SYNOPSIS

Coherent sources of short wavelength radiation are much sought after in view of their applications in a variety of fields such as microlithography, spectroscopic studies etc. Generation of optical harmonics of intense laser pulses using nonlinear crystals is an established method of generating coherent radiation at shorter wavelengths. However, this technique does not enable one to generate radiation at wavelengths < 200 nm due to strong absorption of the radiation inside the crystals. The interaction of intense fs laser pulses with under-dense gaseous media enables generation of harmonics of the laser into soft x-ray region. Nonlinear interaction of the laser with the gaseous medium results in the emission of odd multiples of the laser frequency to very high orders. The phenomenon is known as high order harmonic generation (HHG). No even harmonics are generated in this case because of the inversion symmetry of the process. The HHG has been understood in terms of a three-step model. The model explains HHG from tunnelling of electron from atom, its acceleration under the action of laser field, and its recombination with parent atom. The model correctly predicts the shape and cut-off of the emitted harmonic radiation. The intensity and the cut-off of the emitted harmonic orders depend on the density and the ionization potential of the gaseous medium, and also on the laser parameters such as intensity and pulse duration. Through the optimization of various laser/gas parameters, the cut-off for high order harmonics can be extended to the so called ‘water window’ region (2.3 – 4.4 nm). However, the conversion efficiency of the HHG process remains rather low (~10⁻⁶).

There are several interesting applications of HHG such as measurement of ultrafast photo-recombination cross sections, probing of rotational dynamics of molecules, detection of molecular structure, strong field fast alignment of molecules, achievement of ultrahigh focussed intensities etc. Many of these have been demonstrated,
however for practical utilization of these applications, it is desirable to have higher intensity of the harmonics. Hence, in order to increase the practical applicability of harmonic radiation, it is necessary to increase the conversion efficiency of HHG. Many efforts are being made to increase the conversion efficiency of HHG. It was observed that the conversion efficiency of the HHG process depends greatly on the properties of the nonlinear medium used. However, the number of gases available for this purpose (usually noble gases are used) is rather small, which restricts research investigations to only a few elements. To overcome this limitation, we have generated high order harmonics using low excited under-dense plasma plumes ablated from solid materials through their interaction with low intensity laser pulses. These plasma plumes mainly consist of neutrals, singly charged ions, and in some cases doubly charged ions. The interaction of fs laser with these plumes is similar to that with gases. The use of low-excited plasma plumes enables one to explore the nonlinear properties of HHG in wide variety of materials. This also provides increased possibility of matching atomic/ionic transitions in the medium used with one or more harmonic orders. This may result in resonant intensity enhancement of a particular harmonic order several times compared to the neighbouring ones. Efforts are also made to use novel media to improve on conversion efficiency and cut-off order of harmonics.

In the present research work, we have experimentally studied the HHG from the interaction of intense femtoseconds ($\tau = 45$ fs) laser pulses with low excited plasma plumes of various target materials. The target materials used may be broadly categorized as bulk solids such as: Ag, In, Cr, Mn, C etc. and nanostructured targets such as: fullerenes ($C_{60}$) and nanoparticles of various metals. The plasma plumes were generated by the interaction of low intensity laser pulse (hereafter referred to as ‘pre-pulse’) with the target surface. The pre-pulse was obtained by reflecting part of the uncompressed
chirped laser pulse before the grating compressor stage in the laser system. The high order harmonics were generated by the interaction of the compressed femtosecond laser pulse (hereafter referred to as main pulse) with the preformed plasma plume. The HHG was optimized for intensity and cut-off of the harmonic orders. The enhancement of the intensity of particular harmonic orders due to various atomic/ionic resonances in different plasma plumes was also observed. It was found that nanostructured materials generate high order harmonics with larger conversion efficiency whereas the harmonics generated in bulk materials have a higher cut-off. Various properties of HHG such as the dependence of harmonic intensity on the laser intensity and focussing conditions, length of plasma plume, etc. were studied. Spectral characterization of HHG from bulk and nanostructured targets was carried out and the effect of laser spectral broadening through self-phase modulation of laser pulse on the spectral characteristics of harmonic radiation was studied. The effect of symmetry breaking of HHG process through the use of two-colour laser radiation was also studied. A chapter wise summery of these studies is given below.

