Corrigendum: Noncollinear optical gating (2014 New J. Phys. 16 052001)

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Received 20 August 2014
Accepted for publication 20 August 2014
Published 28 October 2014

New Journal of Physics 16 (2014) 109501
doi:10.1088/1367-2630/16/10/109501

Figure 1. Comparison between NOG (a)–(c) and lighthouse (d)–(f). Normalized spatio-temporal field distribution $\Re(E)^2$ of the generation field before focusing (a), (d), focusing scheme (b), (e) and generated XUV intensity distribution together with the fundamental field (grayscale) at a focal length distance behind the focus (c), (f). In order to achieve comparable spatio-temporal XUV pulse characteristics we consider $\Delta t = \tau$ for NOG, a beam radius before focusing that is a factor of 3.7 larger for the lighthouse scheme than for NOG and laser pulses that have the same spectral bandwidth, corresponding to $\tau = 2T$. The dashed lines in (c), (f) indicate the time-to-space mapping.
The original article was published on 23 May 2014 with a production error in figure 4. One layer of this figure was shifted (figure 4(f)) and another was missing completely (figure 4(c)), leading to incorrect timing information between the fundamental field and the generated XUV field. The corrected figure is shown as figure 1 in this corrigendum.
Noncollinear optical gating

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Received 25 April 2014
Accepted for publication 7 May 2014
Published 23 May 2014

Abstract

We present a novel scheme for high-order harmonic generation, enabling the production of spatially separated isolated attosecond pulses. This can be achieved by driving the generation process with two identical, but temporally delayed laser pulses, which are noncollinearly overlapping in the generation medium. Our approach provides intense attosecond pulses directly separated from the fundamental field, which is left undistorted. The method is therefore ideally suited for pump-probe studies in the extreme ultraviolet regime and promises new advances for intra-cavity attosecond pulse generation. We present a theoretical description of noncollinear optical gating, with an analytical derivation and simulations using the strong field approximation.

Keywords: isolated attosecond pulses, high harmonic generation, attosecond physics, extreme nonlinear optics

1. Introduction

The generation of single attosecond pulses (SAPs) via high-order harmonic generation (HHG) has enabled a multitude of experiments providing insight into attosecond dynamics in atoms [1], molecules [2] and solids [3]. While the generation of trains of attosecond pulses, i.e. high-order harmonics, does not present any technical difficulty with present ultrafast laser
technology, the generation of SAPs remains challenging, requiring carrier-envelope-phase (CEP)-stable few-cycle driving laser pulses and/or advanced gating methods to confine the extreme ultraviolet (XUV) emission to a single half-cycle of the driving field. Commonly-used techniques are based on manipulating the polarization state of the driving field [4, 5], utilizing a high laser intensity to rapidly deplete the nonlinear medium [6] or altering the shape of the driving field via multi-color field synthesis [7]. Vincenti and co-workers recently introduced a conceptually different technique [8], named attosecond lighthouse, utilizing spatio-temporal couplings to angularly streak the generated attosecond pulses. The technique was experimentally implemented both in gases [9] and using plasma mirrors [10].

Most gating techniques imply extensive manipulation of the fundamental field and require spectral filtering of the harmonic radiation, limiting the efficiency and restricting any further use of the laser pulse. Using the fundamental field after generation is of interest for pump-probe experiments and essential for intra-cavity HHG [11, 12], a promising scheme that enables the generation of attosecond pulses at unprecedented power levels and repetition rates, with applications in ultrahigh precision frequency metrology. While the generation of high-order harmonics inside a cavity has been experimentally demonstrated [11, 12], that of isolated attosecond pulses remains an unsolved challenge, requiring broadband enhancement cavities [13] and suitable gating methods [14].

Here, we introduce an efficient gating technique, noncollinear optical gating (NOG), based on driving the HHG process in a noncollinear geometry. Noncollinear HHG has recently been employed for several applications [18, 19], including in situ diagnostics of the generation process [20], and proposed as out-coupling method for intra-cavity HHG [21, 22]. Here, we present a new implementation of noncollinear HHG, which is used to generate angularly streaked attosecond pulse trains, thus providing an efficient gate for SAP generation. Two identical but time-delayed, noncollinearly overlapping laser pulses form a driving field with rotating wave fronts. Consecutive attosecond pulses are emitted in different directions, as illustrated in figure 1. The two driving laser pulses are left unperturbed after generation and are spatially separated from the XUV radiation emitted at the bisector of the two driving field propagation angles. Thus, our method opens new possibilities for attosecond pump-probe experiments [15–17] as well as for frequency-comb spectroscopy studies in the XUV regime.

