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ATTOSECOND PULSE SHAPING OF X-RAY FREE-ELECTRON LASERS AND APPLICATIONS TO COHERENT CONTROL IN QUANTUM SYSTEMS

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Abstract

The development of high-power, attosecond methods at free-electron lasers has led to new possibilities in the probing and control of valence electron dynamics. Beyond simple observation of ultrafast processes, one of the longstanding goals of atomic physics is control of the electronic wavefunction on attosecond timescales. We present a scheme to generate sub-femtosecond pulse pairs from x-ray free-electron lasers with fs-scale separation, few eV energy separation, and a coherent phase relationship. This shaping method can be employed to coherently control ultrafast electronic wavepackets in quantum systems. We study in detail the Auger-Meitner decay process initiated by such a pulse pair and demonstrate that quantum beats of the decaying electronic wavepacket can be shaped by controlling the separation in energy and time of the pulse pair.

INTRODUCTION

Sub-femtosecond soft x-ray pulses are crucial tools to understand the ultrafast quantum mechanical motion of electrons in molecular systems. Traditionally, high harmonic generation (HHG) sources have been the state-of-the-art method by which to produce attosecond pulses, however their pulse energy output does not scale favorably with photon energy. In practice, HHG just barely pushes into the soft x-ray regime, and even then with only 10^{-12} – 10^{-9} J pulse energy [1].The first demonstration of isolated attosecond x-ray pulses at a free-electron laser, performed by the XLEAP collaboration at SLAC, represented a paradigm shift in the world of ultrafast atomic physics [1]. The XLEAP project provides pulses with few hundred attosecond durations, tens of μJ level pulse energy, and photon energies tunable deep into the soft x-ray regime. With this source, single-shot ultrafast measurements became possible, as did the possibility to probe nonlinear physics.

To date, the XLEAP project is largely restricted to the production of isolated gaussian pulses. While these pulses are useful for probing ultrafast dynamics, they do not provide much flexibility in terms of pulse structure. Here we introduce a new method for generating two-color attosecond pulse pairs based on overlapping an isolated attosecond pulse with a fresh part of the electron bunch. We show that this corresponds to FEL frequency pulling, and that there exists a before-now unexplored regime of frequency pulling which yields a time-delayed two-color pulse pair. To demonstrate the utility of such pulse pairs, we examine a possible experiment in which the pulse pairs can be used to not just probe, but control coherent electronic wavepackets observable through Auger-Meitner decay.

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FREQUENCY PULLING IN A TRANSIENT REGIME FOR PULSE PAIR PRODUCTION

The scheme we suggest is based on frequency pulling, which in the FEL community refers to the seeding of an FEL by a broad bandwidth pulse [2–4]. The basic physics are illustrated in Figure 1. In the cartoon, an electron bunch lases first in one undulator segment with undulator strength K_1 and therefore a corresponding photon energy ω_1. The output of the first stage is assumed to have a bandwidth $\sigma_{\omega,\mathrm{seed}}$. The bunch is then sent into a second undulator with a different undulator parameter K_2 and photon energy ω_2. In the frequency domain and in the linear regime, the output of the second stage is the product of the seed spectrum and the FEL gain spectrum shifted to the new resonance at ω_2 with a RMS bandwidth $\sigma_{\omega,\mathrm{SASE}}$. This gives rise to the phenomenon shown in the two plots below the cartoon. The product of the two curves is peaked around whichever of the two spectra is narrower. Thus for a narrow bandwidth, temporally long seed, the second stage emits radiation at ω_1 in spite of the new resonance condition. For a broad bandwidth, temporally short seed on the other hand, the second FEL stage is able to output at its resonance frequency.

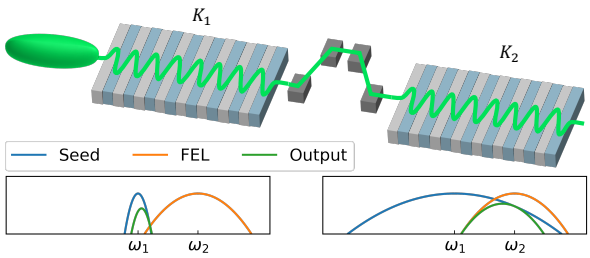

Figure 1: Cartoon demonstration of the concept of frequency pulling in FELs. The output of a seeded FEL in the linear regime is the product of the seed spectrum and the FEL gain spectrum, so the output frequency is determined by a comparison of the two bandwidths.

