Attosecond Cross Correlation Technique

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A technique for the measurement of attosecond, extreme-ultraviolet pulses is proposed and theoretically analyzed that is based on cross correlation of the attosecond pulse with a strong laser pulse in a gas target. Pulse durations on the time scale of a fraction of the optical period can be resolved. The method is linear in the extreme-ultraviolet intensity, which ensures high efficiency and applicability for wavelengths to below 10 nm.

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Recently, a number of methods have been proposed for the generation of attosecond (as) pulse trains [1–6] and single as-pulses [7,8]. The most promising route for the generation of a single as-pulse is high harmonic generation (HHG) with a few-cycle Ti:S laser pulse in a noble gas [8,9]. Both spectral width and profile of harmonic radiation observed in recent experiments with few-cycle laser pulses corroborate theoretical predictions of single as-pulse generation. Unfortunately, as-pulse durations could not be directly measured so far, since commonly used autocorrelation and cross-correlation techniques cannot be extended into the attosecond time and extreme-ultraviolet (XUV) wavelength regime [10–13]. On the one hand, there is a lack of nonlinear mechanisms [10] that are efficient enough to realize autocorrelation techniques at short wavelengths. On the other hand, the time resolution of the more efficient cross correlation techniques, such as laser assisted single photon ionization of gases [13], is limited to a few femtoseconds by the shortest feasible laser pulse duration.

In this Letter we present and theoretically analyze a technique for the measurement of as-pulse durations termed the attosecond cross correlation (ACC) method that is not subject to the above limitations. The as-XUV pulse is cross correlated with the electric field of a strong laser pulse in a gas target. The parameters are chosen in such a way that electrons are set free by a combination of Coulomb barrier suppression by the laser field and single XUV photoionization. The XUV pulse duration is determined by measuring the ionization yield as a function of delay between laser and XUV pulse. The strongly nonlinear character of optical field ionization distinguishes our method from previous techniques working in the perturbative limit of photoionization [11,13]. By cross correlating with the rapidly alternating electric field rather than with the much more slowly varying pulse envelope one realizes a temporal resolution of a fraction of the laser optical period required for the measurement of as-pulses. Finally, the photoionization signal depends linearly on the XUV-as pulse intensity, which provides for high efficiency and therefore allows extension to short wavelengths.

For the ACC technique we assume that the as-pulse is created by HHG with a few-cycle Ti:S laser pulse [8] in a noble gas. The few-cycle laser pulse is split into two parts, where one part is focused into a gas target for harmonic generation. The high energy end of the harmonics corresponds to a single as-pulse [8] and is selected by a suitable combination of filters [8,14] or by a multilayer mirror. The as-pulse and the second part of the laser pulse are then simultaneously focused into a singly ionized He plasma, which is generated by an intense circularly polarized laser prepulse. With circular polarization nonsequential double ionization [15] of He is suppressed. The photon energy of the XUV pulse is chosen smaller than the transition energy between the He^+ $1s$ and $2p$ states; see Fig. 1. The peak electric field strength of the laser pulse must be large enough to suppress the Coulomb barrier below the $2p$ state of the He^+ ion. Then, the as-pulse generates a maximum number of He^{++} ions, when it coincides with a peak of the laser electric field, while He^{++} production is lowest at zero field strength. Without XUV radiation the number of He^{++} ions remains negligible. The depth of modulation of the He^{++} yield gives a measure of the as-pulse duration.

The above picture was confirmed by solving the time-dependent Schrödinger equation in the dipole approximation for a hydrogenlike ion with nuclear charge Z in the

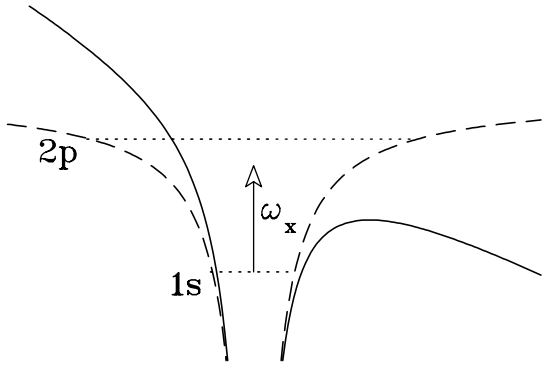

FIG. 1. Principle of the attosecond cross correlation measurement: A strong electric laser field suppresses the Coulomb potential of a He^+ ion. The solid and dashed lines are the ion potential at peak and zero laser field, respectively. At peak field strength an as-pulse can single-photon (ω_x) ionize He^+. The XUV photon energy is chosen smaller than the energy difference between the $1s$ and $2p$ states (40.8 eV).

