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Seeded attosecond-pulse generation in structured media: A road for attosecond optics

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We present an approach for controlling attosecond pulses just like one controls femtosecond pulses nowadays. Using an adequate medium constituted of aligned and pre-dissociated molecules associated with a "seed" attosecond pulse and a control ir field, we propose a scheme for generating secondary attosecond pulses with a high degree of control of their characteristics. By solving the time-dependent Schrödinger equation for a model pre-dissociated molecule (H_2^+) irradiated by the control ir field and seed xuv pulse, we demonstrate that one can generate such well-controlled secondary attosecond pulses by achieving frequency tuning and up-shifting and control on the chirp of the secondary pulses, which can even be significantly shorter than the seed. We also show that their characteristics can be accurately predicted with a simple classical approach.

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Attosecond pulses have received a lot of attention since their first experimental observation in 2001 [1,2]. Today high-order harmonic generation (HHG) sources cover a wavelength range from the vacuum ultraviolet (vuv) to the soft x-ray regime, and pulses of sub-fs duration are now routinely provided in several places worldwide. Attoscience is very young but has already opened new possibilities, and many more are foreseen when new attosecond sources become better controlled [3].

Amplification of high harmonics in free-electron laser sources [4] and in x-ray laser media have been observed [5]. Chirp control and spectral selection techniques have been developed by using metallic filters [6], gas dispersion [7], or chirped mirrors [8]. HHG in prepared molecules [9–15], or xuv-assisted harmonic generation in atoms [16–18] and molecules [19] have also been studied. All these approaches lead to a better control on the attosecond pulses, but developing a real "attosecond optics" especially designed to create fully (and easily) controlled attosecond pulses is the next step that is required for the full development of attophysics.

In this paper, we address this issue and show that highly controlled attosecond pulses can be obtained by considering existing attosecond pulses as a seed for a new type of attosecond optics to generate secondary attosecond pulses. This approach is developed for isolated attosecond pulses but can be extended to trains of attosecond pulses. The proposed attosecond-optical medium is composed of aligned [20,21] and pre-dissociated molecules combined with a strong control ir field that is phase locked with the seed pulse. The key idea [illustrated in Fig. 1 (upper panel)] is that the seed attosecond pulse can release electrons with defined characteristics from one part of the molecule and the control field can force these electrons to acquire energy before interacting with the other part of the molecule. The secondary pulses emitted through this interaction can be finely tailored. For simplicity, we assume that the molecular medium consists of pre-dissociated hydrogen molecular ions (H_2^+) characterized by their internuclear distance R. We assume that a relatively narrow dissociative wave packet in H_2^+ is created by multiphoton ionization of aligned H_2 by an ultrashort laser pulse so that R can be controlled by varying the delay between the ionizing pulse and the seed and control pulses. Other molecular ions could also be used and their internuclear distance can be finely controlled [20], but to ensure coherence it is important that the two nuclei share the same electronic wave function [22].

The system is studied both by solving the three-dimensional time-dependent Schrödinger equation (TDSE) in the fixed-nuclei approximation [23] and by a classical model. As the degrees of freedom are numerous (internuclear distance R and ionization potential I_p of the molecule; central frequency $\omega_{\rm xuv}$, duration $\tau_{\rm xuv}$, chirp, and timing of the seed xuv pulse; intensity and frequency of the control ir pulse), the classical model is an important guide to anticipate the characteristics of the secondary attosecond pulses and is a way to highlight the influence of each parameter. We demonstrate that both approaches agree qualitatively well and that one can generate secondary pulses of attosecond duration and achieve tunability, spectral broadening, and frequency upshifting of these pulses while controlling their chirp.

