Chapter 9

HHG using two-colour laser pulses

In the previous chapters, we have presented an experimental study on HHG from the interaction of ultrashort fs laser pulses with low excited preformed plasma plumes of bulk materials and nanostructured targets [1-3]. Only odd harmonic orders were observed in these studies. As discussed in Chapter 1, even harmonic orders are not generated due to inversion symmetry of the HHG process. However, the inversion symmetry can be broken by simultaneous use of two or more laser pulses of different frequencies. For instance, it was observed in earlier studies that use of two-colour laser pulses for HHG resulted in the generation of even and odd harmonics of comparable intensities [4]. In these experiments, second harmonic of the driving fs laser beam was used as the second laser pulse and its intensity was about 20% of the fundamental laser pulse. In contrast to this situation, in our experiments using two-colour laser pulses, it was observed that a second harmonic beam of even 2% intensity of the fundamental laser beam was sufficient to break the inversion symmetry, resulting in generation of both even and odd harmonics. It was also observed that intensity of second harmonic pulse relative to the fundamental laser pulse affects the relative intensity of odd and even harmonics [5].

In this chapter, we present an experimental study of HHG using two-colour laser pulses comprising of fundamental Ti:sapphire laser pulse (800 nm) and its second harmonic (SH). We first present the particle trajectory analysis which demonstrates the generation of both even and odd harmonics, through the use of two-colour laser pulses in Section 9.1. Section 9.2 gives a brief description of experimental arrangement. The experimental studies are put in two categories viz. 1) weak two-colour laser irradiation in
which the second harmonic laser pulse intensity was ~2% of the fundamental laser intensity, and 2) strong two-colour laser irradiation where the second harmonic laser pulse intensity was ~15% of the fundamental laser pulse. The results of HHG using weak two-colour pulses are presented in Section 9.3. Both, even and odd harmonics with similar intensities were observed [5]. Different experimental investigations such as effect of laser polarization on HHG, resonance enhancement in both odd and even harmonic orders, HHG from nanostructured targets are presented in this section. In section 9.4, results of HHG from strong two-colour pulses are described. In this case it was observed that odd harmonics disappeared near the cut-off. Disappearance of odd harmonics has been predicted theoretically [6]. The present study [7] provides an experimental demonstration of this feature.

9.1. Even and odd harmonic generation through symmetry breaking

When one uses only a single colour laser pulse to generate high order harmonics, only odd harmonic orders are produced due to inversion symmetry of the HHG process. In Chapter 1 we have discussed the process of HHG from the three-step model [8, 9]. According to this model, when an atom interacts with a high intensity laser pulse, the atomic potential well is distorted by laser electric field and the bound electron can tunnel from the parent atom/ion. This electron then accelerates in the laser field. Depending on the laser phase at the instance of tunnelling, this electron may come back to the parent atom/ion. Its kinetic energy at the instance of collision with the parent atom/ion \( (e_{KE}) \) also depends on the tunnelling phase \( (\phi_t) \). The colliding electron may undergo recombination or get scattered. Upon recombination, an XUV photon is generated of energy equal to the sum of ionization potential \( (I_p) \) of the atom and the electron energy \( e_{KE} \). Thus, the XUV photon is phase locked with the laser pulse. It was discussed in
Section 1.2 that for a cosine laser field (wherein the field amplitude is maximum at 0 phase) the electrons which had tunneled between phase (0, $\pi/2$) and ($\pi$, $3\pi/2$) can return to the parent ion. It is worth noting here that during this process there are two values of $\phi$ in each half of laser field for which the kinetic energy of re-colliding electron is same. Interestingly, the electron which had tunneled earlier would return to the parent ion later than the electron which had tunneled later. As mentioned earlier in Chapter 1, the trajectories of these two electrons are known as long and short trajectories respectively, and are depicted as (1) and (2) in Fig. 1.1 (b). All the harmonic orders except for the cut-off harmonic order can be generated by either of the two electron trajectories. For the cut-off order, the two trajectories merge into one. One may note that an electron tunnelling out between phases ($\pi/2$, $\pi$) and ($3\pi/2$, $2\pi$) will follow the trajectory (3) of the Fig. 1.1(b) and it will not return to the parent ion.

