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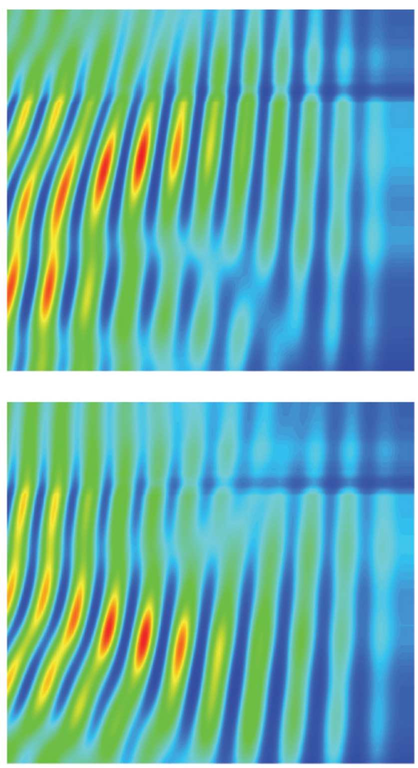

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# Attosecond Stretcher–Compressor Using Aperiodic Multilayer

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**Abstract:** The design of "stretcher–compressor" using aperiodic multilayers (aMLs) for attosecond pulses in the extreme ultraviolet (EUV) region is presented. We use a single-objective temporal optimization method based on a genetic algorithm (GA) to maximize pulse reflection efficiency for a single mirror. Employing this method, we studied the relationships of optimal number of layers, achievable energy reflectivity, and pulse duration. We then apply the findings to a design of an attosecond "stretcher–compressor" configuration with much improved overall throughput for a near-transform-limited attosecond pulse in a multimirror system.

**Index Terms:** Attosecond pulse, aperiodic multilayer (aML), pulse shaping, extreme ultraviolet (EUV).

## 1. Introduction

With rapid progress of attosecond science in recent years [1]–[5], the aperiodic multilayer (aML) in the extreme ultraviolet (EUV) and X-ray region attracts more and more interests because of its abilities of spectral and temporal control for attosecond pulses [6]–[9]. One important role that the aML can play is the dispersion compensator for chirped attosecond pulse, which can be used to achieve effective pulse compression and obtain nearly transform-limited attosecond pulse. This idea was first shown in dispersion control of femtosecond laser [10], and then successfully induced by Morlens *et al.* [11], [12] into attosecond pulse compression by compensating intrinsic chirp of high-order harmonic (HOH) generated by nonlinear interaction of femtosecond laser with rare gas [13], [14].

Efficient pulse compression can be achieved by aML with high total spectral reflectivity coupled with very wide bandwidth, spectral phase compensation, and amplitude reshaping. Traditionally, global searching tools need to be employed in a multilayer design process to find a proper aML structure. As a powerful global optimization method, genetic algorithm (GA) has been used as the optimization tool for aML design in previous papers [6], [15]. Generally, the linear combination with weights of attribute values of different objectives, like the spectral reflectivity and group delay dispersion (GDD), has to be designed in the merit function (MF) of the optimization methods. However, such multiobjective optimization could be problematic in GA because the final solution is usually sensitive to small change in weighting factors in the MF, which may lead to low efficiency [16].

Here, we introduce a single-objective optimization method for aML design where the MF is constructed in time domain. With it, we studied the relationship between compressed pulse characteristics and the number of layers used in optimization process and obtained results with clear physical explanations. We then apply the findings to a proposal of an attosecond stretcher–compressor configuration to greatly enhance the total throughput of a multimirror optical system where both the incoming and outgoing pulses are near transform limited.

## 2. Theoretical Model

Normally, a standard multilayer consists of a periodic structure of two alternative materials. This provides high reflectivity through resonance process, but the relative spectral bandwidth is reduced at the same time. Because of the limit of periodic condition, periodic multilayer has less flexibility of spectral control than aML, which consists of varying thickness for each layer [17]. Usually, the aML structure for specific purpose can be designed by global searching methods, like GA [6], simulated annealing algorithm [18], and needle optimization method [19]. In our paper, GA has been used for aML design, aiming to obtain high-efficiency attosecond pulse reflection and compression.

