Second Order Autocorrelation of an XUV Attosecond Pulse Train

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Temporal widths of an attosecond (asec) XUV radiation pulse train, formed by the superposition of higher order harmonics, have been recently determined utilizing a 2nd order autocorrelation measurement. An assessment of the validity of the approach, for the broadband XUV radiation of asec pulses, is implemented through *ab initio* calculations modeling the spectral and temporal response of the two-XUV-photon He ionization detector employed. The measured width of the asec bursts is discussed in terms of the spectral phases of the individual harmonics, as well as in terms of the spatially modulated temporal width of the radiation, and is found in reasonable agreement with the expected duration.

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The superposition of harmonics of a femtosecond (fsec) laser beam may form a train of pulses with duration in the attosecond (asec) regime [1,2] or even isolated asec pulses [3]. Such pulses provide the ever highest temporal resolution to dynamical studies of ultrafast phenomena in a number of disciplines. Since this extreme temporal localization of light has been demonstrated in the laboratory, its rigorous characterization became a challenging problem that has set off intense experimental and theoretical efforts. Among them, the one targeting the extension of well established methods of optical fsec metrology to the XUV asec regime resulted in the demonstration of a second order autocorrelation (AC) measurement of an asec pulse train [4,5] formed by the superposition of five harmonics. In this experiment, the measured pulse duration was found to be substantially longer than the Fourier transform limited (FTL) duration, in contrast to the findings of other characterization approaches that have resulted durations much closer to their FTL values [2] for several sets of superpositions of five harmonics. Although the harmonics of the two different approaches are not of the same order, this discrepancy has raised important questions as to its origin. The purpose of the present work is a thorough investigation of the parameters that may lead to the considerably longer duration measured by the 2nd order AC and the settlement of the above discrepancy.

The implementation of a 2nd order AC is inherently related to the feasibility of inducing a nonlinear (NL) effect by the radiation to be characterized as well as to the development of a dispersionless AC for asec radiation. The NL process used in the experiment is the two-XUV-photon ionization of He [6]. The XUV pulse train consists of the 7th to the 15th harmonic of the Ti:sapphire laser system of the ATLAS II facility at MPQ, and it is produced in a Xe gas jet with relative intensities 7:9:11:13:15 $\rightarrow$ 0.32:1.0:0.30:0.11:0.01. The successfully implemented two-XUV-photon ionization of He is a particularly important step towards the 2nd order AC, however, in itself is not sufficient for an accurate measurement of the pulse duration of asec pulses. Two further important requirements must be fulfilled in order for the fsec pulse characterization approach to be extendable to asec pulses.

The first requirement is on the spectral response of the two-photon detector. This has to be effectively constant for the entire spectral bandwidth. In case of fsec pulses, because of the much narrower bandwidth, this requirement is generally fulfilled. For asec pulses the bandwidth extends over several eV if not tens of eV. Atomic structure in the bound and continuum spectrum does not *a priori* warrant a flat two-photon ionization cross section. The second requirement is on the practically instantaneous temporal response of the detector. Thus, no retardation in the two-photon ionization process should occur due to the inertia of populations in the intermediate virtual states. Evidently this requirement is again much more easily fulfilled for the longer fsec pulses, but its fulfilment remains an open question for pulses in the asec regime.

The spectral and temporal response of the He two-photon ionization detector has been assessed through *ab initio* quantum calculations. The six-dimensional (three dimensions for each electron) He time dependent Schrödinger equation (TDSE) $i\partial_t\psi(t) = [H_0 + D(t)]\psi(t)$ (atomic units) has been solved numerically [6]. H_0 is the field free Hamiltonian and $D(t) = -\vec{E}(t)\cdot\vec{r}$ the electric dipole interaction with the field $\bar{E}(t)$ of the harmonic superposition used in the experiment, and $\vec{r} = \vec{r}_1 + \vec{r}_2$ the position vector of the two He electrons. The energy-resolved photoelectron spectrum has been calculated and compared to that of a detector having an perfectly flat spectral response for FTL pulses, i.e., with a photoelectron distribution proportional to $[\sum_j \sum_k E_j E_k]^2$, j and k taking appropriate values ranging from 7 to 15 and fulfilling the

energy constraint $j + k = (IP_{\mathrm{He}} + E_{\mathrm{PE}})/\omega_L$, thus accounting for all the possible combinations of the two XUV fields E_i associated with all ionization channels leading to a given photoelectron energy E_{PE}. IP_{He} is the ionization potential of He and ω_L the laser photon energy in atomic units. This comparison is depicted in Fig. 1, where the different energy-resolved channels leading to the two-photon ionization are shown together with the spectral distribution of the ionization yield for the given harmonic intensities calculated solving the TDSE (black bars), and for the hypothetical case of a spectrally perfectly flat ionization cross section (gray bars). The deviation from the flat response is of the order of 30% for the main contributing channels, which has no practical consequences in measuring temporal profiles, as verified below. This calculation is complementary to the previous one [6], where the spectral ionization yield distribution was calculated only for two-photon channels of the same harmonic each time.