We now examine the coherent addition of harmonic photons generated in two halves of the laser pulse. Variation of $e_{KE}$ with $\phi$ for single colour laser pulse is shown in Fig. 9.1a. Since this variation in the two halves is identical, the difference in tunnelling phase of electrons resulting in generation of same energy photons (one in each half) will be $\pi$. The corresponding phase difference in the generated XUV photons of frequency $q$ time laser frequency will be $(q+1)\pi$ where the numeral 1 appearing in the bracket is due to initial phase difference. It follows that the XUV photons will add constructively when $q$ is an odd integer, resulting in production of only odd harmonic orders.

The symmetry of the HHG process can be broken by introducing a very small amount of the second harmonic radiation. For instance, even 1% intensity of second harmonic pulse will alter the electric field profile of the two-colour laser pulse by 10%. Thus the phase difference between two harmonic photons of same energy generated in two halves will no longer be an exact multiple of $\pi$. Thus the even/odd orders do not
interfere completely destructively/constructively at each consecutive half of the laser field. Thus, both even and odd harmonics should be generated. Figures 9.1b and 9.1c show the variation of $e_{KE}$ with $\phi_t$ for two colour laser pulses with 2% and 15% second harmonic intensity respectively. One can see that the two consecutive laser halves are significantly different. It may be noted here that unlike in the case of single-colour laser pulse (Fig. 9.1a) where the electron which tunnel out between phases $(0, \pi/2)$ and $(\pi, 3\pi/2)$ can come back to parent ion, the range of $\phi_t$ for which electron can come back to parent ion, is reduced in the first half and increased in the second half of the laser electric field. This effect is more pronounced in the case of strong two-colour pulses. Moreover, the peak energy of the electrons is also different in the two halves of the electric field.

![Graph](image)

**Fig. 9.1** Electron recombination energy normalized to ponderomotive energy vs. tunelling phase of electron for (a)only fundamental laser pulse (b) two-colour laser pulse with 2% SH field and (c) two-colour laser pulse with 15% SH field.

### 9.2. Experimental description

A schematic of experimental setup for HHG using two-colour laser pulses is shown in Fig. 9.2. The interaction of the ps pre-pulse with the target surface creates a low excited pre-formed plasma plume. The fs laser pulse was focussed inside this plasma plume using a 500 mm focal length lens. A nonlinear crystal (Type-1 phase matched) for
the generation of SH pulse was inserted in the focusing femtosecond laser pulse to generate second harmonic radiation of the laser pulse. The nonlinear optical crystals for the generation of SH pulse used in this study are 1) 1 mm thick KDP crystal and 2) 0.3 mm thick BBO crystal, resulting in SH conversion efficiency of ~2% and ~15% respectively. The intensity of the fundamental radiation inside the SH crystal was kept ~1.5×10^{12} \text{ W/cm}^2, to avoid damage to the crystal. It may be mentioned here that inside the SH crystal, the group velocity for the fundamental pulse is higher than that of SH pulse. The mismatch in the group velocity of these two pulses can result in temporal separation between the two pulses, in a thick crystal, despite the phase matching. Hence crystals of smaller thickness were used to ensure a sufficient temporal overlap of the two laser pulses.

In addition to using different intensities of the SH pulse in the two-colour laser pump, the effect of their polarizations was also studied. In the second harmonic process using Type-1 phase matching, the polarization of SH and fundamental laser pulses are orthogonal. A parallel polarization condition was obtained by inserting a zero-order half wave plate (HWP) (for 800 nm) after the crystal. A BG-39 filter could be inserted in the laser path to cut-off the fundamental radiation for experiments with SH pump only.

![Fig. 9.2 Schematic of the experimental setup](image)
9.3. HHG using weak two-colour pulses