The seed attosecond pulse (linearly polarized along the internuclear axis and centered at the time t_i) had a Gaussian temporal profile:

$$E(t) = E_{\rm xuv}\sqrt{\frac{\tau_0}{\tau_{\rm xuv}}}\exp\left[-\frac{2\ln 2\,(t-t_i)^2}{\tau_0^2(1+\eta^2)}\right] \times \cos\left[\omega_{\rm xuv}(t-t_i)+\frac{2\ln 2\,(t-t_i)^2\eta}{\tau_0^2(1+\eta^2)}\right]. \quad (1)$$

Here, $E_{\rm xuv}$ is the peak electric field amplitude, t_i is the time of ionization (delay of the xuv pulse in the ir field), η is the chirp factor, and $\tau_{\rm xuv} = \tau_0\sqrt{1+\eta^2}$ and τ_0 are the FWHM pulse durations of the chirped and nonchirped pulses, respectively. The central photon energy is fixed at 27.2 eV and the peak intensity is 10^{14} W/cm^2 [24]. The linearly polarized ir field (with a central wavelength of 800 nm) is taken to be a five-cycle sinusoidal field with a sine-square temporal profile and peak intensity 10^{14} W/cm^2.

The spectra of the emitted xuv light are shown in Fig. 1 (lower panel) for three internuclear distances (37, 42, and 48 Å). These spectra show several structures: Atomic-like harmonics are present for xuv energies below 30 eV and are similar to harmonics emitted by isolated H atoms irradiated by the ir field only. With our parameters the atomic-like cutoff,

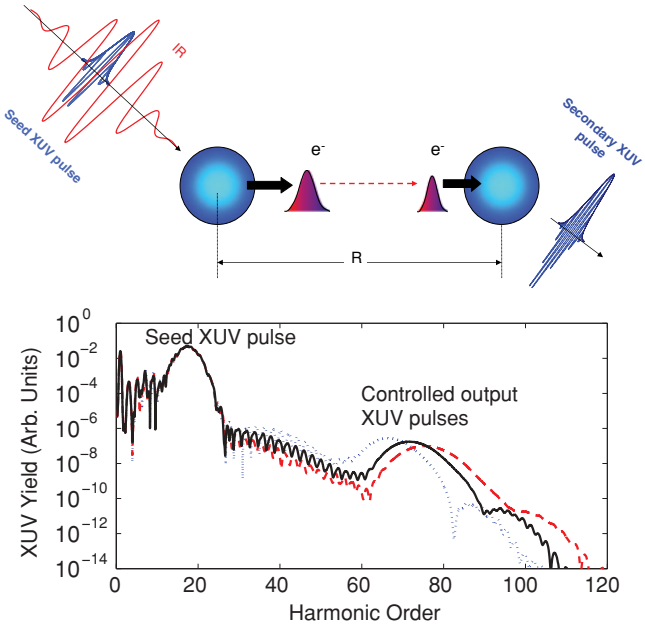

FIG. 1. (Color online) Upper panel: sketch of the present attosecond optics molecular system, where an incident xuv pulse creates an electron wave packet in the vicinity of a nucleus. The wave packet is then accelerated by an ir field toward the neighboring nucleus, where it emits, during collision, a controlled secondary attosecond pulse (see text for details). Lower panel: HHG spectra calculated via the TDSE for $\tau_0 = 270$ as, $\eta = 0$, and $t_i = 0$, and for several internuclear distances, $R = 37$ (dotted blue line), 42 (solid black line), and 48 Å (dashed red line).

given by $I_p + 3.2U_p$, where U_p is the ponderomotive energy, is at the 21st harmonic. The broad continuous structure close to 27 eV is the signature of the seed xuv pulse. Harmonics are visible between the 21st and the 51st harmonic order, even without the seed xuv pulse. These harmonics, extending up to a cutoff energy of $I_p + 10U_p$, correspond to the emission of xuv light arising from the electron wave packet being accelerated from one atom to the neighboring one [9–11,13,14].

Above 75 eV (harmonic order 55), a continuous spectrum appears only if both the xuv seed pulse and the ir control field are present. Classically, the emission of these xuv photons is possible if and only if a seed xuv photon has triggered the emission of an electron from one of the nuclei (with a nonzero initial velocity), followed by an acceleration of the electron in the ir field toward the neighboring nucleus, with subsequent radiative recombination (see upper panel in Fig. 1). As the two nuclei initially share the same electronic wave function, coherence is preserved, resulting in coherent xuv emission [14,22]. Only this specific emission mechanism is considered in this work. Experimentally the corresponding xuv light can be selected by transmitting the xuv light through a Zr filter, thereby selecting only the harmonics above the 55th order.