**Chapter 1** gives a brief introduction to the theoretical background of HHG process and to the status of the various investigations in HHG. The Chapter starts with a discussion on the basic theoretical aspects, followed by a brief review of experimental results with HHG from gaseous media. This is followed by a brief description of the earlier experimental work on various optimization techniques utilized to increase the yield of HHG from gas jet plasmas.

**Chapter 2** describes the experimental arrangement used in present research work. Characterization of the parameters of the driving laser namely: pulse duration, spectral profile etc. were carried out. A chirp pulse amplification based 10 TW Ti:sapphire laser of 45 fs pulse duration was used in this experiment. The harmonics were produced by the
interaction of this ultrashort laser pulse with a low-excited preformed plasma plume. The plasma plumes were created by the interaction of low intensity laser pre-pulse (energy \(~30\ mJ\) and pulse duration \(~200\ ps\)). The pre-pulse was generated by reflecting a part of the amplified uncompressed chirped Ti:sapphire laser pulse before the grating compressor. The extreme ultra-violet (XUV) harmonic radiation was detected by an in-house developed XUV spectrograph based on grazing incidence variable line spacing flat-field grating. The harmonic radiation was dispersed by the grating in the horizontal plane and detected on a microchannel plate (MCP) – charge couple device (CCD) assembly. In the later parts of the experimental study, the sensitivity of the XUV spectrograph was increased by a factor of \(~10\) by focussing harmonic radiation in vertical plane using a grazing incidence gold coated cylindrical mirror placed before the grating.

Plasma plumes of various targets were used for the generation of high order harmonics. Mainly silver, indium, chromium, manganese, GaAs, carbon, silicon were used for HHG. *Chapter 3* describes the HHG from these targets. Optimization of various parameters such as laser intensity, chirp, delay between the two laser pulses (pre-pulse and main pulse), etc. was carried out in order to maximize the yield and cut-off of the HHG process. A brief account of these optimizations is given in this chapter. Next, the tuning of the harmonic frequencies through the variation of laser chirp was studied. It was observed that the harmonic wavelengths shift toward red (blue) with the introduction of positive (negative) chirp in the main laser pulse.

Increasing the conversion efficiency of high order harmonics generation is an important aspect of this research. Recently, it was observed that the conversion efficiency of the harmonics could be increased through various atomic/ionic resonances with particular harmonic orders. In our study it was observed that in certain plasma
the intensity of particular harmonic orders was much higher compared to that of
their neighbouring harmonics, resulting in highly efficient harmonic generation. For
instance, the intensity of 13th H (~61 nm) generated in indium plasma plume was ~200×
higher, and the intensity of the 29th H (~27.5 nm) in chromium plasma plume was ~20×
higher compared to the respective neighbouring harmonic orders. This is ascribed to the
resonance enhancement of a particular harmonic order. In Chapter 4, we discuss
important findings related to the resonance enhancement of the harmonic orders in
various plasma plumes. It was demonstrated that the tuning of the harmonic radiation
with chirp can move a resonantly enhanced harmonic order out of resonance, and also
bring a normal harmonic order into resonance.