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simultaneous fields of an as-XUV pulse and a few-cycle laser pulse

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r}) = \left\{\frac{1}{2}[i\nabla + \mathbf{A}_l(t) + \mathbf{A}_x(t-\delta)]^2 - \frac{Z}{r}\right\} \times \Psi(\mathbf{r}). \quad (1)$$

Three-dimensional coordinates are denoted by $\mathbf{r}$ and ∇ is the corresponding gradient operator. The XUV pulse is delayed by δ relative to the laser pulse. The vector potentials of the laser $\mathbf{A}_l$ and the XUV pulse $\mathbf{A}_x$ are given in atomic units by

$$\mathbf{A}_j(t) = \hat{\mathbf{z}}\frac{I_j^{1/2}}{\omega_j} f_j(t)\cos(\omega_j t), \qquad j = l, x. \quad (2)$$

Linear polarization in the $\hat{z}$ direction is assumed. The parameters I_j and ω_j denote peak intensity and center circular frequency in atomic units, respectively. Further, the laser and XUV pulse envelopes are normalized to give $f_j(0) = 1$. For our investigation we used Lorentzian and sech shaped envelope functions. Equation (2) was solved numerically by complex scaling and L^2 discretization as described in Ref. [16]. Propagation effects do not play a role for ACC measurements and can be neglected [17]. Figure 2 shows the He^{++} yield as a function of delay time δ for the following parameters: $I_l = 10^{15}$ W/cm^2, $I_x = 10^{12}$ W/cm^2, center wavelengths $\lambda_l = 800$ nm and $\lambda_x = 36.5$ nm, and sech envelope functions with full width at half maximum (FWHM) durations of $\tau_l = 5$ fs for the laser and $\tau_x = 600$ as for the XUV pulse. The XUV-pulse parameters used for our calculation are easily realizable with current state-of-the-art tabletop laser systems [18]. Measurements of the high harmonic spectrum close to the cutoff in combination with theoretical calculations [9] show that pulse envelopes described by a smooth function of, for example, sech, Gaussian, or Lorentzian shapes present a reasonable approximation to the actual as-pulse shape. The He^{++} ion yield of $He^{++}/He^{+} > 10^{-4}$ is well above the detection limit. Since the yield decreases only linearly with XUV intensity, significantly lower XUV peak intensities than used in our calculations can be used for ACC measurements.

Ionization by the laser alone was found to be negligible. As the XUV spectrum of HHG beyond the harmonic cutoff frequency drops by several orders of magnitude, the spectral components of the XUV pulse lie below the single photon threshold, and ionization by the XUV pulse in absence of the laser pulse is negligible as well. Therefore, the He^{++} ionization yield is exclusively due to the combined laser and XUV fields. As the delay between the two pulses varies, the ionization yield is modulated at 1/2 of the optical cycle of the laser pulse. Over a delay time of three optical half cycles one sees three equally high peaks of the ionization yield with two equally low minima in between. The ionization yield is nonzero even when the XUV pulse coincides with a node of the laser field, because the XUV pulse extends to either side of the node to laser field strengths that allow for ionization. Outside the range of constant modulation, the laser electric field is too weak to suppress the Coulomb barrier and ionization drops rapidly.

