Method of XUV attosecond pulse characterization using two-photon resonant Raman-type transition

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Abstract

We suggest a new method of attosecond pulse train measurement based on two-photon Raman-type resonant interaction of the XUV with an atom (Attosecond pulse reconstruction using Raman-type Transitions, ART). We show analytically and numerically that the attosecond pulse auto-correlation function and chirp can be measured using this process. Its resonant nature allows obtaining reasonable signal even for relatively low XUV intensity. The interpretation of the results is remarkably simple for the XUV generated by bi-circular field. The experimental realization of the method can be based either on measuring the excitation of the gas or on four-wave mixing fully optical scheme.

1. Introduction

Attosecond XUV pulse measurement is an important and challenging part of the attophysics. A number of methods for such measurements were suggested and realized. These include reconstruction of attosecond harmonic beating by interference of two-photon transitions (RABITT) [1–6], the attosecond streak-camera [7–10], complete reconstruction of attosecond bursts via frequency-resolved optical gating [11–13], spectral phase interferometry for direct electric-field reconstruction [14–17]. Another group of methods is based on auto-correlation measurement of an attosecond pulse [18–22]. In general, the advantage of the auto-correlation approaches is their directness due to the simplicity of the interpretation of the results: measuring the yield of a two-photon process as a function of the delay between two replicas of the attosecond pulse directly provides an auto-correlation function of the pulse. However, measurable yield of the process involving two UV photons requires relatively high intensity. This is the case both for the approach based on the non-resonant two-photon ionization [18] (the process of the two-photon ionization in the UV range was studied in [23, 24]), and that based on the dissociation of a molecule [19, 21, 22]. Using a resonant two-photon process can help to overcome the problem of low efficiency. In [25] two-photon resonant excitation of a doubly excited state of He atom was considered. However, this process involves only photons for which the sum of their energies equals the energy of the doubly excited state so this method can only be applied for a limited frequency range.

In this paper we suggest and study theoretically a new approach for the attosecond pulse train characterization, namely Attosecond pulse reconstruction using Raman-type transitions (ART). The idea of the method is illustrated in figure 1. We consider an interaction of an atom with a number of odd harmonics having frequencies $q\omega_0$, where $\omega_0$ is the fundamental laser frequency. The energy difference of the excited and the ground atomic states should be close to an even multiple of the laser photon energy (atomic units are used throughout):

$$E_2 - E_1 \approx \Delta q\omega_0.$$  \hspace{1cm} (1)

We suppose that the parity of the states is such that a two-photon transition from the ground to the excited state is allowed. Transitions to other bound states are assumed to be non-resonant and so they can be neglected.
The resonant condition (1) can be satisfied by choosing the gas medium and tuning the fundamental frequency. Moreover, for transitions in atomic gases $\Delta q$ is a rather high number (typically from 4 to 10), so a relatively small tuning of $\omega_0$ allows satisfying the condition. Note that a much wider tuning of the fundamental frequency is achieved using mid-IR OPA laser systems for HHG in gases [26–29] and plasma plumes [30–33].

The atom is initially in the ground state and under exposure to XUV some population appears in the excited state due to two-photon Raman-type transitions via continuum. The complex amplitude of the excited state is a sum of contributions due to different pairs of harmonics. (Only the harmonic pairs providing the Raman-type resonance are shown in figure 1; certainly, there are other pairs of harmonics which are dramatically ($2\omega_0$ and more) detuned from the Raman-type resonance and thus they do not contribute to the excitation of the upper state.) The phases of these contributions are determined in particular by the phase difference between the harmonics in the pair. Thus the population of the excited state can be used to evaluate the harmonic phase differences. Very roughly, if all the harmonics are in phase (i.e. in case of a train of Fourier-limited attosecond pulses) their contributions add constructively resulting in a high population of the excited state; if they are not in phase (chirped pulses) the population is lower.

Our method using Raman-type transition has no upper limit for the frequencies of the harmonics involved in the process (because it is applicable for the harmonic frequencies whose difference is equal to the transition frequency). This is an important advantage with respect to the previously suggested methods using two-photon ionization or excitation where the sum of the harmonic frequencies was limited by the ionization potential or the transition frequency and thus the upper frequency of the measured harmonic is limited.

In this paper we develop a theoretical approach connecting the attopulse properties and the excited state population after the passage of the attopulse train. The analytical results are compared with those based on the numerical solution of 3D time-dependent Schrödinger equation (TDSE) for a model atom in an external field. The harmonic intensities required for the signals to be measurable are estimated. We show that the XUV intensity at the GW cm$^{-2}$ level is sufficient for the attosecond pulse measurement using the suggested method. The moderate required intensity in conjunction with the absence of the upper limit for the harmonic frequency and the simplicity of the measurement defines the perspectives of our method such as spatially-resolved characterization of focused attosecond pulses. Finally, we discuss the limitations of our method as well.
2. Theory