In the assessment of the temporal response of the detector, interferometric 2nd order AC traces for a collinear geometry have been calculated and shown in Fig. 2 for three different wavelengths, 790 nm (solid line), 767.62 nm (dashed line), and 759.45 nm (dash-dotted line), i.e., the 13th harmonic being far from resonance, near resonant, and on resonance with the $1s2p$ He intermediate state, respectively. The dipole interaction term in the Hamiltonian is now $D(t) = -[\vec{E}_1(t) + \vec{E}_2(t - \tau)] \cdot \vec{r}$, τ being a variable delay between the two autocorrelating XUV fields. The TDSE equation has been solved for a series of different delays τ, with $0 \leq \tau \leq T_L/2$ and T_L the laser period. The thus calculated AC traces have then been compared to the 2nd order interferometric AC trace $S(\tau) \propto \int_{-\infty}^{\infty}[E_1(t) + E_2(t - \tau)]^4 dt$ of a detector with rigorously instantaneous response (filled gray area). The harmonic fields have amplitudes equal to those used in the experiment and relative phases equal to zero, i.e., FTL pulses with a width of 315 asec [4,5]. The comparison shows that if all harmonics are far from resonance the two AC traces are almost identical. Since 790 nm is essentially the wavelength used in the experiment, it can be concluded that population dynamics do not affect the measured durations. Even when the 13th harmonic is near resonant with the $1s2p$ state the interferometric trace is not substantially affected in contrast to the on resonant case, where the trace shows much longer pulse duration than the real one. Apparently this is due to the remaining excited population in the $1s2p$ state.

Coming back to the measured mean pulse duration, the notable deviation from the FTL value of 315 asec is compatible with the harmonic phases resulting from the atomic response in the generation process. The measured more than twice the FTL value can be attributed to the substantially different emission time t_e [2] associated with each harmonic q and thus to the large phase difference $\Delta\varphi_q$ between subsequent harmonics. Furthermore, the emission times t_e, as a function of the harmonic order, exhibit an extended, almost linear part with a slope $(2\Delta t_e)/\Delta q = 50 \times 10^{14}/I$ (I in W/cm^2) [2], the time shifts Δt_e being the difference between the emission times t_e of two subsequent harmonics. The strong dependence of the harmonic phases on the intensity of the driving field causes some additional broadening of the measured AC peaks, due to the spatiotemporal laser intensity variation,

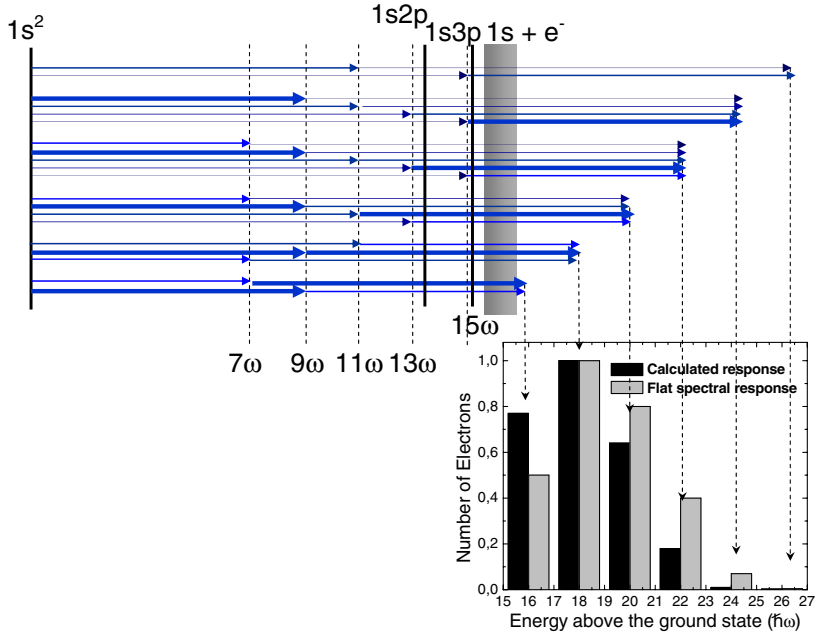

FIG. 1 (color online). All possible two-XUV-photon ionization channels by the superposition of the 7th to the 15th harmonic and the spectral ionization yield distribution calculated by solving the He TDSE (black bars) or calculated for a detector with perfectly flat spectral response (gray bars).

