# Multiple quasi-phase-matching for enhanced generation of selected high harmonics in aperiodic modulated fibers

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**Abstract:** A technique for enhanced generation of selected high harmonics in a gas medium, in a high ionization limit, is proposed in this paper. An aperiodically corrugated hollow-core fiber is employed to modulate the intensity of the fundamental laser pulse along the direction of propagation, resulting in multiple quasi-phase-matched high harmonic emissions at the cutoff region. Simulated annealing (SA) algorithm is applied for optimizing the aperiodic hollow-core fiber. Our simulation shows that the yield of selected harmonics is increased equally by up to 2 orders of magnitude compared with no modulation and this permits flexible control of the quasiphase-matched emission of selected harmonics by appropriate corrugation.

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## 1. Introduction

High-order harmonic generation (HHG) driven by ultrashort laser pulses is a source of extreme-ultraviolet and soft X-ray light with the unique properties of highly spatial and temporal coherence [1-4]. This short wavelength light source has made many new applications possible such as ultrafast spectroscopy and bio-microscopy [4, 5]. However, both the conversion efficiency and the highest achievable photon energy have hitherto been limited by the phase mismatch in the frequency conversion process at high ionization levels. Thus, overcoming the phase mismatch has been a grand challenge to the further development of EUV and Soft X-ray light sources. KM group [3, 6-8] experimentally demonstrated using a periodically modulated hollow waveguide to modulate the intensity of the driving laser to implement quasi-phase-matching (QPM), where the reciprocal lattice vector resulted from the modulation of laser intensity could compensate the phase mismatch. This work succeeded in enhancing the conversion efficiency into soft X-rays region by about one order of magnitude, however, it can provide only one reciprocal lattice vector and thus only the signal that qualified the QPM could build up constructively, whereas the conversion efficiency of other harmonic orders are still relatively low. I.P.Christov et al. [9, 10] introduced a tapered modulated waveguide with a linearly chirped corrugation to provide better conditions for quasi-phase matching of an ultrabroadband x-ray continuum generated in cutoff region. Recently, X. Y. Zhang et al. theoretically proposed using a periodic corrugated and chirped hollow fibers to enhance the extended high harmonic generation from a few-cycle laser pulse [11]. In this paper, we propose a new approach to achieve multiple QPM in the process of HHG by using aperiodically modulated hollow-core fiber which is optimized by Simulated Annealing method. Since the aperiodically modulated hollow-core fiber provides more reciprocal vectors, it can compensate the phase mismatch of different order of high harmonics accurately and make constructive buildup of coherent signal, and thus we can flexibly control the quasi-phase-matched emission of selected harmonics by appropriate corrugation.

## 2. Optimizing method

In order to achieve a predesigned multiple QPM process [12-14], the hollow-core fiber with total length L is divided into N unit blocks with congruent length  $\triangle$  L, as shown in Figure 1. The modulation of fiber inner diameter on each block and the sequence of modulation are determined by the simulated annealing (SA) method to optimally construct the structure [14]. We chose the objective function in SA method as  $F = \sum_{i} G_i - \sqrt{\sum_{i} (G_i - \overline{G})^2}$ , where the

former item is the sum of G, and the latter is the variance of G. To achieve the maximum function F is to make the sum of G be maximum and the variance of G be minimum.



Fig. 1. Schematic diagram of the constructed aperiodically corrugated fiber by SA method in part.

Here, let us consider the process of HHG. Using tunneling ionization rates from ADK theory [15], we can calculate the level of ionization present for a given harmonic generated by the laser pulse. Based on this point, we then calculate the wavevector mismatch between the fundamental laser pulse and the qth harmonic, which is due to the waveguide, plasma, and neutral atoms

$$\Delta k = k_{q\omega} - qk_{\omega} = \frac{qu_{11}^{2}\lambda}{4\pi a^{2}} + P\eta N_{atm}r_{e}(q\lambda - \lambda/q) - \frac{2\pi(1-\eta)Pq}{\lambda}(\delta(\lambda) - \delta(\lambda/q)), \qquad (1)$$

where q is the order of the generated harmonic and  $\omega$  is the fundamental laser frequency,  $\lambda$ ,

