Improving attosecond pulse reflection by large angle incidence for a periodic multilayer mirror in the extreme ultraviolet region

Lin Cheng-You(林承友), Chen Shu-Jing(陈淑静), and Liu Da-He(刘大禾)†

Applied Optics Beijing Area Major Laboratory, Department of Physics, Beijing Normal University, Beijing 100875, *China*

(Received 29 August 2012; revised manuscript received 26 September 2012)

The improvement of attosecond pulse reflection by large angle incidence for a periodic multilayer mirror in the extreme ultraviolet region has been discussed. Numerical simulations of both spectral and temporal reflection characteristics of periodic multilayer mirrors under various incident angles have been analyzed and compared. It was found that the periodic multilayer mirror under a larger incidence angle can provide not only higher integrated reflectivity but also a broader reflection band with negligible dispersion, making it possible to obtain better a reflected pulse that has a higher pulse reflection efficiency and shorter pulse duration for attosecond pulse reflection. In addition, by increasing the incident angle, the promotion of attosecond pulse reflection capability has been proven for periodic multilayer mirrors with arbitrary layers.

Keywords: attosecond pulse, periodic multilayer, large angle incidence

PACS: 42.65.Re, 41.50.+h, 42.30.Kq **DOI:** 10.1088/1674-1056/22/1/014210

1. Introduction

In recent years, with the high-speed development of ultrafast optics, interest has been growing in manipulating attosecond X-ray pulses, typically generated by high order harmonics (HOH) of rare gases.[1–7] Unlike pulses in the spectral range from the near ultraviolet (UV) to the near infrared (IR), the spectrum of HOH is mainly located at the extreme ultraviolet (EUV) or soft X-ray (SXR) region, in which most of the materials are nearly transparent and strongly absorbent. Thus, conventional reflection mirrors, usually consisting of single metallic layer (like Au), cannot achieve a high reflection for an attosecond pulse generated from HOH according to the Fresnel formula.

As a reliable reflector for attosecond pulse in the extreme ultraviolet region, the multilayer mirror consisting of two alternative metallic layers has typically been widely used in reflecting and shaping both a single attosecond pulse[8,9] and an attosecond pulse train,[10] because of its high reflectivity in the EUV and SXR region forming by coherent superposition of reflection light for each layer. However, little research about pulse reflection characteristics of EUV multilayer mirrors had been conducted until Morlens *et al.* proposed the idea of compressing a chirped attosecond pulse by aperiodic multilayer mirrors (or chirped mirrors), which exhibit the powerful capability of controlling the spectrum and phase for an attosecond pulse.[11,12] After that, studies of aperiodic multilayer mirrors applied to ultrafast optics have come out quickly both in theory[13,14] and from experiments.[15–17]

Unlike the aperiodic multilayer, the periodic multilayer cannot achieve flexible phase and spectrum control for incident attosecond pulse because of its limited independent variables of multilayer structure, like bilayer number, period thickness, and thickness ratio. However, periodic EUV multilayer mirrors still showed wide applications in attosecond science, especially in single attosecond pulse generation by filtering and reflecting the cut-off region of the HOH.[10,18] Due to their high reflectivity, large bandwidth, and negligible dispersion in the Bragg reflection band,[19] which all benefit from the reflecting single attosecond pulse, especially the one without a chirp. Furthermore, the simple structure of the periodic multilayer mirror also makes it easy not only for optimization of design but also for realistic manufacture.

In general, normal incidence is often preferred for attosecond pulse reflection, due to its small spatial distortion.[8,18] However, according to our previous research,[20] the bandwidth of the Bragg reflection band, which plays a most important role in determining the duration of the reflected attosecond pulse, strongly depends on the bilayer number of the periodic multilayer under normal incidence. Therefore, if we want to reflect an attosecond pulse with a short duration in a nearly transform-limited way, using the multilayer mirror with few layers seems to be the only solution. However, it will lead to a low pulse reflection efficiency. In other words, there is a tradeoff between pulse duration and pulse reflection efficiency for the periodic multilayer mirror under normal incidence.