The duration of the XUV pulse can be determined by comparing the peak He^{++} yield Y_1 to the nearest minima Y_0 at delay times of ± 0.25 laser optical periods. Figure 3 shows the modulation ratio Y_0/Y_1 as a function of XUV pulse duration in the range $\tau_x = 300$ to 1200 as. The method is robust against fluctuations of the laser parameters. The shaded area covers a variation of laser peak

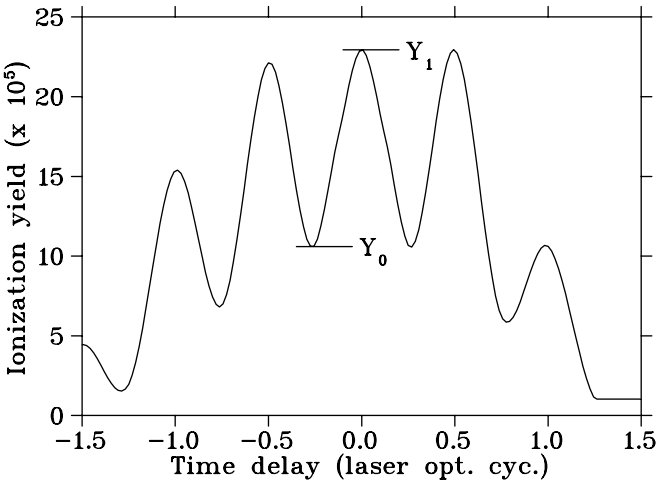

FIG. 2. He^{++} yield versus relative delay between attosecond pulse and laser pulse for the following parameters: 600 as (FWHM) pulse duration, $\lambda_x = 36.5$ nm (corresponding to a photon energy of 34 eV) with $I_x = 10^{12}$ W/cm^2, 5 fs laser pulse with $I_l = 10^{15}$ W/cm^2 and wavelength $\lambda_l = 800$ nm (optical cycle = 2.66 fs). The ratio Y_0/Y_1 depends on the XUV pulse duration.

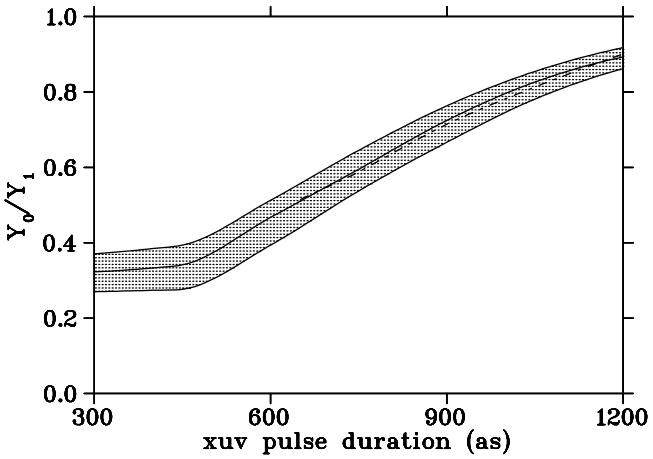

FIG. 3. Dependence of the He^{++} yield modulation on the XUV pulse duration. For the definition of Y_0 and Y_1 and of the remaining parameters, see Fig. 2. Solid line: laser parameters as in Fig. 2, shaded area: variation of the laser intensity by $\pm 20\%$; dashed line: Lorentzian XUV pulse shape. The yield modulation for Lorentzian laser pulse shape and a 7 fs laser pulse coincides with the result for 5 fs and sech shape.

intensity by ±20%, the resulting uncertainty in τ_x amounts to ±75 as for pulse durations between 600 and 900 as. Errors due to variations of laser pulse duration, laser pulse shape, and XUV-pulse shape remain well within that area. In order to achieve reasonable accuracy from ACC measurements, knowledge and stability of the laser parameters within the above assumed uncertainty is necessary. Whereas laser pulse duration and shape can be accurately monitored by spectrogram techniques [9], precise measurement of the laser intensity is a difficult task. Fortunately, the ACC method makes it possible to gauge the absolute laser peak intensity, which is shown in Fig. 4. This can be done by operating the laser at the maximum of the peak ion yield, just above the intensity at which the barrier is completely suppressed.

Assuming that laser pulse parameters can be controlled on the level indicated above, the primary source of error in an ACC measurement of the as-pulse duration will be fluctuations in the XUV peak intensity. Note that the modulation ratio is independent of the XUV peak intensity, as long as it can be assumed to remain constant during a measurement. XUV intensity fluctuations, e.g., between a node and an antinode of the laser pulse, introduce a change of the modulation ratio proportional to the change in XUV intensity. These fluctuations must be corrected for by monitoring the XUV pulse energy changes within the desired accuracy. An experimental error in the modulation ratio Y_0/Y_1 of ±10% causes errors in the pulse duration of the order ±100 as.