2.1. General consideration

The laser resonant coupling of two atomic states via the continuum was widely studied experimentally and theoretically in connection with the effects of laser-induced continuum structures, Raman-type transitions, interference stabilization, etc [34–47]. A generalization of this approach for the multiple-harmonic field case is given in appendix. Below we assume that the detuning from the resonance is small, namely that:

\[ |\Delta q + D_{q+\Delta q}| T \ll 1, \]

where \( T \) is the full irradiation time and \( D_{q+\Delta q} \) is the detuning of the Stark-shifted and ionization-broadened levels from the exact Raman-type resonance given by equation (A.8). Using inequality (2) we find from equation (A.7) that the population of the excited state \( w_2 \) after the irradiation by several harmonics with complex amplitudes \( \tilde{E}_q \) is

\[ w_2(T) = |c_2(T)|^2 \approx \frac{T^2}{16} \sum_q E_q^* E_{q+\Delta q} \alpha_{21}^{(q,q+\Delta q)} \],

where \( \alpha_{21}^{(q,q+\Delta q)} \) is the two-photon transition matrix element (also denoted as the off-diagonal element of the complex polarizability tensor, see [47]) given by equation (A.6).

Figure 2 shows the frequency dependence of the absolute value and the phase of \( \alpha_{21}^{(q,q+\Delta q)} \), calculated for the ground 1s and the excited 2s states in hydrogen for \( \Delta q = 8 \) using equation (A.11).

Below we focus on the practically important region of harmonic frequencies where \( q \omega_0 = |E_1| \) (the electronic energy in continuum) is comparable to the ionization energy \( |E_1| \). This corresponds to \( q \omega_0 \approx 1 \div 3 \) in atomic units. We can see in figure 2 that in this region the phase of \( \alpha_{21}^{(q,q+\Delta q)} \) is almost constant and its absolute value can be well approximated by the following dependence:

\[ |\alpha_{21}^{(q,q+\Delta q)}| \approx \frac{C}{[q(q + \Delta q)]^{7/4}}, \]

where \( C = 0.13/\omega_0^{7/2} \) in atomic units.

Substituting this approximation in equation (3) we obtain:

\[ w_2 = \left| C \frac{T^2}{16} \sum_q \frac{E_q^* E_{q+\Delta q}}{[q(q + \Delta q)]^{7/4}} \right|^2. \]

Thus the fact that the phase of \( \alpha_{21}^{(q,q+\Delta q)} \) is almost constant makes possible the simplified understanding of the suggested method, mentioned in the introduction: when all the harmonics are in phase, their contributions add constructively, and the final population of the excited state is high. Note that in the RABITT approach [1] a similar (but not identical) phase shift also appears, it is usually denoted as the atomic phase.

2.2. Linearly polarized harmonics

To make the auto-correlation measurements the initial signal (its spectrum is denoted below as \( \tilde{E}_q \)) is split into two replicas with a controllable time delay \( \Delta \tau \) between them. After this the spectrum is:

\[ Figure 2. Absolute value (blue) and phase (red) of the two-photon transition matrix element \( \alpha_{21}^{(q,q+\Delta q)} \). Dotted black line shows the approximation of the absolute value given by equation (4). \]
\[ \xi_q = \frac{1}{2} \tilde{\xi}_q [1 + \exp(i q \omega_0 \Delta t)]. \]  

(6)

Substituting these harmonic amplitudes in equation (3) we have:

\[ w_2(\Delta t) = \frac{T^2}{16} \left| \sum_q \alpha_q^{\Delta q} \tilde{\xi}_q^{\Delta} f_q(\Delta t) \right|^2, \]  

(7)

where

\[ f_q(\Delta t) = \frac{1}{4} [\exp(i q \omega_0 \Delta t) + \exp(-i(q + \Delta q) \omega_0 \Delta t) + 1 + \exp(-i \Delta q \omega_0 \Delta t)]. \]  

(8)

Note that the four terms in this equation originate from the two terms in equation (6) corresponding to the two trains (two replicas). Thus, the first two terms in equation (8) correspond to the processes where the two photons come from different trains and the last two terms correspond to those where the photons come from the same train.

Substituting \(|\alpha_q^{\Delta q}\)| given by approximation (4) in equation (7) we have:

\[ w_2(\Delta t) = \frac{C^2 T^2}{16} \left| \sum_q \tilde{\xi}_q^{\Delta q} f_q(\Delta t) \left[ \frac{1}{|q(q + \Delta q)|^{3/4}} \right] \right|^2. \]  

(9)

Below we discuss the behavior of \(w_2(\Delta t)\) for different regions of \(\Delta t\). Note that since \(w_2(\Delta t)\) is periodic with the periodicity of the laser half-cycle, we can consider only the interval \(\Delta t < \pi/\omega_0\).