After the spectral filtering, the spectrum of the secondary pulse can be tuned simply by controlling the internuclear distance, i.e., via the delay between the alignment/ionizing pulses and the seed/control pulse. In the temporal domain, the secondary pulses are also attosecond pulses, typically lasting less than 200 as with the 270-as seed pulse considered here

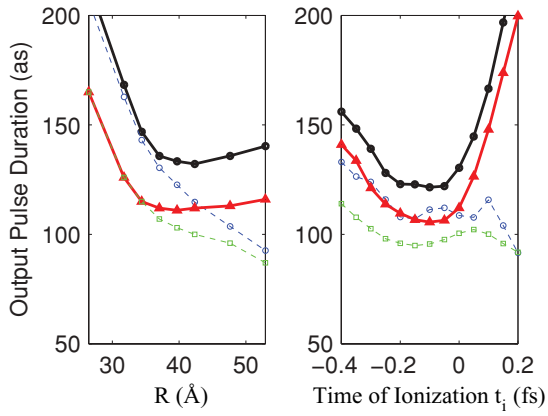

FIG. 2. (Color online) Left panel: FWHM duration of secondary xuv pulse versus internuclear distance by considering a nonchirped ($\eta = 0$) seed xuv pulse with $\tau_0 = 270$ as, centered at a zero of the ir control field ($t_i = 0$). Thick black line (full circles) is the result of the TDSE calculations, and thick red line (full triangles) is the classical result. Dashed blue line (open circles) and dashed green line (open squares) indicate the minimum pulse duration imposed by the Fourier transform of the spectrum (Fourier limit), in the quantum-mechanical and classical simulations, respectively, after spectral selection of the harmonics with order >55. Right panel: Duration of emitted xuv pulse vs time of ionization t_i (delay of xuv pulse) for $R = 42$ Å and $\eta = 0$.

(see left panel in Fig. 2). The short duration of these secondary pulses justifies the use of the fixed nuclei approximation [23] since the nuclei do not move significantly during the 200 as interaction time. We thus demonstrate that it is possible to upshift the frequency of attosecond pulses and tune their central frequency by controlling the internuclear distance (a similar control can be obtained at fixed internuclear distance by varying the ir intensity). It turns out that the secondary pulses typically have a duration close to the Fourier limit imposed by their spectra for internuclear distances less than about 35 Å, but become longer for larger internuclear distances.

From classical considerations, we estimated the duration of the secondary xuv emission by assuming that the xuv photons were emitted during the collision of an accelerated electron with the neighboring atom. To mimic this collision and describe the classical motion of the electron, we considered that an electron can be released at a given time by xuv photoionization with a probability proportional to the intensity profile of the xuv (seed) pulse. Its initial velocity, directed toward the neighboring nucleus, is imposed by I_p and the instantaneous frequency of the xuv light at that time. After the release, the electron is accelerated by the ir field. For each initial condition one can monitor the classical motion and find out whether (and when) this electron will collide with the neighboring nucleus and at which kinetic energy E_c. At the instant of collision the electron may be recaptured, resulting in the emission of a short xuv burst of central photon energy $E_c + I_p$.

To estimate the temporal profile of this emission, we assume that each collision leads to the emission of a 30-as Gaussian xuv burst (emitted at the time of collision) with a spectral width of 15 eV centered at the photon energy $I_p + E_c$. To estimate classically the duration and spectrum

of the secondary attosecond pulse, we assumed that the ionization time, t_i, followed a distribution weighted with the ionization probability. To sample the full temporal profile of the ionizing pulse, we considered 61 possible ionization times within $\pm 1.5\tau_{xuv}$ of the seed pulse. For each of these, the initial velocity also followed a distribution imposed by the input spectrum of the seed pulse and its specific content (instantaneous xuv frequency and spread $2\pi/\tau_{xuv}$) around the considered ionization time. By summing over all initial conditions and performing spectral selection ($q > 55$), we obtained a classical estimate for the duration and spectrum of the secondary xuv pulse.