leading to a spatiotemporal modulation of the duration of the asec bursts. The largest contribution of this effect is from the radial intensity distribution of the laser, due to the increase of the generating surface with increasing radius. t_e has been classically [7,8] calculated and depicted in Fig. 3(a) for four driving laser intensities, I_0, $0.93I_0$, $0.78I_0$, and $0.57I_0$, corresponding to four radial segments of a Gaussian laser beam cross section, I_0 being the experiment peak intensity 0.6×10^{14} W/cm^2. Using these emission times, the experimental harmonic amplitude ratios, a mean harmonic generation nonlinearity of 6 [5], and a 2nd order extrapolated phase for the 7th harmonic, asec bursts have been reconstructed and depicted in Fig. 3(b) for each of the radial segments. At even larger radii the XUV power becomes negligibly small. The reason for using an extrapolated phase value for the 7th harmonic is that it is below the ionization threshold of the generating atom and thus no phase information can be extracted from the rescattering model [7]. As can be seen in Fig. 3(a), for an intensity range from ~0.34 to 0.6 W/cm^2, the time shift Δt_e spans from ~300 to 170 asec, respectively, and the XUV pulses, calculated for a collinear geometry, exhibit a radially strongly varying profile [Fig. 3(b)]. This is because of the harmonic phases dependence on the intensity of the driving field but also because at lower intensities the number of the superimposed harmonics reduces as the cutoff shifts to lower energies. Moreover, propagation is taken into account in this estimation only by assuming that the "long" trajectory is eliminated due to the focusing conditions in the Xe jet [9].

The above describes the expected radial distribution of the asec burst duration at the generation plane. In the experiment [4,5], this plane is imaged by the bisected mirror into the autocorrelator interaction region, thus effectively preserving the spatial phase distribution characteristics. The spatially integrating autocorrelator detector "sees" the sum of the radial autocorrelation ion signals. The sum of the AC traces shown in Fig. 3(b) and the AC trace corresponding to the peak intensity are depicted in Fig. 4 (dashed and solid lines, respectively). The resulting burst durations are 680 and 595 asec, respectively. Inclusion of temporal integration increases these values by ~30 asec. Because of the uncertainty in the phase of the 7th harmonic, the above estimated durations have a

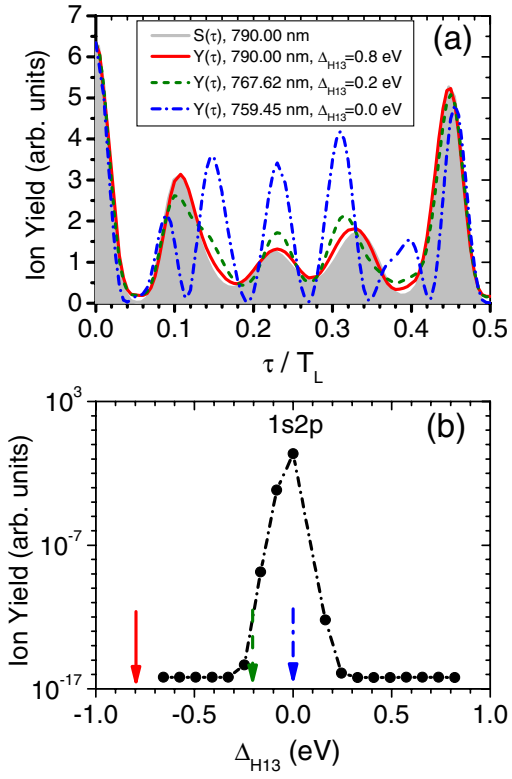

FIG. 2 (color online). (a) Calculated interferometric 2nd order AC traces for three different laser wavelengths, 790 nm (solid line), 767.62 nm (dashed line), and 759.45 nm (dash-dotted line), and compared to the interferometric trace of a detector with instantaneous response (filled gray area). (b) Position of the 13th harmonic of the three laser wavelengths with respect to the $1s2p$ intermediate state of He.

