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These findings are significant as progress in strong-field attosecond physics is currently hampered by the difficulty of obtaining quantitatively accurate and reproducible experimental data. This project provides such data, enabling baseline comparisons across different laboratories. The data can also test theoretical models that are widely used even though their accuracy is poorly characterized. New research questions which came about from this project: How can we transfer calibration standards efficiently to a wider range of laboratories? How can we directly calibrate the stereo-ATI technique for measurement of laser phase?

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Final Report for AOARD Grant FA2386-12-1-4025 "Benchmarking attosecond physics with atomic hydrogen"

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Abstract: Attosecond science promises to revolutionize our understanding of the electronic dynamics of matter, but relies on processes driven by intense few-cycle laser pulses with known carrier-envelope phase (CEP). Progress in both experiments and theoretical models is currently hampered by the difficulty of obtaining quantitatively accurate and reproducible data. In this project, we obtained uniquely reliable and accurate data on atomic interactions with intense few-cycle laser pulses. Our data were calibrated relative to atomic hydrogen, the only species for which exact theoretical simulations are available in this regime. We provided accurate reference data on the photoionization yield and the CEP-dependent photoelectron spectrum for several of the noble gases. From these data, we derived calibration standards for laser intensity at the percent level and CEP at the 100 mrad level. These standards are free of systematic errors and can be readily utilized in other laboratories.

Introduction:

Attosecond science promises to revolutionize our understanding of the electronic dynamics of matter, but relies on the control of highly nonlinear light-matter interactions driven by few-cycle laser pulses. In this regime, retrieving useful measurements from experimental data requires complex, nonlinear numerical simulations.

In this field, the data can be highly sensitive to experimental parameters that are themselves difficult to calibrate, calling the reliability of the data into question. For instance, the laser intensity is typically on the order of 10^{14} W/cm^2, high enough to destroy any material detector, but intensity changes of tens of percent can be sufficient to substantially alter the experimental outcome.

The use of theory to interpret data is also questionable. Exact calculations are only possible for atomic hydrogen (H), but H is difficult to work with experimentally. More commonly used targets, such as the noble gases, can only be simulated approximately. Tremendous theoretical effort has gone into developing approximate theories such as ADK, PPT, SAE-TDSE, and others. These theories underpin the interpretation of data from current experiments, but the magnitude of the errors arising from the approximations are not well known.

These two difficulties – the lack of calibration for experiment, and the lack of calibration for

theory – render quantitative progress in strong-field attosecond science virtually impossible at present. The recent controversy over the so-called attoclock experiment [1] is a prime example of this difficulty. This experiment claimed to show that, contrary to current understanding, the photoionization of an atomic electron is not an instantaneous process but takes several tens of attoseconds. However, it was recently shown [2] that the measurement of this time delay was simply an artifact of the theoretical approximations necessary to interpret the data. Such controversies will recur until attosecond science can be put on a solid quantitative footing.

Before the beginning of this project, the PI's group had demonstrated an unprecedented degree of quantitative agreement between experiment and theory in the laser interaction regime relevant to attosecond science. The key innovation was the use of atomic hydrogen, the only atom that can be simulated ab initio in this regime. The experimental data could therefore be regarded as uniquely reliable: in the specific case of hydrogen, the theory was unimpeachable and the agreement between data and theory was excellent.

The thrust of this project was to exploit the provable accuracy of the PI's experiments toward two aims. (1) To provide accurate reference data on strong-field processes, using atomic hydrogen to verify that the experimental conditions were adequately controlled. (2) To derive, from this reference data, calibration standards for experimental parameters that would be usable in other laboratories.

Experimental and theoretical methods:

The experiments in this work all followed a basic pattern. An intense few-cycle laser pulse was used to ionize a beam of atomic hydrogen. The resulting ions and/or electrons were collected in a charged-particle detector. Figure 1(a) shows a schematic of the apparatus.
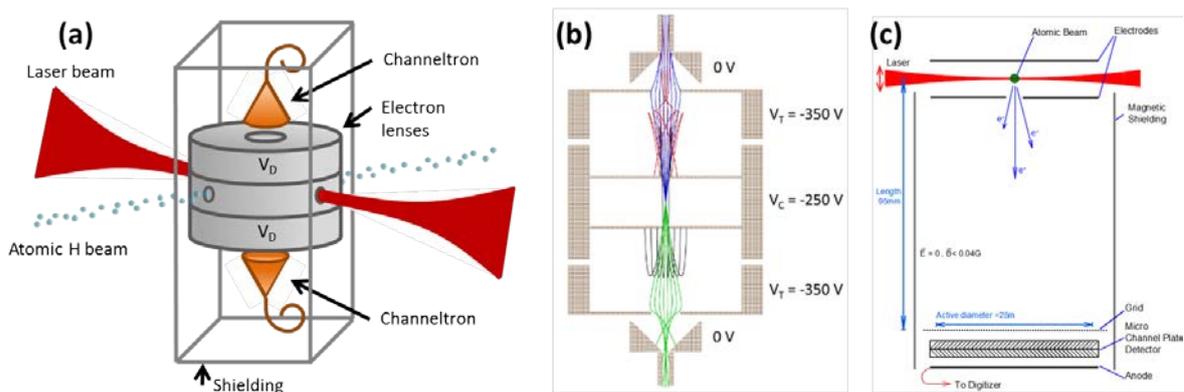
Figure 1. Apparatus for the experiments undertaken in this project. (a) Schematic overview of interaction region. Here, the repeller detector is shown, comprising electron lenses and channeltrons. (b) Schematic of the repeller detector showing electron trajectories from the interaction region at center. (c) Schematic of the time-of-flight detector.

Few-cycle laser source.

Our few-cycle pulses were produced by a commercial laser system (Femtolasers Femtopower CE-Pro). We obtained pulse durations as low as 5.5 fs and pulse energy up to 400 µJ, with pulse

repetition rate of 1 kHz. When focused with a parabolic mirror, these pulses produced intensities in the $10^{14} - 10^{15}$ W/cm^2 range. This intensity range is characteristic of strong-field attosecond physics experiments. The carrier-envelope phase (CEP) of the laser could also be stabilized using an $f - 2f$ interferometer. This capability let us probe dynamical effects occurring faster than the period of the optical electric field (2.7 fs for our laser).

Charged-particle detection.

Initially, we used a repeller-type electron detector, with electron-optical properties illustrated in Fig. 1(b). Details of this detector can be found in [P1 – P3]. During the course of this project, we constructed a time-of-flight spectrometer (Fig. 1(c)) which was used for both ion and electron detection (publications [C1 – C4, S1, S2]). This spectrometer was of a standard type, with a 10 cm magnetically shielded flight tube and multichannel plate (MCP) detection. The yields of all relevant ionic species could be accurately and independently measured by simple digitization of the MCP current. For electron detection, the energy resolution of the spectrometer was approximately 1 eV and the MCP was operated in particle-counting mode.

Theoretical analysis.

Accuracy of the theoretical simulations for H was of paramount importance, since this in turn certified the accuracy of the experimental results. Highly accurate calculations of H dynamics can be obtained by direct numerical integration of the nonrelativistic time-dependent Schrödinger equation for a single electron in a Coulomb field (referred to below as the "3D-TDSE" method). These calculations are extremely numerically intensive. Approximately 1000 CPU-hours are required in order to obtain a complete simulation set that can be used for the prediction of experimental data. The exact 3D-TDSE method cannot be applied to any species other than H, as the computational requirements become too demanding.

We collaborated with several theoretical research groups, each of which had developed their own 3D-TDSE code independently. Cross-checking the results from each group provided confidence that all codes were running correctly. Our earlier work [3] provided additional assurance that 3D-TDSE calculations were sufficient for describing experimental results.

Single-electron 3D-TDSE calculations are not directly applicable to the other atomic and molecular species that we studied, since these species had more than one electron. Our theory collaborators provided simulations based on approximate methods, in particular single-active-electron (SAE) TDSE, to study these cases. SAE-TDSE employs a modified Coulomb potential that is meant to account for the electric potential arising from the multielectron charge distribution. We also performed our own simulations of simple theoretical models, such as the Ammosov-Delone-Krainov (ADK) model [4] and the empirically corrected ADK model of Tong and Lin [5], for additional theory-experiment comparisons.