During the recursive optimization process in GA, an optimized aML structure can be found by maximized the MF. A valid and efficient MF is the key point of the whole optimization procedure. In this paper, we use a single-objective MF in time domain in the form of

$$\mathrm{MF} = \frac{R_{\mathrm{power}}}{\tau} \tag{1}$$

where $R_{\mathrm{power}}$ is the power reflection coefficient of multilayer reflection system, and $\tau$ is the duration of pulse [20], [21]

$$R_{\mathrm{power}} = R_I(\tau_0/\tau) = I_{\max}/I_{0\max} \tag{2}$$

$$\tau = \int_{-\infty}^{\infty} I(t)\, dt / I_{\max}. \tag{3}$$

In (2), $R_I$ describes the pulse reflection coefficient and can also be written as the reflected–incident pulse energy ratio

$$R_I = \int_{-\infty}^{\infty} I(t)\, dt \Bigg/ \int_{-\infty}^{\infty} I_0(t)\, dt. \tag{4}$$

Above, pulse parameters can be calculated from the intensity envelope of pulse $I(t)$, in which $\tau$ and $\tau_0$ describe the pulse durations for reflected and incident pulse, and $I_{\max}$ and $I_{0\max}$ denote the peak intensity of reflected and incident pulse, respectively.

The underlining calculation of the pulse response of aML is similar to other standard numerical approaches [6], [21]. Simply put, the incident pulse is Fourier decomposed to frequency components. Each component is then modified by a complex amplitude reflectivity $r(\omega)$ of the multilayer. An inverse Fourier transform then generates the reflected pulse in time domain. For any aperiodic structure, $r(\omega)$ can be computed by the method of transfer matrix method (TMM) if the optical constants of the materials used in multilayer and their thicknesses are known. In our paper, Mo/Si pair is chosen because of its high reflectivity and stable physical properties in the EUV region, in accord with the spectral area of attosecond pulse used in our simulation. The optical constants of Mo and Si are available from the Center for X-Ray Optics at Lawrence Berkeley National Laboratory [22].

## 3. Single-Mirror Optimization

To study the pulse compression characteristics of a single aML mirror, a series of chirped incident pulses with Gaussian intensity distribution are modeled in our simulation. The pulses are all centered at 80 eV with 30-eV full-width at half-maximum (FWHM). The chirp is assumed to be linear, with amplitudes of 5 as/eV, 10 as/eV, and 15 as/eV. They were chosen to match the intrinsic chirp in the broadband plateau region of high-order harmonics generation (HHG) under commonly used laser intensities [11], [15]. The situation of incident pulse without chirp (0 as/eV) is also

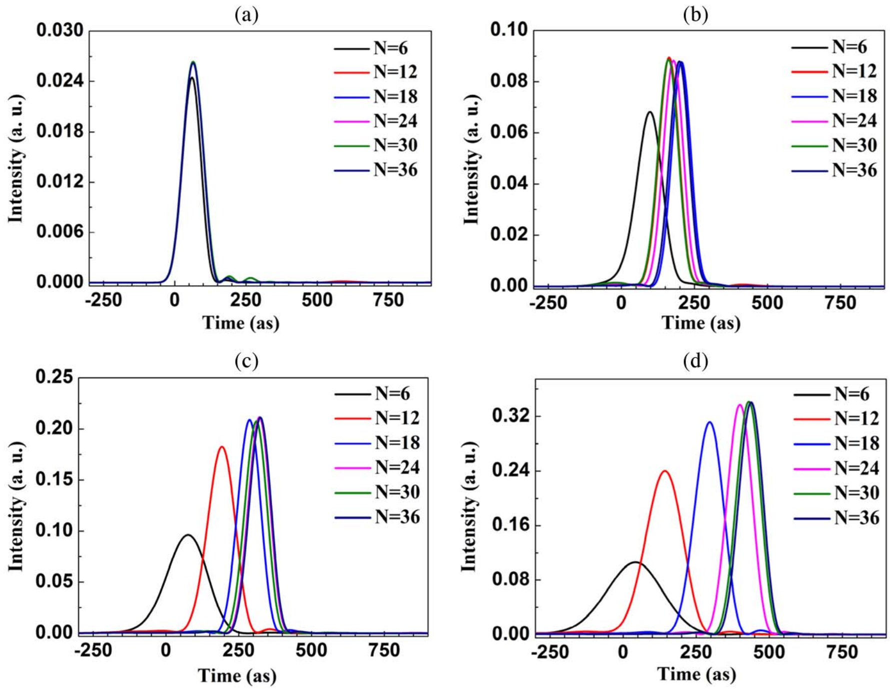


Fig. 1. Pulse responses of aML with different layers for four incident pulses with (a) 0 as/eV, (b) 5 as/eV, (c) 10 as/eV, and (d) 15 as/eV chirp, respectively.

