Optimized waveforms for enhancing high-harmonic yield by synthesizing multi-color laser fields

Cheng Jin[1], **Guoli Wang**[1,2], **Hui Wei**[1], **Anh-Thu Le**[1], **C. D. Lin**[1]

[1] *J. R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas, 66506, U.S.A.*

[2] *Key Laboratory of Atomic and Molecular Physics and Functional Materials of Gansu Province, College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou 730070, China*

Abstract: High harmonics favoring phase-matching conditions can be enhanced by one to two orders of magnitude if the laser's waveform is optimized by synthesizing two or three-color fields without an increase in the total energy.

OCIS codes: 190.2620, 020.2649.

1. Introduction

High-order harmonic generation (HHG) is an extreme nonlinear optical process when atoms and molecules are exposed to strong femtosecond laser pulses. HHG light has been limited to the extreme ultraviolet ($\sim$100 eV) with widely available Ti:sapphire lasers operating at a 0.8-μm wavelength. Advance of mid-infrared lasers with wavelengths of a few microns makes the harmonic cutoff easily extending to the X-ray region [1]. Today the main limitation that prevents HHG from emerging as a useful light source is its low conversion efficiency [2].

HHG enhancement can be achieved by altering the driving electric field at the sub-cycle level to modify the single-atom response. This was investigated by two-color phase control as first shown experimentally by Watanabe *et al.* in 1994 [3]. More recently, it is possible to perform coherent wavelength multiplexing of ultra-broadband pulses with full phase and amplitude control and to achieve any optical waveforms [4–6].

In this work, we demonstrate how to enhance HHG yields by one to two orders of magnitude by modifying the laser waveform to increase the harmonic emission from each atom that are favorable for macroscopic phase-matching. This is accomplished by synthesizing "only" two- or three-color laser fields without increasing the total laser power, much easier to perform in comparison with an early study of an ideal waveform [7].

2. Results and Discussions

High harmonics generated from an atom within each optical cycle can be described by a three-step model: electron tunneling ionization, propagation in the laser field, and recombination with the parent ion. The last step is the property of target, which cannot be altered by the external laser field. According to the quantitative rescattering (QRS) theory [8], enhancing single-atom harmonic emission is carried out by manipulation of the waveform to optimize the returning electron wave packet. Since high harmonics measured in the laboratories should be properly phase-matched in the gas medium [9], macroscopic effects should also be considered in the optimization. Therefore, our strategy for optimal macroscopic HHG is: (a) to increase laser's electric fields at ionization times to release more electrons for harmonic emission, (b) to enhance "short"-trajectory electrons over "long"-trajectory ones since the former can efficiently phase-matched in the medium, (c) to maintain ionization level in each cycle to be less than a few percent, thus avoiding ground-state depletion and plasma defocusing of laser beam, and (d) to maintain nearly constant total energy of the synthesized laser pulse.

Figure 1(a) shows two waveforms over one optical cycle: the single-color (SC) sinusoidal wave of the fundamental (1600 nm) (black) and the optimized waveform (Opt. WF) synthesized from the fundamental and its 3rd harmonic(red). Optimization was performed such that the cutoff and total laser energy for the two waves were about the same. To illustrate the enhancement of the Opt. WF, consider an electron returning with a kinetic energy of 2 U_P, where U_P is referred to the SC field. The ionization time and the recombination time for the "long"- (open circles) and "short"-trajectory electrons (solid circles) are indicated for each waveform. The inset gives the electric fields at ionization times versus the returning electron energies. Comparing the two waveforms, it is clear that (i) the optimized one has higher

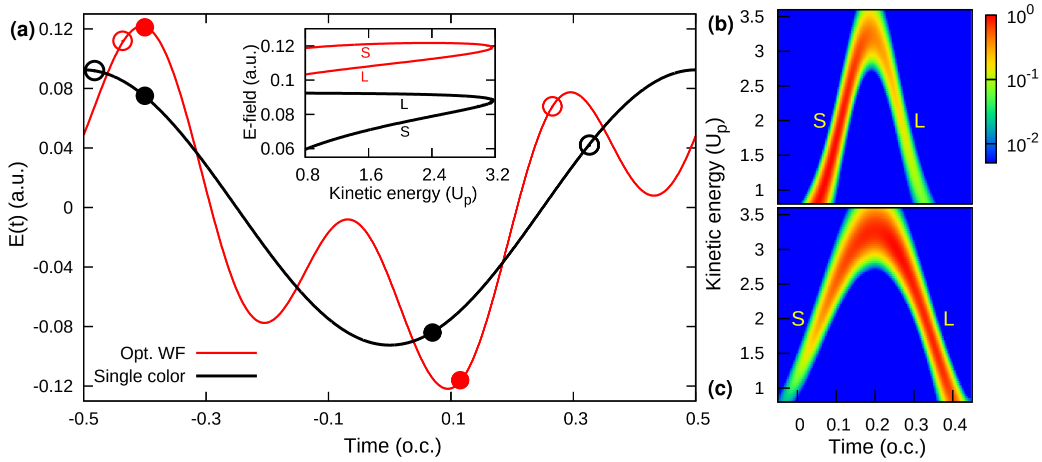

Fig. 1. Principle of the optimized waveform for enhancing the yields of high harmonics. (a) Comparison of electric fields over one optical cycle for a SC wave and a synthesized WF. The ionization and recombination times of 2 U_p electron are shown in open ("long"-trajectory) and filled ("short"-trajectory) circles. The inset shows the electric fields at ionization times for two waves. Time-frequency analysis of high harmonics are shown for WF and SC in (b) and (c), respectively.

electric fields at ionization times, thus leading to more returning electrons, and (ii) there are more "short"-trajectory electrons than "long" ones. Figures 1(b) and (c) show the time-frequency wavelet analysis of HHG calculated using QRS theory [8]. The strong enhancement of "short"-trajectory electrons and the smaller attochirp for the optimized wave are clearly seen. In the simulation, the target was Ne. For the 1600 nm alone, we chose an intensity of 3×10^{14} $\mathrm{W/cm^2}$. The carrier-envelope phase of the fundamental was set as 0. The optimization returned the peak intensity for the fundamental as 1.98×10^{14} $\mathrm{W/cm^2}$, and 1.32×10^{14} $\mathrm{W/cm^2}$ for its 3rd harmonic laser, which had the optimized phase of $1.36\,\pi$.

We have performed macroscopic propagation calculations in a typical gas medium and showed that the harmonics generated with optimized waveform were further enhanced in comparison to those generated from the single-color pulse. Optimization using three-color waves have also been carried out. While the additional gain in harmonic yields was moderate, it favored the generation of single attosecond pulses.

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