a,  $u_{11}$ ,  $\eta$ , P, N<sub>atm</sub>,  $r_e$  and  $\delta$  are the fundamental wavelength, waveguide radius, first zero of Bessel function  $J_0$ , ionization fraction, gas pressure in atmospheres, number density in 1 atm, classical electron radius, and index of refraction of the neutral gas at 1 atm, respectively. Under the conditions of low pressure and high level of ionization, the contribution of the neutral gas to the phase mismatch can be neglected [6], and the dominant terms in Eq. (1) are

due to the waveguide and plasma. Making the assumption that  $q\lambda >> \lambda /q$ , the phase mismatch is then given by

$$\Delta k(\lambda) = \frac{q u_{11}^2 \lambda}{4\pi a^2} + \frac{q n_e e^2 \lambda}{4\pi m_e \mathcal{E}_0 c^2},\tag{2}$$

where  $n_e$  is the electron density, other physical parameters are the same with those in Eq. (1).

In a simplified model of harmonic generation [6], the field of harmonic order q, after propagating a distance L in a nonlinear medium, is related to the phase mismatch  $\triangle$  k by

$$E_{q\omega}(L) \propto \int_{0}^{L} E_{\omega}^{q}(z) d(z) \exp(-i\Delta kz) dz , \qquad (3)$$

where  $E_{\omega}(z)$  is the fundamental laser field, d(z) is the nonlinear coefficient for HHG, and  $\triangle k$  is the phase mismatch calculated above. Eq. (3) can also be expressed as

$$E_{q\omega}(L) \propto E_{\omega}^{q} d_{eff} L \left| \frac{1}{L} \int_{0}^{L} g(z) \exp(-i\Delta k(\lambda)z) dz \right| = E_{\omega}^{q} d_{eff} L \times \left| G(\Delta k) \right|$$
(4)

where  $d(z)=d_{eff}g(z)$ ,  $d_{eff}$  is the effective nonlinear coefficient, and g(z) represents the inner diameter modulation of each block taking binary values of 1 or the modulation depth. The laser intensity is sufficient to generate the cutoff harmonics only in regions where the inner diameter is small, i.e. g(z) is small. Here, we define the last term in Eq. (4) as a new physical parameter  $G(\triangle k)$  which can scale the conversion efficiency of HHG by Fourier

transformation. So the conversion efficiency from driving laser pulse to HHG depends completely on the fiber structure and is proportional to  $|G(\triangle k)|^2$ .

Thus, the original question turns into how to construct an inner diameter-modulated fiber structure so that multiple QPM HHG process can be obtained with high efficiency simultaneously, i.e.  $G(\triangle k)$  is optimal and equal to each other. Since the fiber is divided into N uniform blocks, so  $G(\triangle k)$  can be expanded by

$$G\left[\Delta k(\lambda)\right] = \left|\frac{1}{L}\int_{0}^{L}g(z)\exp(-i\Delta k(\lambda)z)dz\right| = \frac{1}{N\cdot\Delta L}\left|\sum_{p=0}^{N-1}g(z_{p})\int_{z_{p}}^{z_{p}+\Delta L}\exp(-i\Delta k(\lambda)\xi)d\xi\right|,$$
(5)

where  $\triangle k(\lambda)$  depends on the order of high harmonics and can be calculated from Eq. (2). In order to achieve the largest conversion efficiency for the specific order of high harmonic, we should make  $G(\triangle k)$  be maximum, i.e.  $g(z_p)$  should be optimized by SA algorithm to make

$$\left|\sum_{p=0}^{N-1} g(z_p) \int_{z_p}^{z_p + \Delta L} \exp(-i\Delta k(\lambda)\xi) d\xi\right| \text{ maximum while } \frac{1}{N\Delta L} \text{ is a constant.}$$

