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Isolated 170 as Pulse Generation in the XUV

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We demonstrate generation of isolated XUV attosecond pulses in the sub-200 as regime (1 as = 10^{-18} s). They are generated utilizing waveform-controlled few-cycle laser pulses and advanced broad-band XUV multilayer optics.

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1. Introduction

Macroscopic properties of matter constitute the manifestation of ultrafast phenomena evolving on a microscopic scale in atoms, molecules, and solids. Insight into these phenomena calls for experimental techniques endowed with temporal resolution commensurable with their characteristic time scale. This time scale of microscopic phenomena is predictable via the laws of quantum mechanics. For example, in a molecule, the sub-meV spacing of the rotational energy levels and the sub-eV spacing for the vibrational ones correspond to picosecond (1 ps = 10^{-12} s) and femtosecond (1 fs = 10^{-15} s) time scales, respectively. Phenomena evolving on a femtosecond time scale have been resolved in real time owing to the development of femtosecond laser pulses which marked the era of femtosience [1].

However, the resolution that these pulses provide is inadequate for tracking and controlling the motion of electrons in atoms and molecules. Based on similar arguments as above, an electronic wave packet formed by the coherent superposition of lower electronic states of an atom or a molecule (typically spaced in energy by few eV) will evolve on an attosecond time scale (1 as = 10^{-18} s). In many cases the evolution of electron wave packets results in electronic charge redistribution

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that can be regarded as an ultrafast “hopping” of the electron from one atom to another on an attosecond time scale ($T_p \approx 400$ as) [2]. Simulations of more complex systems of biological interest have recently revealed that electron dynamics might manifest themselves as a rapid charge migration (positive or negative) that follows site selective ionization [3].

Interrogating the role of electron dynamics on the macroscopic behavior of matter as well as steering chemical reactions by controlling the electronic motion, falls within the scope of attoscience and calls for new tools with attosecond resolution [4].

Here we present sub-200 as XUV pulses at high photon energy (≈ 95 eV) and flux and with the potential to enable studies of electron dynamics in a vast variety of microscopic systems as the above mentioned.

2. Isolated attosecond pulses: generation and measurement

2.1. Generation of isolated XUV pulses

Attosecond pulses are generated in the extreme ultraviolet part (XUV) of the electromagnetic spectrum, utilizing a process referred to as high harmonic generation (HHG). HHG takes place when ultraintense laser pulses interact with atoms in the gas phase. The process can be described by a quite intuitive model [5] in which an electron driven by the oscillating electric field of the laser pulse, (i) is set free around the maximum of the field (tunnel ionization), (ii) follows a quasi-classical motion in the field and acquires kinetic energy that can reach many hundreds of eV, and (iii) emits its excess energy upon recombination with the parent ion as high energy photons. The maximum energy of these photons is given by $E_{\max} = I_p + 3.17U_p$ with I_p being the atomic ionization potential and U_p the ponderomotive energy of the electron in the oscillating electric field of the laser.

In case that the HHG process is driven by multi-cycle laser pulses, the electrons encounter the parent ion every half-cycle of the electric field which results in a series of XUV emissions that manifest themselves in the spectral domain as high harmonics [6–8]. In contrast, when few-cycle laser pulses are employed, the energy of the emitted light wave packet from consecutive recombination events close to the peak of the pulse, can vary substantially from the maximum emitted energy. By setting the carrier envelope phase (CEP) of a few-cycle laser pulse such that the field maximum coincides with the maximum of the pulse envelope (resulting in a substantial modification of U_p for consecutive recombination events) — referred to as cosine pulse — it is possible to restrict the emission of the most energetic light wave packets to take place only once per laser pulse [9]. By contrast, in a sinusoidally-shaped laser field a couple of re-colliding electron wave packets leads to emission at the highest photon energies and results into two XUV bursts separated by half a period of the laser field. This is shown in Fig. 1.

