High-Order Harmonic Generation based on Femtosecond Dynamic Grating
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Abstract- The high-order harmonic generation (HHG), if phase matched, opens a unique way to produce high flux coherent ultrafast extreme-UV (XUV). We describe an efficient and self-phase matched method for scaling and narrowing the bandwidth of high-order harmonics in different transversal modes based on femtosecond dynamic grating in a gas jet. The grating pattern introduces a phase modulation in the dipole emission imprint such pattern in the gas medium. This technique has potential of self-phase controlled XUV generation by adjusting the intersect angle and the ratio-intensity of interfering fundamental beams. The limitations of using high intensity laser fundamental pulses in gas, such as non-negligible magnetic field, self-focusing, and plasma dispersion in the generated harmonics beam path can be addressed by using this technique. Another advantage of this technique is perfect phase-matched hard X-ray generation using mid-IR driving laser where using long wavelength dramatically decreases the HHG intensity.

Key Words: High-order harmonic generation, Femtosecond dynamic grating, Ultrafast extreme-UV, Transversal modes, Self-phase matched.

1. INTRODUCTION

When an intense laser pulse on the order or above of 1014 W/cm2 is focused into a noble gas or onto solid surface, high-order harmonics of the fundamental driving field can be produced [1, 2]. Phase-matched high harmonic generation (HHG) process can produce high flux coherent ultrafast/attosecond extreme-UV or soft x-ray light sources [3-5], make it possible to directly access the fastest time scales relevant to electron dynamics in atoms, molecules and materials [6, 7], ultrafast transient holography [8], ultrafast probe of atomic and molecular systems [9, 10], coherent diffraction surface imaging [11], or developing a new generation of laser systems, i.e. attosecond seeded free electron lasers (seeded FELs) [12]. Noticeably, soft x-ray in the water-window, which covers the spectral range between the carbon K-edge (4.4 nm) and oxygen K-edge (2.34 nm), can be used for high resolution imaging of organic materials [13].

So far, most of the investigations of HHG from gaseous media has been focused on production of harmonic radiation using single driving laser beam. In such process, an intense femtosecond laser pulse is focused into a gas jet to generate XUV or soft X-ray radiation collinearly with respect to the driving laser beam [14]. However, efficient HHG has been demonstrated only for photon energies <130 eV [15], and large effort has been devoted to improve conversion efficiency of HHG in soft x-ray with photon energies >130 eV. The most significant barrier to generate efficient HHG with such photon energies is the low conversion efficiency which limits achievable output flux. It is because of the fact of phase mismatch since the driving laser and harmonic fields in general have different phase velocity in the medium. For photon energies <130 eV, the perfect phase matched HHG can be achieved by balancing dispersion of the neutral atoms and the free-electron plasmas. It is worth mentioning that an increase in laser intensity to increase photon flux from harmonic radiation is limited by depletion of the medium via ionization of neutral as well as plasma dispersion which degrades HHG gain and also distorts the laser pulse in the plasma.

To increase the observable harmonics and to overcome the phase mismatch factors which severely reduce the efficiency, many efforts have been made and different techniques have been proposed [16-23]. For instance, Yakovlev et al. [16] have theoretically shown that conversion efficiency can be improved by orders of magnitude and phase-matched build-up of harmonics can be achieved in a jet with a high gas pressure by using mid-infrared (IR) driving fields. In the absence of perfect phase-matching, quasi-phase matching (QPM) is an alternative method to overcome phase matching limitation and consequently improves conversion efficiency [20, 21, 24]. QPM was first proposed by Armstrong et al. [24]. In their work, the phase mismatch is periodically corrected by introducing a periodicity corresponding to twice the coherence length in the nonlinearity of the medium, 2Lc=2π/Δk, where Lc is coherence length and Δk=k(qω)-k(ω) is wave-vector mismatch in general nonzero, k(ω) is the wave-vector of radiation of angular frequency ω and q is the harmonic order. A modulated waveguide can be also used to create a periodic change in the fundamental laser intensity [23, 25]. With the purpose of improving the phase matching, a weak counter-propagating pulse has been used together with driving laser beam [17, 22]. In this method, the counter-propagating field induces both a standing amplitude and phase modulation on the driving laser field. Recently, generation of XUV optical vortex is implemented by interfering a strong Gaussian driving pulse and a weak Laguerre Gaussian (LG) perturbing one [18]. Moreover, by using two-color orthogonally polarized driving IR pulses, the high harmonic signal is shown to be increased [19]. In these works and in similar papers, the effect of
transient gratings produced by interfering of two femtosecond pulses of same or different wavelengths considering the transversal variation of intensity on generation of phase matched high-order harmonics narrow bandwidth with different transversal modes, to the authors’ knowledge, has not been investigated.

