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Frequency-resolved optical gating for complete reconstruction of attosecond bursts

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We describe a method for the complete temporal characterization of attosecond extreme ultraviolet (xuv) fields. An electron wave packet is generated in the continuum by photoionizing atoms with the attosecond field, and a low-frequency dressing laser pulse is used as a phase gate for frequency-resolved-optical-gating-like measurements on this wave packet. This method is valid for xuv fields of an arbitrary temporal structure, e.g., trains of nonidentical attosecond pulses. It establishes a direct connection between the main attosecond characterization techniques demonstrated experimentally so far, and considerably extends their scope, thus providing a general perspective on attosecond metrology.

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The generation of isolated attosecond (as) pulses [1,2] and trains of attosecond bursts [3–5] has recently been demonstrated experimentally, opening the route to the time-domain study of electronic dynamics in matter [6]. Characterizing such attosecond fields is challenging, not only due to the extremely short time scales involved and the large associated bandwidth, but also because these fields are in the extreme ultraviolet (xuv) range, where no efficient nonlinear medium is available. It is thus extremely difficult to directly apply the conventional methods of ultrafast optics. Several schemes have now been demonstrated [1–3,5] or proposed [7–11] to circumvent these problems. Most of these methods consist of converting the as pulses into continuum electron wave packets, through the photoionization of atoms, and in using the femtosecond oscillations of an infrared laser field to gain information on the temporal structure of these wave packets. However, only a few of these methods enable a complete characterization, and most of them are restricted to specific and simple temporal structures, e.g., single isolated as pulses [1,2,7–10] or trains of identical as bursts [3,4]. Moreover, no clear connection has been established yet between the main demonstrated techniques [1–3].

We describe a simple and general method allowing for the complete characterization of arbitrary as fields: frequency-resolved optical gating for complete reconstruction of attosecond bursts (FROG CRAB [12], hereafter called CRAB). Introducing this technique allows us to transpose some of the most efficient tools of the very mature field of ultrafast optics to attosecond metrology. It also merges the main techniques demonstrated so far [1–4] into a common and more general framework, thus providing a general perspective of attosecond measurements.

CRAB is inspired from frequency-resolved optical gating (FROG), a widely used technique for the full temporal characterization of visible pulses [13]. FROG consists of decomposing the pulse to be characterized in temporal slices thanks to a temporal gate $G(t)$, and then measuring the spectrum of each slice. This provides a two-dimensional set of data, called a spectrogram or FROG trace, given by

$$S(\omega,\tau) = \left| \int_{-\infty}^{+\infty} dt G(t) E(t-\tau) \mathrm{e}^{i\omega t} \right|^2, \qquad (1)$$

where $E(t)$ is the field of the pulse to be characterized, and τ is the variable delay between the gate and the pulse. The gate may either be a known function of the pulse, as in most implementations of FROG, or an unrelated—and possibly unknown—function (blind FROG) [14]. Various iterative algorithms, such as the very efficient principal component generalized projections algorithm (PCGPA) [14], can then be used to extract both $E(t)$ and $G(t)$ from $S(\omega,\tau)$. The FROG technique is best understood intuitively by considering measurements performed with pure *amplitude* gates $G(t)=f(t) \in \mathcal{R}$. However, femtosecond pulses metrology shows that pure *phase* gates $G(t)=e^{i\phi(t)}$ can also be used [15].

As most other attosecond measurement techniques, CRAB is based on the photoionization of atoms by the as field, in the presence of a dressing laser field. We consider an atom with ionization potential I_p, photoionized by an xuv electric field $\mathbf{E}_X(t)$, in the presence of a low-frequency laser field $\mathbf{E}_L(t)=-\partial \mathbf{A}/\partial t$ shifted by a variable delay τ [$\mathbf{A}(t)$ being the vector potential of this laser field]. The transition amplitude to the final continuum state $|\mathbf{v}\rangle$ with momentum $\mathbf{v}$, is given, within the strong field approximation, by [8,9,16]

$$a(\mathbf{v},\tau) = -i \int_{-\infty}^{+\infty} dt e^{i\phi(t)} \mathbf{d}_{\mathbf{p}(t)} \mathbf{E}_X(t-\tau) e^{i(W+Ip)t}, \qquad (2)$$

$$\phi(t) = -\int_{t}^{+\infty} dt' [\mathbf{v}\cdot\mathbf{A}(t') + \mathbf{A}^2(t')/2]. \qquad (3)$$

$\mathbf{p}(t)=\mathbf{v}+\mathbf{A}(t)$ is the instantaneous momentum of the free electron in the laser field. $\mathbf{d}_{\mathbf{p}}$ is the dipole transition matrix element from the ground state to the continuum state $|\mathbf{p}\rangle$. $W=\mathbf{v}^2/2$ is the final kinetic energy of the electron.

