Tunnel ionization as a high-dynamic range at-focus ultrafast pulse measurement

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Introduction

Ultrafast laser pulses of the order of femtoseconds (1 fs = 10^{-15} seconds) in duration are a highly adaptable tool for observing and controlling the fastest motions of atoms within molecules, even electrons within atoms. To produce exotic states of matter (for example excited nuclear and rotational states or Rydberg orbits) or influence the geometrical evolution of a molecule, high peak powers are a desirable route to generating non-perturbative responses. Arguably the most successful technique for producing ultrafast intense laser pulses is through chirped-pulse amplification (CPA), whereby a low-power ultrafast pulse, typically from a titanium-sapphire (Ti:S) oscillator is spectrally dispersed in time (chirped), amplified in further externally pumped Ti:S crystals, then temporally compressed to the original pulse duration by reversing the chirp. Many research groups internationally are taking advantage of the availability of laboratory-scale off-the-shelf systems generating sub-30 fs, 800 nm few-millijoule energy pulses at high repetition rates (order of kHz) to investigate the behaviour of matter under these unique conditions. Prominent examples include the 'quantum control' experiments by the authors and co-workers, to be published in this Report, and the new frontier of 'attophysics', as pioneered by the groups of Krausz, L'Huillier, Corkum and Marangos among others.

CPA systems generally do not generate sufficient bandwidth to support pulses shorter than 20 fs (~ 40 nm); compression below this transform limit therefore requires additional bandwidth, often generated through self-phase modulation in a Noble gas-filled hollow fibre, and further temporal compression on reflection from pairs of multilayer chirped mirrors. Pulse durations of the order of 5 fs are the current state of the art, as demonstrated by a number of groups.

A number of highly successful optical techniques exist to measure the parameters of near-few-cycle pulses (at 800 nm, one cycle = 2.667 fs, thus of the order of ~20 fs), popular examples include frequency-resolved optical gating (FROG)[1], along with the user friendly derivative GRENOUILLE[2], and spectral phase interferometry for direct electric-field reconstruction (SPIDER)[3], both of which yield far more information than a traditional autocorrelation. Both FROG and SPIDER can be used to recover the pulse duration, spectrum, etc., however the non-iterative nature of SPIDER results in far faster measurement, and can directly recover the spectral phase.

However, in making the transition from near- to true-few-cycle pulse metrology, it is generally necessary to minimize the amount of additional dispersion introduced by transmission optics. Therefore, the robust measurement of such pulses relies on a well-characterized measurement technique, often challenging in the case of autocorrelation, FROG or SPIDER, as considerable uncertainty can exist within such setups. Furthermore, measuring the duration of a free-propagating pulse is no guarantee of focussing to a high intensity, maintaining the temporal duration We present a straightforward method producing reliable measurement of the shortest infra-red laser pulses available. This novel method uses two pieces of apparatus often available in an Ultrafast Science laboratory: a time-of-flight mass spectrometer and a co-linear interferometer.

Laser System

The laser source consists of an oscillator (FemtoSource Pro, FemtoLasers GmbH) pumped by a solid state laser (Verdi, Coherent Inc), delivering sub-10 fs pulses of a few nJ in energy. This pulse is dispersively stretched to ~20 ps and amplified to ~2 mJ in a multi-pass amplifier, pumped with a second diode-pumped solid state laser (Jade, Thales Laser S.A.). The amplified pulses are compressed by two pairs of Brewster-angled fused silica prisms: by changing the prism pair separation, the dispersion of the compressor can be accurately controlled, allowing an optimal pulse duration of <30 fs to be measured. To produce sufficiently short pulses to temporally probe quantum dynamics[4,5], the pulse duration is further decreased. The performance of the laser system is such that the compressed pulses are near-transform limited, thus we introduce additional bandwidth to support a shorter pulse duration by self-phase modulation in a Noble gas-filled hollow fibre. Following transmission, the bandwidth is increased from ~30 nm to ~100 nm, and recompressed to a near-transform limited duration of 10 fs by a series of multi-layer chirped mirrors[6].