The article is organized as follows: the method is described in sections 2 and 3. Numerical simulations are presented in section 4. In section 5, we discuss the influence of macroscopic propagation effects. Finally, in section 6, we compare our gating scheme with the attosecond lighthouse technique.

2. Principle of the gating method

We consider two identical laser pulses, which are overlapped in time and superimposed noncollinearly with an angle $2\gamma$ at the focus, thus forming a spatial intensity grating in the focal plane [19]. The far-field angular distribution of the harmonics generated in such a geometry is determined by the interference of multiple sources. In general, the harmonics are emitted in different directions [23], and consequently the corresponding attosecond pulses are angularly distorted. The angular emission characteristics simplify considerably at small noncollinear

1 Perturbations intrinsic to the HHG process, e.g. due to plasma formation as well as possible perturbations induced when splitting an initial laser pulse into two pulses might of course occur.
angle, when the intensity grating in the generation medium becomes a single maximum with weak satellites, as discussed in more detail below. In these conditions, the XUV emission follows the instantaneous direction of propagation of the total driving field (the $z$-axis in figure 1(c)), defined by the bisector of the two driving field propagation angles.

The wave front orientation in the focus of two noncollinearly overlapping laser fields with field envelope $E_1$ and $E_2$, propagating along the wavevectors $k_1$ and $k_2$ respectively, can be defined through the composite wavevector at the point of intersection:

$$k_{tot} = \frac{(E_1 k_1 + E_2 k_2)}{\sqrt{E_1^2 + E_2^2}}.$$  

Figure 1. Principle of noncollinear optical gating. (a) A temporal delay between two few-cycle pulses leads to a rapidly changing amplitude ratio between subsequent half-cycles. (b) Overlapping these pulses noncollinearly causes a temporal rotation of the wavefronts (the figure displays $\Re(E^2)$ of the total field $E$). Attosecond pulses generated from subsequent half-cycles are therefore emitted in different directions. (c) Illustration of the experimental scheme: two noncollinearly overlapping driving pulses generate spatially separated isolated attosecond pulses in the angle sector between the fundamental propagation directions. The displayed spectrogram was calculated using a quasi-classical approach, considering a pulse duration and temporal delay of $2T$, where $T$ is the field-cycle period. The observed continua, labelled 1–5, are characteristic of spatially separated attosecond pulses.
By taking the scalar product $\hat{x} \cdot \mathbf{k}_{\text{tot}}/|\mathbf{k}_{\text{tot}}| = -\sin(\beta)$, in which $\hat{x}$ is the unit vector in lateral direction (see figure 1(b), (c)) and $\beta$ is the angle between $\mathbf{k}_{\text{tot}}$ and the $z$-axis, we obtain for small $\gamma$:

$$\beta = \gamma \frac{1 - \xi}{1 + \xi}. \quad (2)$$

A variation of the amplitude ratio of the laser fields, $\xi = \mathcal{E}_2/\mathcal{E}_1$, results in a macroscopic tilt of the wave fronts and a change in the emission direction $\beta$. The introduction of a temporal delay $\Delta t$ between two driving laser pulses leads to an amplitude ratio that changes rapidly from one half-cycle to the next (figure 1(a)), defining a unique orientation angle of the corresponding wave fronts as a function of time. The consequence is an ultrafast wave front rotation (WFR) in the focus (figure 1(b)), with attosecond pulses emitted in different directions [8]. The emission of XUV light is thus angularly streaked, mapping time into spatial position in the far-field [9]. A conventional imaging-XUV spectrometer displaying the spatially resolved spectral profile will therefore provide a spectrogram of the generated attosecond pulse train, i.e. frequency versus time, as illustrated in figure 1(c). The time-to-space mapping is defined through equation (2) where $\xi$ varies with time depending on the envelope and separation of the driving fields.

Considering two Gaussian laser pulses with $E_{1,2} = \exp[-2 \ln(2)(t \pm \Delta t/2)^2/\tau^2]$ and pulse duration $\tau$ (FWHM), we can deduce the emission angle as a function of time. The time-to-space mapping is approximately linear around $t = 0$, i.e. at the temporal center of the total driving field, where maximum WFR speed is achieved, with

$$\beta(t) = 2 \ln(2) \gamma \frac{\Delta t}{\tau^2} t. \quad (3)$$

In the temporal wings of the pulses, the WFR speed is reduced. The direction of the time axis itself is determined by the sign of the temporal delay, i.e. the order of the driving pulses. The WFR velocity increases linearly with $\Delta t$. Choosing $\Delta t \approx \tau$ ensures that the total field intensity does not exhibit a local minimum at $t = 0$.