Equations (2) and (3) show that the main effect of the laser field is to induce a temporal phase modulation $\phi(t)$ on the electron wave packet $\mathbf{d}_{\mathbf{p}}\mathbf{E}_X(t)$ generated in the continuum by the xuv field. Qualitatively, the trajectory of a photoelectron from its parent ion to the spectrometer depends on its time of ionization within the laser field optical cycle [8]: the phase it accumulates along this trajectory is thus temporally modulated by the dressing field. Because of the scalar product $\mathbf{v}\cdot\mathbf{A}$ in Eq. (3), the photoelectrons have to be observed in a given direction for the phase modulation to be well defined.

Different ways of using this ultrafast electron-phase modulator for the characterization of xuv fields have already been demonstrated or proposed. In the limit of a single many-laser-cycle-long xuv pulse, $\phi(t)$ corresponds to a periodic phase modulation on the photoelectron wave packet. This leads to the appearance of sidebands in the photoelectron energy spectrum [17], which have been used to characterize fs to ps xuv pulses, either by cross correlation with the envelope of fs laser pulses [17], or by FROG measurements [18,19]. In the other limit of an as xuv pulse significantly shorter than the dressing field optical period, depending on the choice of the delay τ, attosecond spectral shearing interferometry [10] or streak-camera [2,8,9] measurements can be performed. In the latter, τ is chosen in such a way that the phase modulation is quadratic in time: the electron wave packet then experiences a linear streaking in energy $dW/dt = -\partial^2\phi/\partial t^2$, and the resulting distortion of the photoelectron spectrum provides direct information on the duration of the as pulses.

CRAB provides another, much more versatile, way of using this electron-phase modulator. Its principle can be derived from the FROG technique, by comparing the expression of $S(\omega,\tau)$ given by Eq. (1), which describes an optical FROG, and the expression of the photoelectron spectrum $|a(\mathbf{v},\tau)|^2$ in a given observation direction, obtained from Eqs. (2) and (3). This comparison shows that, by scanning the delay τ, the dressing laser field can be used as a temporal phase gate $G(t)=e^{i\phi(t)}$ for FROG measurements on electron wave packets generated in the continuum by attosecond fields. The full characterization of these wave packets provides all the information on the temporal structure of the generating as fields.

To demonstrate that this electron phase modulator is well suited for attosecond measurements, we consider the particular case of a linearly polarized dressing laser field $\mathbf{E}_L(t) = \mathbf{E}_0(t)\cos(\omega_L t)$, long enough for the slowly varying envelope approximation to apply. $\phi(t)$ is then given by $\phi(t)=\phi_1(t) + \phi_2(t)+\phi_3(t)$, with

$$\phi_1(t) = -\int_t^{+\infty} dt U_p(t),$$

$$\phi_2(t) = (\sqrt{8WU_p}/\omega_L)\cos\theta\cos\omega_L t, \tag{4}$$

$$\phi_3(t) = -(U_p/2\omega_L)\sin(2\omega_L t).$$

$U_p(t)=E_0^2(t)/4\omega_L^2$ is the ponderomotive potential of the electron in the laser field at time t. The observation angle θ is the angle between $\mathbf{v}$ and the laser polarization direction. $\phi_2(t)$ and $\phi_3(t)$ oscillate, respectively, at the laser field frequency and its second harmonic. Due to the fast oscillations in $\phi(t)$ and to the large amplitude of the phase modulation, this electron-phase modulator has a bandwidth $|\partial\phi/\partial t|_{\max}$ of 5 fs^{-1} ($\approx$20 eV) for realistic parameters (W=100 eV, θ=0, I_{laser}=10 TW/cm^2 at 800 nm), which makes it adequate for attosecond measurements. On the other hand, as we will show, the slow variations of all terms of $\phi(t)$ associated to the envelope $E_0(t)$ of the laser pulse, allow us to simultaneously determine the femtosecond temporal structure of trains of as pulses.