For the detection of as-XUV pulses at shorter wavelengths the ion charge must be increased, such that the ground state energy remains below the single photon ionization threshold. The necessary laser parameters and expected ion yields can be estimated by scaling our numerical findings for He^+ (nuclear charge $Z = 2$) and $\lambda_x = 36.5$ to higher Z. The Schrödinger equation (1) of a hydrogenlike ion with a slowly varying laser field ($\omega_l \ll 1$) and a rapidly alternating XUV field is approximately invariant under the following scale transformation:

$$\begin{aligned} \lambda_x &\to \lambda_x/\alpha, \qquad Z \to \sqrt{\alpha}\, Z\,, \\ \mathbf{r} &\to \mathbf{r}/\sqrt{\alpha}, \qquad t \to t/\alpha\,, \\ I_0 &\to \alpha^3 I_0, \qquad I_x \to \alpha^3 I_x\,. \end{aligned} \tag{3}$$

Time and length coordinates are denoted by t and $\mathbf{r} = (x, y, z)$, respectively. The scaling holds rigorously, when the time dependence of the laser field is treated in the quasistatic approximation [9]. The ionization yield, which is a dimensionless quantity, is invariant under the change of scale. For example, at a quarter of the wavelength used in Fig. 2 ($\alpha = 4, \lambda_x \to \lambda_x/\alpha = 9.125$ nm) the ionization yield remains constant, when the nuclear charge is increased to $Z = 4$, the laser intensity I_0 and the XUV intensity I_x are increased by a factor of 64, and the pulse duration is decreased by a factor of 4. For fixed Z and fixed laser parameters the single XUV photoionization yield is proportional to I_x and to λ^4.

Attosecond pulses may be generated by HHG not only as single pulses, but also as pulse trains consisting of two or more comparably strong pulses separated by multiples of one-half of the fundamental laser period. Although the spectrum of the XUV pulse contains information on the number of pulses, an unambiguous determination of the number of as-pulses requires additional evidence. ACC provides a unique signature that distinguishes a single pulse from a pulse train. Figure 5 shows the ion yield by a pulse train consisting of one, two, and three equally strong as-XUV pulses separated by half a laser period at a laser intensity where the peak ionization yield becomes saturated; see Fig. 4. Single pulses create a central sequence of three equally high ionization peaks, which corresponds to the three times, when the barrier is suppressed by the given 5 fs laser pulse. In contrast, a triplet of pulses creates a central dominant peak with two smaller side peaks. This at first sight somewhat counterintuitive behavior is due to the fact that maximal yield for a triplet of pulses can be achieved only when all three XUV pulses coincide with the strongest laser electric field peaks. By a similar argument a pair of pulses creates two equally high peaks. From Fig. 5 we find that to discriminate a single as-pulse from a pulse train, an experimental resolution of the He^{++} peak heights of 20% or better is required.

On theoretical grounds the generation of XUV pulse trains with more than 3–4 pulses from HHG by a few-cycle laser pulse can be excluded. If needed, one can also discriminate a single pulse from a longer pulse train by using a multicycle laser pulse. Here a single as-pulse would create a long plateau of equally high peaks, while a

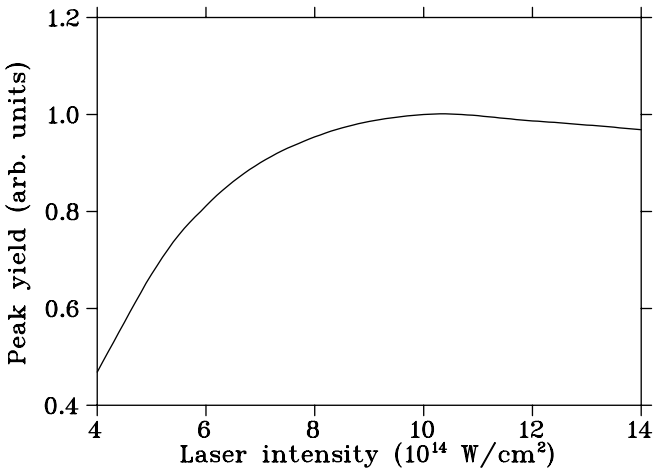

FIG. 4. Maximum ion yield versus laser peak intensity. XUV pulse parameters and laser pulse shape and duration as in Fig. 2. The ion yield reaches a maximum at the laser intensity for which the electron can escape above the barrier after absorption of a single XUV photon. Resolution of the peak ion yield within an accuracy of ±10% allows determination of the laser peak intensity within an accuracy of ±20%.