At-focus pulse measurement technique

To measure the electric field of a FCP, we employ the technique illustrated in Fig. 1(a). Importantly, such a measurement is made at the laser focus: the vast majority of ultrafast high-field physics and chemistry is performed with focused laser pulses, so ability to recover information about the optical conditions atoms and / or molecules are exposed to is a major advantage. The only requirements are the ability to interferometrically split initial FCP into
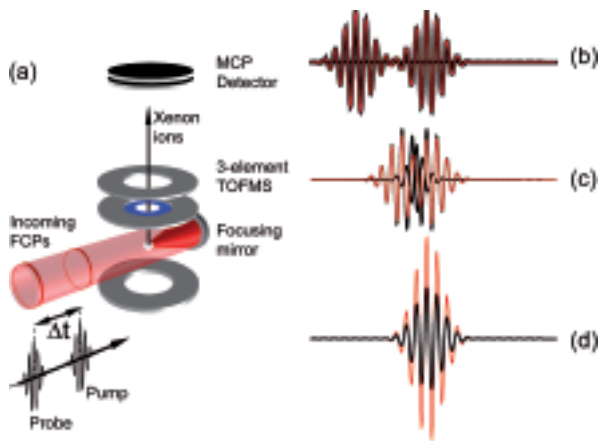
Figure 1. (a) Illustration of the proposed measurement principle. The two co-propagating pulse replicas from an interferometrically split few-cycle pulse (FCP) are delayed by time Δt, and focused into the source region of our time-of-flight mass spectrometer. The yield of multiply-charged xenon ions Xe^{q+} (q = 1 to 5) from xenon gas is measured as a function of Δt. (b) - (d) The interaction of the focused FCPs (black) is apparent from the ion yield, and reflects the resulting temporal intensity distribution (red). A relative separation of (b) 6 cycles, (c) 4 cycles and (d) zero cycles is shown.
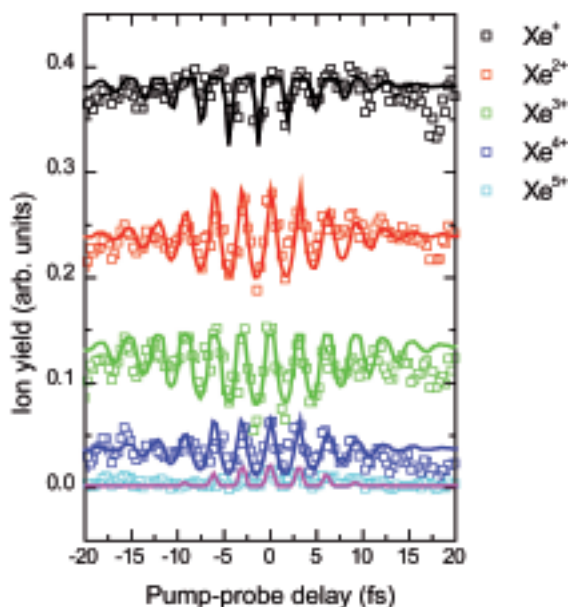

Figure 2. Direct measurement of the electric field and pulse envelope of two 12 fs pulses by observing the delay-dependence of the ionization of xenon. The data markers are the experimental results for Xe^{q+} (q = 1 to 5), and the curves are a theoretical fit using modified tunnel ionization theory and a numerical integration over the focal volume to predict the Xe^{q+} ion yield for all Δt.

two replicas, delay one pulse with respect to the other in controlled and stable manner, and be able to select a small region of the laser focus over which to measure ionization. As the information which can be learnt from a single ultrafast pulse is limited, most laboratories engaged in this type of research will employ some form of interferometer. Common methods for interfering ultrafast pulses are using

two thin beam splitters in the Mach-Zehnder configuration, or a multi-element focussing mirror, where independent concentric rings of the mirror can be relatively translated, delaying part of wavefront en route to the focus. We prefer to spatially select a region of the laser focus where the intensity is well-described, thus this technique is most successful with two completely spatially overlapping (co-linear) pulse replicas.