A new idea based on a large angle incidence has been proposed in this paper, which can guarantee not only nearly transform-limited pulse reflection, but also high pulse reflection efficiency at the same time for periodic multilayer mirrors. According to our analysis, with the increase of incident angle, pulse duration and pulse reflection efficiency of the re-

†Corresponding author. E-mail: dhliu@bnu.edu.cn

© 2013 Chinese Physical Society and IOP Publishing Ltd

flected attosecond pulse will be improved impressively when using periodic multilayer mirrors to reflect a single attosecond pulse.

2. Numerical approach

In order to simulate the process of reflecting an attosecond pulse by periodic EUV multilayer mirrors, a common calculation method, discussed in detail by Beigman *et al.*,[21,22] has been used in our paper. This method needs to calculate the complex amplitude reflectivity $r(\omega)$ based on the transfer-matrix method (TMM) at first, and then, to obtain the Fourier components of reflected pulse $E(\omega)$ by multiplying $r(\omega)$ with each Fourier component $E_0(\omega)$ of the incident pulse, which can be obtained from the incident pulse field $E_0(t)$ by a Fourier transformation. Finally, inverse Fourier transforms with the object of $E(\omega)$ to obtain the reflected pulse field $E(t)$ and intensity $I(t)$ ($I(t) = E(t)E^*(t)$). The whole calculation procedure can be expressed simply as follows:

$$E_0(\omega) = \int_{-\infty}^{\infty} E_0(t)\exp(-\mathrm{i}\omega t)\,\mathrm{d}t, \tag{1}$$

$$E(\omega) = E_0(\omega)r(\omega), \tag{2}$$

$$E(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} E(\omega)\exp(\mathrm{i}\omega t)\,\mathrm{d}t. \tag{3}$$

In addition, on behalf of describing the features of the attosecond pulse quantitatively, three temporal parameters are induced. The first one is the pulse duration τ, which describes the effective width of intensity envelope $I(t)$ of the pulse:

$$\tau = \int_{-\infty}^{\infty} I(t)\,\mathrm{d}t / I_{\max}. \tag{4}$$

The second one is the pulse reflection coefficient R_I, which is defined as the reflected-to-incident pulse energy ratio

$$R_\mathrm{I} = \int_{-\infty}^{\infty} I(t)\,\mathrm{d}t \Big/ \int_{-\infty}^{\infty} I_0(t)\,\mathrm{d}t. \tag{5}$$

Lastly, the power reflection coefficient R_p is defined as

$$R_\mathrm{p} = R_\mathrm{I}(\tau_0/\tau) = I_{\max}/I_{0\max}, \tag{6}$$

which will be used to evaluate the ability of the reflecting pulse for multilayer mirrors, and τ_0 and $I_{0\max}$ denote the duration and peak intensity of the incident pulse, respectively.

In Fig. 1, the geometry of the periodic multilayer mirror for an attosecond pulse reflection has been given. In order to study how the incident angle θ affects the attosecond pulse reflection in a periodic multilayer reflection system. An unchirped Gaussian pulse with 250 as the duration ($\tau_0 = 250$ as) centered at 93 eV has been used as the incident pulse in our simulation, according to Kienberger *et al.*'s research.[8] In addition, the famous periodic Mo/Si multilayer mirror has been chosen for attosecond pulse reflection, because of its great reflection performance in the EUV region over the years. In Fig. 1, d_1 and d_2 are the thickness of the Si layer and Mo layer, respectively, in periodic multilayer structure, and N denotes the bilayer number of periodic multilayers. The optical constants of air, silicon, and molybdenum ($\tilde{n}_0$, $\tilde{n}_1$, and $\tilde{n}_2$), which are necessary for calculating the complex amplitude reflectivity $r(\omega)$ of periodic Mo/Si multilayer mirrors, come from Henke *et al.*[23] Then, we can study the characteristics of attosecond pulse reflection for periodic multilayer mirrors under different incident angles θ, by comparing the temporal parameters (τ, R_I, and R_P) of each reflected attosecond pulse. To make our simulation more realistic, the interdiffusion effect between the Mo layer and the Si layer in a multilayer structure has also been considered in our simulation by using Aquila *et al.*'s model.[24] which has been verified effectively by the realistic design of the Mo/Si multilayer mirror.