considered, as a reference for other chirped pulses. We note that Gaussian-shaped pulse is not a good approximation for a real attosecond source. However, this simplifies the calculation, and, as can be seen later, our main conclusion of this section does not depend on this assumption. All simulations in this paper assumed normal incidence.

As described above, to determine the optimal distribution of layer thickness for a chirped multilayer used for reflecting and compressing attosecond pulse, we use GA to search for optimized aML structure by maximizing the MF defined by (1). During our search, a series of layer number $N$ has been used in aML design. In this paper, $N = 6, 12, 18, 24, 30, 36$ are considered for each incident pulse. The search region of thickness for every layer in aML is from 1 nm to 10 nm. After enough generations, GA will pick out the optimized aML structures with maximum MF.

In Fig. 1, responses of final optimized aMLs with different number of layers are plotted for the four chirped incident pulses. We can see that the four incident pulses are all reflected and compressed nicely by optimized aMLs picked out by GA, which demonstrates the validity of our MF. However, different incident pulse needs different number of layers to achieve maximum compression and optimized reflection. In Fig. 1, it seems that more layers are wanted for compressing broader incident pulses for chirped incident pulses. In our simulation, the aML with 30 layers is enough for full compression and reflection of all four incident pulses.

In Fig. 2, pulse duration (measured in FWHM) and the power reflection coefficient $R_{\text{power}}$ of all reflected pulses showed in Fig. 1 are compared. From Fig. 2(a), we can see that all FWHM values of reflected pulses are under 100 as after reflected by optimized aML with enough layers. For different chirped incident pulses, the number of layers needed for maximum pulse compression is different. For example, only 12 layers are needed for maximum pulse compression for the incident pulse with 5-as/eV chirp, while 30 layers are needed for the incident pulse with 15-as/eV chirp. This means the incident pulse with larger chirp (or broader pulsewidth) needs more layers to achieve maximum pulse compression and reflection.

In Fig. 2(b), the power reflection coefficients $R_{\text{power}}$ of final optimized aML with different layers are plotted. It is interesting to see that all four curves will all go to saturation after certain number of

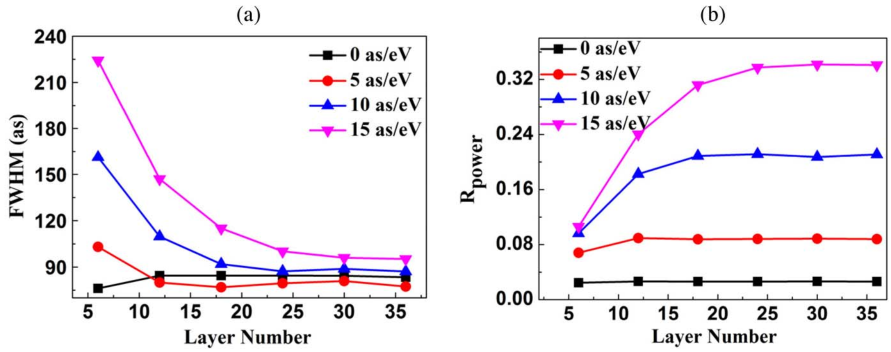


Fig. 2. (a) Pulse duration (in FWHM) and (b) $R_{\text{power}}$ of all reflected pulses showed in Fig. 1.

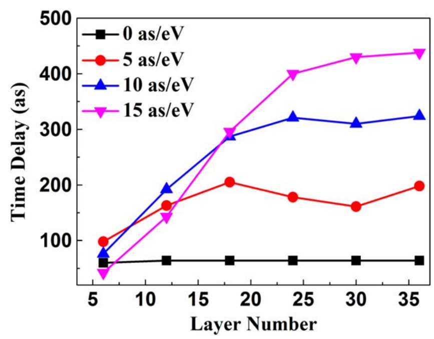


Fig. 3. Time delay of pulse peak under different chirped incident pulses versus the optimized number of layers $N$ in optimized aML.

layers. In other words, the value of $R_{\text{power}}$ for a fixed incident pulse will remain unchanged when the maximum $R_{\text{power}}$ is obtained.