The above models all predict the behavior of a single atom in a strong field, but connecting these predictions to experiment is also a significant challenge. For instance, atoms at different positions in the atomic beam lie at different points in the laser focal volume and therefore experience different laser intensity. These issues are reviewed comprehensively in our publication [P1]. We developed a significant amount of Matlab code for post-processing the theoretical simulations from our collaborators into predictions of the experimental results. Again,

the reliability of this code was verified by the match between predictions and data for our work on H.

Results and Discussion:

A number of experiments were undertaken toward the stated aims of the project. Each successive experiment aimed to resolve ever finer features of attosecond atomic dynamics by applying different detection techniques. Hence, progressively more sophisticated and sensitive reference data was produced. Our work is described in chronological order.

Intensity calibration using electron yield measurements in H.

In this work [P3], we re-analyzed data on the yield of high-energy electrons that was taken prior to the commencement of the project. We demonstrated that a highly accurate intensity calibration standard could be derived from this data. The 1% calibration uncertainty was an order of magnitude better than previously demonstrated techniques. In addition, the calibration was shown to be free from the systematic effects that plague those other techniques.

This work showed that our basic methodology was sound: new and accurate calibration information could, in fact, be extracted from measurements on H.

Strength of carrier-envelope phase effects in H.

In this experiment, we measured the yield of high-energy electrons from H as a function of the laser CEP, showing that the data agreed with 3D-TDSE simulations. This experiment was the first to report such quantitative agreement for CEP effects. The experiment is summarized briefly here: more details can be found in [P2].

The repeller detector was used to measure the electron yield in this experiment, so that the electron energies could not be directly determined. Rather, all electrons with energies exceeding the repeller cutoff voltage were detected. By varying the repeller voltage, we could vary the electron energy cutoff. The yield oscillated sinusoidally with CEP, as observed in other work on CEP-dependent ionization. We compared the amplitude of this yield with 3D-TDSE calculations and observed quantitative agreement between experiment and theory over a wide range of electron energy cutoffs.

Accurate measurement of total photoion yield and transferrable intensity calibration.

The dependence of photoionization probability on laser intensity is one of the most fundamental observables in strong-field physics and is therefore of key importance for attosecond science. Strong-field processes are highly nonlinear in laser intensity, but the measurement of laser intensity on target is plagued by systematic errors that can easily amount to tens of percent. Therefore, one of the main goals of this program was to establish an intensity calibration method that was both accurate and readily usable in other laboratories.

The measurement of total yield of ions from photoionization is one of the simplest and most common measurements in strong-field physics. We performed this measurement for H and several noble gases. The agreement of theory and experiment for H certified the accuracy of the

data for the other gases. The data provide a percent-level intensity calibration standard that can be readily transferred to other laboratories. We anticipate publication of this work [S1] in late 2015.

We obtained highly accurate data on the intensity-dependent ionization yield of H and several of the noble gases (Ar, Kr, Xe). Agreement with 3D-TDSE predictions was obtained at the few percent level for H, certifying the accuracy of the data (Fig. 2). As usual for such measurements, the quantum efficiency of ion detection is not known and so the units of yield remain arbitrary. Again, as usual in strong-field measurements, the intensity on target can only be reliably estimated to within several tens of percent. Our fit to the 3D-TDSE therefore uses multiplicative rescaling of yield and intensity as the two free fit parameters. From the intensity rescaling, we can see that the actual intensity is equal to $I_{act} = (0.64 \pm 0.01)I_{est}$. Since the measurements agree with theory, we can rule out systematic errors in this intensity calibration. Hence, these measurements represent a primary intensity calibration standard with an inaccuracy below 2%.
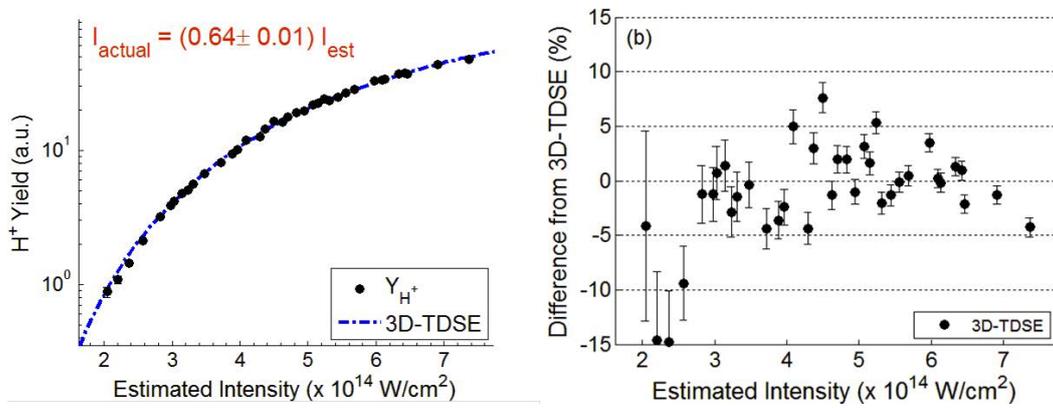
Figure 2. Measurement of photoion yield for H as a function of estimated intensity. Left: Log-scale yield (points, arbitrary units) and fit to 3D-TDSE theory (dot-dashed line). Error bars are smaller than the points. The actual intensity is retrieved from the data with an error below 2%. Right: Residuals of the fit, showing accuracy of the data at the few percent level.