The extension of the XUV/X-ray spectrum associated with a single re-collision event and hence with the potential to form an isolated attosecond pulse

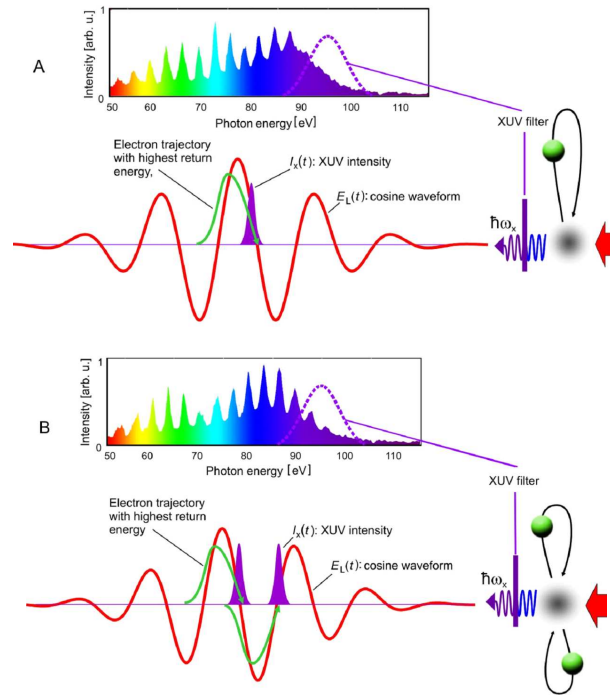


Fig. 1. Generation of isolated attosecond pulses utilizing few-cycle laser driving fields. (A) With the CEP set to zero a single highly energetic electron wave packet can be generated. Upon re-collision with the parent ion, a single burst of light is emitted and results in a spectral continuum at high photon energies. (B) Two highly energetic electron wave packets are generated when the CEP is adjusted to $\pi/2$. This is manifested in the spectral domain by modulated spectra. A spectral filter (cyan) is used to isolate this continuum. Experimentally this is realized with metal filters [10] and advanced multilayer mirrors [11].

is precisely linked to the intensity contrast between consecutive field maxima and therefore the *duration* of the driving pulse. In the case of $\approx 4.5\text{--}5$ fs pulses utilized in the experiments presented in the next sections, this contrast is expected to result in a continuum of approximately 20 eV [12]. This bandwidth — if properly manipulated — can support isolated XUV pulses shorter than 150 attoseconds.

2.2. Extending ultrafast metrology techniques — the atomic transient recorder

In electron-optical chronoscopy realized by image-tube streak cameras, an electron “replica” of a light pulse generated when the latter impinges on a photosensitive cathode, is deflected by a controlled electric field that projects its temporal structure on a screen. These techniques have permitted accurate characterization of sub-picosecond light pulses. Recently the extension of these ideas into the attosecond regime has been possible [13, 14] by applying a key modification; the

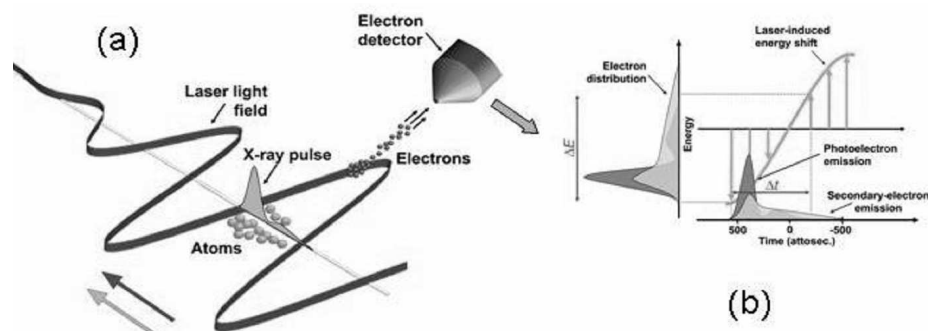


Fig. 2. The atomic transient recorder: (a) impulsive ionization of atoms by an XUV pulse releases its electron replica into a laser field; (b) electrons detected along the laser polarization vector, suffer a strong transient shift of their momentum. A systematic variation of the timing of the electron release with respect to the laser streaking field, allows mapping of an initial time-momentum distribution onto a series of different final momentum distributions.

electric field of a laser pulse is used to streak electrons that are released within a small fraction of its oscillation ($T_{\text{laser}} \approx 2.5$ fs) (Fig. 2a).

