Control of High-Order Harmonic Generation through Shaped Pulse Optimization

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Abstract: We demonstrate that using phase-only control of ultrashort pulses, we can selectively enhance the brightness of high-harmonic generation by over an order of magnitude, and generate near transform-limited soft x-ray pulses.

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In this work, we demonstrate for the first time coherent control of a very high-order nonlinear process[1]. By altering the shape of an ultrashort laser pulse using phase-only control with a deformable mirror, and by adaptive control of the pulse shape using an evolutionary algorithm, we can both enhance the brightness and control the spectral characteristics of high-order harmonic generation (HHG). We demonstrate an enhancement in peak brightness by over an order of magnitude, as well as enhancing a single harmonic order compared to its adjacent orders. We can also spectrally narrow a given harmonic order, resulting in the probable generation of ultrashort x-ray pulses with near transform-limited temporal pulse shape. This work represents the first time that adaptive or "learning" control of a high-order nonlinear process has been experimentally demonstrated. Surprisingly, the optimal laser pulse shapes differ only slightly from the transform-limited case (21fs versus 18fs), but have significant phase variations on the leading edge of the optimized pulse.

Fig. 1: Optimization of a single (27th) harmonic in argon while suppressing adjacent harmonics. The high-order harmonic spectrum for the best pulse shape solution for each iteration of the evolutionary strategy is plotted.

For our experiment, pulses are generated by an amplified Ti:Sapphire laser system capable of generating transform-limited 16fs pulses with mJ energy levels.[2] A zero-dispersion stretcher folded by a micro-machined deformable mirror (MMDM) manipulates the phase of the amplified pulses. When the mirror is deformed, the spectral phase (and thereby the temporal phase and profile) are modified. The shaped pulses are injected into a
hollow-core fiber and generate HHG radiation in the XUV region. The generated light is coupled into a flat-field grating spectrometer and the spectral data is collected by an x-ray CCD camera.

Optimization of the HHG is accomplished using an evolutionary strategy (ES) feedback algorithm. The ES begins with a set (population) of random solutions (pulse shapes generated by a particular mirror deformation). The resulting x-ray spectrum from each member of the population is then measured. A fitness function (or cost function) is applied to the set of HHG spectra to evaluate how well each member of the population meets the target spectral characteristics (e.g., optimize a particular high-harmonic order). Those solutions with a large fitness value are retained and mutated copies (children) are made. The best solutions and the children then form the population for the next iteration of the algorithm and the process is repeated until the fitness value saturates. Prior to each optimization of the HHG radiation, the output laser pulses are compressed to the transform-limit using SHG intensity as a fitness function. The HHG spectrum generated by the transform-limited pulse is used as a baseline for comparison of the optimized HHG spectra. The result of an experimental test of the optimization is shown in Fig. 1. Iteration number 0 shows the spectrum from the transform-limited laser pulse. As the optimization proceeds, we can see the brightness of the 27th harmonic in argon is enhanced by a factor of 8 without increasing the brightness of the neighboring harmonic orders.

Figure 2 shows the laser pulse shapes corresponding to the initial (iteration number 0) and final (iteration number 94) HHG spectra shown in figure 1. The initial pulse is quite smooth and nearly transform-limited while the optimized pulse is slightly broader with some additional structure. Thus, a very slight change in the pulse used to drive the HHG process can result in a substantial and beneficial change in the output energy, brightness, and spectrum of the HHG radiation.