9.3.1. HHG from plasma plumes of bulk materials

The HHG spectra from the interaction of single-colour (only fundamental laser pulse) and weak two-colour laser pulse with low excited plasma plume of silver are shown in Fig 9.3. Fig. 9.3a shows the harmonic spectrum from fundamental laser pulse, which comprises of only odd harmonic orders. In the case of weak two-colour laser pulse, both even and odd harmonic orders with comparable intensities were produced (Fig. 9.3b). After insertion of the HWP in the path of the laser pulse (with it optic axis 45° w.r.t. the fundamental beam polarization), the polarization of the fundamental and the second harmonic laser pulses become parallel. This is because, for the fundamental radiation, it acts as a HWP and rotates the polarization by (2×45°) 90°, and acts as a full wave plate for the second harmonic, thereby doing nothing to its polarization. The high order harmonic spectrum from this two-colour laser pulse is shown in Fig. 9.3c. It may be seen from this figure that the cut-off order for even harmonics was smaller compared to that for the odd harmonics in the case of parallel polarized two-colour pump. During the experiments, it was observed that if the laser intensity inside the plasma plume is increased, the cut-off order for odd harmonics increases, whereas that for even harmonics decreases. These observations may be understood from the temporal separation of the two pulses due to their different group velocities inside the KDP crystal and half wave plate. As explained earlier due to dispersion there is only a partial overlap between fundamental and SH pulse. The fundamental pulse emerges ahead of the SH pulse. As one introduces the HWP these pulses get further separated due to GVD in HWP and the influence of the second harmonic pulse decreases. Same thing happens if one increases the laser intensity of the fundamental radiation inside the medium. As explained in
Chapter 4, in a high intensity laser pulse, the HHG comes predominantly from the leading edge of the laser pulse. With increase in laser intensity, HHG predominantly occurs from initial part of the leading edge of the pulse. This decreases the effective overlap of the SH pulse, which in turn reduces the intensity of even harmonics.

The HHG from two-colour laser pulse with circularly polarized fundamental laser was also studied. It was observed earlier, in HHG studies [2] with single colour laser pulses that the harmonic emission vanishes when the laser polarization was made circular. However, with the introduction of SHG crystal after the circularly polarized laser light, both even and odd harmonics appeared (Fig. 9.4a). Since the SHG crystals are birefringent in nature the circularly polarized laser pulse gets divided into ordinary and extraordinary waves inside the crystal. These waves travel with different group velocities and hence they get separated in time when they come out of the crystal. So one now has two plane polarized laser pulses, separated temporally, with perpendicular polarizations. As these orthogonal components cannot form circularly polarized light (due to lack of

![Fig. 9.3 HHG spectra from Ag plasma at: (a) single-colour (800 nm) pump, (b) orthogonally polarized two-colour pump, and (c) parallel polarized two-colour pump. Side lobes of the odd harmonics in the (a) correspond to the second-order diffraction lines of strong high-order harmonics.](image-url)
temporal overlap), the harmonic emission does not vanish. Out of these two pulses, only one pulse satisfies the phase matching condition for SHG. Hence, one of the laser pulses is a single-colour pulse (fundamental) and the other is two-colour laser pulse. The results of HHG from these pulses (Fig 9.4a) are compared with the HHG from two-colour pulses generated by linearly polarized fundamental laser light (Fig. 9.4b). One can see that the maximum even harmonic order visible in the first case (Fig 9.4a) is 30th order, whereas in the latter case it is 38th order (Fig 9.4b). The vanishing of even harmonic orders near cut-off generated from circularly polarized fundamental laser pulses can be understood from reduced intensity of SH pulse as only one pulse is satisfying the phase matching conditions.

The variation in HHG with the change in laser chirp was also studied. It was explained in Chapter 4 that the use of positively (negatively) chirped pulses results in shifting of harmonic peaks towards red (blue) wavelengths. In the case of two-colour pump also, the change of chirp of fundamental radiation resulted in wavelength shifting of both odd and even harmonics. The HHG from negatively and positively chirped pulses are shown in Fig. 9.5. It may be noted that while the intensity of odd harmonics (11th and 13th) is comparable the intensity of even orders (12th) was higher in the case of negatively chirped pulses compared to that with positively chirped radiation. The reason behind such observation is not fully understood.
Occurrence of new resonances

Resonant intensity enhancement in plasma plumes was observed in studies discussed in Chapter 4. For example, the intensity of the 13th harmonic of indium was ~200 times more compared to neighbouring harmonics (Fig 4.1). Since the use of fundamental radiation generates only odd harmonics, one will neither observe even harmonic orders nor any resonance enhancement in them. However, a two-colour pump results in generation of both even and odd harmonics. Hence, use of two-colour laser pulses will be helpful in searching resonance enhancement in even harmonic orders as well. Harmonic generation from indium plasma plume was studied with two-colour laser pulses. As shown in Fig. 9.6, it was found in the case of HHG from In plasma plume that a very intense 12th harmonic appears alongside the strong 13th harmonic (Fig. 9.6). Thus the use of two-colour laser pulses is also useful in increasing the HHG conversion efficiency through resonance enhancement.