The field is applied along the direction of motion of the detected electrons and therefore does not result in deflection but rather in modification of their final kinetic energy. This capability renders possible the projection of the initial time-momentum distribution of the electron emission onto a series of different final (accurately measurable) momentum distributions. The idea, in its most generic case, where primary (photoionization) and secondary electron emission (e.g. Auger decay) can be temporally resolved is illustrated in Fig. 2b. We have dubbed this scheme of atomic transient recorder [9]. The resolution attainable with this technique is of the order of 100 as and constitutes the briefest time interval accessible to date.

In its simplest form the technique can be used to characterize attosecond XUV pulses by interrogating the temporal structure of an electron replica that is generated by impulsive ionization of atoms in the presence of the streaking field. In this case, the technique is closely related to frequency resolved optical gating (FROG) with the field oscillation in the role of the gate [15]. This entails that reconstruction of the pulse can be realized utilizing advanced algorithms which are readily available. Reconstruction is not only possible for the attosecond pulse but for the gate at the same time. In the latter case the attosecond pulse has the role of a sub-femtosecond sampler that tracks the oscillation of light waves [4].

3. Experimental apparatus

The apparatus employed in the experiments is discussed in detail in Ref. [16]. A simplified schematic is shown in Fig. 3. Briefly, phase-stabilized few-cycle laser

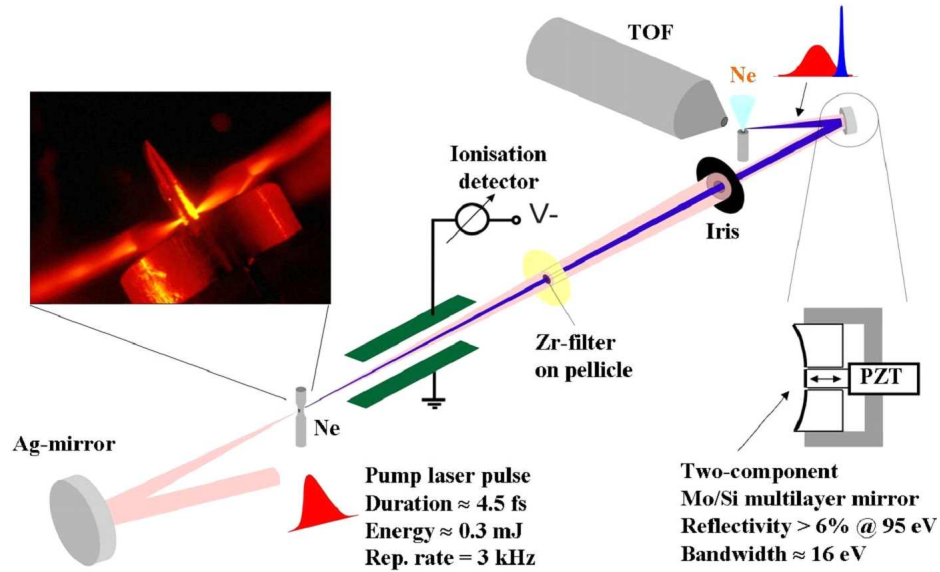


Fig. 3. Attosecond generation and measurement apparatus. For details see text and Ref. [16].

pulses [17, 18] are focused into a Ne gas target to generate XUV light. The remaining near infrared and the XUV beam are focused into a second gas (Ne) target by a double mirror assembly installed ≈ 1.5 m downstream the source. The inner part of the assembly is a MoSi aperiodic multilayer mirror that supports a 16 eV bandwidth in reflection centered at about 95 eV whilst the outer part of the mirror focuses the laser light [19]. The delay between the XUV and the laser pulse can be adjusted with high accuracy (better than 100 as) using a piezo translational stage. Electron spectra are recorded by a time of flight detector (TOF) that collects electrons that are emitted along the direction of the laser polarization vector.