Predictions of the widely used approximate ADK theory [4] differ from the 3D-TDSE predictions at the 20 – 50% level for H under our experimental conditions. Hence, these approximate theories do not provide an adequate quantitative guide for, e.g., intensity calibration.

Figure 3 shows calibrated reference data for the photoion yields of the noble gases. These reference data represent a unique benchmark for testing the various approximate theories developed for multielectron systems, such as ADK, PPT, SAE-TDSE, and others. The data were taken using the same apparatus as the H data in Fig. 2, during the same experimental run. The only difference in conditions was the substitution of gases at the atomic beam gas inlet. Hence, these data inherit the percent-level accuracy of the H data. The intensity scale in Fig. 3 is calibrated using the intensity rescaling derived from the H data.
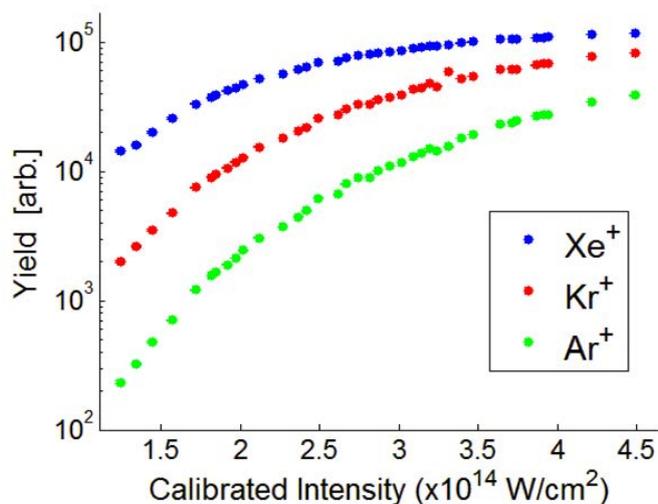
Figure 3. Calibrated photoion yields for the noble gases Ar, Kr, and Xe. The accuracy of these data is certified by the agreement between theory and experiment for H, and the intensity is calibrated relative to the H measurement in Fig. 2.

The data in Fig. 2 will enable us to construct intensity calibration standards that inherit the percent-level accuracy of the H standard. In contrast to our previous calibration standard based on H, such standards can be directly used in many laboratories. All that is necessary is a few-cycle 800 nm laser system and an ion mass spectrometer – standard tools in attosecond physics. For each of the noble gases in Fig. 2, a calibration standard can be constructed by fitting a phenomenological reference curve to the data. To use the standard, the remote laboratory measures the ion yield as a function of their estimated intensity. Since relative intensities can be estimated very precisely, they need only find the scaling factor between estimated and actual intensity. This scale factor, and thus the calibrated intensity values, can be found by fitting their experimental yield curve to the reference curve.

As discussed in previous reports, we also obtained yield data on H_2 using the same procedure as for the noble gases. The data themselves were accurate given the experimental conditions. However, it now appears that the data were much more sensitive to the temporal profile of the laser pulses than we believed at first. In the experiment, we can only measure yields of ionic species (H_2^+ and H^+), but an undetectable change in the trailing edge of the laser pulse will convert a significant fraction of H_2^+ to H^+ through the so-called enhanced ionization mechanism [6]. Moreover, the presence of a competing mechanism, dissociation, prevents us from measuring and correcting for enhanced ionization.