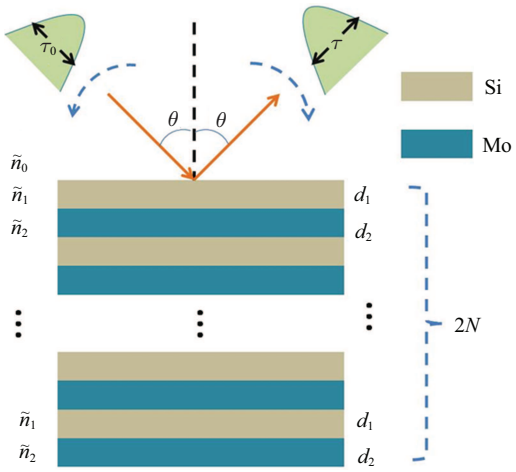


Fig. 1. (color online) The geometry of the periodic Mo/Si multilayer mirror for attosecond pulse reflection.

3. Results and discussion

3.1. Spectral response of periodic Mo/Si multilayer mirrors

To study how a large angle incidence can affect attosecond pulse reflection in a periodic multilayer reflection system, reflective characteristics of periodic Mo/Si multilayer mirrors under various incident angles have been studied. In our simulation, incident angles of 0° (normal incidence), 15°, 30°, 45°, 60°, and 75° have been considered. Besides, the situation of total external reflection in the EUV region has also been considered for an attosecond pulse reflection, due to its extremely high reflectivity across the broad reflection band. Also, because the total external reflection only happens at the surface of the interface, a 100-nm Si layer is used for reflecting the attosecond pulse in a total external reflection way.

To achieve total external reflection, the incident angle should be larger than the critical angles θ_c of the single Si

layer, which can be calculated by:

$$\theta_c = \sqrt{2(1-n)}, \tag{7}$$

where n is the real part of a complex index of refraction for Si.[25] For EUV radiation near 93 eV, θ_c is about 89°. Thus, an incident angle of 89.7°, which has been used for attosecond X-ray free electron laser pulse reflection,[26] is designed for total external reflection, although such a large angle incidence can cause serious wavefront aberration of the focused pulse which is essentially a temporal broadening due to the spatial inhomogeneity.[27]

Furthermore, a series of periodic Mo/Si multilayer mirrors with different thickness of period d are designed, to make sure their Bragg reflection peaks all center at 93 eV under different incident angles, according to the Bragg law

$$2d\cos\theta = m\lambda, \tag{8}$$

where d, θ, m, and λ represent a period of multilayer, incident angle of beam, order of Bragg reflection, and wavelength of incident light, respectively. Aiming to obtain maximum pulse reflection efficiency, the highest reflectivity should be achieved at the center energy of incident pulse for each periodic multilayer. Thus, the bilayer number N of each periodic multilayer should be designed properly to guarantee its maximum peak reflection located at 93 eV. In Fig. 2, the dependence of peak reflectivity R_{peak} of a periodic Mo/Si multilayer on bilayer number N has been shown for each incident angle θ.

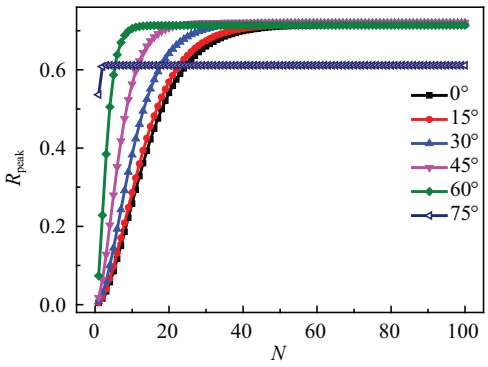


Fig. 2. (color online) The peak reflectivity R_{peak} of the periodic multilayer mirrors versus the bilayer number N for each incident angles θ (from 0° to 75°).