In order to understand the effect of laser and plasma conditions on the
propagation of harmonics through the medium, a detailed knowledge of various phase-
matching factors on the intensity of high order harmonics is required. In Chapter 5, we
address the influence of these phase matching factors on HHG. For instance, the
propagation of the high order harmonics in elongated silver plasma plume was studied.
Since HHG is a coherent process, under perfect phase matching conditions, the intensity
of high order harmonics \(I_H\) is expected to increase with medium length \(L\) as \(I_H \propto L^2\).
It was observed that the scaling exponent \(p\) of harmonic intensity on medium length
\(I_H \propto L^p, p \sim 0.7-0.9\) was much smaller than the predicted value of \(p = 2\) under perfect
phase matching conditions. The smaller value of \(p\) occurs due to deviation from perfect
phase-matching between the laser and the harmonic radiation, and the reabsorption of the
harmonic radiation inside the plasma plume through its photo-ionization. The dispersive
elements responsible for the phase mismatch are: atomic dispersion, plasma dispersion,
Gouy phase shift, and intensity dependent dynamical phase shift. The phase-mismatch
factors for these are calculated and included in the propagation equation along with re-
absorption factor. The intensity scaling laws with length for HHG are then calculated taking standard density and ionization conditions. A comparison of these scaling laws with the observed intensity scalings of various harmonic orders brings out the relative roles of various phase-mismatch factors on HHG.

Improvement in the stability and the cut-off of the HHG is important for their use as a coherent x-ray source for practical applications. These improvements through the optimization of laser and plasma parameters are discussed in Chapter 6. It was observed that after various optimizations of laser and plasma conditions, one can continuously generate high order harmonics for about 5 minutes from plasma plumes produced from silver target, without much change in their intensity and spectrum. During the experiment, the laser operated at 10 Hz repetition rate and pre-pulse irradiated the same spot on the target surface. Next, a second plateau of harmonics was observed in Mn plasma plumes. It was seen that HHG spectrum from Mn plasma consists of a plateau, followed by a sharp cut-off at ~29th order. The spectrum again starts at 33rd harmonic and the intensity of the higher orders falls rapidly. Next, it was experimentally observed that if one increases the excitation of the pre-plasma, and optimizes the intensity of fs pulse, the harmonic orders below 33rd order are suppressed, and the HHG cut-off extends to 73rd H order. The spectrum looks as if the HHG plateau starts from 33rd harmonic order. Details of observation of second plateau and physical understanding of its occurrence are presented in this chapter.

In order to increase the conversion efficiency of HHG process, we have tried out novel targets to explore the effect of the enhanced non-linear optical properties on the harmonic conversion. For instance, the nanoparticles, due to their small size exhibit increased optical nonlinearity and high absorption at surface plasmon resonance. It was anticipated that they would also generate high order harmonics with greater efficiency. In
Chapter 7 we present our study of HHG from plasma plumes of nano-structured targets. Nano-structured targets such as fullerene and metal nano-particles of Ag, Au, etc. were used. These targets were made from the dried mixture of nanoparticles of various materials with organic matrix or with simple glue. It was observed that the intensity of the lower order harmonics generated from these targets is much enhanced compared to that of the corresponding harmonics from bulk materials. For example, the intensity of the 9th harmonic from the plume of silver nano-particles is ~200 times higher compared to that from plasma plume of bulk silver. Apart from laser and plasma conditions, the intensity of harmonics from nano-structured targets also depends on the target preparation technique. This aspect is also discussed in this chapter. The HHG from nanoparticles is not as stable as that from the bulk materials. This is primarily due to the fact that unlike in the bulk materials, only a thin layer of nano-structured material is available as target. Hence, the stability of the HHG from such targets is limited by the depletion of the target. The stability of HHG from these targets can be increased by increasing the target thickness and also by optimizing plasma formation conditions. It was observed that if one uses lower intensity of laser pre-pulse to produce plasma plumes, the harmonic generation could continue without appreciable loss of intensity for up-to ~200 laser shots fired at the same spot.