Overall, this problem leads to systematic errors on the order of 10% in the data. Such errors are still not large by the standards of attosecond science. However, to reproduce our results, other laboratories would have to undertake exceptional scrutiny of their own laser systems. Therefore, we no longer regard H_2 as a viable target for the collection of useful reference data.

Accurate measurement of CEP-dependent electron spectra and calibration of CEP.

The control of laser carrier-envelope phase (CEP) is a key technology for attosecond science, as it enables the laser electric field waveform to be shaped on timescales below an optical cycle. A key signature of CEP effects is that the photoelectron emission along the laser polarization direction becomes asymmetric. For example, if the polarization lies along the "up-down" direction, more electrons may be emitted "up" than "down" at a particular value of CEP. The size and polarity of this asymmetry also varies as a function of the electron kinetic energy.

Photoelectron asymmetry furnishes the operating principle for the so-called stereo-ATI phase meter [7], which has become a widely used instrument for measuring CEP. However, as usual, phase metering relies on approximate theoretical models to retrieve CEP from the observed asymmetry. Therefore, unknown systematic errors plague the retrieved CEP.

In this experiment, we measured the CEP-dependent electron spectrum for H and several of the noble gases. Our strategy was similar to the intensity calibration experiments discussed above. We observed that the H data matched the 3D-TDSE predictions, certifying the accuracy of data collection. This process enabled us to accurately determine our laser CEP, rejecting systematic errors. Our data will calibrate measurement of laser CEP for other noble gases. We anticipate publication of this work [S2] in late 2015 or early 2016.

Our experimental configuration allows us to measure the yield of electrons, $Y(E, \varphi)$, as a function of E, the electron energy, and $\square$, the CEP relative to an instrument offset phase $\square_{inst.}$. Like many laboratories, we use an $f - 2f$ interferometer for CEP-resolved measurements. this instrument can only stabilize the CEP, not measure it in an absolute sense, although the corresponding offset phase is stable over a period of days. The yield varies by orders of magnitude over the accessible range of E. We therefore examine a normalized, background-subtracted CEP signal, defined as $S(E, \varphi) = \dfrac{Y(E, \varphi) - \overline{Y}(E)}{\overline{Y}(E)}$, where $\overline{Y}(E)$ is the CEP-averaged value of $Y(E, \varphi)$.

Figure 4 shows example data and simulations for H and the noble gases at an intensity of $2 \square 10^{14}$ W/cm^2. The simulations based on the 3D-TDSE for H and the SAE-TDSE for the noble gases. Theory and experiment agree well for H over the entire range of electron energies and CEP, indicating that accurate data are being obtained. A characteristic "checkerboard" pattern at low electron energy gives way to a strong, nearly energy-independent "bar"-shaped feature at higher energy.
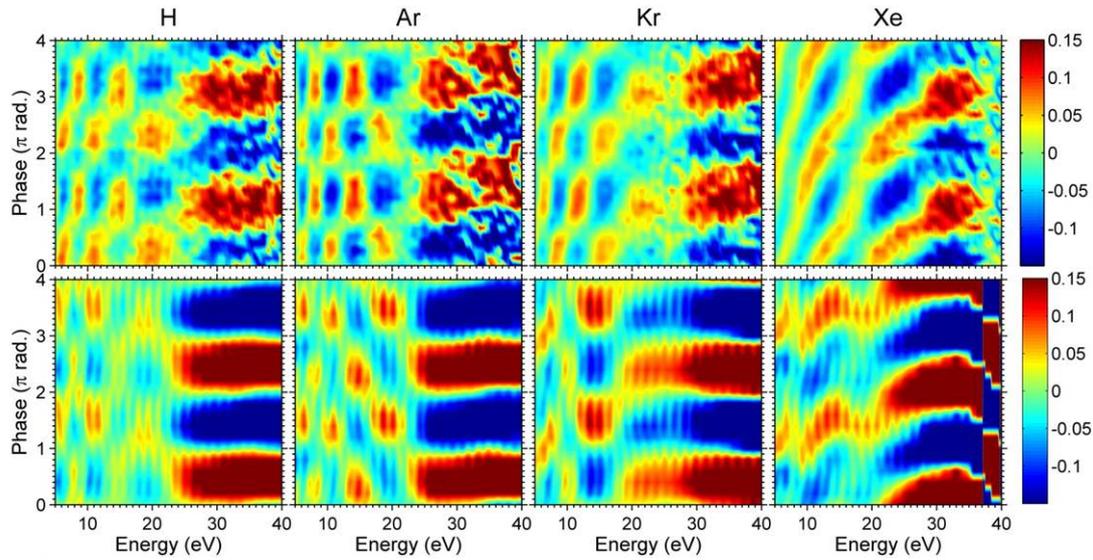
Figure 4. CEP signal maps for H and several noble gases at a laser intensity of 2×10^{14} W/cm^2. Top: experimental data. Bottom: Simulations using 3D-TDSE for H and SAE-TDSE for the noble gases.