From Fig. 2, we can see clearly that each R_{peak} curve rises with N at first, and then, achieves saturation after a certain bilayer number N_{eff}, which denotes the effective number of bilayers contributing to the reflectivity in the multilayer. In addition, the periodic multilayer under a larger incident angle needs a smaller N_{eff} to achieve maximum peak reflectivity R_{peak}. In particular, for $\theta = 0°$, 15°, 30°, 45°, 60°, and 75°, $N = 50$, 50, 40, 30, 15, and 3 are needed to obtain a maximum R_{peak}, respectively, which will be used in our simulation.

In Fig. 3, the reflectivity curves of periodic multilayer mirrors with enough layers have been exhibited in the frequency domain for each incident angle (solid line). Furthermore, the reflectivity curve of the total external reflection achieved by a 100-nm Si layer has also been plotted (the dashed line). The dotted line in Fig. 3 denotes the spectrum of incident attosecond pulse. The structure parameters of these periodic Mo/Si multilayer structures are listed in Table 1, together with the center photon energy E_c, the full wave at half maximum ΔE, and the peak reflectivity R_{peak} of the reflection band, which have been used to describe the reflective characteristics of periodic multilayer mirrors.

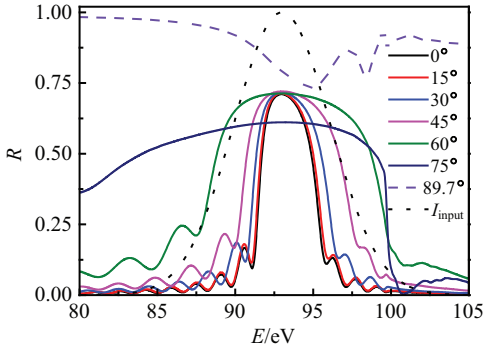


Fig. 3. (color online) Reflectivity R of Mo/Si periodic multilayer mirrors versus incident photon energy E under different incident angles θ (from 0° to 89.7°). Dotted line of I_{input} denotes the spectral distribution of an incident pulse.

Table 1. The structural and spectral parameters of periodic Mo/Si multilayer mirrors under different incident angles.

θ/(°)	d/nm	N	E_c/eV	ΔE/eV	R_{peak}
0	7.41	50	93	3.70	71.07%
15	7.66	50	93	3.80	71.37%
30	8.51	40	93	4.78	71.66%
45	10.40	30	93	6.69	72.02%
60	15.03	15	93	11.02	71.32%
75	37.9	3	93	25	61.13%
89.7	100	1 (Si)	93	> 30	79.17%

From Table 1, we can see that all peaks of reflectivity curves are shown at 93 eV except the one under total external reflection. Besides, the broadening of the reflection band is obvious (from 3.7 to 25 eV) with the increase of incident angles (from 0° to 75°). Moreover, for different incident angles, all maximum R_{peak} have nearly the same value (around 71%) except the one under 75° incidence (about 61%), probably due to the competition between Bragg reflection and total external reflection in the periodic multilayer structure, which can be verified by its irregular reflection band shown in Fig. 3.

In addition, when the incident angle reaches 60°, the bandwidth of the reflection curve becomes larger than the one of the incident pulse, which makes it possible to obtain a transform-limited reflected pulse. This broadening effect mainly comes from less effective bilayer number N_{eff} of periodic multilayer under a larger incident angle, due to the inversely proportional relationship between the bandwidth of the

Bragg reflection band and the effective bilayer number $N_{\rm eff}$of periodic multilayer mirrors.[28] On the other hand, a single Si layer under an incident angle of 89.7° also showed an extremely broad reflection band and high reflectivity across the spectral range of the incident pulse, due to the total external reflection effect.

However, only broadband reflection cannot guarantee a short pulse duration for the reflected attosecond pulse. The effect of dispersion caused by reflective mirrors, which results from the nonlinear relationship between the reflection phase of the multilayer and the angular frequency of the incident light, can also produce serious pulse broadening for the incident pulse.[21,22] The dispersion of the multilayer, also called the group delay dispersion (GDD), can be written as[29]

$$\mathrm{GDD} = \frac{\partial^2 \phi}{\partial \omega^2}, \tag{9}$$

where ω is the angular frequency of the incident light, and φ is the reflective phase of multilayer, which can be easily obtained by calculating the argument of complex amplitude reflectivity $r(\omega)$

$$\phi = \arctan\left(\frac{\mathrm{Im}(r(\omega))}{\mathrm{Real}(r(\omega))}\right). \tag{10}$$

For comparing dispersion characteristics of periodic multilayer mirrors under different incident angles, the GDD of periodic Mo/Si multilayer mirrors under 0° and 60° incidence, together with a single Si layer under 89.7° incidence, have been plotted in Fig. 4.