Fig. 9.5 Harmonic spectra obtained from carbon plasma plume using the two-colour pump scheme in the case of negatively chirped (thick line) and positively chirped (thin line) pulses of fundamental radiation.

9.3.2. Occurrence of new resonances

Resonant intensity enhancement in plasma plumes was observed in studies discussed in Chapter 4. For example, the intensity of the 13th harmonic of indium was ~200 times more compared to neighbouring harmonics (Fig 4.1). Since the use of fundamental radiation generates only odd harmonics, one will neither observe even harmonic orders nor any resonance enhancement in them. However, a two-colour pump results in generation of both even and odd harmonics. Hence, use of two-colour laser pulses will be helpful in searching resonance enhancement in even harmonic orders as well. Harmonic generation from indium plasma plume was studied with two-colour laser pulses. As shown in Fig. 9.6, it was found in the case of HHG from In plasma plume that a very intense 12th harmonic appears alongside the strong 13th harmonic (Fig. 9.6). Thus the use of two-colour laser pulses is also useful in increasing the HHG conversion efficiency through resonance enhancement.
9.3.3. Harmonic intensity enhancement in plasma plumes of nanostructured targets

Efficient HHG using fullerene (C_{60}) targets was observed in our studies described in Section 7.2. It was observed that the harmonic intensity in the range of 9\textsuperscript{th} to 19\textsuperscript{th} order was higher by a factor of \(~25\) in comparison to harmonics from plasma plumes of bulk silver. In this Section, we present results of HHG through the interaction of two-colour laser pulses with plasma plumes containing fullerenes. It was observed that under the same experimental conditions, using two-colour laser pulses, the intensity of the HHG radiation could be further increased by a factor of 4-8 depending on the harmonic order [10]. The harmonic spectrum, generated from plasma plumes containing fullerenes, using single-colour and weak two-colour laser pulses are shown in Fig. 9.7a and 9.7b.

![Fig. 9.6 Harmonic spectrum from indium plasma using the two-colour pump](image)

![Fig. 9.7 CCD images of the harmonic spectra generated in C_{60} plasma using: (a) single-colour fundamental pump (800 nm), (b) two-colour pump (800 nm + 400 nm), and (c) single-colour SH pump (400 nm). The data were collected under similar experimental conditions.](image)
respectively. We have also compared these two HHG spectra with the HHG spectrum generated from only SH laser pulse (Fig. 9.7c). One can see that in the case of Fig. 9.7c only the odd harmonics of the SH laser pulses are generated (i.e. 5th and 7th H of the SH pulse), but in the case of two-colour laser pulse both even and odd harmonic orders could be seen. One may note that when only fundamental or SH laser pulse is used, 12th H of the fundamental laser (which is also 6th H of the SH pulse) is not produced. This comparison demonstrates that the HHG from two-colour laser pulse is indeed a symmetry breaking phenomena and not the sum of HHG from individual laser pulses. Although the mechanism of HHG enhancement in fullerene targets through the use of two-colour laser pulse is not well understood, the method would be useful in increasing the conversion efficiency of HHG process.

9.4. HHG using strong two-colour pulses

HHG using strong two-colour laser pulses generated from plasma plumes of bulk silver is shown in Fig 9.8a. Both, even and odd harmonics are generated by the strong two-colour laser pulses. The harmonic emission from single colour laser pulse (Fig 9.8 b) is shown for comparison with that from strong two-colour pulses. It is seen that these two spectra reveal the suppression of odd harmonic orders near the cut-off [7]. The situation is contrary to the case of HHG from weak two-colour laser pulses where even harmonics disappeared near the cut-off. The suppression of the odd harmonics in the cut-off may be due to larger intensity of SH pulse. In order to check on this point, we decreased the intensity of SH pulse to ~5% by tilting the BBO crystal vertically by ~5°. The HHG spectrum for this situation is shown in Fig 9.8c. It is observed from Fig 9.8c that with the reduction of SH intensity all the even and odd harmonics reappeared. The
same results were also observed when the duration of the laser pulse was increased to 
\(\sim 150\) fs to reduce the laser intensity and hence the intensity of SH pulse.

![Fig. 9.8 HHG spectra from excitation with a) strong two-colour pump (b) single-colour fundamental pump (800 nm), and (c) two-colour pump with reduced efficiency through tilting the SH crystal. One may see that odd harmonics reappear with the reduction in SH intensity. The spectra are normalized and shifted vertically to facilitate visual comparison](image)

**Bibliography**