Focusing on the strong high-energy "bar" feature and comparing theory to experiment for H, we find that the instrument offset phase ϕ_{inst} can be calibrated to within 100 mrad. Again, by using H, we can be sure that this calibration does not suffer from systematic errors. However, the data show that the "bar" features for the noble gases are significantly shifted from that for H. To make this observation quantitative, we perform a statistical analysis of the "bar" features for each atomic species. First, we coarsely bin the high-energy electron yield with respect to E to obtain a high signal-to-noise ratio. Then we fit the electron yield in each E-bin to a sinusoidal function of ϕ with a offset phase coefficient ϕ_0.

Figure 5 shows these phase coefficients as a function of electron energy for each atomic species. The species-specific phase shift can be as large as a radian and appear to be quite a bit larger than theoretical predictions. Hence, an uncritical use of the position of these "bars" to calibrate ϕ_{inst} would lead to large systematic errors. The phase shift is found to be much larger at intensity of 2×10^{14} W/cm^2 than at 1×10^{14} W/cm^2. The energy dependence of the phase shift is also substantial. Theoretical analysis is ongoing, but as usual in strong-field physics, it is difficult to give a heuristic interpretation of the data.
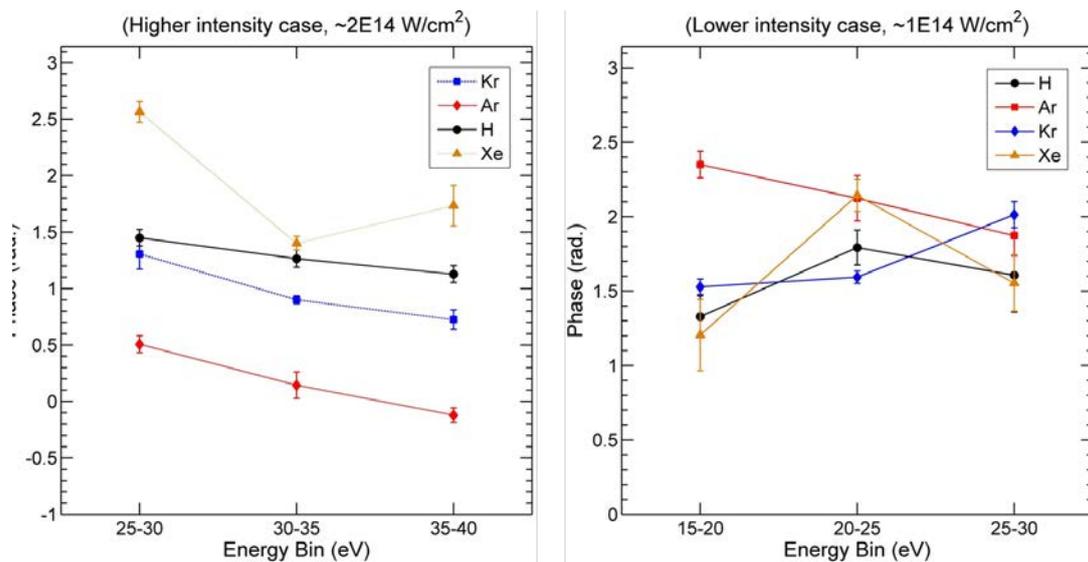
Figure 5. Species-specific phase shifts for the noble gases at energy bins within the "bar" region. Lines are guides to the eye. Note that the "bar" feature shifts to lower energies at lower intensity, as predicted by theory.