In its few mentions in the FEL literature, frequency pulling is generally considered in the regime of full conversion from seed photon energy to new resonant photon energy. In reality, this process happens gradually, and in particular there exists

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a regime in which the second undulator is just long enough to match the power of the two frequency components. We refer to this regime as transient frequency pulling, because in this transient regime one finds two pulses at the two different frequencies which are time delayed with respect to each other. Based on this, we suggest the scheme in Figure 2. We propose to use half of the undulators to generate an isolated attosecond pulse via the typical XLEAP chirp-taper method, hence the linear undulator strength parameter in the first stage. The typical e-beam current employed in the XLEAP setup naturally consists of an isolated current spike with a large chirp, and a flattop current ahead of it. By sending the beam through a chicane after the first undulator section, we can delay the e-beam so as to overlap the isolated attosecond pulse generated in the first stage with the flat portion of the e-beam current. The isolated attosecond pulse then acts as a seed for the second stage, in which by choosing the undulator length correctly we can have a pulse pair as the output.

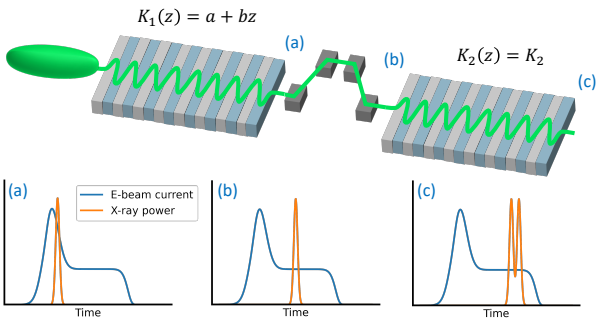

Figure 2: Schematic showing a scheme to generate attosecond pulse pairs. An isolated attosecond pulse is generated in a first stage. After a delay chicane the isolated pulse is overlapped with a fresh portion of the electron bunch in a detuned undulator, leading to frequency pulling. For a short enough second undulator, we observe a transient regime where two pulses coexist at the two frequencies.

We show an example in Figure 3 generated using GENESIS with a start-to-end LCLS beam [5]. In the first stage, the undulators are set to have a linear taper around a photon energy of 287 eV to generate the isolated attosecond pulse. After delaying the electron beam, the pulse overlaps with a flat part of the beam current with roughly 1.5 kA average current. By tuning the second undulator stage with no taper, and setting the undulator length appropriately, we arrive at pulses like those shown in Figure 3. We see now two distinct attosecond pulses, each with 30 GW peak power. The spectrum shown in the middle plot indicates clearly that there are two distinct frequency components in the spectrum, and the Wigner distribution on the bottom identifies the earlier pulse as having roughly 5 eV smaller photon energy than the later one.

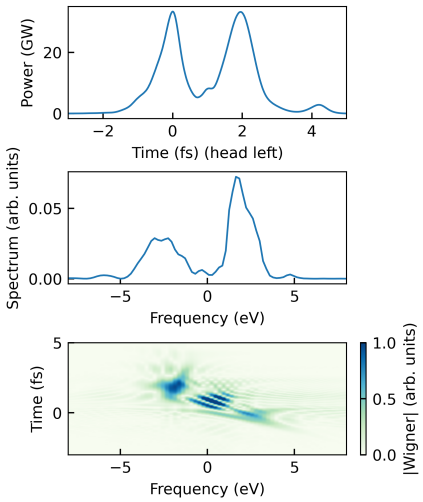

Figure 3: Example start-to-end GENESIS simulation of a frequency pulled pulse pair centered at 287 eV.