We assume that the train of the XUV pulses is formed by approximately \(\delta q\) harmonics, and therefore the duration of every pulse is \(t_D = \bar{t}(\Delta q \omega_0)\). Let us rewrite equation (9) using equation (8) as

\[ w_2(\Delta t) = \frac{C^2 T^2}{16} \left| a_1(\Delta t) + a_2(\Delta t) + b(\Delta t) \right|^2, \]  

(10)

where

\[ a_1(\Delta t) = \frac{1}{4} \sum_{q = -\delta q}^{\delta q} \tilde{\xi}_q \tilde{\xi}_q^{\Delta q} \exp(i q \omega_0 \Delta t), \]  

(11)

\[ a_2(\Delta t) = \frac{1}{4} \exp(-i \Delta q \omega_0 \Delta t) \times \sum_{q = -\delta q}^{\delta q} \tilde{\xi}_q \tilde{\xi}_q^{\Delta q} \exp(-i q \omega_0 \Delta t), \]  

(12)

\[ b(\Delta t) = \frac{1}{4} [1 + \exp(-i \Delta q \omega_0 \Delta t)] \times \sum_{q = -\delta q}^{\delta q} \tilde{\xi}_q \tilde{\xi}_q^{\Delta q} \exp(-i q \omega_0 \Delta t). \]  

(13)

For the long delays \(\Delta t \gg \tau\) the sums (11) and (12) vanish because of the fast oscillations of the exponents. Thus, we assume that

\[ w_2(\Delta t) \bigg|_{\Delta t \gg \tau} = \frac{w_2(0)}{16} \left| 1 + \exp(-i \Delta q \omega_0 \Delta t) \right|^2 = \frac{w_2(0)}{8} [1 + \cos(\Delta q \omega_0 \Delta t)] \]  

(14)

oscillates from 0 to \(w_2(0)/4\) with the period of the resonant transition \(T_{21} = 2\pi/(\Delta q \omega_0)\). This result can be understood as follows: for \(\Delta t \gg \tau\) the attosecond pulses from different replicas do not superimpose. Thus some population is transferred to the excited state by the attopulse from replica 1, then after time \(\Delta t\) the same amount of population is transferred by the attopulse from replica 2. Depending on the phase \(\Delta q \omega_0 \Delta t\) these two contributions interfere constructively or destructively.

Calculating \(w_2(\Delta t)\) for the short delays we assume that

(i) the delay is shorter than the transition period: \(\Delta t \ll T_{21}\) and

(ii) the pulse spectral width is much broader than the transition frequency, but much less than the carrier frequency:

\[ \Delta q \ll \delta q \ll q. \]  

(15)
The first part of this inequality means that the pulse duration is shorter than the resonant transition period:

$$\tau \ll T_{21}.$$  \hspace{1cm} (16)

Also the first part of inequality (15) means that we can assume \(\hat{\xi}_{q+\Delta q} \approx \hat{\xi}_q\) in sums (11)–(13). The second part of (15) means that calculating these sums we can assume \(q'(q+\Delta q) \approx q^2\).

Using the fact that the pulse auto-correlation function \(A(\Delta t)\) is an inverse Fourier transform of its power spectrum \(A(\Delta t) = \sum_q \hat{\xi}_q \hat{\xi}_q^* \exp(iq\omega_0 \Delta t)\) (the simplest version of the Wiener–Khinchin theorem) we present equation (10) as:

$$w_2(\Delta t)|_{\Delta t \leq T_{21}} = \frac{C^2 T^2}{64 q^2} \left[\text{Re} A(\Delta t) + A(0)^2\right].$$ \hspace{1cm} (17)

Under the used approximations from equation (9) we have \(w_2(0) = C^2 T^2 A(0)^2/(16 q^2)\). Thus equation (17) can be rewritten as:

$$w_2(\Delta t)|_{\Delta t \leq T_{21}} = w_2(0) \left[\frac{\text{Re} A(\Delta t)}{A(0)} + 1\right]^2.$$ \hspace{1cm} (18)

Note that under the condition (16) the regions of applicability of equation (14) and equation (18) superimpose.

Thus, for \(\Delta t, \tau \ll T_{21}\) and \(\delta q \ll q\) there is a direct correspondence between the measured value \(w_2\) and the auto-correlation function. If these conditions are not fulfilled, the correspondence is not so straightforward, and it is more convenient to reconstruct the pulse parameters directly from \(w_2(\Delta t)\) (without reconstructing \(A(\Delta t)\)).

2.2.1. Circularly polarized harmonics

When the harmonics are generated by a bi-circular field, they are circularly polarized, namely when harmonics are generated by fields with frequencies \(\nu_q\omega_0\) and \(\pm \nu_q\omega_0\) circularly polarized in opposite directions, harmonics with frequencies \(q \nu_q\omega_0 = m(r + s)\omega_0\) are generated with helicities of \(\pm 1\) (in the former equation corresponds to \(+\) in the latter one), \(m = 0, 1, 2, \ldots\) (see, for instance, recent review [18]). We consider the simplest case of \(r = 1\) and \(s = 2\) (the fundamental field and its second harmonic), so that harmonics \(q = 3m + 1\) have helicity \(+1\), harmonics \(q = 3m - 1\) have helicity \(-1\), and harmonics \(q = 3m\) are absent. We assume that both the ground and the excited state are \(s\) states. Then the transition is allowed only for harmonics \(q = 3m + 1\) having the same helicity. Now let us assume that the replica 2 is not only delayed with respect to the replica 1, but also has the opposite helicity, and that \(\Delta q\) is a multiple of 4. In this case the transition is possible only if harmonics from different replicas are involved. This allows the background-free measurement of the auto-correlation function, as we will show below.

