13]. A schematic of this setup is shown in Fig 2.11. A cylindrical grazing incidence mirror was introduced before the flat field grating which focuses the dispersed spectral lines to points, thereby making the setup ~10 times more sensitive than the earlier one. A typical spectrum of the high order harmonic radiation from the silver plume recorded using this setup is shown in Fig. 2.12.

![Image of a schematic of the improved XUV spectrograph]

**Fig. 2.11: A schematic of the improved XUV spectrograph**

![Image of a spectrum with exposure time 2 sec]

**Fig. 2.12: Typical spectrum from the improved XUV spectrograph**

From the next chapter onwards, we shall discuss our experimental findings of HHG from plasma plumes in the plateau region, and the various processes used for the optimization of the HHG yield and cut-off.
Bibliography