Such behavior is not surprising. A multilayer provides high reflectivity through constructive interference between the reflections from multiple interfaces. In the cases of attosecond pulses, the spatial spread of the pulse is quite limited, only 30 nm in space for 100-as pulse. This should limit the number of layers that can be used in a multilayer. For the same reason, a chirped pulse, with its longer pulse duration, could use more layers and thus enjoy higher total reflectivity. We can see more evidence of this in Fig. 3, where the time delay for each optimized aML is plotted. It is clear that the pulse with larger chirp takes more time to accomplish the whole reflection process and penetrate deeper in multilayer structure when the complete pulse compression achieves.

## 4. Attosecond Stretcher–Compressor

From the last section, we concluded that a pulse with larger chirp (or longer pulse duration) could use more layers of multilayer to reach higher reflection efficiency. We note that this conclusion should hold for either the incoming or reflected pulse, i.e., if the reflected pulse is allowed to have larger chirp (or pulse duration), then more layers can also be used and more pulse energy be reflected. This leads us to a proposal of an attosecond stretcher–compressor configuration with improved total throughput.

In attosecond experiments, the preservation of the short pulse duration is usually the paramount requirement, while, at the same time, higher optics efficiency is greatly desired due to the extremely limited number of photons available from source. Imagine a double-mirror reflection system, for example in an interferometer [23], to be used with an attosecond light source. The overall throughput of the system would be quite low if each mirror was designed to preserve the short pulse

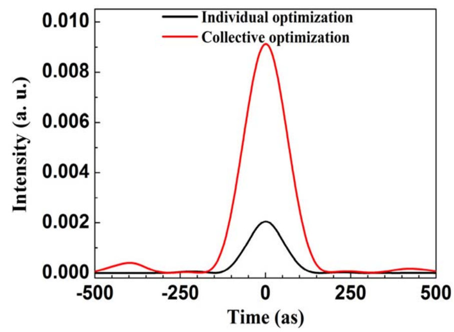


Fig. 4. Final reflected pulses obtained by "individual optimization" method and "collective optimization" method.

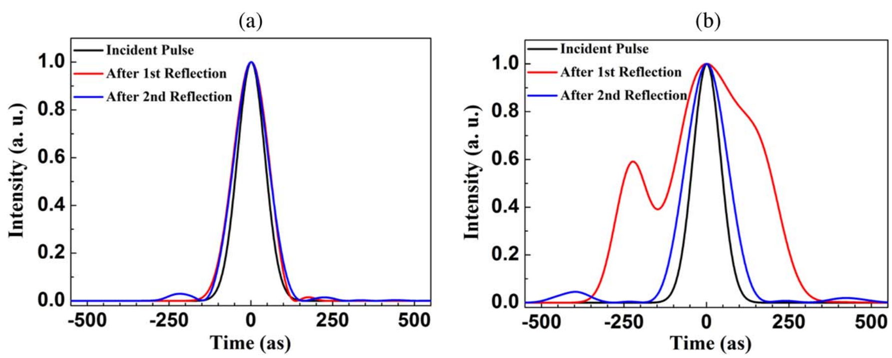


Fig. 5. Incident pulse, the pulse after the first mirror reflection, and the pulse after the second mirror reflection under (a) the "individual optimization" method and (b) the "collective optimization" method, respectively.

duration, since each mirror will have to use few layers, thus having low reflectivity. However, if our theory is correct, we can use the first multilayer mirror to broaden the narrow incident pulse by importing additional dispersion and then use the second one to compress it by compensating the dispersion introduced by the first reflection mirror. Since we could deploy a multilayer with more layers in both steps, this would provide great potential of achieving higher overall pulse reflection efficiency. We note that, although the attosecond pulse is stretched and recompressed, there is no amplification process involved, unlike the case for the well-known femtosecond pulse stretcher–compressor configuration. Rather, the analogy is that a stretched attosecond pulse can reserve more energy by utilizing more reflection interfaces in space, while a stretched femtosecond pulse can gain more energy by utilizing more amplification in time.