3. Angular separation of the attosecond pulses

In order to isolate attosecond pulses emitted from subsequent half-cycles, the angle difference $\Delta \beta \approx \beta(T/2)$ has to be larger than the divergence angle of a SAP. We define this angle as $\Theta_q = W_q(z)/z$, with $W_q(z)$ denoting the beam radius in the far field at a distance $z$ from the focus for a beam with central frequency $q\omega$. To estimate $\Theta_q$, we consider the intensity grating of the fundamental field in the focal plane with a Gaussian envelope at the beam waist and radius $W_0$. All spatial beam widths are defined at $1/e^2$ of the intensity profile. For small $\gamma$, the intensity distribution in the focal plane can be written as:

$$I(x, z = 0) \propto \cos^2[kx \sin(\gamma)] \exp\left(-\frac{2x^2}{W_0^2}\right). \quad (4)$$

In order to avoid interference effects due to multiple harmonic sources formed by the interference grating in the focus, we consider a degenerated grating with only one central maximum at $x = 0$ and weak satellites. Limiting the intensity of the first satellites to $1/e^2$ of the
central maximum implies that $\gamma$ must be smaller than $\gamma_{\text{opt}} = \lambda/2W_f$ with $\lambda$ denoting the carrier wavelength. Using Gaussian optics, we have $\lambda = W_f$ with $f$ the focal length and $W_f$ the beam radius before focusing. Defining $x_0$ as the center-to-center separation of the two generation beams before focusing and $\delta = x_0/W_f$, the required intensity grating is obtained when $\delta \leq \pi/2$.

For small $\gamma$, the cosine$^2$-intensity distribution caused by the beam interference can be approximated with a Gaussian function, $I(x, z = 0) \propto \exp(-2x^2/W_f^2)$, with $W_f^2 = 1/[k^2\gamma^2/2 + 1/W_0^2]$, as illustrated in figure 2(a). With the scaling relation $\Theta_q \propto 1/W_f$, this leads to an expression for the divergence angle $\Theta_q = \Theta_q(\gamma)$ in units of $\Theta_q(0)$:

$$\frac{\Theta_q(\gamma)}{\Theta_q(0)} = \frac{W_0}{W_f} = W_0 \sqrt{\frac{k^2\gamma^2}{2} + \frac{1}{W_0^2}}.$$ (5)

In order to determine whether the condition for SAP emission $\Delta\beta > \Theta_q$ is satisfied, we look at the scaling of $\Delta\beta$ and $\Theta_q$ with increasing noncollinear angle, or equivalently beam separation $\delta$ (figure 2(b)). $\Delta\beta$ is plotted as a function of $\gamma$ for three different pulse lengths while $\Theta_q$ is shown for two different values of the ratio between the fundamental and the attosecond pulse divergence $\eta = \Theta_q/\Theta_q(0)$. While $\Delta\beta$ increases linearly with $\gamma$, $\Theta_q$ follows a nonlinear scaling relation. The conditions for SAP generation improve with increasing $\gamma$ until $\Theta_q$ approaches an asymptotic linear dependence. Figure 2(b) shows that efficient gating requires short driving pulses and collimated harmonic beams. For $\eta = 10$ and $\lambda = 800$ nm, gating is achieved for
τ \leq 10 \text{ fs} \quad \text{and} \quad \delta \geq 1, \quad \text{while if} \quad \eta = 5, \quad \text{gating needs pulses of the order of} \quad 5 \text{ fs} \quad \text{or less and similarly} \quad \delta \geq 1. \quad \text{The gating conditions improve until} \quad \delta \approx \pi/2, \quad \text{which corresponds to the maximum allowed noncollinear angle} \quad \gamma = \gamma_{\text{opt}}. \quad \text{As an example, for} \quad f = 0.5 \text{ m} \quad \text{and} \quad W_f = 2 \text{ mm} \quad \text{we obtain} \quad \gamma_{\text{opt}} = 6.3 \text{ mrad.} \\

In summary, optimal conditions for SAP generation, which ensure minimized temporal satellite pulses as well as suppression of interference effects due to multiple sources in the focus, are obtained when the beam separation before focusing is approximately \pi/2 \text{ times the} \quad 1/e^2\text{-diameter of the intensity profile. This condition is valid for loose and tight focal geometries, since the NOG scheme does not depend on focusing as long as} \quad \eta \quad \text{remains the same (see figure 2). With decreasing focal length,} \quad W_0 \quad \text{decreases and the SAP divergence increases. However,} \quad \Delta \beta \quad \text{increases accordingly, and identical temporal SAP characteristics are obtained.} \\

