

Attochirp-free High-order Harmonic Generation

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Abstract

A method is proposed for arbitrarily engineering the high-order harmonic generation phase achieved by shaping a laser pulse and employing xuv light or x rays for ionization. This renders the production of bandwidth-limited attosecond pulses possible while avoiding the use of filters for chirp compensation. By adding the first 8 Fourier components to a sinusoidal field of $10^{16}\text{W}/\text{cm}^2$, the bandwidth-limited emission of 8 as is shown to be possible from a Li^{2+} gas. The scheme is extendable to the zs-scale.

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I. INTRODUCTION

High-order harmonic generation (HHG) is the key technology for attosecond science. The shortest pulse durations currently achieved are below 100 as [1–3]. Highly efficient sources are available having a bandwidth of hundreds of electronvolts [4] being large enough to produce pulses of only 10 as duration — in case they can be generated without chirp in the future. Many properties of HHG radiation emitted from a gas target can be understood by studying a single atom. The three-step model [5] is the simplest model to describe the single-atom dynamics. The process starts when a strong laser field liberates the electron of an atom and subsequently drives it in the continuum. If ionization happened at the right time, the electron can be accelerated back towards the parent ion after the field has changed its sign and can recombine along with the emission of an energetic photon.

One prominent feature of HHG is that the emitted light has an intrinsic chirp, the so-called attochirp [6, 7]. The origin of the attochirp can be understood from the classical electron trajectories in the laser field: trajectories with different energies recollide at different times. Due to the widespread classical recollision times in a usual sinusoidal laser field, the emitted harmonic pulses have a longer duration than their bandwidth limit, that is the minimum pulse duration for a given spectral bandwidth reached when the spectral phase of the harmonic pulse is constant. To compress the emitted pulse, dispersive elements of either chirped multilayer x-ray mirrors [8], thin metallic films [9, 10] or gaseous media [3, 11] are employed and even the use of grating compressors is attempted [12]. However, these techniques suffer from losses, rely on the material properties and the chirp is not well-controlable in these schemes. Additionally, it is required to select radiation from the positively chirped short trajectory via phase-matching before the compensation element. Other approaches [4, 13] try to exploit the reduced time window of recollision when increasing the wavelength of the driving laser at a constant bandwidth and field maximum. However, this way, it cannot be taken advantage of the increased cutoff. Without selection of a certain trajectory, it is possible to partially reduce the attochirp as shown in [14, 15] by adding a weak second-harmonic or subharmonic field. The attochirp problem is expected to get more significant and demanding when photon energies spanning far into the soft x-ray domain are reached in the future.

In this paper, we propose a way to engineer the attochirp in a determined manner by

altering the harmonic generation process. In terms of the wave function, the following scenario is realized: the wave function localized in the binding potential is continuously freed resulting in a large spread in space and momentum. The electronic quantum dynamics is tuned in a way that after a certain time of propagation, the wave function spatially recompresses at least along the propagation direction of the wave packet. It has its minimum width exactly at the time of recollision but with a large energy bandwidth gained during propagation in the continuum. We show that when the laser pulse is shaped by adding a small number of low-order harmonics and employing soft x rays for ionization, attosecond pulses with arbitrary chirp can be formed including the possibility of attochirp-free HHG and bandwidth-limited attosecond pulses.

The benefit of assisting the HHG process in a strong laser field with a weak high-frequency field has been demonstrated mainly for the purpose of enhancing the single-atom yield [16–20], improving phase matching [20] or suppressing the relativistic drift [21, 22]. On the other hand, femtosecond pulse shaping has been used to shape the HHG spectrum [23], to increase the HHG cutoff [24] or for relativistic HHG [25, 26]. Here, we employ x rays to ionize the electron with non-zero velocity and combine it with femtosecond pulse shaping to control the spectral phase of the harmonic spectrum.