To verify this idea, two optimization methods for double-mirror system reflection have been tested by our GA, respectively. In the first optimization method, we optimize the two multilayer structures one by one, which we call "individual optimization." Both multilayer mirrors should guarantee small pulse duration for each reflection. On the other hand, in the second optimization method, we optimize the two reflection mirrors together, and only limit the final reflected pulse. We call it "collective optimization." In the simulation, we used a chirp-free Gaussian pulse with 100-as FWHM as the incident pulse. The center photon energy of this incident pulse has been set to 93 eV.

After enough generations, GA gives out two optimized multilayer structures for each optimization method, respectively. In Fig. 4, the final reflected pulses for two different optimization methods are compared. The "collective optimization" method showed a $>$ 4x higher throughput than "individual optimization" (0.91% versus 0.21%), while keeping the final pulse duration almost identically small (148 as versus 123 as).

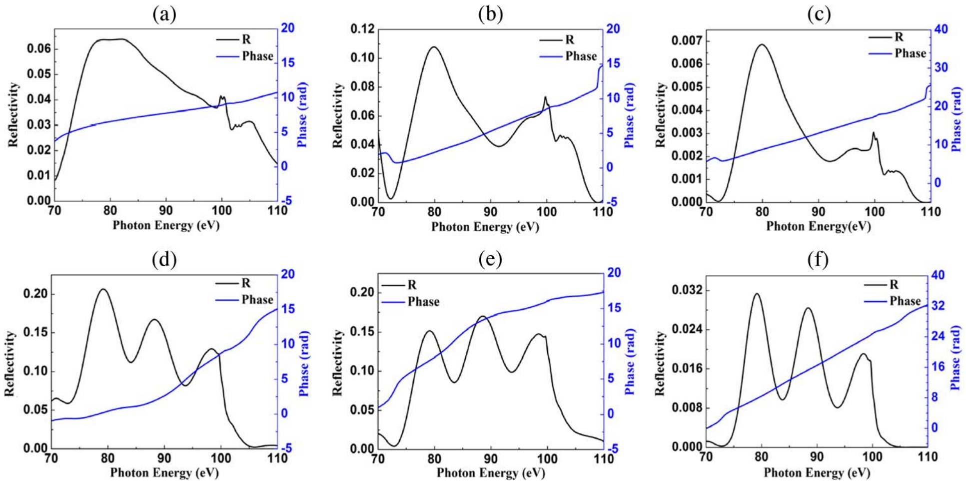


Fig. 6. Reflectivity and phase characteristics of (a) the first mirror, (b) the second mirror, and (c) two mirrors together under the "individual optimization" method and the reflectivity and phase characteristics of (d) the first mirror, (e) the second mirror, and (f) two mirrors together under the "collective optimization" method.

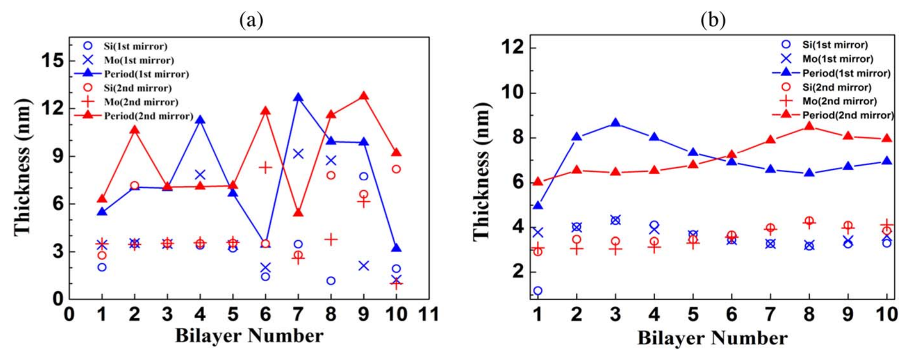


Fig. 7. Thickness distributions of two multilayer mirrors in double-mirror reflection system designed by (a) individual optimization method and (b) collective optimization method, respectively.