Let us denote harmonics from replica 1 and 2 as \(\hat{\xi}_q^{(1)}\) and \(\hat{\xi}_q^{(2)}\), respectively. If \(\Delta q = 6n - 2, n = 1, 2, \ldots\) then the harmonics with orders \(q = 3m + 1\) are involved in the process (because for \(q = 3m - 1\) the harmonic \(q + \Delta q\) is absent) and for \(\Delta q = 6n + 2, n = 0, 1, 2, \ldots\) the harmonics with orders \(q = 3m - 1\) are involved. In both cases the harmonic from the second replica is \(\hat{\xi}_q^{(2)} = \hat{\xi}_q^{(1)} \exp(iq\omega_0 \Delta t)\). In equation (10) terms \(a_1\) and \(a_2\) are the same as for the linearly-polarized field, and the term \(b\) is absent. So, under condition (15), instead of (18), we have the following expression:

$$w_2(\Delta t) = w_2(0) \left[\frac{\text{Re} A(\Delta t)}{A(0)}\right]^2.$$ \hspace{1cm} (19)

Thus the direct correspondence between the population of the excited state and the auto-correlation function of the attosecond pulse is remarkably clear for the XUV generated by a bi-circular field.

2.3. Numerical TDSE solution

The 3D TDSE is solved numerically for a single-electron model atom in an external linearly-polarized ultraviolet field. The atomic potential is a soft-Coulomb one:

$$V(R) = -1/\sqrt{R^2 + R_{sm}^2},$$ \hspace{1cm} (20)

where \(R_{sm} = 0.15\) is chosen to reproduce the xenon ionization energy. The numerical approach is described in [49]. The energy of the transition between the ground state and the lowest excited state with the same parity (so that a two-photon transition is allowed) is 8.72 eV. We use the harmonics of the 1.14 \(\mu\)m radiation which provide the Raman-type resonances with \(\Delta q = 8\).
3. Results

3.1. Two-photon Raman excitation

Let us consider now an XUV field having the form of a train of chirped Gaussian pulses. Its spectrum is written as:

\[
\tilde{\mathcal{E}}_\omega = \tilde{\mathcal{E}} \exp \left( \frac{q^2 - q_0^2}{\sigma^2} \right) (-1 + i\sigma),
\]

(21)

where \( q_0 \) is the carrier frequency, \( \tilde{\mathcal{E}} \) is its amplitude. Parameter \( \sigma \) describes the pulse chirp: \( \sigma = 0 \) for a Fourier-limited pulse, and positive and negative \( \sigma \) correspond to positive (growing instant frequency) and negative chirp, respectively. The pulse duration (the full width at half maximum intensity) is \( \tau = \tau_{\text{FL}} \sqrt{\sigma^2 + 1} \), where \( \tau_{\text{FL}} = 2\sqrt{2\ln(2)}/(\delta q_0) \).

In the analytical studies we use a spectrum given by equations (21) and (6) which corresponds formally to an infinite field. In the numerical TDSE solution we have to use the field which has a limited duration and some temporal envelope. We use the following expression for the temporal dependence of the XUV field:

\[
E(t) = \frac{1}{2}E_0(t)\text{Re} \sum_q \tilde{\mathcal{E}}_q [\exp(-iq_0 t) + \exp(-iq_0 (t - \Delta t))],
\]

(22)

where \( E_0(t) \) is a slowly varying envelope. This envelope describes the turn-on and the turn-off of the XUV field on the femtosecond time scale, thus it can be understood as the envelope of the train of the attosecond pulses.

In the calculations discussed below we use linearly-polarized XUV pulses with the carrier frequency \( q_0 = 3500 \). This corresponds to the carrier period of 109 as.

Figure 3 shows the normalized population of the excited state calculated numerically and analytically using equations (7) and (9). One can see the good agreement between the ‘exact’ equation (7) and the approximation (9). Moreover, the analytical results agree with the numerical ones. This confirms the acceptability of the two-level approach neglecting all the bound states except the two involved in the resonance. Also this shows that the one-photon ionization does not affect the results dramatically. Let us discuss the latter issue in more detail. In our calculations the intensity of the central (i.e. most intense) harmonic in the XUV spectrum is \( 3 \times 10^{12} \) W cm\(^{-2}\) and the duration of the attosecond pulse train is 35 fs. Under these conditions the depletion of the ground and the excited states due to the one-photon ionization is negligible: the ground state ionization probability is less than 0.1 and for the excited state it is even lower. So the ionization damping of the excited state does not influence the results. For a higher XUV intensity or a much longer irradiation time the depletion due to the photoionization could play crucial role. However, the study of such intense harmonic fields is outside the scope of this paper.

The inset in figure 3 shows that for long delays the population oscillates from 0 to \( w_2(0)/4 \), in agreement with equation (14). The period of these oscillations under our conditions is 476 as. For shorter delays there is a pronounced maximum for values close to the carrier cycle. Figure 4 shows that the shorter is the pulse, the lower is the maximum.
The calculated dependence, as shown in figure 4, is different for the pulses having the same spectral width ($\delta q = 14.1$ corresponding to $\tau_{FL} = 100$ as) but different duration values. Thus the method is in fact sensitive not only to the spectral, but also to the temporal properties of the pulses. Moreover, the dependence is different for the pulses with the same duration ($\tau = 150$ as) but different chirp. This means that the method allows characterization of both the duration and the chirp.