The FCP is split into two pulse replicas in a Mach-Zehnder interferometer. To minimise stretching of the pulses, 4-micron thick pellicle beamsplitters are employed to split and recombine the beams. The delay between the two replicas (Δt) is controlled using a high-resolution translation stage (Newport UTM-25CC), with a repeatable resolution of ~ 150 nm, equivalent to ~ 300 attoseconds double-pass. The pump-probe pulses are reflection focused in an ultra-high vacuum (UHV) ion time-of-flight mass spectrometer (TOFMS), Fig. 1(a), individually generating an intensity in excess of 10^{14} Wcm^{-2}. The laser intensity is defined by controlling the position of the focusing optic with respect to the source region of the TOFMS. Ionization yield depends nonlinearly on the laser intensity and pulse duration, thus by measuring the ionization yield as a function of pump-probe delay, the pulses can be cross-correlated, and compared to a modified ADK model. In essence the optically 'active' medium that replaces the SHG crystal is ionization in a Nobel gas.

The source region of the TOFMS is filled with xenon, at a number density low enough to avoid space-charge effects. Space charge effects become apparent when the focusing optic is translated parallel to the direction of laser propagation as a broad, featureless distribution of ions. As the pressure is lowered, the intensity dependence of ionization reveals a rich structure as the optic is translated, for example see the article on non-recollisional diabatic excitation of krypton[7] in this Report. As the temporal separation of the pump and probe pulses is varied, the resultant multiply charged ions are separated by charge-to-mass ratio in the TOFMS. A representative electron flux from each ion is generated in a pair of high gain (> 10^7) micro-channel plates, averaged on a digital storage oscilloscope.

The measured ion yield recorded as Δt is varied is shown in Fig. 2: the data markers indicate the integrated yield. The temporal structure is the result of the vector addition of the electric field in the pump and probe pulses, illustrated in Fig. 1(b)-(d). The 2.7 fs structure is clear: if the ionization yield depended purely on the modulus of the electric field, the temporal structure would have halve the period. Also apparent is the pulse envelope.