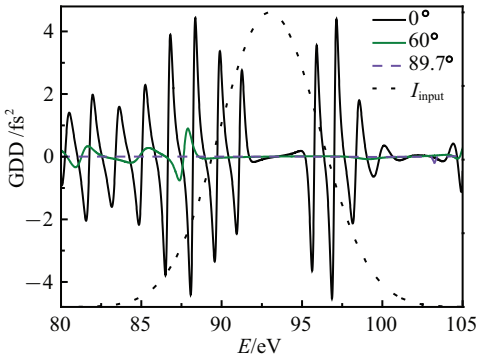


Fig. 4. (color online) Group delay dispersion GDD of periodic Mo/Si multilayer mirrors under 0° and 60° incidence together with group delay dispersion of a single Si layer under 89.7° incidence versus incident photon energy E. The dotted line of $I_{\rm input}$ denotes the spectral distribution of the incident pulse.

In Fig. 4, improvement of dispersion has been exhibited clearly with increasing incident angle. Although three GDD curves possess nearly zero dispersion at their reflection band, GDD of the reflection mirror under 60° incidence and single Si layer under 89.7° incidence show a much wider zero-dispersion range than the one under 0° incidence (normal incidence) in the spectral region of incident attosecond pulse (dotted line), due to its broader Bragg reflection band. Moreover, stronger dispersion oscillation at the band sides of the reflection mirror under 0° incidence, contributed by more effective reflection layers $N_{\rm eff}$, will lead to more serious pulse broadening for the incident attosecond pulse than other situations.

3.2. Temporal responses of periodic Mo/Si multilayer mirrors

Due to our previous discussion, the periodic multilayer mirror under a larger incident angle can afford not only a wider reflection band with high peak reflectivity, but also better dispersion features in the Bragg reflection band, providing the potential of obtaining a high-efficiency reflected pulse without obvious broadening. By using the simulation method described in Section 2, the temporal responses of periodic multilayer structures have been studied for different incident angles.

In Fig. 5, reflected pulses from periodic multilayer mirrors under different incident angles have been calculated for the incident Gaussian pulse with 250 as duration. The situation of total external reflection has also been considered in this simulation (the dashed line). All periodic multilayer structures are the same as the ones showed in Fig. 3. It is clear to see that, with the increase of the incident angle, the peak intensity of the reflected pulse becomes higher, and the duration becomes shorter, showing an obvious improvement in attosecond pulse reflection.

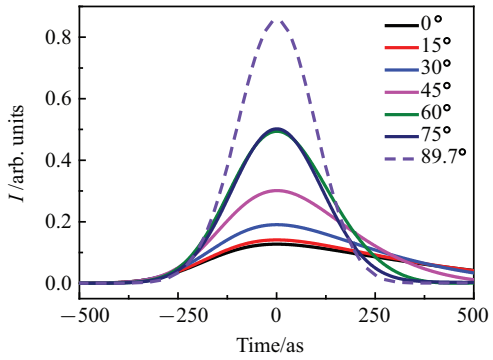


Fig. 5. (color online) The intensity envelope I of reflected pulses under different incident angles θ (from 0° to 89.7°) in time domain.

By using Eqs. (4)–(6), the temporal parameters of reflected pulses can be calculated, and have been listed in Table 2.

Table 2. The temporal parameters of the reflected pulse for periodic multilayer mirrors under different incident angles.