In our calculations the condition (15) is fulfilled. So, having the experimentally measured population as a function of the delay we can reconstruct the auto-correlation function. (Note that for the circularly polarized fields considered in section 2.2.1 this reconstruction is remarkably simple, see equation (19).) However, it is well known that the field cannot be uniquely defined from the auto-correlation function; one has to assume some model (for instance, a Gaussian pulse) for the field. Having this in mind, the reconstruction of the auto-correlation function itself is hardly useful. One should better use the experimentally measured harmonic intensities and assume some model describing their phases, then calculate the population as a function of the delay using equation (9) and fit the model parameters to reproduce the measured dependence. For instance, one could characterize the harmonic phases with a model including linear and quadratic chirps with certain values (the model’s fitting parameters).

Experimentally the chirp of the attosecond pulses can be controlled within a certain range when the XUV passes a dispersing filter [5]. In particular, some filter dispersion is necessary to compensate the ‘natural’ chirp of the attosecond pulses (the chirp which is intrinsically present when the train of attosecond pulses is obtained from HH). Such compensation can be easily controlled via measuring the population of the excited state (without splitting the attosecond pulse train into two replicas) as a function of the additional chirp added by the filter. From equation (9) we find that the population as a function of the chirp of the pulse given by equation (21) can be approximated as:

$$w_2(\sigma) \propto |\hat{E}|^4 \exp\left[-(\sigma/\sigma_{th})^2\right].$$

where $\sigma_{th} = \delta q/\Delta q$. Thus, assuming that the quadratic chirp is negligible, the Fourier-limited pulse is obtained under the filter parameters maximizing $w_2/|\hat{E}|^4$ (note that the UV intensity $|\hat{E}|^2$ also depends on the filter parameters because of the filter absorption). When a Fourier-limited pulse is obtained, its duration can be found from the spectral width.

The other parameter which can be varied in the considered Raman-type transition is the detuning from the resonance:

$$\Delta = E_2 - E_1 - \Delta q \omega_0.$$

It can be controlled via finely tuning the fundamental frequency $\omega_0$. In the analytical theory presented in section 2 we assumed that the detuning is small, namely, that inequality (2) is satisfied (note that if the Stark shift and the ionization width is negligible then $D_{q+\Delta q} = \Delta$, see appendix for more details). To consider the non-negligible detunings we use the numerical TDSE solution. The results are shown in figure 5. The spectral width of the pulses corresponds to $\tau_{FL} = 100$ as, the central frequency is $\bar{q} \omega_0 = 50 \omega_0$ and the intensity of this spectral
component is $3 \times 10^{12} \text{ W cm}^{-2}$ (here we also do not split the XUV pulse into two replicas). One can see that the positive and the negative chirps lead to opposite shifts of the maximum in this dependence, so the presence of the chirp in the attosecond pulse can be recognized via the shift of the maximum from zero detuning.

3.2. Four-wave mixing

Up to now we assumed that the upper state population after the UV irradiation is measured to study the auto-correlation. The other approach to realize our concept can be a fully optical scheme using some nonlinear optical process. However, the direct one-photon transition from the upper to the ground state is forbidden, that is why the additional field should be used to achieve the nonlinear optical process involving (an intermediate) resonance. So this scheme should be based on a four-wave-mixing process. Below we consider an experimental technique where the gas having the resonant transition is irradiated with the two replicas of the attosecond train and a (relatively weak) IR field (note that ionization by combining optical and bichromatic XUV pulses was considered recently in [50]). A pair of high harmonics and IR field generate the UV at the frequency of a lower-order harmonic as it is shown in the inset to figure 6 with green and orange arrows. The phase of the generated field is controlled by the dephasing between the high harmonics in the pair. Other pairs of high harmonics with the orders differing by $\Delta q$ provide their contributions to the fields at the frequencies $(\Delta q \pm 1)\omega_0$. The further line of reasoning is the same as in the Introduction: if the dephasing of all the HH pairs is the same, their...
contributions add constructively, so the generation efficiency is high; if the dephasings are different, the efficiency is lower.

Below the four-wave-mixing process is studied numerically. Namely, we solve numerically the TDSE for an atom in external fields to find the electronic wavefunction $\psi(r, t)$ and calculate the quantum–mechanical average of the dipole moment:

$$P(t) = -\langle \psi(r, t)|r|\psi(r, t)\rangle,$$

see [49, 51] for more details.

