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Obtaining two attosecond pulses for X-ray stimulated Raman spectroscopy

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ABSTRACT

Attosecond X-ray pulses are an indispensable tool for the study of electronic and structural changes in molecules undergoing chemical reactions. They have a wide bandwidth comparable to the energy bands of valence electronic states and, therefore, are well suited for making and probing multiple valence electronic excitations using core electron transitions. Here we propose a method of creating a sequence of two attosecond soft X-ray pulses in a free electron laser by optical manipulation of electrons located in two different sections of the electron bunch. The energy of each X-ray pulse can be of the order of 100 nJ and the pulse width of the order of 250 as. The carrier frequency of each X-ray pulse can be independently tuned to a resonant core electron transition of a specific atom of the molecule. The time interval between the two attosecond pulses is tunable from a few femtoseconds to a hundred femtoseconds with better than 100 as precision.

1. Introduction

Chemical bonds form, break or change on a time scale of femtoseconds [1]. The advent of extreme ultraviolet attosecond pulses produced with the technique of high harmonic generation in a gas (see Ref. [2] and references therein) have opened up the possibility for direct study of these processes. Promising ideas for the generation of intense soft X-ray and hard X-ray attosecond pulses using free electron lasers (FELs) [3–11] were also proposed. More recently, another scheme for the generation of attosecond X-ray pulses taking advantage of the echo-enabled harmonic generation (EEHG) [12] was published [13]. In this paper, we propose a slightly different use of the EEHG that allows us to generate two attosecond X-ray pulses at two different wavelengths using the same electron bunch. To the best of our knowledge, no other methods of attosecond pulse generation can readily produce this pulse structure with a controlled time delay. A strong scientific motivation for utilizing two colors for X-ray pump–probe experiments has been recently presented in Ref. [14], with the goal of measuring the evolution of valence electronic wave packets using stimulated X-ray Raman spectroscopy. The proposed experiments would exploit the wide bandwidth of attosecond X-ray pulses, which is comparable to the energy width of valence electronic states in molecules, to create localized excitations in the valence electrons. The first attosecond X-ray pulse with the carrier frequency tuned to a ground state transition of one atom of the molecule creates an electronic wave packet of valence electrons that is later probed by the second attosecond X-ray pulse with the carrier frequency tuned to a ground state transition of another atom of the molecule. The atom specificity helps to define where the wave packet of valence electrons is created and where it is probed, which simplifies the analysis of the experiment and aids in understanding the spatial distribution of the valence electron wave packets. Evidently, individual measurements done with each pair of the X-ray pump and the X-ray probe pulses and with various and well-controlled time delays between the pulses combine into a motion picture showing the dynamics of changes in chemical bonds.

Here we propose a method of production of a sequence of two attosecond X-ray pulses with two different carrier frequencies. Also implemented is a possibility to adjust the time delay between the first and the second pulse with a high precision. In the first part of the paper we describe the basic idea of the method and illustrate it in the second part with a numerical example. In this example, we demonstrate the feasibility of generation of two X-ray pulses with $\sim$250 as FWHM tuned to the K-edges of oxygen and nitrogen atoms, each with energy of $\sim$100 nJ.

2. Method

In the scheme shown in Fig. 1, we combine two recent ideas, current enhanced self amplified spontaneous emission [15] and echo-enabled harmonic generation [12]. We begin from the interaction of an electron bunch with a long laser pulse having frequency ω_1 inside a wiggler magnet W1 containing approximately 10 periods. The magnet has period λ_{w1} and

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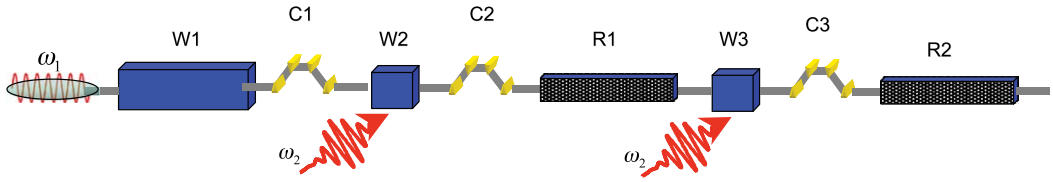

Fig. 1. A schematic of the generation of two attosecond X-ray pulses, where W1, W2 and W3 are wiggler magnets, C1, C2 and C3 are magnetic chicanes, R1 and R2 are X-ray undulator radiators, ω_1 is the carrier frequency of the long laser pulse and ω_2 is the carrier frequency of the short few-cycle laser pulses.

wiggler parameter $K_{w1}=eB\lambda_{w1}/(2\pi mc)$, where B is the peak magnetic field, e and m are the electron charge and mass, c is the speed of light, and the wiggler parameter is chosen to satisfy the FEL resonance condition $\lambda_1\equiv 2\pi c/\omega_1=\lambda_{w1}(1+K_{w1}^2/2)/2\gamma^2$; here, $\gamma=E/mc^2$ and E is the electron bunch energy. We also define the wave number $q_1=\omega_1/c$. This interaction produces a modest sinusoidal energy modulation of electrons with a normalized amplitude $a_1=\Delta E_1/\sigma_E$ that is slightly greater than unity, where ΔE_1 is the peak electron energy gain in the wiggler due to interaction with the laser and σ_E is the original rms energy spread in the electron bunch. It is assumed, in all wigglers where a seed laser interacts with electrons, that the cross-section of the laser light in the wiggler is several times larger than the transverse rms sizes of the electron bunch, and thus all electrons at the same location along the electron bunch receive equal energy change according to the phase of the laser light at the beginning of the interaction. It is also assumed, but only for this particular wiggler W1, that the laser pulse is longer than the electron bunch, so that the entire electron bunch is energy modulated independent of jitter in the relative timing of the electron bunch and the laser.