θ/(°)	τ/as	$R_{\rm I}$	$R_{\rm p}$
0	621	31.62%	12.71%
15	590	33.28%	14.09%
30	510	38.86%	19.05%
45	410	48.92%	30.13%
60	314	62.10%	49.42%
75	290	58.15%	50.2%
89.7	245	83.88%	85.56%

In Table 2, when the incident angle increases from 0° to 89.7°, the duration τ of the pulse response reduces from 621

to 245 as (transform-limited pulse). Furthermore, the power reflection coefficient R_p, which is used to evaluate the ability of the reflecting pulse for multilayer mirrors, also shows impressive development with the increasing incident angle (from 12.71% to 85.56%), proving that the periodic multilayer under a larger incident angle owns a better pulse reflection capability. The singular point of R_I at 75° results from the competition of Bragg reflection and total external reflection in the multilayer, which leads to the decrease of integrated reflectivity for the incident attosecond pulse.

To further verify the better performance of large angle incidence for attosecond pulse reflection, temporal pulse reflection parameters of periodic multilayer mirrors with different layers have been displayed in Fig. 6.

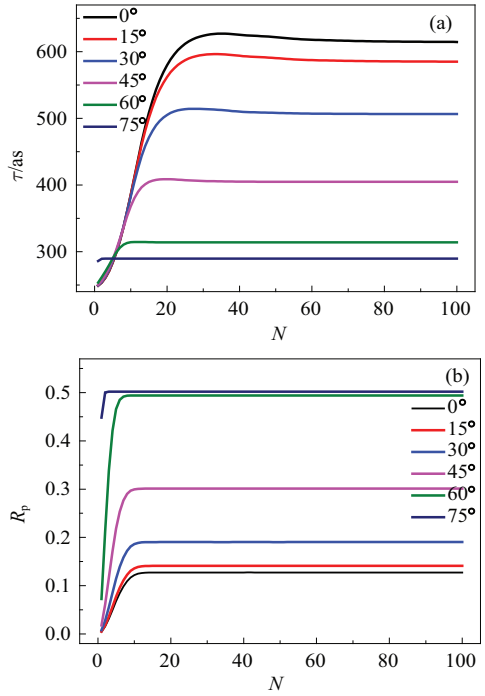


Fig. 6. (color online) (a) Duration τ and (b) power reflection coefficient R_p of reflected pulses versus the bilayer number N of periodic multilayer mirrors under different θ (from 0° to 75°).

In Fig. 6, duration τ and power reflection coefficient R_p of reflected pulses in the periodic multilayer reflection systems have been plotted with a changing bilayer number N of periodic multilayer mirrors. The incident angles from 0° to 75° have been considered. From Fig. 6, we can see that all curves go up with N at the beginning, and then, get to saturation at certain bilayer number. The power reflection coefficient R_p always shows a better performance with a larger incident angle θ for arbitrary N. When N is small, the duration τ of reflected pulses for different incident angles are almost the same, because of broad enough reflection bands for an incident pulse afforded by periodic multilayer mirrors. Then, when N becomes larger, multilayers under large incident angles will give out smaller τ due to its relatively large bandwidth.

4. Conclusion

In summary, the improvement of attosecond pulse reflection by a large angle incidence for periodic Mo/Si multilayer mirrors has been proven in this paper. The broader reflection band with a high reflectivity has been obtained by the periodic multilayer mirror under a larger incident angle in the frequency domain, which guarantees a better pulse reflection capability for attosecond pulse reflection in the time domain. Furthermore, such an effect of improving the attosecond pulse reflection has also been verified effectively for periodic multilayer mirrors with arbitrary layers, exhibiting its wide application in single attosecond pulse reflection.