Figure 2 (left panel) shows the duration (FWHM) of the emitted xuv pulse versus internuclear distance, obtained for both the classical model and the full TDSE calculations with $t_i = 0$. The classical result clearly follows the trend of the *ab initio* one, and even quantitatively the numbers are not far from each other.

Another important parameter is the time of ionization t_i as it affects both the time of collision and the electron energy at recollision. Classically one would expect that if some electrons are released before $t_i = 0$, they will first be decelerated by the ir field and it will take them a longer time to reach the neighboring atom as compared to electrons released later. This implies that it is possible to calculate classically an optimal (negative) delay of the seed xuv pulse, ensuring that electrons ejected at different times all reach the other atom around the same recollision time, leading to a shorter xuv burst. To the contrary, for positive ionization times ($t_i > 0$), the xuv emission will go on for a longer time, leading to longer secondary pulses.

This is illustrated in Fig. 2 (right panel) which shows the output pulse duration as a function of t_i with $R = 42$ Å. The TDSE and the classical results agree well and the results confirm that shorter secondary pulses can be emitted for negative values of the ionization time ($t_i < 0$). Furthermore, the results are clearly asymmetric around $t_i = 0$, demonstrating that a negative delay of the seed xuv pulse is favorable to obtain short pulses as expected classically. Secondary pulses with duration of about 120 as, i.e., about 10% above the Fourier limit, are obtained for $t_i = -0.1$ fs, showing further control on the characteristics of the attosecond pulses.

To highlight this control, it is worth noting that the spectrum of the xuv light (and thereby the Fourier limited pulse duration) did not vary much with t_i, while the xuv output pulse duration clearly changed (right panel in Fig. 2). This implies that changing t_i is a way to directly control the chirp of the attosecond pulses. To mimic realistic experimental conditions, simulations were also performed with a chirped seed xuv pulse, and the results are shown in Fig. 3. Imposing a positive chirp (with $t_i = 0$) has an effect similar to that of choosing negative values of t_i, i.e., faster electrons are emitted later during the pulse in such a way that they may catch up with the first ones. Hence, the control can be increased in this way and very short secondary pulses were obtained even with $t_i = 0$ (left panel in Fig. 3). For positive values of the chirp we could achieve output pulse durations that were very close to the Fourier limit imposed by the xuv spectrum, the shortest ones being about 90 as for $\eta \sim 1.75$. When both the chirp and the ionization times are varied (Fig. 3, right panel), an increased control

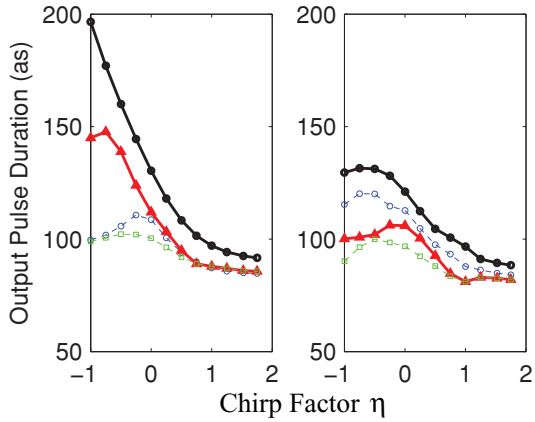

FIG. 3. (Color online) Duration of the emitted xuv pulse vs chirp η of the seed xuv pulse (same labels as in Fig. 2) for $\tau_0 = 270$ as and $R = 42$ Å. Left panel: $t_i = 0$. Right panel: $t_i < 0$. In the right panel, both the chirp and the value of t_i have been varied, i.e., the "optimal" value of t_i, as determined classically, has been selected for each value of the chirp.

appears, leading to even shorter pulses that are very close to their Fourier limit for a large range of values of η.