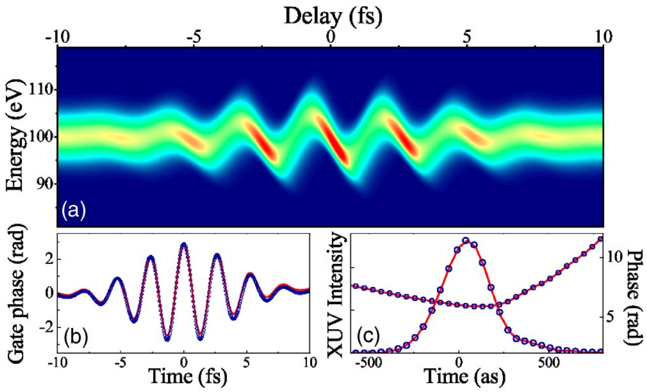

FIG. 1. (a) CRAB trace of a single 315 as pulse [full width at half maximum (FWHM) of intensity], having second- and third-order spectral phases (Fourier limit=250 as), gated by a Fourier-limited 6-fs 800 nm laser pulse, of 0.5 TW/cm^2 peak intensity. The electrons are collected around θ=0 with an acceptance angle of ±30°. (b), (c) A comparison of the exact as pulse and the laser-induced gate phase $\phi(t)$ (full line) with the corresponding reconstructions (dots) obtained from the CRAB trace after 100 iterations of the PCGPA algorithm [20]. The gate modulus $|G(t)|$ is constant and equals to 1.

We now use Eqs. (2) and (3), and the iterative PCGPA algorithm developed for the optical blind FROG, to simulate an experiment and demonstrate different schemes of CRAB for a linearly polarized laser pulse. We first show how CRAB extends existing methods for single as pulses [1,2] and trains of as bursts [3], and finally we present the case of an as field that, as far as we know, no other existing method allows to characterize.

Figure 1(a) shows a CRAB trace calculated around the θ=0 direction using Eqs. (2) and (3), for a single 315 as pulse. From a classical point of view, this trace can be understood qualitatively as resulting from the oscillations of a suddenly freed electron in the dressing laser field. Figures 1(b) and 1(c) show the as pulse, and the gate phase $\phi(t)$, retrieved from this trace using PCGPA [20], and compares them with the exact profiles. For both signals, the agreement is excellent.

We emphasize the striking similarity of the CRAB trace of Fig. 1, with Figs. 4 of [1] and [2], which provided experimental evidences of the generation of a single as pulse. Our approach provides a systematic and straightforward procedure for the full retrieval of the laser pulse and the as burst from such measurements. This procedure has many advantages which are inherited from optical FROG [13]. Due to the high redundancy of information in the CRAB trace, it is very robust against noise, and is unlikely to properly converge if experimental flaws exist, e.g., shot-to-shot variations in the as pulse temporal structure. The retrieval of the laser pulse offers an additional opportunity to check the validity of the measurement, by comparison with the results of the standard methods for visible pulses.

For any given delay τ_0 in Fig. 1(a), the spectrum $S(\omega,\tau_0)$ can be considered as an attosecond streak-camera measurement [8], each delay corresponding to a different streaking speed: in this scheme of CRAB, the information on the temporal structure of the pulse is also obtained by streaking the electron energy. The ultimate temporal resolution is thus determined by the maximum streaking speed that can be achieved, i.e., by the maximum laser intensity that can be applied to the atoms. This leads to a limit of ≈70 as for $W=100$ eV for near Fourier-limited pulses, as demonstrated in [8].