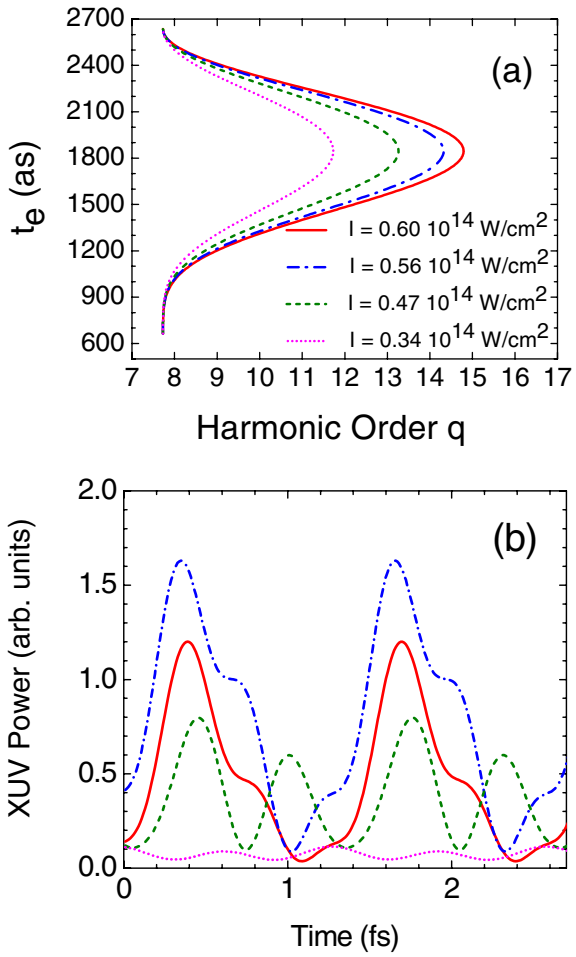

FIG. 3 (color online). (a) Classically calculated emission times t_e for four different driving laser intensities across the radial laser beam profile. (b) Attosecond pulse trains estimated using the experimental harmonic amplitudes, and the phases resulting from the calculated emission times t_e.

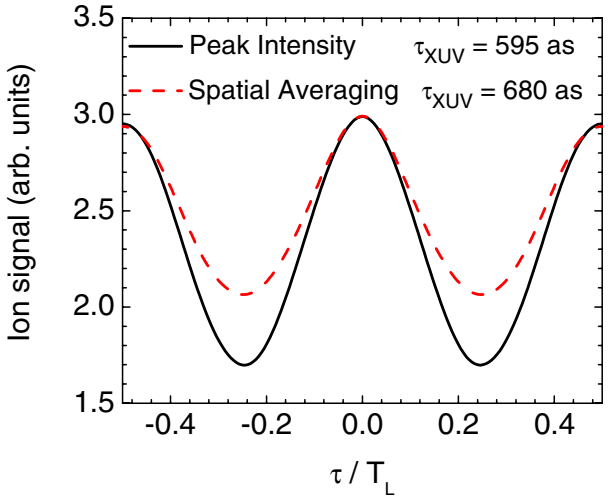

FIG. 4 (color online). Calculated peak intensity (solid line) and radially averaged (dashed line) 2nd order interferometric AC traces of a superposition of harmonics with amplitudes equal to the amplitudes used in the experiment [4,5], and phase distributions classically calculated [6,7].

large uncertainty with a lowest limit of 550 asec in the averaged trace.

These estimations indicate that the measured pulse duration of (780 ± 80) asec is in good agreement with the expected XUV pulse duration, as this results from the atomic response taking into consideration the spatiotemporal intensity variation of the generating laser field. Furthermore, it shows that the duration of the asec pulses is sensitive to the exact driving intensity, and thus they exhibit spatiotemporally modulated duration that is far from its FTL value. The lower the driving intensity is, the more pronounced the latter becomes because of the $1/I$ dependence of Δt_e. This modulation leads to a more increased measured duration than the duration at the peak intensity when the measurement is through a 2nd order AC. In contrast, in approaches that measure spatiotemporally averaged spectral phases, such as those using XUV-IR cross correlations [2], the same effect results in a reduced phase difference between subsequent harmonics, and therefore to a shorter measured burst duration [10].

Concluding, the individual pulse widths of an asec pulse train have been determined through 2nd order AC measurements. The measured width of the ultrashort bursts within the train is found to be much larger than the FTL width and reasons for that have been investigated. Artificial broadening due to the spectral and temporal response of the NL detector used has been excluded through rigorous *ab initio* calculations. The measured duration is found justifiable in terms of the spectral phases of the individual harmonics as resulted from the atomic response of the harmonic generating medium, as well as of the spatial phase modulation effects of the XUV radiation due to the intensity distribution of the generating field. Improved approaches based on 2nd (or higher) order AC of the asec radiation to be characterized are a requisite for a rigorous determination of asec structures. Towards this goal, intense harmonic generation is vital.

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