For \quad \gamma \approx \gamma_{\text{opt}}, \quad \text{the influence of the Gaussian envelope on the radial dimension of the harmonic source can be neglected, and the divergence can be approximated by its asymptotic value} \\
\Theta_\eta \approx \Theta_\eta(0) W_0 \frac{k\gamma}{\sqrt{2}} = \frac{\lambda k}{\pi \sqrt{2} \eta} \gamma = \frac{\sqrt{2} \gamma}{\eta}. \quad (6) \\

Using equation (3) this leads to a simple condition for the maximum pulse duration allowing an angular separation of the generated attosecond pulses: \\
\tau \leq \frac{\ln (2) \eta \lambda}{\sqrt{2} c}, \quad (7) \\
\text{c denoting the speed of light. If the above condition is fulfilled, the emission direction between consecutive SAPs changes by at least the divergence angle of the same pulses. This is equivalent to limiting the on-axis intensity of pre- and post pulses to} \quad 1/e^2\text{ of the main pulse. For} \quad \lambda = 800 \text{ nm, and} \quad \eta = 10, \quad \text{we obtain} \quad \tau \leq 13 \text{ fs.} \\

The factor \quad 1/\eta \quad \text{can also be interpreted as a lower limit for the angular divergence of the harmonic emission at frequency} \quad \eta \omega, \quad \text{defined in units of the fundamental beam divergence. Thus, equation (7) leads to an approximate limit for the low-energy cutoff} \quad E_{\text{low}} = \hbar \omega \eta \quad \text{of the generated spectral continua:} \\
E_{\text{low}} \approx \frac{\hbar \omega^2 \tau}{\pi \sqrt{2} \ln (2)}, \quad (8) \\
\text{the high-energy limit being determined by the fundamental intensity. Using the above parameters and considering} \quad \tau = 5 \text{ fs} \quad \text{at} \quad \lambda = 800 \text{ nm and a peak intensity of} \quad 2 \times 10^{14} \text{ W cm}^{-2}, \quad \text{we have} \quad E_{\text{low}} = 5.9 \text{ eV, leading to SAPs with up to} \quad 48 \text{ eV bandwidth if Ar is used for generation.} \\

4. Simulations \\
We performed numerical calculations in order to simulate HHG in a noncollinear geometry. The single atom response was obtained by solving the time dependent Schrödinger equation within the strong field approximation [24], selecting the short trajectory contribution. The harmonic dipole response was calculated numerically on a finite temporal grid for all points
along a one dimensional grid in the focal plane. The far-field emission was computed using Fraunhofer diffraction integrals. Propagation effects (see section 5) were not taken into account.

Figure 3 presents simulations of HHG in neon for two-cycle driving pulses ($\tau = 2T$), considering $\delta = \pi/2$. The first row (a) shows the focused laser field $\Re (E)^2$, the attosecond pulse train in the far field (b), the far field spectrum (c) and a spectral line-out at $\beta = 0$ (d) when the two pulses are superposed with no time delay at the focus. The far-field intensity distribution is shown as a function of the propagation angle $\beta$, normalized to the divergence of the focused fundamental laser pulses. The generated attosecond pulse train includes approximately four pulses, which interfere in the far field, leading to a discrete HHG spectrum. The second and third rows present the same quantities for temporally-delayed driving pulses, with $\Delta t = 2T = \tau$ and $\Delta t = 2.5T$ respectively. The WFR ((e) and (i)) induced by the temporal separation streaks the attosecond pulses angularly as shown in (f) and (j). With increasing temporal delay, the discrete spectrum becomes a characteristic XUV spectrogram with discrete harmonic peaks at low energies and spectral continua towards the high-energy cut-off. The spatially overlapping contributions from consecutive attosecond pulses interfere, leading to a fringe pattern between
the spectral continua. The slope of the interference fringes, which increases with energy, reflects the harmonic dipole phase variation with driving field intensity [25].

A variation of the CEP $\phi_{CE}$ of the driving fields moves the wavefronts in time and leads to a spatial shift of the corresponding XUV spectrogram [9, 10], providing direct access to the CEP of the driving laser field. For the simulations presented in figure 3, $\phi_{CE}$ was chosen in order to emit a SAP at $\beta = 0$. Changing $\phi_{CE}$ by $\pi/2$ leads to the emission of two pulses at $\beta \approx \pm \Delta \beta/2$, thus providing two synchronized but spatially separated SAPs, of interest for attosecond-pump-attosecond-probe studies.