As illustrated by the results of Fig. 1, CRAB has a large angular acceptance at $\theta=0$: approximating $\phi(t)$ by the long-pulse expression Eq. (4), and given that $U_p \ll W$, $\phi_2(t)$ is the dominant term of $\phi(t)$ for all angles except $\theta \approx \pi/2$, and has a slow angular dependence around $\theta=0$. However, one specificity of $\phi_2(t)$ is that it depends on the final electron energy W. This dependence is not taken into account by the existing reconstruction algorithms, thus introducing systematic errors in the reconstructed pulses. We have checked numerically that these errors are negligible provided the bandwidth of the as pulse is small compared to its central frequency. Besides, such systematic errors do not occur at $\theta=\pi/2$, where $\phi_1(t)$ and $\phi_3(t)$ dominate, but measurements then have to be carried out with a much smaller collection angle, typically of a few degrees.

We now turn to the complete characterization of trains of as pulses using CRAB. Such trains are naturally generated by high-order harmonic generation (HHG) on gaseous targets with intense many-cycle laser pulses, and their accurate characterization is essential for their future use in attosecond pump-probe experiments. CRAB requires no specific relationship between the periods T of the train and T_ϕ of the laser-induced phase-gate oscillations. However, in the most general case, the resulting CRAB trace is complicated, which makes it difficult to determine how the temporal information is encoded in the trace. Two particular schemes enable us to get some insight into how CRAB works for trains.

The first one corresponds to $T=T_\phi$, i.e., the train and the gate oscillations have the same period. The as field generates a train of continuum electron wave packets, which experience almost identical energy streakings by the laser field. Due to the resulting temporal periodicity of the dressed train, the obtained CRAB trace is similar to the single-pulse trace of Fig. 1(a), but is now discretized along the energy axis, with a sampling step of $1/T$. Thus, in this scheme, the temporal information on each as burst is still obtained through an energy streaking, resulting in an intensity-dependent temporal resolution. This streaking now leads to the appearance of "outer" sidebands, below or above the field-free spectrum.

The second instructive scheme corresponds to $T=T_\phi/2$, i.e., the period of the train is half that of the gate oscillations. This situation is naturally encountered experimentally when the same laser pulse is used both to generate HHG in a gas and to characterize the resulting superposition of harmonics [3]. Figure 2 shows a CRAB trace obtained in this scheme,

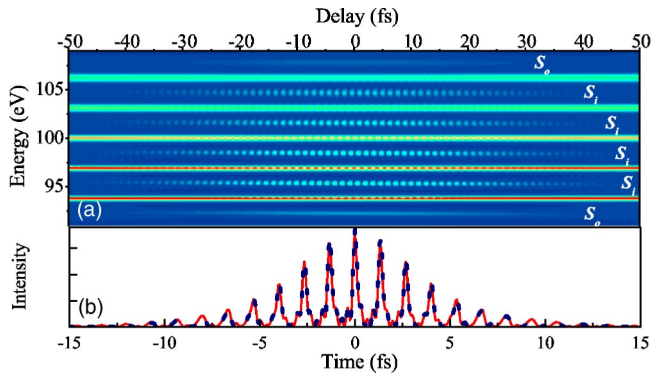

FIG. 2. (a) CRAB trace at $\theta=0$ of a 12-fs-train of nonidentical as pulses, of period $T/2=1.3$ fs, gated by a 30-fs-800 nm- ($T=2.6$ fs) laser pulse, of 0.05 TW/cm^2 peak intensity, assuming a spectrometer resolution of 100 meV. The as pulses are shorter in the center of the train (≈250 as), than in the edges (≈400 as). The outer and inner sidebands are respectively labelled S_o and S_i. (b) A comparison of the exact as train (red line) and the reconstruction (dotted blue line) obtained from the CRAB trace after 750 iterations of the PCGPA algorithm.

and the train of nonidentical pulses retrieved from this trace. Although this configuration can also be entirely analyzed in the time domain [21], it is more easily understood in the frequency domain: the information on the temporal structure of the bursts is partly obtained through the same process as in resolution of attosecond beating by interference of two-photon transitions (RABBITT [3,4,11]), which is the following. Due to the very specific ratio of T and T_ϕ, the upper first-order sideband of the harmonic n overlaps and interferes with the lower first-order sideband of the harmonic $n+1$. As the delay is scanned, these interferences lead to an oscillation of these "inner" sideband amplitudes with a period of T [Fig. 2(a)]. RABBITT measurements are performed in the perturbative intensity regime: the phase of these oscillations then provides the relative phase between neighboring harmonics, which suffices to retrieve trains of identical as bursts.