Figure 6 shows the spectrum of this atomic response calculated for the two cases: the atom is irradiated by

1. the IR field only and
2. the IR field, H19 and H27 (below for brevity we denote 19th harmonic as H19 and so on). In the second case we can see the pronounced spectral components at frequencies $\omega_7$ and $\omega_9$ (here we denote $\omega_{q} = \bar{q}\omega_0$) which can be attributed to the third-order nonlinear process $H27 - H19 \pm IR$. Though the nonlinear optical processes in the UV domain are usually weak, here the process is resonant, so the polarization at the low-order harmonic frequency is not so low. Namely, it is much higher than the polarization at the tripled IR frequency due to the non-resonant third-order nonlinearity. The process involving the resonance is non-stationary, so the responses at $\omega_7$ and $\omega_9$ frequencies depend not only on the driving fields’ intensities, but also on the pulse durations. In the TDSE solution we used 20 fs pulses (for all frequencies). For this pulse duration the calculated effective resonant third-order nonlinearity $\chi^{(3)}(\omega_7 = \omega_9 - \omega_0)$ is approximately 4 times higher than the non-resonant one for the IR field only $\chi^{(3)}(\omega_3 = 3 \times \omega_0)$. Note that H7 and H9 can also be generated in both cases due to resonant 9th order process $\omega_{7(9)} = 8 \times \omega_0 - \omega_0$ and $\omega_{9(9)} = 8 \times \omega_0 + \omega_0$, respectively. This process is the only (resonant) one which leads to H7 and H9 generation in the case of one-color IR field. Figure 6 shows that the polarization at H7 and H9 in this case is much weaker than in the three-color field (though the IR intensity is higher for the one-color case), so this process is weak and thus should not affect the auto-correlation measurement.

Using numerical TDSE solution we have verified that for the used intensities the third-order nonlinearity can be described within the perturbation approach (in agreement with [52]). Moreover, we have checked that the behavior of nonlinearity $\chi^{(3)}(\omega_{2q\pm1} = \omega_q \pm \Delta\omega$: for all frequencies $\Delta\omega = \omega_q - \omega_{2q}$ is very similar to that of $\chi^{(3)}(\omega_{2q\pm1})$ (this is natural because the resonant part of $\chi^{(3)}(\omega_{2q\pm1})$ is defined by the upper state amplitude). Thus the conclusions made above for the auto-correlation measurement using upper state population are directly generalized for the method based on the four-wave mixing.

4. Discussion

In the numerical TDSE solution we used quite a high harmonic intensity to obtain substantial population of the excited state. Namely, the intensity of the central harmonic $\bar{q}$ in every replica was $3 \times 10^{12}$ W cm$^{-2}$ (note that experimentally such (or even higher) harmonic intensities can be obtained [18, 20, 22, 53–55], see also [56] for a review) and duration of the train was 35 fs. The highest upper state population was about $10^{-3}$. Experimentally populations several orders of magnitude lower can be measured. Namely, let us assume that the XUV is focused in a gas target with an atmospheric pressure, the XUV focal volume is 0.1 mm$^3$ and that $10^6$ excited atoms are sufficient for the measurement (for some methods of the excited atoms registration the sensitivity can be orders of magnitude better, see below). Taking into account the quadratic dependence of the excited state population on the XUV intensity we find that the required harmonic intensity is $1.5 \times 10^7$ times lower than the one used in our calculations, thus it is $2 \times 10^9$ W cm$^{-2}$.

The experimental realization of our method can be relatively simple because it does not require the photoelectron spectrum measurement which is necessary in several other methods of the attosecond pulse characterization. Experimentally, the population of the excited state after the passage of the attosecond pulse train can be measured via the fluorescence from this state (to the ground or some other state). For measuring the metastable state population the secondary emission from an upper selectively excited state can be detected. Another method of the excited atoms detection is described in [57, 58]. The method demonstrates very high sensitivity, namely even single excited atoms can be detected. Moreover, the spatial distribution of the excited atoms was measured in [57]. Application of this measurement in our approach will make possible a spatially-resolved characterization of focused attosecond pulses. The order-dependent high harmonic wavefronts measured in [59, 60] suggest that one can expect a non-trivial spatio-temporal structure of the focused XUV field.
Estimating the four-wave mixing efficiency is more complicated because it depends on the phase-matching. However, the fact that the nonlinearities $\chi^{(3)}(\omega_3)$ and $\chi^{(3)}(\omega_{\Delta q\pm 1})$ are comparable shows that using the four-wave mixing should be feasible and its efficiency should be comparable to that of the well-known process of the third-order harmonic generation by IR field caused by $\chi^{(3)}(\omega_3)$. This is a remarkable result because the nonlinearities in the UV range are usually much weaker than in the IR range. Note that though nonlinearity $\chi^{(3)}(\omega_{\Delta q\pm 1})$ involves an intermediate resonance, there is no resonant absorption at the generated frequency $\omega_{\Delta q\pm 1}$ as it is usual for the four-wave mixing processes.

The calculations presented in the paper show that the method is applicable for typical durations and carrier frequencies of the attosecond pulses. Let us discuss in more detail the requirements for these parameters ensuring the applicability of the method. The first requirement is that the transition frequency should be less than the attosecond pulse spectral width: $\Delta q \ll \delta q$. If this is not the case, only harmonics from the spectral wings participate in the Raman-type transition, thus the central part of the spectrum is not characterized. This requirement is usually well-satisfied because the transition frequency is less than the ionization energy and the attosecond pulse spectrum is usually broader. The other requirement originates from the fact that harmonic pairs from the blue and red parts of the attosecond pulse spectrum should provide comparable contributions to the upper state population. Having in mind the independence (4) we find that the spectral width should be less than the central frequency: $\delta q < \bar{q}$. Thus we derive the requirement which is similar to (15), but less stringent:

$$\Delta q < \delta q < \bar{q}. \tag{25}$$

Note that the second requirement is not satisfied for few-cycle attosecond pulses for which $\delta q \approx \bar{q}$. However, there are many cases when it is satisfied, for instance, for attosecond pulse generation using resonantly-enhanced harmonics [61].