References

[1] Lan P F, Lu P X, Cao W, Li Y H and Wang X L 2007 *Phys. Rev.* A **76** 011402(R)
[2] Hong W Y, Lu P X, Li Q G and Zhang Q B 2009 *Opt. Lett.* **34** 2102
[3] Peng L Y, Gong Q H and Tan F 2009 *Chin. Phys.* B **18** 4807
[4] He H P and Ge Y C 2010 *Chin. Phys.* B **19** 103302
[5] Hong W Y, Yang Z Y, Lan P F, Zhang Q B, Li Q G and Lu P X 2009 *Acta Phys. Sin.* **58** 4914 (in Chinese)
[6] Luo M H and Zhang Q J 2011 *Chin. Phys.* B **20** 085201
[7] Zhang G T, Bai T T and Zhang M G 2012 *Chin. Phys.* B **21** 054214
[8] Kienberger R, Goulielmakis E, Uiberacker M, Baltuska A, Yakovlev V, Bammer F, Scrinzi A, Westerwalbesloh T, Kleineberg U, Heinzmann U, Drescher M and Krausz F 2004 *Nature* **427** 817
[9] Hofstetter M, Aquila A, Schultze M, Guggenmos A, Yang S, Gullikson E, Huth M, Nickel B, Gagnon J, Yakovlev V S, Goulielmakis E, Krausz F and Kleineberg U 2011 *New J. Phys.* **13** 063038
[10] Kienberger R, Hentschel N, Uiberacker M, Spielmann C, Kitzler M, Scrinzi A, Wieland M, Westerwalbesloh T, Kleineberg U, Heinzmann U, Drescher M and Krausz F 2002 *Science* **297** 1144
[11] Morlens A S, Balcou P, Zeitoun P, Valentin C, Laude V and Kazamias S 2005 *Opt. Lett.* **30** 1554
[12] Morlens A S, Lopez-Martens R, Boyko O, Zeitoun P, Balcou P, Varju K, Gustafsson E, Remetter T, L'Huillier A, Kazamias S, Gautier J, Delmotte F and Ravet M F 2006 *Opt. Lett.* **31** 1558
[13] Wonisch A, Neuhausler U, Kabachnik N M, Uphues T, Uiberacker M, Yakovlev V, Krausz F, Drescher M, Kleineberg U and Heinzmann U 2006 *Appl. Opt.* **45** 4147
[14] Suman M, Frassetto F, Nicolosi P and Pelizzo M G 2007 *Appl. Opt.* **46** 8159
[15] Hofstetter M, Schultze M, FieB M, Dennhardt B, Guggenmos A, Gagnon J, Yakovlev V S, Goulielmakis E, Kienberger R, Gullikson E M, Krausz F and Kleineberg U 2011 *Opt. Express* **19** 1767
[16] Bourassin-Bouchet C, de Rossi S, Wang J, Meltchakov E, Giglia A, Mahne N, Nannarone S and Delmotte F 2012 *New J. Phys.* **14** 023040
[17] Bourassin-Bouchet C, Diveki Z, de Rossi S, English E, Meltchakov E, Gobert O, Guénot D, Carre B, Delmotte F, Salières P and Ruchon T 2011 *Opt. Express* **19** 3809
[18] Drescher M, Hentschel M, Kienberger R, Tempea G, Spielmann C, Reider G A, Corkum P B and Krausz F 2001 *Science* **291** 1923
[19] Loch R A, Dubrouil A, Sobierajski R, Descamps D, Fabre B, Lidon P, van de Kruijs R W E, Boekhout F, Gullikson E, Gaudin J, Louis E, Bijkerk F, Mevel E, Petit S, Constant E and Mairesse Y 2011 *Opt. Lett.* **36** 3386
[20] Lin C Y and Liu D H 2012 *Chin. Phys.* B **21** 094216
[21] Beigman I L, Pirozhkov A S and Ragozin E N 2001 *JETP Lett.* **74** 149
[22] Beigman I L, Pirozhkov A S and Ragozin E N 2002 *J. Opt.* A **4** 433
[23] Henke B L, Gullikson E M and Davis J C 1993 *At. Data Nucl. Data Tables* **54** 181
[24] Aquila A L, Salmassi F, Dollar F, Liu Y and Gullikson E 2006 *Opt. Express* 14 10073
[25] Attowood D T 2003 *Soft X-rays and Extreme Ultraviolet Radiation: Principles and Application* (1st edn.) (Beijing: Science Press) p. 61 (in Chinese)
[26] Hau-Riege A P and Chapman H N 2007 *Rev. Sci. Instrum.* **78** 013104
[27] Pirozhkov A S, Daido H, Bulanov S V and Ragozin E N 2005 *Springer Series in Chemical Physics: Ultrafast Phenomena XIV* **79** 85
[28] Underwood J H and Barbee T W 1981 *Appl. Opt.* **20** 3027
[29] Chen R, Wang F L and Wang Z S 2008 *Chin. Opt. Lett.* **6** 310