4. Experiments — results

For the characterization of the XUV pulses with the atomic transient recorder the XUV continuum is reflected, filtered and focused by the multilayer mirror into a Ne gas nozzle along with the IR field. Series of electron spectra as a function of the delay between the XUV and laser pulse (delay step 300 as) are shown in false color representation in Fig. 4.

For the retrieval of the duration of the attosecond pulse we have utilized tools based on principles discussed in [20, 21] in detail. The retrieval accounts for possible satellite pulses, while all key parameters including the chirp of the attosecond pulse and the temporal shape of the laser field, are left free to be reconstructed invoking the power of a genetic algorithm. The retrieved pulse duration by our analysis was 170 as ± 10 as approximately 5% longer compared

to the transform limited pulse supported by the reflected spectral bandwidth (≈ 160 as). The satellite pulse was found to contain about 17% of the total photon number.

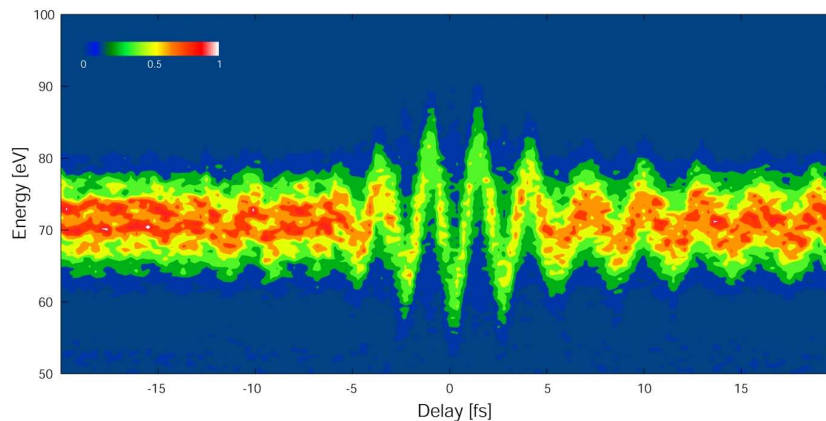


Fig. 4. Atomic transient recorder spectrogram of a 170 as XUV pulse. A series of electron spectra generated in Ne have been recorded as a function of the delay between the XUV and the few-cycle laser pulse in steps of 300 as. The electron spectrometer is oriented such as to collect electrons emitted only in the direction of the polarization vector of the laser field utilizing the geometry presented in [16].

As alluded, the electric field of the XUV attosecond pulse and that of the laser pulse can be reconstructed from the same spectrogram. This is however only possible if the attosecond pulse samples at least 5–10 field oscillations for a few-cycle laser pulse, necessary to probe its built-up and disappearance. The retrieved duration (FWHM) of the near-infrared laser pulse was 4.7 fs.

Furthermore, estimations on the photon flux yielded $\approx 3.6 \times 10^8$ photons/s for the attosecond pulse delivered into the target. The flux at the HHG source was more than an order of magnitude higher over the same spectral range, suggesting that further improvements in XUV multilayer optics afford promise for substantially higher photon flux delivered to experiments.

5. Conclusions

In conclusion, we have demonstrated isolated attosecond XUV pulses in the sub-200 as regime with high photon flux and energy that renders possible a broad range of investigation in atoms, molecules, and solids. Utilizing shorter driving fields [22] will allow generation of XUV pulses with duration well below 100 as to become available in the near future.

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