As we mentioned in the introduction, the resonant condition (1) can be satisfied by tuning the fundamental wavelength in the OPA laser system used for HHG. The other option could be using the Ti–Sapp laser fundamental frequency for which the transition from the ground to the Rydberg states in argon corresponds to $\Delta q = 10$. Note that the analysis in this case is less straightforward because several upper levels can be populated.

5. Conclusions

Thus in this paper we suggest and investigate analytically and numerically a new method for attosecond pulse train characterization: attosecond pulse reconstruction using Raman-type transitions (ART). Being based on two-photon resonant interaction of the XUV with an atom, this method assumes fine tuning of the fundamental wavelength so that an even number of the fundamental quanta are in resonance with the transition frequency. To find the attosecond pulse auto-correlation one should split the attosecond pulse train into two replicas and measure the upper state population as a function of the delay between them; under certain conditions the auto-correlation function can be directly reconstructed from this dependence. The interpretation of the results is remarkably simple for the XUV generated by bi-circular field. The sign of the chirp can be measured separately by varying the detuning from the resonance. Even without splitting the attosecond pulse train into two replicas (which could be experimentally challenging) our approach can be utilized to obtain train of the Fourier-limited pulses: the latter can be obtained when the train passes the dispersive filter; we show that the proper filter can be found measuring the upper state population.

The suggested method can be practically realized via measuring the excitation of the gas by the fluorescence signal. Alternatively, one can realize the four-wave mixing scheme involving the same Raman-type transition.

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Appendix

Figure 1 shows the schematic of transitions in a two-level atomic system in the multi-harmonic field

$$E(t) = \text{Re}\sum_{q} E_{q} e_{q}\exp(-i\omega_{q} t), \tag{A.1}$$

where $E_{q}$ is the complex field strength amplitude in a set of the odd harmonics $\omega_{q} = q\omega_{0}$ with the polarization vector $e_{q}$.
In the framework of RWA the solution of TDSE for the wavefunction $\Psi(t)$ transforms (after eliminating non-resonant bound and continuum states) to the pair of coupled equations for the population amplitudes $c_n(t) = \langle n | \Psi(t) \rangle \exp(iE_n t)$ of the stationary atomic states $| n = 1 \rangle$ and $| n = 2 \rangle$ with the eigenenergies $E_n$

$$\left( \frac{d}{dt} - H \right) \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = 0 \tag{A.2}$$

for the initial conditions $c_1(0) = 1$ and $c_2(0) = 0$.

The effective non-Hermitian two-level Hamiltonian $H$ in (A.2) is

$$H = \frac{1}{4} \left( \sum_{q'q} \exp\left[i(q' - q)\omega_1 t\right] E_{q'} E_{q}^* \alpha_{12}^{(q'q)} + \sum_{q'q} \exp\left[-i\Delta_q q' t\right] E_q E_{q'}^* \alpha_{21}^{(q,q')} \right) \tag{A.3}$$

where

$$\Delta_q q' = \omega_21 - (q' - q)\omega_0 \tag{A.4}$$

is the Raman transition frequency detuning from the resonance for the $q$ and $q'$ harmonics, $\omega_{nm'} = E_n - E_{n'}$ is the atomic level energy difference (transition frequency),

$$\alpha_{nm}^{(q,q')} = \sum_{n'} \frac{(n|d_{eq'}|n') \langle n'|d_{eq}^*|n \rangle + \langle n|d_{eq}^*|n' \rangle \langle n'|d_{eq'}|n \rangle}{\omega_{n'n} + i\omega_q - \omega_q - 0i} \tag{A.5}$$

is the dynamical polarizability of the states $n = 1$ and $2$ in the field of the $q$ and $q'$ harmonics, $d$ is the atomic dipole operator, the set of intermediate states $n'$ includes discrete levels and continuum states,

$$\alpha_{21}^{(q,q')} = \sum_{n} \frac{(2|d_{eq'}|n) \langle n|d_{eq}^*|1 \rangle + (2|d_{eq}^*|n) \langle n|d_{eq'}|1 \rangle}{\omega_{n1} + i\omega_q - \omega_q - 0i} \tag{A.6}$$

is the two-photon transition matrix element, and $\alpha_{12}^{(q'q)} \approx \alpha_{21}^{(q,q')}$ is obtained by interchange $1 \leftrightarrow 2$ and $q \leftrightarrow q'$ in (A.6).

Adiabaticity of the effective Hamiltonian (A.3) corresponds to neglecting contributions from terms with high frequency oscillations during the pulse. Thus, for pulse trains of several tens of femtoseconds only $q' = q$ and $q' = q + \Delta_q q$ should be accounted for diagonal and non-diagonal elements, correspondingly, in (A.3).