The "interferometric" version of CRAB shown in Fig. 2 extends RABBITT in several respects. (i) By scanning the delay until the two fields no longer overlap, and thus exploiting the envelope of the laser pulse in a similar way as in [18,19], trains of nonidentical pulses can now be retrieved. (ii) To obtain the reconstruction of Fig. 2(b), we not only use the amplitude of the inner sidebands [3,4], the full photoelectron spectrum is injected in the PCGPA algorithm. (iii) Using this procedure, there is no more restriction on the intensity of the dressing field. At high intensity, the information on the as pulses temporal structure is encoded both in the sideband interference pattern and a streaking effect [leading to outer sidebands, Fig. 2(a)]. Thanks to the interference effect, the temporal resolution of this scheme does not depend on the laser intensity: by taking advantage of the gaps in between the harmonics, only a small bandwidth is required for the electron phase modulator, whatever the number of harmonics involved. Measurements of trains of arbitrarily short as bursts can thus be carried out without necessarily using very high laser intensities, a major advantage over the streaking scheme previously described. From an experimental point of

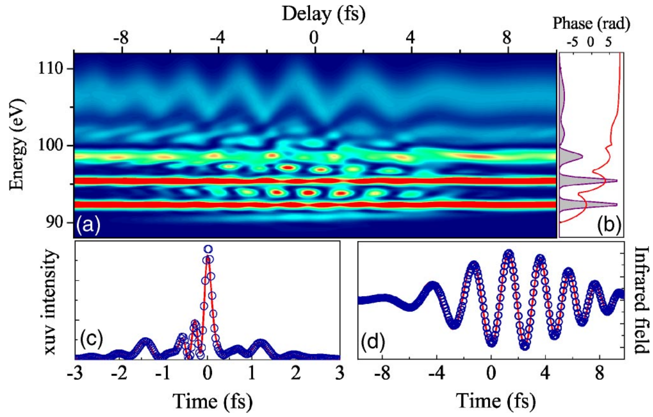

FIG. 3. (a) CRAB trace at $\theta=0$ of a complex as field, whose spectrum (shaded curve) and spectral phase (red line) are shown in panel (b). The spectrum consists of discrete peaks spaced by ≈ 3 eV in its lower part, and a continuous component in its upper part. In (c) and (d), the exact intensity profile of the as field and the laser electric field (full lines) are compared to the ones retrieved from this trace (dots) after 300 iterations of PCGPA.

view, the finite resolution of electron spectrometers can introduce systematic errors in CRAB traces of trains if the harmonics are too narrow. We have determined numerically that for an accurate retrieval, a resolution of 100 meV sets an upper limit of about 8 fs on the Fourier-limited duration of the as train.

We now illustrate the universality of CRAB by treating the case of a rather complicated as field (Fig. 3). The spectrum of this field qualitatively corresponds to what would be obtained by selecting the end of the plateau and the cutoff of the high-harmonic spectrum generated in a gas by a few-cycle laser pulse [Fig. 3(b)]. As in HHG in gases, each harmonic peak has a different chirp, and the relative phase of these peaks does not vary linearly [Fig. 3(b)]. Figure 3(a) shows the CRAB trace obtained when an 800–nm 7-fs chirped laser pulse ($\lambda = 800$ nm and $I = 0.05$ TW/cm^2) is used as a phase gate. The temporal intensity profile of the as field retrieved from this CRAB trace is in excellent agreement with the exact profile [Fig. 3(c)]. Based on the previous analysis of simpler CRAB traces, two main relevant features can be identified in Fig. 3(a): the overall oscillations of the continuous upper part, and the oscillating sidebands in the discrete lower part. As the energy varies, a gradual transition between these two regimes is observed. CRAB is the only existing method for the full characterization of such complex as fields. This will, for instance, allow us to study the transition regime between as trains and single as pulses generated by few-cycle laser pulses or through polarization gating [22].

In a conclusion, FROG CRAB is a general method for the complete temporal characterization of arbitrary attosecond fields, which consists of adapting the concepts of optical FROG to electron wave packets. It is simple, systematic, and robust against noise and experimental flaws. All the experimental tools are available for its implementation, thus opening the way to the routine characterization of attosecond fields.

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