# 3. Results and discussion

Here, we pay much attention to the HHG in "water window" ( $\lambda = 2.33$  nm  $\sim 4.37$  nm, where carbon-containing biological objects absorb radiation efficiently but water is comparatively transparent), since light sources at this waveband can be used to observe microscopic biological structures in the living state by means of holography [4]. In our calculation, a strong laser pulse with wavelength of 800nm is focused into the fiber with extremely high intensity. The fiber is filled with Ne gas with pressure of 10 torr. Since the focused laser intensity is high enough to fully ionize Ne gases in low pressure, the electron density  $n_e$ resulted from the laser-gas interaction is the same with the atom density  $N_{atm}$ . Here, we just consider a very simplified model, whereas the absorption loss of the driving laser, refraction, mode beating, group velocity dispersion are not taken into account. The harmonics are chosen with order of 181<sup>th</sup>, 183<sup>th</sup>, 185<sup>th</sup>, 187<sup>th</sup>, 189<sup>th</sup>, and 191<sup>th</sup>. The length L of the fiber is 1cm. Firstly, we theoretically investigated the optimal length of unit block. Under a modulation depth of 0.1,  $G(\triangle k)$  was calculated from the 171<sup>th</sup> to 201<sup>th</sup> harmonics, with unit block length  $^{\text{A}}$  L=50µm, 20µm, 10µm, respectively. The calculated harmonic intensity scales  $|G(^{\text{A}} k)|^2$ . As shown in Figure 2, the calculated results show that the yield of selected harmonics in "water window" by aperiodically modulated fiber is enhanced 10-100 times compared with no modulation. The intensity of the selected 6 harmonics with fiber modulation are apparently higher than those of  $171^{\text{th}} \sim 179^{\text{th}}$ ,  $193^{\text{th}} \sim 201^{\text{th}}$  harmonics, and the shorter  $\triangle$  L is, the more intense the generation of the selected harmonics. This is partly because increasing block numbers can provide much more probability to search the most optimum structure to make the conversion efficiency highest. And partly because the shorter  $\triangle$  L is, the more favorable and feasible reciprocal vector the modulated fiber would provide, which can compensate the phase

feasible reciprocal vector the modulated fiber would provide, which c mismatch more accurately.



Fig. 2. Enhancement factor over no modulation as a function of harmonic order, using aperiodically modulated fibers with different unit block length  $\triangle$  L. All the modulation depth is 0.1.

It is interesting to investigate the effect of modulation depth on the harmonic yield. Here, we calculated the harmonic intensity with modulation depth of 0.1 and 0.2 under the same unit block length. In Figure 3, the square dots are harmonic intensity in a straight fiber, while the circle and triangle dots are the intensity calculated with modulation depth of 0.1 and 0.2, respectively. The average intensity of 181th~191th harmonics with fiber modulation depth of 0.1 has been enhanced nearly 100 times than the counterpart from straight fiber, so it can be clearly seen that, through small modulation on the fiber inner diameter by appropriate sequence, an increment of harmonic intensity by up to two magnitude can be easily achieved. With the modulation depth of 0.2, the enhancement factor over no modulation becomes approaching 400, 4 times higher than that with modulation depth of 0.1. The deeper the modulation of fiber inner diameter of the fiber is, the smaller and tighter the laser intensity is. This is because stronger field of laser radiation caused much higher flux of the high harmonics.



Fig. 3. Enhancement factor over no modulation as a function of harmonic order, using aperiodically modulated fibers with modulation depth of 0.1 and 0.2. The unit block length  $\triangle$  L is 10 $\mu$ m.

We also calculated the intensity of harmonics from the whole harmonic order of 25th to 301th, as shown in Figure 4. The intensity of selected 6 harmonics in "water window" is like a high flat narrow ladder standing on a widest grassland, which reveals that predesigned aperiodically modulation of a fiber by SA algorithm greatly improved the intensity of harmonics we selected while suppressed other orders of harmonic generation. This ensured most energy of the driving laser pulse can be turn into the selected harmonics, avoiding energy waste to other harmonics. Thus, we can obtain any certain orders of high harmonic we need with high conversion efficiency through an aperiodically modulated fiber which can be designed by SA method.



Fig. 4. Harmonic emissions from the aperiodically modulated fibers through orders of  $25^{th}$  to  $301^{th}$ , with a modulation depth of 0.1 and unit block length of  $10\mu m$ .

## 4. Conclusion

In conclusion, we have proposed an aperiodically modulated waveguide designed by SA algorithm to achieve multiple quasi-phase-matched HHG simultaneously with equal enhancement factor. The calculated results show that the yield of selected harmonics in "water window" by aperiodically modulated fiber is enhanced 100 times compared with no modulation. With shorter block length and greater modulation depth, the conversion efficiency can be improved much more, because it can provide more favorable reciprocal vectors to compensate the phase mismatch. From the discussions above, this technique is really feasible and provides us a new promising route to select appropriate high harmonics to approach the attosecond frontier.

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