Spectral characterization of harmonic radiation is important especially for the matching of harmonic wavelengths with resonance transitions for the generation of high efficiency harmonic radiation. In this regard, the generation of broadband harmonics can be helpful for easier matching of harmonic wavelengths with atomic/ionic resonances. Spectrally broadened harmonics are also useful in various spectroscopic applications for the same reason. Chapter 8 describes our study on broadband HHG. The spectrally broadened harmonics were observed when spectrally broadened fs laser was used. The
latter was accomplished by passing the fs laser pulse through a 5 mm thick glass plate while it was being focussed by the lens. The intensity of the laser at glass surface was kept low enough to avoid filamentation and white light generation. The self-phase modulation of the laser pulse inside the glass medium results in spectral broadening of the laser pulse. The bandwidth of the laser pulse was increased for ~18 nm to ~32 nm. It was observed that the bandwidth of the lower harmonic orders gets increased. For example, the bandwidth of the 17th harmonic generated from plasma plumes of Ag increased from ~0.5 nm to ~0.9 nm. Since the laser pulse, after passing through the glass plate, has inherent positive chirp, the harmonic orders are also shifted towards red side. It was also observed that the bandwidth of the high order harmonics generated from nanostructured targets can be increased by increasing the intensity of fs laser pulse inside the plasma plume. For example, the bandwidth of the 11th harmonic generated from plasma plumes containing Ag nanoparticles increased from ~1.4 nm to ~5 nm when the intensity of laser was increased from $1.8 \times 10^{15}$ W cm$^{-2}$ to $3.5 \times 10^{15}$ W cm$^{-2}$. In this case, the spectral broadening of harmonics was only towards the blue side. This phenomenon is similar to that observed in our previous study on the spectral blue broadening of picosecond laser pulses towards blue side during their interaction with rare gas clusters. A theoretical model was earlier proposed to explain the observed blue broadening of the spectrum of the scattered laser pulse ($\tau \sim 27$ ps) due to its interaction with rare gas clusters. The model explains that the interaction of intense laser pulse with expanding clusters results in blue broadening of the laser pulse through self-phase modulation (SPM). This SPM is prominent when the cluster is passing through its resonance phase at $3n_{cr}$, where $n_{cr}$ is the critical density of the plasma. The same mechanism results in the blue broadening of the laser spectrum during its interaction with metal nanoparticles. The harmonics generated by this blue broadened laser pulse are also blue broadened. Salient
features of this model are described in this chapter for the understanding of underlying process.

The interaction of the femtosecond laser pulses with the plasma plume resulted in the generation of only odd harmonic orders. No even harmonic order was observed due to the inversion symmetry of the process. However, this inversion symmetry of the process could be broken by the introduction of small amount of second harmonic radiation in the main pulse. This led to the generation of both even and odd harmonics with similar conversion efficiency and thereby increasing the conversion efficiency by factor of ~ 2. Chapter 9 presents our experimental study on HHG from two-colour excitation. The Ti:sapphire laser pulse with its second harmonic was used for HHG. The second harmonic of the Ti:sapphire laser pulse was generated by passing the laser beam through a KDP crystal. The conversion efficiency of the second harmonic was kept low to ~ 2.5%. When HHG was carried out using the fundamental laser radiation together with this small intensity second harmonic radiation, the harmonic spectrum showed both even and odd harmonic orders with comparable intensities. These observations indicated that although the relative intensity of the second harmonic pulse was very small, it was sufficient for symmetry breaking of the HHG process. It was observed that if one increases the intensity of the fs pulse inside the plasma plume, the cut-off for odd harmonic orders increases, whereas the cut-off for even harmonics decreases. The fundamental and second harmonic pulses got partially separated in time due to dispersion in SHG crystal, the separation depends on the thickness of the crystal. The observed reduction of cut-off order of even harmonics with increasing intensity is explained by HHG from leading edge of fundamental pulse. The HHG comes from the leading edge where intensity remains below saturation intensity. With increasing focussing, the HHG comes from progressively earlier part of the leading edge thereby reducing the overlap.
between fundamental and second harmonic pulse resulting in disappearance of even harmonic orders.

Finally, the thesis concludes in Chapter 10 with a summary of the results, and a brief discussion on possible future work.