II. CLASSICAL ANALYSIS

It is known [27] that the HHG process can be analyzed by taking only a few quantum orbits into account that correspond to classical trajectories. Therefore, a classical analysis is able to give first insight into our idea. In the first part of our paper, we start out by considering classical trajectories in a tailored laser field [see Fig. 1 (a)] and find the condition when trajectories ionized at different times recollide at the same time. For the considered laser intensities, the classical trajectories have only a component along the polarization direction of the laser field, i.e. 1-dimensional trajectories are shown. We describe the principle of our method by discussing two example trajectories marked by α and β in Fig. 1 (b) being ionized separated by a small time span δt_i . We first focus on the point in time when trajectory β just starts. At that time, α has already been driven slightly away from the origin. The distance between the trajectories is $\delta x_i \approx p_i \delta t_i$ where p_i is an initial momentum e.g. mediated via one-photon-ionization (atomic units are used throughout unless stated otherwise). The momentum

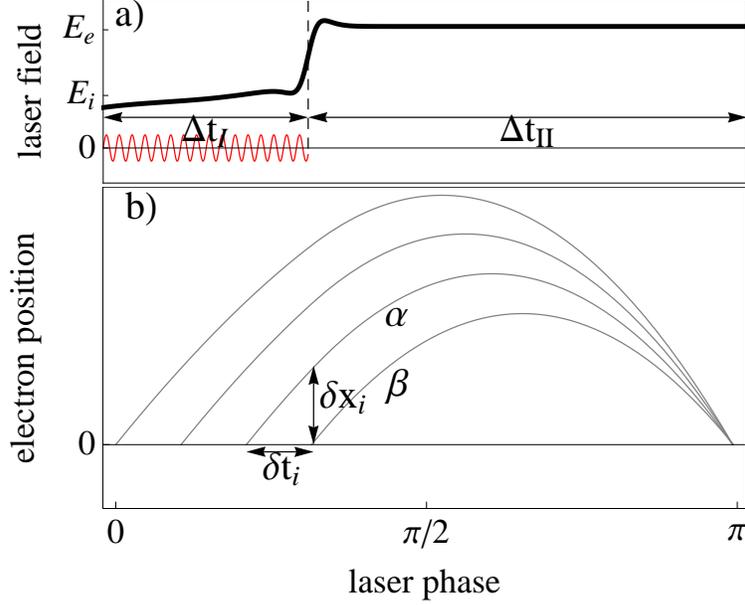


FIG. 1: Schematic of the recollision scenario: a) A half cycle of the tailored laser field (black). The red line is the assisting x-ray pulse. b) Different classical trajectories in the field of (a) which start into the continuum at different times but revisit the ionic core at the same time.

difference at that time is given by $\delta p_i \approx -E_i \delta t_i$ because α has already been decelerated by the laser field E_i . From now on, the momentum difference $\delta p = \delta p_i$ is conserved during the whole propagation time. The separation of both trajectories at recollision after a time τ is therefore given by $\delta x_e \approx \delta x_i + \delta p \tau = (p_i - E_i \tau) \delta t_i$. $\delta x_e = 0$ reflects the condition on the initial momentum and on the electric field at ionization necessary for simultaneous recollision of the two trajectories:

$$E_i \approx p_i / \tau \quad (1)$$

The previous discussion applies only to the case of two single trajectories ionized with an infinitely small time separation but is sufficient for demonstration of our principle idea of simultaneous recollision for trajectories ionized from a finite time window.

In Fig. 1, we chose the simplest possible laser field to fulfill condition (1): a first plateau of strength E_i and duration Δt_I which is followed by a second higher plateau of duration Δt_{II} . In this procedure, the field strength E_e of the second plateau is chosen first. It determines τ for trajectories β . Then, the first plateau is found having a small slope determined by optimizing a polynomial expression to have simultaneous recollision of all classical trajectories

(approximately condition (1)).

Crucial for the previous scheme is the ionization with a non-zero velocity. The condition can be reached by using x rays of frequency ω_X which are responsible for ionization. In this case, the initial momentum is $p_i = \sqrt{2(\omega_x - I_p)}$ where I_p is the binding potential. The x rays co-propagate with the laser field and are spectrally filtered out before the harmonic radiation reaches the detector. ω_X can be chosen in a way (with the appropriate laser field according to Eq. (1)) to meet a resonance of a suitable absorber at ω_X . Alternatively, the x rays could propagate with a tiny angle to the propagation direction of the laser. In Fig. 1, we only consider trajectories ionized within Δt_I and having a direction pointing upwards to the potential (starting in positive direction in Fig. 1). The initial momentum could also be directed downwards but in this case, the classical electron would not recollide and no harmonics would be emitted. A second branch of trajectories that are ignored in the figure are those trajectories emerging from the second plateau during Δt_{II} . They re-encounter the core region but all at different times and would break the desired scenario of simultaneous recollision. Therefore, ionization during the second plateau has to be suppressed which demands for the use of an x-ray pulse [sketched as red wiggled line in Fig. 1 (a)] rather than a cw x-ray field. The x-ray pulse must have a non-vanishing field strength only during Δt_I being of order of 1 fs. Moreover, as medium, we choose a gas of ions because their high ionization potential is necessary to suppress tunnel ionization. The plasma could be generated via laser ionization with a strong pre-pulse.