In the second step, we send the energy-modulated electron bunch through a dispersive magnetic chicane C1 with a rather large $R_{56}^{(1)}$ and achieve a characteristic electron distribution in longitudinal phase space in which narrow bands of electrons are interleaved with similarly narrow bands of empty phase space (see illustration in Fig. 2). This pattern in phase space is hidden in the sense that it yields minimal current modulation, but is a critical step that prepares the electrons for subsequent microbunching at wavelengths much shorter than λ_1 via the transformation of narrow spacing along the energy axis into short microbunches along the coordinate axis.

As proposed in Ref. [12], this transformation is achieved by employing a second energy modulation of the electrons using the electron beam interaction with the laser field with a frequency ω_2 in a second short wiggler magnet W2. The wiggler period λ_{w2} and wiggler parameter K_{w2} satisfy the FEL resonance condition for $\lambda_2=2\pi/q_2$, where $q_2=\omega_2/c$. This time the normalized amplitude of energy modulation $a_2=\Delta E_2/\sigma_E$ is supposed to be large, of the order of 10–20. This second seed laser is used to prepare the electron bunch for the production of the first of the attosecond pulses, and so we use a few-cycle laser pulse with carrier–envelope phase stabilization (see, for example, Ref. [2]) and a wiggler magnet with only one period, to apply the energy modulation in as short a section of the electron bunch as possible. The phase for the electric field with respect to the envelope is adjusted so that there is zero field at the center of the laser pulse (see the insert in Fig. 3).

After the second wiggler, the electron bunch passes the second dispersive magnetic chicane C2 whose strength $R_{56}^{(2)}$ is much smaller than $R_{56}^{(1)}$. As a result, we obtain the pattern of current enhancement shown in Fig. 3 which is large at the central peak and smaller at two side peaks (and absent everywhere else) [15]. At the same time, the bands of electrons seen in Fig. 2 rotate in longitudinal phase space and appear on the coordinate axis as shown in Fig. 4. This indicates an ultra-fine microbunching

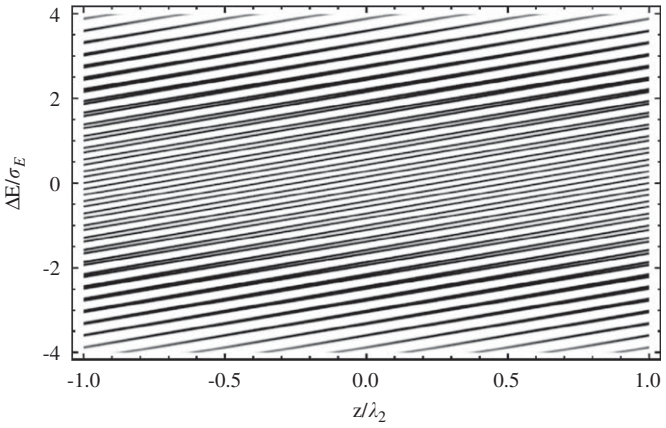

Fig. 2. A fragment of the electron bunch longitudinal phase space after C1. Here and on other similar plots the horizontal axis is the distance along the bunch normalized to λ_2 and the vertical axis is energy deviation from the equilibrium energy normalized to the rms energy spread in the electron bunch.

structure of electrons inside the spikes of the peak current. For this to happen, one should carefully choose the combination of values of $R_{56}^{(1)}$, $R_{56}^{(2)}$, and a_2. According to Ref. [12], the microbunching with a small period λ_{x1} corresponding to a harmonic number $h_1=\lambda_2/\lambda_{x1}=|n_1+q_1/q_2|$ is strong when:

$$R_{56}^{(1)}q_1\frac{a_2\sigma_E}{E}\simeq|n_1|+0.809|n_1|^{1/3} \tag{1}$$

$$R_{56}^{(2)}=-\frac{R_{56}^{(1)}q_1-E/\sigma_E}{q_2n_1+q_1}. \tag{2}$$

Here n_1 is a large positive or negative integer number. We note that for a short seed pulse, as above, the harmonic number h_1 does not strictly need to be an integer because of the wide bandwidth. This allows for more freedom in the choice of q_1/q_2, although in the example below we will take this ratio to be 4 for simplicity. The conditions of Eqs. (1) and (2) can be used to define $R_{56}^{(1)}$, $R_{56}^{(2)}$ as a function of a_2 for a given h_1, q_1, q_2 and σ_E. The parameters are selected to maximize the microbunching only inside the central spike of the peak current where we achieve the result that the narrow bands of electrons take a perfect upright position (see Fig. 4a). At the same time, the same a_2, $R_{56}^{(1)}$ and $R_{56}^{(2)}$ are not optimal for the side peaks (see Fig. 4b) and the microbunching there is much weaker than the microbunching inside the central current spike.

Following the generation of a narrow current spike, the electron bunch enters the undulator radiator R1 with period λ_{u1} and undulator parameter K_{u1} tuned for the FEL resonance at the wave length $\lambda_{x1}=\lambda_{u1}(1+K_{u1}^2/2)/2\gamma^2$. This undulator is relatively short because the central spike in the electron peak current is rather narrow, of the order of $\Delta z_1=\lambda_2 a_1/2a_2$, and in the case of a large a_2 and a modest a_1 it cannot provide a sustained support for the FEL process due to slippage of the radiation field relative to electrons. All electrons radiate there, but electrons in the central