![Figure 2: Amplitude and phase of the laser pulses corresponding to Fig. 1: (a) pulse intensity profile and (b) phase of the transform limited reference pulse (dashed) and the optimized pulse (solid). The pulses are the same energy.](image)

By selecting different criteria (i.e, modifying the fitness function that evaluates the spectral characteristics), the ES algorithm can optimize different spectral characteristics. Figure 3 shows the results of an optimization that caused substantial narrowing of the HHG spectral peaks and increased the brightness of the 27th harmonic by a factor of 11. The measured width of the 27th harmonic is 0.24 eV, and is limited by the resolution of our x-ray spectrometer. Thus the actual enhancement of the brightness may be substantially higher than we have measured. The fact that the spectral width of the harmonic decreases, in addition to the peak enhancement, implies that the time-bandwidth product for the harmonic x-ray pulse is closer to the fourier-transform limit. For a 20 fs laser pulse, the predicted pulse duration of the x-ray pulse is ≈ 5 fs. Thus, the measured bandwidth of the 27th harmonic of 0.24 eV is consistent with a transform-limited 5fs-duration x-ray pulse at a wavelength of 29 nm. Further work is in progress to confirm this experimentally.

HHG control was implemented in a majority of the rare gasses at pressures between 2-95 torr, under conditions with and without phase matching of the HHG process. The highest enhancements were observed for conditions close to phase-matching,[3] but enhancements of at least a factor of 2 were observed for all conditions and all harmonics tested. For example, in the case of Krypton gas, the absorption and nonlinearity resulted in ~6 peaks of comparable intensity with a transform-limited pulse. Optimization of any of these peaks resulted in brightness enhancements of ~3-4x, while immediately adjacent peaks saw enhancements of ~1.5-2x, and other peaks saw negligible or negative enhancements. The conversion efficiency of the x-ray generation process optimizes for a non-transform-limited pulse. This is in stark contrast to recent experimental evidence from a number of groups that has shown low-order,
perturbative, non-linear processes (i.e. second-harmonic generation) have a maximum conversion efficiency for a transform-limited pulse. [2,4]

To explain the unexpected result that very slight changes in laser pulse shape can so dramatically enhance and select individual harmonics, we consider the semi-classical rescattering model of HHG. From a classical point of view, electrons are ionized and accelerated away from the core during one half-cycle of the laser field, and can be subsequently driven back to the core when the laser field reverses. Some fraction of the ionized electrons can recombine with the parent ion and give off their energy in the form of high harmonics. This process occurs over a few optical cycles of the laser field, resulting in a ≈ 5fs x-ray burst. Particular harmonics are generated by electrons returning to the core with a specific return energy that is related to the exact time within the optical cycles when the electron is initially ionized. Our simulations show that at a semiclassical level, the mutated phase at each successive cycle of the laser field adjusts the action acquired by the electron along the most relevant trajectory, which corresponds to the phase of the induced dipole moment. Thus, the electron phase is adjusted by the laser field in such a way that the destructive interferences are reduced and total harmonic energy output is increased. This effect is analogous to macroscopic phase matching in the bulk where matching phase velocities eliminates destructive interferences. This coherent control can lead to channeling and redirection of energy between different high-order nonlinear interactions. These findings therefore demonstrate a new type of intra-atomic “phase matching” between the laser field and the wavefunction of the ionized electron. From a quantum point of view, our optimized laser pulse can adjust the quantum phase of the electron wavefunction which returns to the core, to optimize it for a particular harmonic feature.

![Graph showing soft-x-ray intensity vs harmonic order]

Fig. 3: Optimization of a single harmonic in argon with a spectral window at longer wavelengths than in Fig. 1 and without suppressing adjacent harmonics. The harmonic peak is enhanced by over an order of magnitude. Harmonics before and after optimization are shown in blue and red respectively.

In summary, this work represents the first demonstration of control of a very high-order nonlinear process. We demonstrate significantly increased enhancement and selectivity of high-order harmonic generation, as well as the generation of near-transform-limited harmonics. We achieve this using a new type of phasematching, which allows us to channel energy into a selected harmonic while increasing total x-ray flux. This result has immediate utility in the probing of dynamics of chemical and material systems, because it provides a way to select a harmonic without temporally broadening it. The result is a bright, quasi-monochromatic, and highly spatially-coherent soft x-ray light source for use in techniques such as photoelectron spectroscopy and spectromicroscopy, time-resolved x-ray studies of material and chemical systems, and time-resolved holographic imaging.