In Fig. 5, the incident pulse, the pulse after the first mirror reflection, and the pulse after the second mirror reflection under both two optimization methods are shown, respectively. For easy comparison, all pulses have been normalized to their own maximum intensity, and all pulse peaks are set to zero time. It is clear that the pulse durations were kept at almost the same value during the whole double-mirror reflection process in "individual optimization," while the pulse has been broadened after reflected by the first multilayer mirror, and then, compressed after reflected by the second mirror in "collective optimization." This proved our "stretcher–compressor" proposal indeed worked.

To show more details, all reflectivity and phase characteristics of mirrors under the two optimization methods are given in Fig. 6. For "individual optimization" method, nearly linear phase is shown near the center photon energy of incident pulse (93 eV) for both mirrors, to avoid pulse broadening caused by dispersion of reflection mirrors. However, because of the limited pulsewidth, the reflectivity of both mirrors is relatively low across the spectrum region of incident pulse, due to the less contribution layers. For "collective optimization" method, the first mirror and the second mirror show a positive and a negative dispersion, respectively, together with relative higher reflectivity. Using this pair of positive and negative dispersion, the incident pulse can be broadened first and then compressed. During such broadening and compressing process, the chirped mirrors provide more layers manipulating the pulse, which contribute to higher pulse reflection efficiency. From Fig. 6(f), we can see that the linear phase is also shown after reflection by two chirped mirrors, which exhibits great dispersion compensation between these two chirped mirrors. The thickness distributions of the two mirrors under both optimizations are shown in Fig. 7. It can be seen that in "collective optimization" method [see Fig. 7(b)], there are two clear trends of the multilayer's periods to decrease or increase in the two mirrors, which generated the opposite chirps, while there is no such trend in both mirrors under "individual optimization" method [see Fig. 7(a)].

## 5. Conclusion

In this paper, a time-domain optimization method for aML structures is presented. We found a chirped pulse could effectively use more layers to reach higher energy reflectivity. We proposed an attosecond "stretcher–compressor" configuration, where we intentionally introduced a chirp and subsequently compensated it, achieving 4x higher overall system throughput.