We briefly comment on the energy distribution of the recolliding trajectories. The classical trajectory marked by β has a distinct role because it experiences approximately a constant laser field. Therefore, it is symmetric to the turning point and also recollides with same momentum p_i as it started. Trajectories starting prior to β recollide with a higher energy but at the same time as β assuming the first plateau-like structure is chosen in agreement with Eq. (1) as it is in Fig. 1. Trajectory β plays the role of the trajectory with the lowest energy ω_X . The duration of the first plateau Δt_I determines the velocity difference $\Delta p \approx E_i \Delta t_I$ and the energy bandwidth $\Delta \omega_q \approx \frac{1}{2}(\Delta p + p_i)^2 + I_p - \omega_X = \frac{1}{2}\Delta p^2 + \Delta p p_i$.

In the optimum case shown in Fig. 1, all trajectories recollide simultaneously. However, under real experimental conditions, deviations of the laser and x-ray field from the optimal conditions result in a non-zero time window Δt_e of recollision. The classical model employed a discrete x-ray frequency ω_X rather than a finite bandwidth as a real pulse. The

maximum allowed bandwidth can be deduced from the former model. The x rays ionize the electron with an initial velocity and arrange this way the initial displacement δx_i between two trajectories. If ω_X deviates from its optimal value by $\delta\omega_X$, the initial momentum will deviate by $\delta p_i \sim \delta\omega_X/(2p_i)$ and result in a additional displacement $\delta x_i = \delta p_i \delta t_i$ which is not compensated for. Therefore, the final wave packet size is of order $\delta p_i \Delta t_I$ resulting in a time spread of

$$\Delta t_e^{BW} = \delta p_i \Delta t_1 / p = \frac{\delta\omega_X \Delta t_I}{2p_i p} \quad (2)$$

which has to be smaller than the envisaged pulse duration.

III. STRONG-FIELD APPROXIMATION MODEL

So far, purely classical dynamics was considered. In order to model the single-atom HHG yield, we use the strong-field approximation (SFA) and include the laser field within the dipole approximation. This way, the Fourier transformed dipole matrix element is given by [19, 22]

$$\tilde{d}_q = i \int_{-\infty}^{\infty} dt \int_{-\infty}^t dt' \int d^3\mathbf{q} \langle \Phi_0(t) | x | \mathbf{p} + \mathbf{A}(t)/c \rangle \mathbf{E}_X(t') \langle \mathbf{p} + \mathbf{A}(t)/c | x | \Phi_0(t') \rangle e^{-iS_q(\mathbf{p}, t, t')} \quad (3)$$

where $\Phi_0(t)$ is the ground state wave function of the employed zero-range potential, $S_q(\mathbf{p}, t, t') = \int_{t'}^t \{ [\mathbf{p} + \mathbf{A}(t'')/c]^2 / 2 + I_p \} dt'' + \omega_X t' - q\omega t$ the classical action and q the harmonic number. In the long wavelength regime the highly oscillating integral can be evaluated by using the saddle-point approximation. Expression (3) is approximated by a sum

$$\tilde{d}_q = -i \sum_s \sqrt{\frac{(-2\pi i)^5}{\det(\tilde{S}_s)}} \langle \Phi_0(t_e) | x | \mathbf{p}_s + \mathbf{A}(t_e)/c \rangle \mathbf{E}_X(t_i) \langle \mathbf{p}_s + \mathbf{A}(t_i)/c | x | \Phi_0(t_i) \rangle e^{-iS_q(\mathbf{p}_s, t_e, t_i)} \quad (4)$$

over the saddle points $s = (\mathbf{p}_s, t_e, t_i)$ defined by

$$\int_{t_i}^{t_e} [\mathbf{p}_s + A(t'')/c] dt'' = 0 \quad (5)$$

$$[\mathbf{p}_s + A(t_i)/c]^2 / 2 = \omega_X - I_p \quad (6)$$

$$[\mathbf{p}_s + A(t_e)/c]^2 / 2 + I_p = q\omega \quad (7)$$

for $\tilde{S}_{(i,j)} = \partial_i \partial_j S$ where $i, j \in \{p_x, p_y, p_z, t, t'\}$. In case $\omega_X - I_p$ is positive all saddle points are real and, therefore, t_i and t_e are the ionization and recollision times, respectively, being also a solution of the classical equations of motion.