Here we have shown that how a dynamic grating, generated by the interference of two coherent femtosecond IR/visible or UV pulses, can be used to control the intensity distribution and phase of the produced XUV pulses which has application in microscopy down to nano-metric scale and manipulating the detecting object with complex rotator-vibrational moving. In addition, the dynamic grating effect on the temporal and spatial properties of the HHG generated by Gaussian or LG beam has been investigated.

2. INTERFERENCE OF TWO FEMTOSECOND PULSES

The transient grating is created by two mutually coherent light beams. We consider the general case that two coherent light pulses with wavelength $\lambda_1$ and $\lambda_2$ and polarization $e_1$ and $e_2$ propagate in x-z plane and intersect at an angle $\theta$ (see Fig. 1). The electric field of two pulses in paraxial approximation can be written as

$$E_{1,2}(r, t) = e_{1,2} E_{1,2}(r_1) f_{1,2}(t) \times$$

$$\exp \left\{ -i \left[ \omega_{1,2} t + \Theta_{1,2}(r) + \Phi_{1,2}(t) \right] \right\}$$  \hspace{1cm} (1)

where $\Phi_{1,2}$ and $E_{1,2}$ are envelope phase and transversal electric field of the interfering laser beams, respectively, with

$$f_{1,2}(t) = \exp \left[ -2 \ln 2 \left( \frac{t}{\tau_{1,2}} \right)^2 \right]$$  \hspace{1cm} (2)

and

$$\Theta_{1,2}(r) = k_{1,2} \cdot r + \frac{k_{1,2} \tau_{1,2}^2}{2R_{1,2}(z)} + \varphi(z)$$  \hspace{1cm} (3)

Here $\omega_{1,2}, k_{1,2}, R_{1,2}(z)$ and $\varphi(z)$ are the carrier frequency of the interfering laser pulses, the wavevector of the fields, radius of curvature of the wavefront and Gouy phase of the interfering beams, respectively.

Assuming that there is no delay between two pulses, the intensity distribution of the total field in space-time gives a dynamic pattern with the intensity distribution over time and space as

$$I(r, t) = E_1^2(r_1) f_1^2(t) + E_2^2(r_2) f_2^2(t) +$$

$$2E_1(r_1) E_2(r_2) f_1(t) f_2(t) \cos(\delta)$$  \hspace{1cm} (4)

where ignoring the Gouy phase and the phase due to the radius of curvature of the wavefront, $\delta = \Delta k \cdot r - \Omega t$ with $\Delta k = k_2 - k_1$ and $\Omega = \omega_2 - \omega_1$ is the phase different between the beams. In this condition, the interfering beams make an intensity modulation which depends on both x and z coordinates as shown in Fig. 1.

In a spatial case of interfering two beams with different wavelength, the interference pattern will move with velocity defined by

$$V = \frac{\Omega}{|\Delta k|}$$  \hspace{1cm} (5)

Fig. 1: Schematic of interference of two beams with different frequency and wave vectors. The fringe pattern produced by two beams moves in the direction of $\Delta k$.

Fig. 2: Moving velocity of the grating produced by two beams with wavelength 800 nm and 400 nm as a function of intersect angle (up) and wavelength detuning at a fixed intersect angle $4^\circ$ (down). c is speed of light.
Fig. 2 shows moving velocity of the grating pattern versus intersect angle and detuning wavelength of the interfering beams. It can be seen that the grating period and moving velocity of the pattern initially reduces with intersect angle. According to Eq. (5), moving velocity oscillates with changing \( \theta \). In addition, at a fixed intersect angle, the moving velocity of the pattern increases very fast with detuning wavelength and this trend gets slower and for \( \Delta \lambda >400 \text{ nm} \) it reaches velocity of light.

