Compression of attosecond harmonic pulses by extreme-ultraviolet chirped mirrors

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In the race toward attosecond pulses, for which high-order harmonics generated in rare gases are the best candidates, both the harmonic spectral range and the spectral phase have to be controlled. We demonstrate that multilayer extreme-ultraviolet chirped mirrors can be numerically optimized and designed to compensate for the intrinsic harmonic chirp that was recently discovered and that is responsible for temporal broadening of pulses. A simulation shows that an optimized mirror is capable of compressing the duration from ~260 to 90 as. This new technique is an interesting solution because of its ability to cover a wider spectral range than other technical devices that have already been proposed to overcome the chirp of high harmonics.

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High harmonics (HHs) generated by nonlinear interaction of an ultrashort 800-nm Ti:sapphire laser with rare gas is currently the most promising way to generate attosecond pulses. It was both theoretically[1] and experimentally[2] demonstrated that high-order harmonics form a train of attosecond pulses with a periodicity of half the IR laser period and a burst duration near a few hundreds of attoseconds (~250 as). The characteristic comb spectrum indicates that, by phase locking several HHs, one should be able to achieve pulses as short as 100 as. These ultrashort pulses are the result of the spectral interference of many high-order harmonics that gives rise in the temporal domain to an attosecond burst of radiation. The shortest pulses are obtained when many harmonics interfere in phase (the Fourier limit duration, t, is given by $t = 1300/N$ as, where N is the number of harmonics considered). As recently demonstrated both theoretically[3] and experimentally,[4] the phase relation between harmonics is not linear but there exists an inherent group-delay dispersion (GDD) between harmonics that broadens the pulse duration. Technical solutions are to be found to overcome this fundamental limitation and reach the 100-as frontier. Some, such as using the negative GDD of a thin x-ray filter,[5] have already been proposed. This method is easy to implement but can compensate for only a small chirp while maintaining good transmission. Moreover, adapted material can be found for only a small wavelength range, giving this technique poor versatility.

In this Letter we propose an alternative and more general solution consisting of compensating for the intrinsic chirp with an aperiodic multilayer mirror. This method allows for the compensation of a large range of chirps over a large spectral bandwidth. This chirped mirror has the capability of bandpass filtering the harmonics spectrum, too, and so it eliminates the undesirable harmonics, which could interfere destructively. Moreover, the multilayer coating can be made on a curved, low-aberration-level mirror, allowing a single element to both focus and compress the extreme-ultraviolet (XUV) pulse. Chirped mirrors have been widely used in the visible and near-IR ranges, and previous work[6] has shown the possibility of extending this technology to the XUV range. Aperiodic XUV multilayer stacks, necessary for chirped mirrors, have already been made that show high control of individual layer thickness.[7] The work presented here goes a step further in precisely adapting the mirror characteristics to realistic HHs with a quadratic chirp. This is made possible by new advances in the understanding of the physical origin of the intrinsic HH chirp combined with the use of a powerful optimization algorithm for mirror design.

The first step to design the chirped mirror, which will be able to compress harmonic pulses, is to predict the GDD as well as the intensity of each individual harmonic. For predicting the intensity of the harmonics, a simplified spectrum that reproduces the characteristic behavior when focusing an ultrashort laser pulse into a rare-gas cell is used. After a rapid decrease of intensity for the first orders, the harmonic conversion efficiency remains constant up to relatively HH orders (this spectral region is called the plateau) and drops in the cutoff region.

For predicting the GDD, we use the semiclassical three-step model for HH generation.[8] This theory is widely accepted and allows one to reproduce and understand the harmonic chirp behavior in the plateau

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Fig. 1. (a) Recombination time and (b) chirp versus harmonic order assuming neon as the generating gas and an IR laser intensity of 7.5×10^{14} W/cm^2 (solid curves) and 5×10^{14} W/cm^2 (dashed curves).

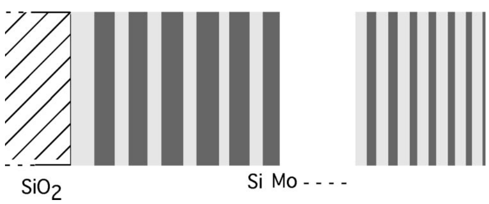

Fig. 2. Designed chirped mirror consisting of alternating Si (light gray) and Mo (dark gray) deposited on a SiO_2 substrate.

region efficiently.[3] The model considers the tunnel ionization at a time t' of an electron by a linearly polarized laser electric field with pulsation ω, $[E_0\cos(\omega t)]$. The laser field then accelerates the electron. Its trajectory, $x(t,\ t')$, i.e., the distance between the electron and its parent ion can be calculated from ionization time t' as follows:

$$x(t,\ t') = \frac{eE_0}{m\omega^2}[\cos(\omega t) - \cos(\omega t')] + \frac{eE_0}{m\omega}\sin(\omega t')(t - t').$$