Theoretical analysis
To fit the measured Xe^{q+} (q = 1 to 5) ion yield as a function of Δt, two considerations are necessary. Firstly the theoretical ionization rate over a wide (10^{11} to 10^{15} Wcm^{-2}) intensity range is calculated. The contribution of recollision ionization to the Xe^{2+} and higher yield is quantified using the technique described in Bryan et al.[8], whereby the contribution from recollision ionization is related to the tunnel ionization[9] of lower-order charge states. The outcome of such calculations are presented in Fig. 3, whereby the dependence of the temporal evolution of the ionization yield on intensity dependence can be seen. A
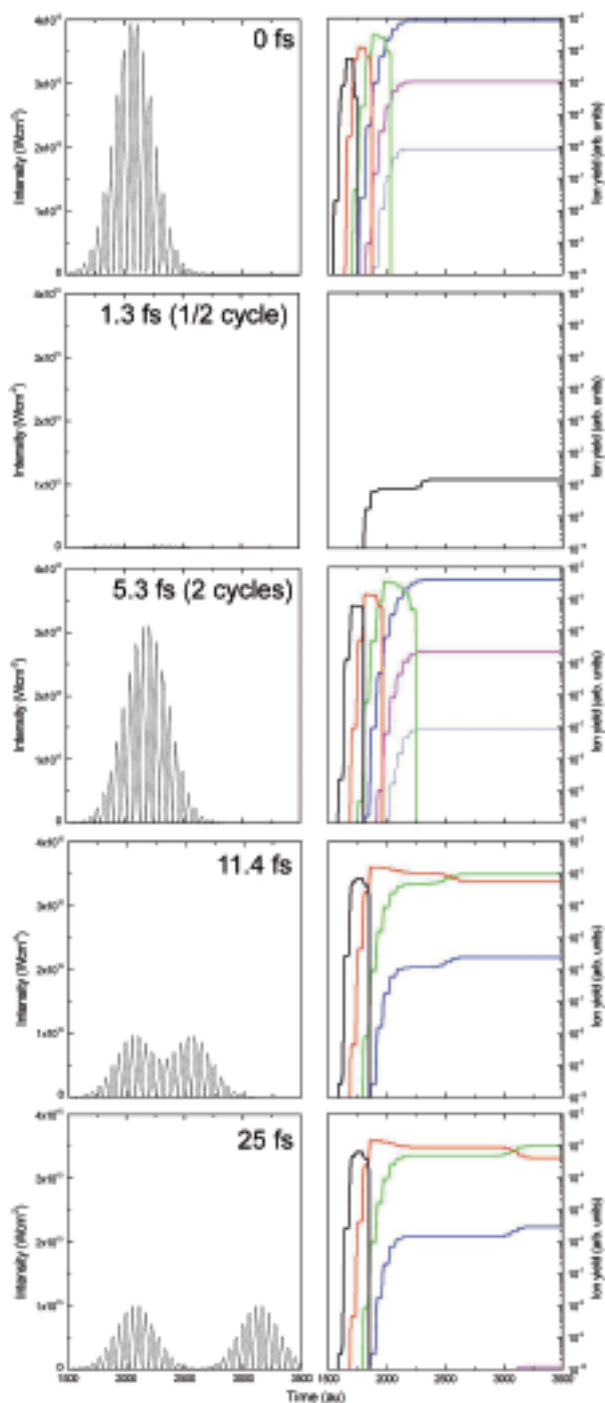
Figure 3. Examples of theoretical ionization yield as a function of time for five different pump-probe delays, Δt, showing how ionization depends on the temporal distribution of laser intensity. Here, time is in au (1 au = 24.2 attoseconds), and the same curve identification is used as in Fig. 2. Also, the final ion yield presented in Fig. 2 is the time-integrated yield.

range of pump-probe delays are presented, from totally overlapped (Fig. 3, top) to separate (Fig. 3, bottom).

Secondly, the degree of spatial integration over the focal volume by our instrument is quantified. At this point, we turn again to the powerful technique of intensity selective scanning. Measurements of ion yield as a function of detector / focal volume overlap has allowed the

quantification of the laser focus, not presented here. The combination of this measurement and the spatial selectivity of this technique will be the subject of a forthcoming publication.

To match the experimentally measured ion yield as a function of (t, for each time delay, our numerical model generates a temporal integral of the volume-independent ionization rate from -300 fs to +300 fs. This is then scaled by a spatial integral over the known focal volume. The result of such modelling is presented in Fig. 2 as the solid curves. The remarkable fit to the experimental data illustrates that our assumption of a 12 fs FWHM laser pulse is very accurate.

Conclusion

We have proposed a novel technique to recover the temporal intensity profile of a few-cycle pulse (FCP) by measuring the ionization yield from two co-propagating replicas derived from the same initial FCP. By using the highly nonlinear dependence of atomic ionization rate on *focused* laser intensity as the optically-active medium, a broad dynamic range is possible. As the relative delay between the replicas is varied interferometrically, the intensity is the square of vector sum of the electric fields of the two replicas. By modelling multiple ionization, a direct measure of the time profile of the laser intensity is possible. This technique allows an straightforward yet accurate measure of the laser intensity at focus, and is only limited by the spatial resolution of the interferometer, thus can potentially be used to diagnose attosecond-scale pulses: in the present work, a temporal resolution of 300 attoseconds is achieved.

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