To control the secondary attosecond pulse, the bandwidth of the seed attosecond pulse is of prime importance. Its effect is shown in Fig. 4, where the output pulse duration is plotted for seed-pulse durations varying from 150 to 550 as. With $t_i = 0$, $R = 42$ Å, and $\eta = 0$, it clearly appears that the longest secondary pulses are obtained with the shortest seed pulses, while much shorter pulses can be obtained when the seed xuv pulse is long. This can be explained classically as a smaller spread in the initial velocity of the electron is compatible with a better control of its trajectory. This even demonstrates that postcompression of attosecond pulses is possible, as here secondary pulses five times shorter than the seed xuv pulse can be obtained (see inset in Fig. 4).

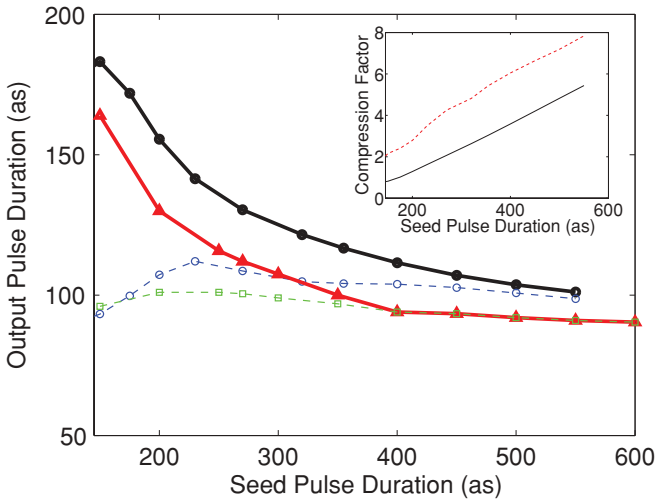

FIG. 4. (Color online) Duration of the emitted xuv pulse vs the duration τ_0 of the seed xuv pulse (same labels as in Fig. 2) for $t_i = 0$, $\eta = 0$, and $R = 42$ Å. The inset shows the degree of compression in the TDSE (black line) and classical (dashed red line) cases, respectively.

In conclusion, we have demonstrated that a seed attosecond xuv pulse can be used in combination with a control ir field and a prepared molecular medium to produce short secondary attosecond pulses with controlled spectra and chirp. We show that with a simple spectral filtering, it is possible to shift the frequency of the attosecond pulse, broaden its spectrum, and reach a pulse duration close to the Fourier limit imposed by the spectrum. With a moderate intensity of 10^{14} W/cm^2 and a 27-eV seed pulse, we show that it is possible to generate sub-100-as pulses at a photon energy of about 150 eV. It seems also rather straightforward to upscale these results to produce even shorter pulses simply by using a higher intensity and/or a longer wavelength of the control field. Thus, by increasing the intensity and wavelength of the control field and even increasing the seed pulse duration, one should be able to produce very short and well-controlled attosecond pulses, even within the x-ray domain, and far below the 100-as limit.

These results open numerous perspectives. With control pulses of longer wavelength one could even get short attosecond pulses of high photon energy with femtosecond seed pulses. Thus, the seed pulses do not need to be extremely short to be useful in this context, which is another big experimental advantage. To go further, one could even imagine relativistic harmonic generation with such an approach. Harmonic generation at relativistic intensities is usually prevented in atoms as the Lorentz force drives the electron away from the nucleus. If the molecular ion is pre-aligned in a direction different from that of the control field polarization, one can locate the neighboring atoms so that the Lorentz deflection will make the collision possible and thereby go around this limitation. Seeded attosecond pulse generation could also be extended to more complex systems, such as bigger engineered molecules or even nano objects if coherence is created by a strong field coupling [22], or more simple systems like atoms or ions. For instance, using atoms and an elliptically polarized driving field can also lead to a reasonable control of the secondary attosecond pulses and an automatic spectral selection as unseeded HHG is prevented by ellipticity. On a broader perspective, the emission of controlled attosecond pulses is a signature of highly controlled electron wave packets, and such wave packets could be used to trigger or probe other ultrafast phenomena. For instance, they can be used to perform electron diffraction on neighboring molecules. In this respect, seeded attosecond optics in structured media is also a key for developing electron diffraction with attosecond temporal resolution.

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