We analyze the condition required for (near-) bandwidth-limited emission of high harmonics. The time-dependent intensity of the emitted light bursts is given by

$$I(t) \propto \left| \sum_j \omega_{2j+1}^2 |\tilde{d}_{2j+1}| e^{-i \operatorname{Re} S_{2j+1}(\mathbf{p}_s, t_e, t_i)} e^{-i\omega_{2j+1}t} \right|^2 \quad (8)$$

where the phase $\operatorname{Re} S_q(\mathbf{p}_s, t_e, t_i) \approx \alpha_0 + \alpha_1(\omega_q - \omega_c) + \frac{1}{2}\alpha_2(\omega_q - \omega_c)^2 + \dots$ is crucial for the duration of the attosecond burst. We Taylor-expanded $\operatorname{Re} S_q(\mathbf{p}_s, t_e, t_i)$ about the central harmonic frequency ω_c . As outlined in [6, 7] the linear coefficient of the expansion, the group delay, is simply given by

$$\alpha_1 = \left. \frac{d}{d\omega_q} \operatorname{Re} S_q(\mathbf{p}_s, t_e, t_i) \right|_{\omega_q=\omega_c} = - \left. \operatorname{Re} t_e(\omega_q) \right|_{\omega_q=\omega_c} \quad (9)$$

because the saddle-point equations lead to vanishing partial derivatives. The linear chirp, the group-delay dispersion (GDD), is given by

$$\alpha_2 = - \left. \operatorname{Re} \frac{d}{d\omega_q} t_e(\omega_q) \right|_{\omega_q=\omega_c} \approx - \frac{\Delta t_e}{\Delta \omega_q} \quad (10)$$

being in first approximation responsible for the duration of the harmonic pulse. Hence, the pulse can be considered to be bandwidth-limited if the quadratic term in the Taylor-expansion fulfills the following demand $|\frac{1}{2}\alpha_2(\Delta\omega_q)^2| \ll 2\pi$. This yields a criterion for the classical recollision time window Δt_e resulting from the bandwidth of the trajectories:

$$\Delta t_e \ll \frac{4\pi}{\Delta\omega_q} = \frac{4\pi}{\alpha} \Delta t_p \quad (11)$$

with the bandwidth-limited pulse duration $\Delta t_p = \alpha/\Delta\omega_q$ and the parameter α of order of unity determined by the spectrum. This reflects that only an approximate condition $\Delta t_e \approx 0$ is necessary for bandwidth-limited HHG emission. In the previous section, we discussed in detail how the condition of simultaneous recollision of classical trajectories $\Delta t_e \approx 0$ can be fulfilled. We will show that it is sufficient to add only a few low-order Fourier components of the fundamental laser frequency to the laser field in order to achieve an optimized field which fulfills Eq. (11). Moreover, Eq. (11) evidences that the chirp compensation becomes more difficult for larger bandwidth.

In our classical and quantum descriptions, we used the common assumption [16–20] of neglecting the x-ray field for the continuum propagation of the electron. This means dropping the term $S_x(\mathbf{p}, t, t') = \int_{t'}^t d\tau ([p_x + A(\tau)/c] A_x(\tau)/c + \frac{1}{2}A_x^2(\tau)/c^2)$ in the definition of

$S_q(\mathbf{p}, t, t')$. We derive an approximate condition for the maximum electric field strength of the x rays being allowed that this approximation holds and that the HHG pulse duration is not influenced by the x-ray field. When evaluating S_x at the previously determined saddle points, the additional shift of the recollision times by the x-ray field is:

$$t_{e,x} = \frac{d}{d\omega_q} S_x(p_s, t_i, t_e) = \frac{\partial S_x}{\partial t_i} \frac{\partial t_i}{\partial \omega_q} + \frac{\partial S_x}{\partial t_e} \frac{\partial t_e}{\partial \omega_q} + \frac{\partial S_x}{\partial p_s} \frac{\partial p_s}{\partial \omega_q} \quad (12a)$$

$$\approx -[p + A(t_i)/c] A_x(t_i)/c \frac{\partial t_i}{\partial \omega_q} \approx -\frac{E_x}{\sqrt{\omega_X}} \frac{\partial t_i}{\partial \omega_q} \quad (12b)$$