CONTROL OF AUGER-MEITNER DECAYING WAVEPACKETS WITH PULSE PAIRS

With the basic physics of attosecond pulse pair production elucidated, we now move to an application. Li *et al* was the first group to make use of the isolated attosecond x-ray pulses produced by SLAC in an ultrafast experiment [6]. They studied coherent electronic wavepacket dynamics by exciting nitric oxide (NO) using a broad bandwidth attosecond pulse centered around 533 eV photon energy. This photon energy was chosen to be near the 1s→ π resonance in NO which decays primarily by the Auger-Meitner mechanism. They time-resolved the emission of electrons from the system using angular streaking, and found that instead of a pure decay they measured a revival consistent with coherent interference effects between closely spaced energy levels in the system.

We propose to use our pulse pairs to add another layer of complexity to this process. By controlling the temporal separation of two pulses at slightly different photon energies, we can in effect control the time delay between population of the different energy levels. This should then change the phase of the ensuing quantum beats, indicating that we have exerted some amount of control over the transient state of the system. We propose to do this in H_2CCF_2, which has two resonant states around 287 eV excitation energy spaced 4 eV apart. We can accomplish this control in several ways, a summary is presented in Figure 4. The top show shows a set of simulations in which we've scanned the average photon energy of the pulse pairs for a second stage detuning of -0.4%, -0.1%, and 0.2%. The plots themselves show the ionization rate of the molecular system. We see that by changing the average photon energy we can change the ionization dynamics dramatically, and furthermore we find that for different detunings (and therefore different pulse pair geometries) we can change the phase of the oscillations. In particular for average photon energies of 285.5, 286, and

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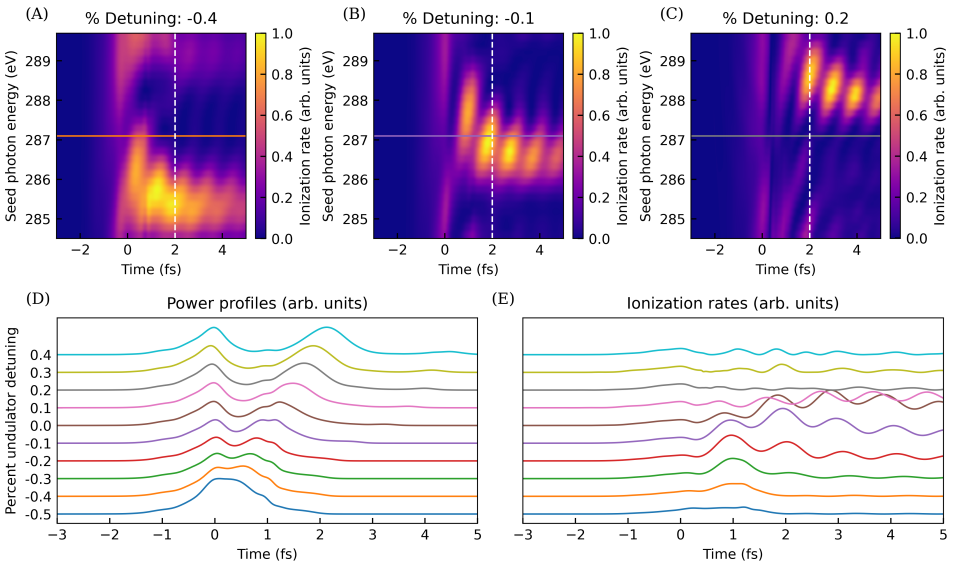

Figure 4: Impact of pulse pair shape and central photon energy on the ionization rate of H_2CCF_2. In the top row we scan the average photon energy of the pulse pair for different second stage detuning values. On the bottom we show a set of power profiles generated by different detunings on the left and the corresponding ionization rate profile on the right.

288.5 eV (left to right) we find strong oscillations, but the phase of those oscillations shift by a full period as we move from -0.4% detuning to 0.2%. The bottom plots show, on the left, the pulse pair power profiles for different second stage detunings, and on the right, the ionization rate output for that detuning with fixed seed photon energy. These oscillations can be measured using the same angular streaking technique. We plan to perform this experiment at the LCLS.

CONCLUSIONS

We have presented a scheme to shape isolated attosecond XFEL pulses into pulse pairs with tunable energy and temporal separation. Such a capability will greatly broaden the scope of possible attosecond experiments performed at XFELs. We highlight one such possibility in the form of control of electronic wavepacket dynamics.

ACKNOWLEDGEMENTS

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