## References

[1] T. Popmintchev, M. Chen, P. Arpin, M. M. Murnane, and H. C. Kapteyn, "The attosecond nonlinear optics of bright coherent X-ray generation," *Nature Photon.*, vol. 4, no. 12, pp. 822–832, Dec. 2010.
[2] F. Krausz and M. Ivanov, "Attosecond physics," *Rev. Mod. Phys.*, vol. 81, no. 1, pp. 163–234, Feb. 2009.
[3] P. B. Corkum and F. Krausz, "Attosecond science," *Nature Phys.*, vol. 3, no. 6, pp. 381–387, Jun. 2007.
[4] H. C. Kapteyn, O. Cohen, I. Christov, and M. Murnane, "Harnessing attosecond science in the quest for coherent X-rays," *Science*, vol. 317, no. 5839, pp. 775–778, Aug. 2007.
[5] U. Keller, "Femtosecond to attosecond optics," *IEEE Photon. J.*, vol. 2, no. 2, pp. 225–228, Apr. 2010.
[6] A. Wonisch, U. Neuhäusler, N. M. Kabachnik, T. Uphues, M. Uiberacker, V. Yakovlev, F. Krausz, M. Drescher, U. Kleineberg, and U. Heinzmann, "Design, fabrication, and analysis of chirped multilayer mirrors for reflection of extreme-ultraviolet attosecond pulses," *Appl. Opt.*, vol. 45, no. 17, pp. 4147–4156, Jun. 2006.
[7] M. Suman, F. Frassetto, P. Nicolosi, and M.-G. Pelizzo, "Design of aperiodic multilayer structures for attosecond pulses in the extreme ultraviolet," *Appl. Opt.*, vol. 46, no. 33, pp. 8159–8169, Nov. 2007.
[8] M. Hofstetter, M. Schultze, M. Fiess, B. Dennhardt, A. Guggenmos, J. Gagnon, V. S. Yakovlev, E. Goulielmakis, R. Kienberger, E. M. Gullikson, F. Krausz, and U. Kleineberg, "Attosecond dispersion control by extreme ultraviolet multilayer mirrors," *Opt. Exp.*, vol. 19, no. 3, pp. 1767–1776, Jan. 2011.
[9] C. Bourassin-Bouchet, Z. Diveki, S. de Rossi, E. English, E. Meltchakov, O. Gobert, D. Guénot, B. Carré, F. Delmotte, P. Salieres, and T. Ruchon, "Control of the attosecond synchronization of XUV radiation with phase-optimized mirrors," *Opt. Exp.*, vol. 19, no. 4, pp. 3809–3817, Feb. 2011.
[10] R. Szipocs, K. Ferencz, C. Spielmann, and F. Krausz, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," *Opt. Lett.*, vol. 19, no. 3, pp. 201–203, Feb. 1994.
[11] A. S. Morlens, P. Balcou, P. Zeitoun, C. Valentin, V. Laude, and S. Kazamias, "Compression of attosecond harmonic pulses by extreme-ultraviolet chirped mirrors," *Opt. Lett.*, vol. 30, no. 12, pp. 1554–1556, Jun. 2005.
[12] A. S. Morlens, R. Lopez-Martens, O. Boyko, P. Zeitoun, P. Balcou, K. Varjú, E. Gustafsson, T. Remetter, A. LHuillier, S. Kazamias, J. Gautier, F. Delmotte, and M.-F. Ravet, "Design and characterization of extreme-ultraviolet broadband mirrors for attosecond science," *Opt. Lett.*, vol. 31, no. 10, pp. 1558–1560, May 2006.
[13] S. Kazamias and P. Balcou, "Intrinsic chirp of attosecond pulses: Single-atom model versus experiment," *Phys. Rev. A*, vol. 69, no. 6, pp. 063416-1–063416-6, Jun. 2004.
[14] Y. Mairesse, A. de Bohan, L. J. Franzinski, H. Merdji, P. Monchicourt, P. Breger, M. Kovacev, B. Carré, H. G. Muller, P. Agostini, and P. Salières, "Attosecond synchronization of high-harmonic soft X-rays," *Science*, vol. 302, no. 5650, pp. 1540–1543, Nov. 2003.
[15] R. Chen, F. Wang, and Z. Wang, "Design of chirped Mo/Si multilayer mirror in the extreme ultraviolet region," *Chin. Opt. Lett.*, vol. 6, no. 4, pp. 310–312, Apr. 2008.
[16] J. Horn, N. Nafpliotis, and D. E. Goldberg, "A niched Pareto genetic algorithm for multiobjective optimization," in *Proc. 1st IEEE Conf. Evol. Comput.*, Jun. 1994, vol. 1, pp. 82–87.
[17] N. N. Kohlachevskii, A. S. Pirozhkov, and E. N. Ragozin, "Broadband X-ray optical elements based on aperiodic multilayer structures," *Quantum Electron.*, vol. 30, no. 5, pp. 428–434, May 2000.
[18] X. Cheng, Z. S. Wang, Z. Zhang, F. Wang, and L. Chen, "Design of X-ray super-mirrors using simulated annealing algorithm," *Opt. Commun.*, vol. 265, no. 1, pp. 197–206, Sep. 2006.
[19] M. Singh and J. J. M. Braat, "Design of multilayer extreme-ultraviolet mirrors for enhanced reflectivity," *Appl. Opt.*, vol. 39, no. 13, pp. 2189–2197, May 2000.
[20] I. L. Beigman, A. S. Pirozhkov, and E. N. Ragozin, "Reflection of atto- and femtosecond X-ray pulses from aperiodic multilayer mirror," *JETP Lett.*, vol. 74, no. 3, pp. 149–153, Aug. 2001.
[21] I. L. Beigman, A. S. Pirozhkov, and E. N. Ragozin, "Reflection of few-cycle X-ray pulses by aperiodic multilayer structures," *J. Opt. A, Pure Appl. Opt.*, vol. 4, no. 4, pp. 433–439, Jul. 2002.
[22] [Online]. Available: http://henke.lbl.gov/optical_constants/getdb2.html
[23] E. Goulielmakis, G. Nersisyan, N. A. Papadogiannis, D. Charalambidis, G. Tsakirislink, and K. J. Witte, "A dispersionless michelson interferometer for the characterization of attosecond pulses," *Appl. Phys. B, Laser Opt.*, vol. 74, no. 3, pp. 197–206, 2002.