3. **HHG BASED ON DYNAMIC GRATING**

The transient grating can be created by two mutually coherent light beams. In HHG based on femtosecond transient grating a fundamental linearly polarized femtosecond Gaussian laser beam (e.g. 800 nm from Ti:Sapphire laser) with intensity on the order of \( 10^{14} \text{ W/cm}^2 \) and pulse duration of few 10 fs is superimposed in time and space at a tunable angle with a pulse with controllable transversal shape and polarization. The duration of the pulses should fulfill the criteria of the maximum modulation depth. The temporal overlap of two pulses can be achieved by a translation stage in the perturbing beam path (see Fig. 3). By introducing a desired beam shaper in the interfering beam path, the transversal mode can be engineered to create a desired transversal shape. For example, by passing the perturbing beam through a q phase plate it can be converted to LG mode with an orbital angular momentum [5].

The polarization is controlled by introducing birefringence \( \lambda/2 \) and \( \lambda/4 \) retarding plates.

Both laser beams are focused with the estimated intensity ratio \( I_1/I_2 \) and the intersect angle into a noble gas medium. The superposed coherent driving pulses and the interfering pattern yields a spatially modulated distribution of the energy density. The grating pattern is static if both beams have same color, whereas the moving grating can be produced by the detuning wavelength of the interfering beam or by interfering the fundamental beam and its harmonic (for instance, second harmonic of 800 nm which can be generated in BBO). In this condition, the interfering beams make an intensity modulation which depends on both \( x \) and \( z \) coordinates and moves with velocity which defines by Eq. (5).

The interaction of the created dynamic grating with atoms imposes a suitable grating shape for desired harmonic mode onto the plane of emitting dipoles which leads to create a periodic sources for generating of harmonics of the fundamental beam to carry the information of interfering pattern. The period and the velocity of the grating can be tuned by adjusting the intersect angle and wavelength detuning for constructive interference of the HHG signals generated by the sources in dynamic grating.

4. **RESULT AND DISCUSSION**

As mentioned above, the period and the velocity of the grating can be tuned by adjusting the intersect angle and wavelength detuning. Therefore, a spatiotemporal interference of HHG can be achieved which increases the HHG efficiency by a factor which depends on the number of bright points of driving laser pulse within interacting area. Adjusting interference of pulses should result in shorter bandwidth XUV pulses. To synchronize perfectly the generated pulses, it is possible to use diffraction pattern of two LG beams with opposite angular momentum. This pattern contains several maximum loops on a circle in the transversal plane respect to the propagation direction.

Fig. 3: Schematic of possible optical setup for engineered transversal mode HHG by moving grating. Refer to the text for details.

The phase matching condition in HHG is related to the momentum conservation in the total interaction process. In Fig. 4, contribution of the transient grating in HHG is shown versus a two-dimensional momentum geometry of the fundamental and interfering beams in \( x-z \) surface. In photon picture, the combination of \( m_1 \) photons from fundamental laser pulse with frequency \( \omega_1 = \omega \) and \( m_2 \) photon from interfering laser pulse with frequency \( \omega_2 = \omega \) for stationary grating or \( \omega_2 = 2\omega \) for moving grating, contribute in high harmonic radiation with \( q= \omega_0 \) where \( q= m_1 + m_2 \) or \( q= m_1 + 2m_2 \) for stationary and moving grating, respectively. Then, for small intersect angles, the diffraction angle of the generated harmonic can be given by:

\[
\beta_{q,m_2}(\theta) = 2m_2 \theta / q
\]

(6)

The angle and correspondingly the phase of the \( q \)th harmonic depends on the number of interfering photons that contribute in HHG, and intersect angle of interfering beams. Therefore, the constructive interference of the generated harmonics can be achieved by adjusting the relative intensity
of the interfering and fundamental beams together with intersect angle.

5. CONCLUSION

In HHG based on dynamic grating, an enhancement in conversion efficiency can be achieved by self-phase matching due to the modulation of geometrical parameters and tuning phase of the harmonics with modulating intensity of the interfering beams as well as intersect angle. The method introduced here can be extended for any combination of driving lasers to produce different grating patterns with various tunable properties to create phase matched XUV pulses with engineered shapes. A control over the spin as well as orbital angular momentum of the femtosecond interfering beams can pave the way toward tailoring more complex XUV light sources for fundamental studies and applications. In addition by interfering a mid-IR laser beam and its suitable harmonic combined with favorable phase matching condition, it is possible to extend the applications of this technique from XUV to soft X-ray with reasonable intensity for application in microscopy and biology.

REFERENCES