The radiative recombination occurs when $x(t,\ t')=0$ and produces a harmonic photon whose energy comes from the kinetic energy acquired by the electron in the electric field of the laser. The periodicity of the phenomenon is twice the pump laser period and explains why attosecond bursts occur twice per laser cycle. The solution of the equation $x(t,\ t')=0$ provides the recombination time (t_{rec}). Consequently, the GDD of the harmonics comes from the fact that the recombination times are not exactly the same for different harmonic orders, as can be seen from Fig. 1(a) (here we represent only the shortest trajectory, which is the one that produces the shortest attosecond bursts). The harmonic coefficient of the chirp, α [Fig. 1(b)], is then obtained from the first derivative of the recombination time versus the harmonic energy ($\alpha = \partial t_{rec}/\partial \omega_{HHG}$). From Fig. 1(b) it appears that the chirp is nearly constant over the plateau, extending from H25 to H61 from a pump laser intensity of 5×10^{14} W cm^{-2} and from H25 to H101 at 7.5×10^{14} W cm^{-2}. The phase of the harmonics, φ, is deduced from the recombination time ($\varphi=\omega t_{rec}$). The GDD is then expressed by GDD$=\partial^2\varphi/\partial^2\omega$. In the conditions shown in Fig. 1 the GDD is nearly constant near 10,000 as^2 ($\alpha\approx 100$ fs^{-2}) at 5×10^{14} W cm^{-2} and near 6500 as^2 ($\alpha\approx155$ fs^{-2}) at 7.5×10^{14} W cm^{-2}.

From the considerations mentioned above it appears that the two main parameters for designing a chirped mirror that can compress a HH pulse are a large bandwidth and a negative GDD. The materials of the multilayer are dependent on the harmonic central frequency determined by experimental conditions. For example, the spectrum generated in neon is centered near 12 nm (harmonic H67), and efficient reflective multilayers can be constructed by alternating molybdenum (Mo) and silicon (Si). The material indices of refraction are from the Center for X-Ray Optics.[9] Note that, unlike with IR mirrors, the imaginary part is nonvanishing in the target spectrum, and thus attenuation is always present. The computation of the spectral reflectivity and the phase of a stack of plane layers with complex refractive indices can be performed with the usual matrix methods[10] based on the Maxwell equations. Chirped mirrors designed in femtosecond lasers in the visible and near-IR spectral domains are based on the alternation of two supposedly lossless materials with high contrast index. As a result, chirped mirrors can be designed with high reflectivity (e.g., 99.9%) over large bandwidths. Such is not the case in the XUV spectral domain, and reflectivity has to be traded off against GDD, because of simultaneous absorption and low index contrast. Consequently, the reflectivity of the chirped mirrors is lower than the reflectivity of the corresponding constant period multilayer structures. Because of the high number of constraints one needs to consider while designing a multilayer stack (HH intrinsic chirp, absorption, variation of the indices of refraction with wavelength, etc.), we prefer to use a Gibbs sampler algorithm[10] that is known to easily converge in such a multiparameter calculation. The optimization algorithm, which belongs to the class of simulated annealing algorithms,[11] was thus used to obtain an optimal sequence of layer thicknesses. The

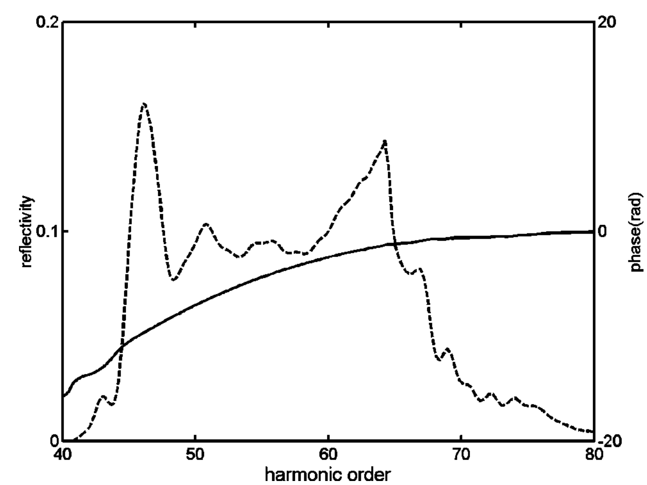

Fig. 3. Reflectivity (dashed curve) and phase (solid curve) at normal incidence for the chirped mirror represented in Fig. 2.