A variation of the relative phase between the two driving fields moves the fundamental intensity grating across the focal plane. A zero relative phase at the point of intersection ensures a central intensity maxima at $x = 0$, as shown in figure 3 (first column) while a phase shift of $\pi$ leads to two intensity maxima in the focal plane and a phase singularity at $x = z = 0$ [26]. In the latter case, interference effects between the two harmonic sources in the focus lead to angularly distorted attosecond pulses, thus preventing efficient gating.

5. Propagation effects

Propagation effects in the nonlinear medium can be described in a similar way as for a collinear geometry. The noncollinear geometry adds a geometrical component to the total wavevector missmatch between fundamental beam and generated XUV field with central frequency $\omega$ [19]. In the focus, the geometrical wave vector mismatch can be taken into account by replacing the component arising from the Gouy phase shift for one beam $\Delta k^q = -q/z_0$ ($z_0$ denotes the Rayleigh length) by:

$$\Delta k^q_{nc} = \Delta k^q \left(1 + \frac{\pi^2}{4} - \frac{\lambda \pi}{8z_0}\right).$$  \hspace{1cm} (9)

The second term in equation (9) can be attributed to the noncollinear geometry considering plane waves (equivalent to the Gouy phase shift of a Gaussian beam with an initial beam radius of $W/\sqrt{\pi}/2$), the last term is caused by the off-axis components of the Gouy phase shift.

$\Delta k^q_{nc}$ is equivalent to the wave vector mismatch for a collinear geometry with tighter focusing, which does not imply a lower conversion efficiency if the generation parameter are selected adequately [27]. As for collinear HHG, phase matching can be achieved by minimizing the total wave vector mismatch, i.e. adjusting gas pressure, medium length and laser intensity [28]. Our numerical simulations confirm the robustness of the NOG scheme even for a long medium, which is consistent with the findings presented in the supplementary information of [9] for the case of the lighthouse technique (see section 6).

6. Comparison with the attosecond lighthouse

Finally, we compare NOG with the attosecond lighthouse technique [8]. Both methods are based on WFR of the fundamental field, introducing an angular streaking effect. In the lighthouse configuration, WFR is achieved by focusing an angularly chirped laser pulse while an ultrafast amplitude modulation induces a rotating wave front in the NOG scheme. The lighthouse can also be seen as the continuous analog of NOG. The two techniques are compared
in Figure 4 for a two-cycle fundamental pulse and maximum WFR speed for the lighthouse scheme. Figures 4(a) and (d) show the spatio-temporal representation of the driving field distribution before focusing, while (c) and (f) present the angularly streaked attosecond pulse train in the far field together with the driving field after generation (in gray). Considering $\Delta t = \tau$ for the NOG scheme, both methods lead to angularly streaked attosecond pulse trains with very similar spatio-temporal characteristics and identical streaking efficiency at $t = 0$. Interestingly, the NOG scheme allows even faster WFR for $\Delta t > \tau$ (Figure 2, third row) and thus a better spatial separation of the generated attosecond pulses while the WFR speed is limited in the lighthouse scheme [8]. Another important difference emerges for the fundamental field characteristics after generation. In the lighthouse scheme the fundamental field is chirped and distributed over the angle sector in which attosecond pulses are emitted. In the NOG scheme, two, nearly transform limited fundamental pulses (see footnote 1), leave the interaction process after generation and isolated attosecond pulses are emitted in the angle sector between the two driving fields.

7. Conclusion and outlook

In conclusion, we have introduced a new method for streaking attosecond pulse trains, which gives access to multiple, perfectly synchronized SAPs. NOG constitutes the first gating scheme that allows a direct spatial separation of fundamental driving field and generated attosecond pulses. It is therefore ideally suited for attosecond-pump-attosecond-probe studies, which
require high pulse energies. Finally, NOG can be applied to intracavity HHG, leading to the generation of isolated attosecond pulses at unprecedented average power levels and MHz repetition rates, promising advances in XUV-frequency-comb spectroscopy.

Acknowledgments

This research was supported by the Marie Curie program ATTOFEL (ITN), the European Research Council (ALMA, PALP), the Swedish Research Council, the Swedish Foundation for Strategic Research and the Knut and Alice Wallenberg Foundation.

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