Note, that for spherically symmetric states $|1\rangle$ and $|2\rangle$ harmonic polarization dependence in (A.5) and (A.6) reduces to the scalar product of corresponding vectors, while dipole matrix elements transform to the reduced ones.

The imaginary part in (A.5) and (A.6) emerges from the pole in energy integrals over the continuum states (indicated by zero shift from the real energy axis). The real and the imaginary parts in (A.5) correspond, respectively, to the level energy shift and photoionization width by the field ($H_{nm} = \Delta E_n - \frac{i}{2} \Gamma_n$), while the phase of the complex transition amplitudes appeared in (A.6) should be accounted for in (A.3) together with the phase difference of corresponding harmonics.

Equations (A.2) can be approximately solved omitting the coupling matrix element $H_{12}$ in (A.3):

$$c_1(t) \approx \exp(-iH_{11} t) \quad \text{and} \quad c_2(t) \approx \exp(-iH_{22} t) \times \sum_q \frac{\exp(iD_q q' t) - 1}{4D_q q'} E_q^* E_{q'} \alpha_{21}^{(q,q')} \tag{A.7}$$

where $q' = q + \Delta q$,

$$D_q q' = \Delta_q q' + H_{22} - H_{11} \tag{A.8}$$

is the field-induced level energy difference in the complex plane.

In the case of the $1s$ and $2s$ coupled states of hydrogen the scalar values of (A.5) and (A.6) can be obtained analytically using the Coulomb Green’s function (see, e.g., [62])

$$\alpha_{q,q'}^{(q,q')} = \frac{1}{\omega_q} \left[ T_n(\nu_+) + T_n(\nu_-) - 1 \right], \tag{A.9}$$

where $\nu_+ = \sqrt{2(\omega_q + E_n)}$, $\nu_- = \sqrt{2(\omega_q - E_n)}$, 

$$T_n(\nu) = \frac{2n^2\nu^2}{(2 - \nu)(n + \nu)^2} \times \text{$_2F_1$} \left( 1, 1 - \nu, 3 - \nu, \frac{n - \nu}{n + \nu} \right)^2, \tag{A.10}$$

and $\text{$_2F_1$}(a, b; c; x)$ is the hypergeometric Gauss function [63].
Similarly,
\[ \alpha_{21}^{(qq')} = -\frac{1}{\omega_q \omega_{q'}} [M(\nu_q) + M(\nu_{q'})], \]
where \( \nu_q = \sqrt{2(\omega_q + E_i)} \), \( \nu_{q'} = \sqrt{2(\omega_{q'} - E_i)} \), and
\[ M(\nu) = \frac{32\sqrt{2}}{81} \frac{\nu}{2 - \nu} \times \left[ 1 - \frac{4}{2 + \nu} \right]^{2F1(1, -2 - \nu; 3 - \nu; y)}, \]
where \( y = \frac{1 - \nu - 2}{1 + \nu + 2 + \nu} \).

Actually, the imaginary parts of (A.9) and (A.11) are expressed directly through the dipole radial integrals
\[ \text{Im} \alpha_n^{(q)} = \frac{\pi}{3} [R_0(ns, E_q p)^2], \text{Im} \alpha_{21}^{(qq')} = \frac{\pi}{3} R_0(2s, E_{q'}p)R_0(1s, E_q p), \]
where \( E_q = E_n + \omega_q \) and \( E_{q'} = E_i + \omega_{q'} \) is the ‘resonant’ photoelectron energies for the \( q \) and \( q' \) harmonics, and
\[ R_0(1s, Ep) = \frac{2\sqrt{2} \exp\left(-\frac{\sqrt{2}}{\sqrt{E}} \arctan \frac{\sqrt{2E}}{\pi}\right)}{\left(E + \frac{1}{2}\right)^{\frac{1}{2}} \left(1 - \exp\left(-\frac{\sqrt{2}}{\sqrt{E}} \arctan \frac{\sqrt{2E}}{\pi}\right)\right)}, \]
\[ R_0(2s, Ep) = \frac{\sqrt{E + \frac{1}{2}} \exp\left(-\frac{\sqrt{2}}{\sqrt{E}} \arctan \frac{\sqrt{2E}}{\pi}\right)}{\left(E + \frac{1}{2}\right)^{\frac{1}{2}} \left(1 - \exp\left(-\frac{\sqrt{2}}{\sqrt{E}} \arctan \frac{\sqrt{2E}}{\pi}\right)\right)}. \]

Note, that the high-frequency behavior of (A.9) and (A.11) follows after expansion of the hypergeometric function at \( \nu \to 0 \). The first expansion term dominates at rather high frequencies \( \omega_q \gtrsim 10 \) au. At lower frequencies either the next-term contributions should be added or an analytical fitting function should be applied as shown in figure 2. Generally, the positive in sign imaginary part in (A.9) and (A.11) decreases more rapidly than the negative real one, thus the phase of \( \alpha_{21}^{(qq')} \) goes monotonously to \( \pi \).

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