In Eq. (12a) we dropped the second term because $\frac{\partial t_e}{\omega_q}$ is negligible due to the simultaneous recollision and the third term because it contains an integration over the heavily oscillating $A_x(\tau)/c$. Thus, the overall additional recollision time spread

$$\Delta t_{e,x} = \frac{E_x}{\sqrt{\omega_X}} \frac{\Delta t_I}{\Delta \omega_q} \ll \Delta t_e \quad (13)$$

has to be much smaller than the recollision time window to justify in our case the neglect of S_x .

In summary, bandwidth-limited HHG emission can be reached when the recollision time window determined by the classical dynamics in the laser field fulfills Eq. (11) and the x-ray field satisfies Eq. (2) and Eq. (13).

IV. GENERATION OF ATTOSECOND PULSES

In the following, we describe an implementation of the scheme. We start from an optimal field shape determined in the way described above with a fundamental frequency of $\omega = 0.06$ a.u. Then, we represent the field as a Fourier series and only take the $N_F = 8$ lowest frequency components of its spectrum into account. Following this procedure, the pulses in Fig. 2 a) (solid black line) were found. Its relevant classical trajectories ionized by a one-photon transition are shown as solid red lines. We can observe that $t_e(\omega_q)$ is approximately constant and, therefore, we can expect bandwidth-limited harmonic emission. We compare it to the traditional case of a sinusoidal laser field with similar parameters as in [1] (dashed lines). Evaluating the dipole moment within the SFA [Eq. (4)], we find the attosecond pulse generated by the fields in Fig. 2 (a). The results are shown in Fig. 2 (b). The duration of the pulse generated by our method (solid line) is 86 as at full-width half maximum (FWHM) only being slightly longer than a pulse generated by a sinusoidal field in combination with a

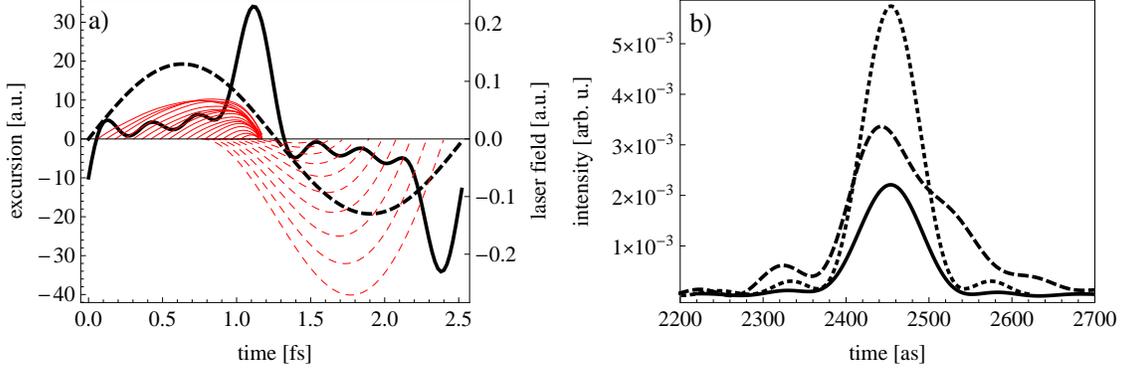


FIG. 2: a) Classical electron trajectories (red lines) are shown for two different field configurations (thick black lines): The dashed curves are for a conventional sinusoidal laser field whereas the full lines are for a shaped laser field consisting of $N_F = 8$ harmonics that is assisted by continuous wave x-rays ($\omega_x = 60$ eV, $I_x = 9 \times 10^{12}$ W/cm²) triggering ionization. b) Attosecond pulses emitted from the laser fields proposed in a). The solid line shows the intensity for the case of the proposed scheme whereas the dotted and dashed lines correspond to the attosecond pulses for a conventional field with a 40 eV bandwidth either with or without phase compensation, respectively.