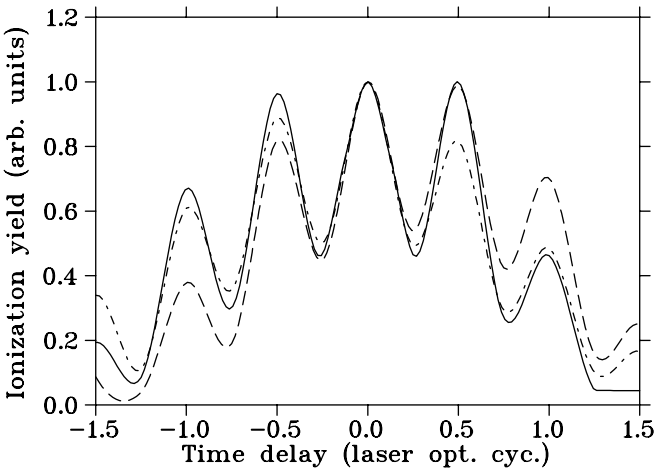

FIG. 5. Modulation of the He^{++} yield caused by a single as pulse (solid line) compared with a pair of pulses (dashed line), and a triplet of pulses (dot-dashed line). The separation between multiple pulses is assumed to be $1/2$ the laser optical cycle. Laser pulse parameters and XUV duration as in Fig. 2. A resolution of better than 20% is sufficient to distinguish a single pulse creating three equally high ion peaks from multiple pulses.

pulse train will yield a continuous rise and fall of the peak heights as one scans through the delay times.

Finally, in an ACC measurement of as-pulses the relative delay between the two pulses must be controlled on a sub-fs time scale from shot to shot, which requires mechanical stability on a sub-μm-length scale. Recent experiments [14] demonstrated that this is possible. The ACC technique is insensitive to time jitters caused by fluctuations of the laser pulse parameters, since the same laser pulse is used for both as-pulse generation and cross correlation. In particular, a critical parameter for as-pulse generation is the carrier phase of the laser pulse. The as-pulse position and peak intensity depend sensitively on the carrier phase of the laser pulse [19,20]. Recent experiments have demonstrated that the carrier phase can be stabilized [21], which greatly facilitates application of as-pulses.

In conclusion, we have presented and theoretically analyzed a cross-correlation technique for the measurement of as-pulse durations based on ionization of atoms in the combined field of the XUV pulse and a strong laser pulse. The method is linear in the XUV intensity guaranteeing high efficiency and therewith can easily be scaled to shorter wavelengths. Measurement of the harmonic pulse duration down to the few-nm-wavelength range should be possible. This opens the way towards applications of attosecond pulses, such as the investigation of dynamical processes in matter with an unprecedented resolution.

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[17] We assume a fully ionized gas with a density of 10^{17} cm^{-3}; the free electron contribution to dispersion is stronger than the contribution arising from nonresonant, bound electrons and, therefore, presents an upper estimate for dispersive effects. The phase velocity mismatch between laser and XUV pulse would shift the laser phase by $\pi/2$ over a propagation distance of 1 cm, thus destroying the subcycle time resolution. The dispersive length $L_d = (1.76\tau)^2/\beta_2$ over which the pulse is broadened by a factor of 2 is $\approx$0.5 cm and 100 cm for the 5 fs laser pulse and the 0.6 fs XUV pulse (sech shape) used in our calculations, respectively. Here β_2 is the free electron group velocity dispersion. ACC measurements can be performed for much shorter propagation distances of the order of 100 μm, thus showing that propagation effects do not play a role.
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