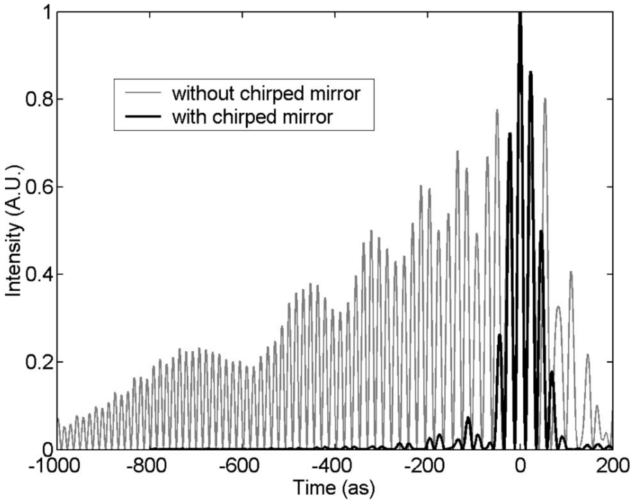

Fig. 4. Attosecond pulse before (gray line) and after compression by the chirped mirror (black line). Each pulse has been artificially centered in time for better comparison. The pulse repetition rate is 1.33 fs in both cases.

initial stack can be a quarter-wave mirror design, but this choice is not critical. The global convergence criterion is a linear combination of the reflectivity averaged over the target bandwidth and the achieved GDD compared with the target dispersion.

The example presented in this Letter is a mirror that corrects for the intrinsic harmonic chirp generated by an IR pump laser focused onto a neon gas cell. As long as the Ti:sapphire laser intensity remains below the suppression barrier intensity (8.2×10^{14} W cm^{-2} for neon), it is favorable to use the highest intensity to extend the plateau as much as possible and to produce intense harmonic emission. We then choose an IR intensity of 7.5×10^{14} W cm^{-2}. In this case the harmonics present a large intrinsic chirp (GDD of 6500 as^2). If we want to correct this chirp with a Sn filter, for example, we must use a thickness of 3800 nm, and the transmission must be 4×10^{-6}. The optimized multilayer alternates 60 layers of Mo and Si, and each layer is 2–9 nm thick. The structure is displayed in Fig. 2(b). Figure 3 shows the reflectivity (R) and the phase (ϕ_m) of this mirror, simulated by IMD software,[12] whose performance has already been demonstrated. The average reflectivity of the mirror is 11%, and the group-delay standard deviation (departure from the desired chirp compensation) is less than 10 as from harmonics H45 to H65. If we represent the amplitude of a particular harmonic before the mirror as $A(K) = A_0(K)\cos[\omega(K)t + \Phi(K)]$, we obtain after the mirror $A(K) = A_0(K)\sqrt{R(K)}\cos[\omega(K)t + \Phi(K) + \Phi_m(K)]$. The intensity of the final attosecond pulse is $I = |\Sigma_K A(K)|^2$ and is displayed in Fig. 4. The width of the pulse has been reduced from 260 to 90 as while a reasonable peak intensity has been maintained. Another important practical attribute of the chirp-compensating mirror is its ability to perform with pulses derived from a variety of pump intensities (with a pump intensity of 5×10^{14} W/cm^2, the mirror provides pulse compression from 450 to 170 as with a pump intensity of 10^{14} W/cm^2, a pulse compression from 120 to 90 as). It is also important to consider the feasibility of the chirped mirror. Because of technological constraints, the values of the uncertainties considered in this Letter have already been obtained with a similar aperiodic multilayer, deposited by ion-beam sputtering, with *in situ* control of the thickness of each layer by a quartz microbalance. Assuming a readily achievable layer thickness precision of ±0.2 nm, the reflectivity uncertainty is ±15% and the GDD uncertainty is ±3%. Furthermore, considering that, at each interface there is a region where the two materials overlap, which is simulated by introducing at each interface a roughness of 0.5 nm, the reflectivity is reduced by 20% but the phase is not affected. Under the constraints of these uncertainties, the output pulse width would not be significantly affected.

We have demonstrated compression of high harmonics below 100 as, which is required for optimizing the global system composed by the harmonics (chirp) and a GDD-compensating optic. By linking a simulation code of the HHs chirp and an optimization program for XUV chirped mirrors, we have shown that the designed mirror is capable of reducing the pulse from 260 to 90 as. Moreover, this mirror is efficient over a large range of pump laser intensities (one decade). Realization of such a mirror is feasible under current fabrication constraints. Chirped mirrors also have the advantage of being able to adapt to other wavelength ranges by changing the multilayer materials. The possibility of depositing a chirped mirror on a curved surface opens the way to new applications in attosecond metrology.

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