perfect chirp compensation (78 as) (dotted line). The dashed line shows the uncompensated pulse with a duration of 130 as. In all three cases, the same frequency window between 60 eV and 110 eV was employed and the x-ray field ($\omega_x = 60$ eV, $I_x = 9 \times 10^{12}$ W/cm²) was chosen to have the same average ionization rates as by tunnel ionization in the sinusoidal field. The two pulses from the sinusoidal pulse were obtained from Neon using only the short trajectories whereas in our case He⁺ has to be employed. The laser frequency is in all cases $\omega = 0.06$ a.u. Although the single-atom emission rate of both examples are on the same order, the photon yield of our setup will be lower because the gas density is restricted by a maximum value of 5×10^{16} /cm⁻³ as determined later. We estimate an emitted photon number per half cycle of 10^7 in the traditional case (density 10^{19} /cm⁻³) and 10^3 in our case from a volume of $200 \mu\text{m} \times 200 \mu\text{m} \times 1$ mm.

So far, the attosecond pulse production with durations little below 100 as was considered. The time difference of the uncompensated (~ 130 as) and compensated (~ 80 as) pulses were moderate. Now, we discuss our proposal for future experiments where the bandwidth is of order of keV and Eq. (11) imposes a stronger constraint, demanding for a larger phase compensation over a larger frequency spectrum. In the following, we present two examples

with production of pulses below 10 as and 1 as, see Fig. 3.

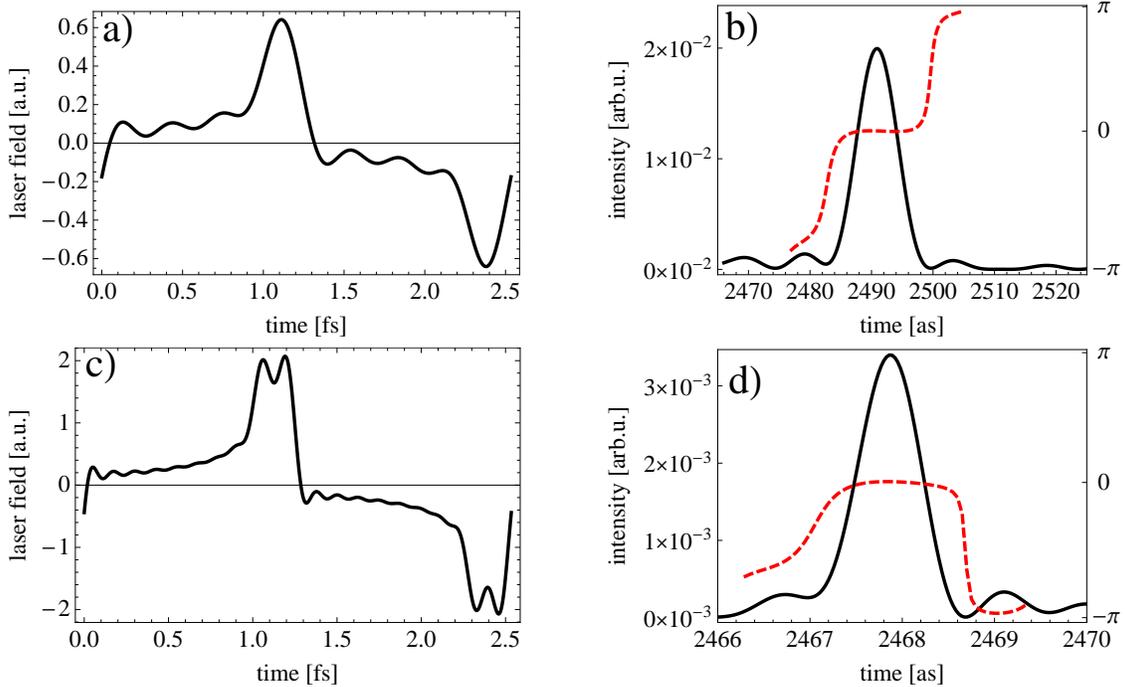


FIG. 3: a) shows the laser field composed of only 8 Fourier components that illuminates an Li^{2+} atom with $I_p = 4.5$ a.u.. The parameters of the additional x-ray field are indicated in the first row of Tab. I. The created 8 as pulse is shown in b). c) displays the laser field needed to create a pulse of 800 zs duration from Be^{3+} atoms with $I_p = 8$ a.u. and the parameters indicated in the second row of Tab. I. The respective pulse is shown in d). The dashed red lines are the temporal phases of the pulses.

The laser field strength and the x-ray frequency have to be increased compared to the example before to obtain a much larger bandwidth and are indicated in Tab. I. This way, attosecond pulses with a FWHM of 8 as and 800 zs are formed. The pulses are almost bandwidth-limited as can be seen from the constant phase (red dashed line) in the main part of the pulse. Here, the linear phase term given by the central frequency of the pulse was subtracted from the phase.

However, the photon yield emitted from the target is very low for several reasons: In the two examples, $\omega_X \gg I_p$ is valid because a much larger initial momentum is required, however, the ratio renders ionization inefficient. We limited the x-ray intensity according to Eq. (13). On the other hand, one-photon ionization exhibits a larger angular distribution

N_F	I_L [W/cm ²]	ω_x [eV]	I_X [W/cm ²]	$\Delta\omega_q$ [eV]	T_P [as]	ρ_{max} [cm ⁻³]	N_{ph}
8	10^{16}	218	3.5×10^{14}	470	8	2.5×10^{16}	10^1
20	10^{17}	996	1.4×10^{15}	4.9×10^3	0.8	7×10^{14}	10^{-6}

TABLE I: Parameters for the two examples in Fig. 3: N_F represents the number of Fourier components contained in the fundamental pulse, I_L its peak intensity, ω_X the x-ray frequency employed for ionization, E_X its field strength, $\Delta\omega_q$ the achieved HHG bandwidth, T_P the HHG pulse duration, ρ_{max} the estimated maximum gas density and N_{ph} an estimate of the emitted HHG photon number per half cycle emitted from a volume of $200 \mu\text{m} \times 200 \mu\text{m} \times 1 \text{mm}$ having the maximum density.

resulting in a large spread of the electron as soon as the initial momentum becomes larger. Finally, the gas density is limited to a small value which will be discussed shortly.

V. MACROSCOPIC EFFECTS

In the remainder of this paper, we discuss consequences of applying the scheme to a macroscopic gas target. Due to dispersion, the initially optimal pulse shape will be deformed during propagation. We estimate the impact of the dispersion by investigating the pulse shape after 1-dimensional propagation through a plasma of length L and refractive index $n_q = \sqrt{1 - \frac{4\pi\rho_e}{\omega_q^2}}$ where ρ_e is the electron density. In this case, each Fourier component of frequency ω_q propagates with the phase velocity $v_{ph} = c/n$ and an analytic expression can be obtained:

$$I(t) \propto \left| \sum_j \omega_{2j+1}^2 \tilde{d}_{2j+1} e^{-i\omega_{2j+1}[t+(n_1-n_{2j+1})L/c]} \right|^2 \quad (14)$$

The influence of the different atomic transition lines of the medium on the dispersion is omitted because the free electron background forms the largest contribution to the dispersion. When calculating the driving pulse shape of Fig. 3 a) after a propagation length of $L = 1 \text{mm}$, we find a maximum ion density of $5 \times 10^{16}/\text{cm}^3$ and $7 \times 10^{14}/\text{cm}^3$ to maintain a duration of the harmonic burst below 10 as and 1 as, respectively. Similarly, we can specify the precision of the phase of the different Fourier components that is required. The allowed fluctuations of the different components in terms of time delay is of the order of 25 as and 2.5 as, respectively, in agreement with the time delays caused by the plasma dispersion

discussed before. The sensitivity is lower for harmonics with lower energies.

Apart from causing a deviation from the optimized pulse form, dispersion can also lead to phase mismatching. Due to the rather low gas density, we do not expect a dramatic phase mismatch. To achieve phase matching, we propose either to exploit the geometry of the laser focus or to use quasi-phasematching schemes as employing a weak counterpropagating IR field [28, 29], weak static fields [30] or modulated wave guides [31–33].

So far, the recollision scheme was discussed in the spotlight of bandwidth-limited emission of attosecond pulses. The scheme can also be applied as a new type of pump-probe technique where the atom is excited or probed at a precise time by the recolliding electron. The recollision time can be controlled via shaping the driving pulse. Moreover, the spectral diversity of the simultaneously recolliding trajectories is an excellent condition for the observation of continuum-continuum harmonics [34].

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