These results represent a significant step toward calibration of the currently popular stereo-ATI phase meter technique. It is now possible for other laboratories to reproduce our measurements and therefore obtain phase information that is referenced to H. However, the current data were obtained at substantially lower electron energy than is usual for stereo-ATI phase metering. Our apparatus was designed to yield data of unquestionable accuracy. To achieve this end, we were forced to compromise on the density of the atomic beam [P1]. The yield of high-energy electrons in our apparatus is correspondingly low, too low for adequate statistics in the region typical of stereo-ATI. Another cross-calibration step, comparing low-energy and high-energy CEP effects for a noble gas target, is necessary to complete the calibration of stereo-ATI phase metering. This additional step would require substantial alterations in the atomic beam apparatus.

Control of few-cycle pulse shape.

In addition to the calibration and benchmarking goals of this program, we also aimed to investigate the control of attosecond dynamics by shaping the laser electric field waveform. This part of the work was performed concurrently with other experiments.

We were able to demonstrate extremely flexible waveform control of few-cycle pulses at pulse energies suitable for attosecond science (>40 µJ). Such control had previously only been reported in the literature for nJ pulse energies. These experiments were performed using a customized spectral pulse shaper (MIIPS 640, Biophotonic Solutions). The shaper used liquid crystal (LCD) arrays to provide both amplitude and phase control over 640 spectral pixels. The relatively low damage threshold of the LCDs presented the principal obstacle to the use of high-energy pulses. It was essential to ensure that the bandwidth of the few-cycle pulses remained high at all times, so that the pulse energy was always properly spread out across all of the spectral pixels.

We demonstrated the generation of pairs of pulses with tunable time delay by spectral shaping. The resulting waveforms were independently characterized using a few-cycle autocorrelator as a cross-check of the generation process. A typical autocorrelation signal is shown in Fig. 6. With appropriate compensation of the spectral phase, we were able to maintain the duration of each pulse at 6 fs, near the transform limit. The time delay could be set in fs steps up to a maximum of ≈50 fs. A significant level of undesired background intensity was observed between the pulses. However, since the waveform is meant to drive highly nonlinear processes, this relatively weak background is unlikely to compromise future experiments. At delays over ≈50 fs, the pulse contrast degraded rapidly relative to background.
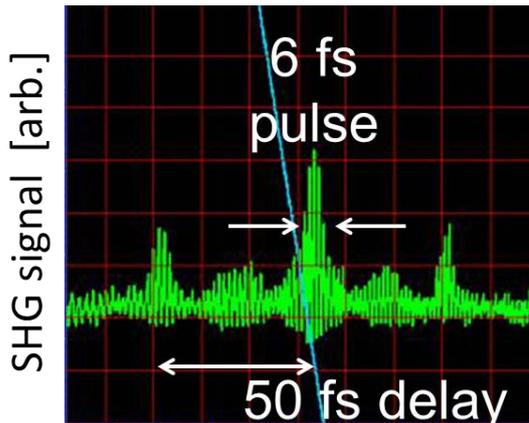
Figure 6. Autocorrelation trace of a high-energy few-cycle pulse pair, generated using spectral shaping. The pulse intensities are equal, resulting in the typical three-peaked autocorrelation trace with a 2:1 peak height ratio. An undesirable, but tolerable, level of background intensity is visible at intermediate time delay.

Fourier transform considerations show that control over finer and finer spectral features is required as one increases the total temporal control range. We therefore believe that the limit to time delay arises from the pixelization of the ideal spectral amplitude/phase pattern. However, it should be possible to control the waveform almost arbitrarily within the observed timespan limit of 50 fs.

List of Publications and Significant Collaborations that resulted from your AOARD supported project:

a) papers published in peer-reviewed journals

[P1] Benchmarking strong-field ionisation with atomic hydrogen. D Kielpinski, RT Sang, and IV Litvinyuk. J Phys B 47, 204003 (2014). Invited review article.

[P2] Carrier-envelope phase effects in above-threshold ionization of atomic hydrogen. WC Wallace, MG Pullen, O Ghafur, H Xu, DE Laban, AJ Palmer, GF Hanne, K Bartschat, AN Grum-Grzhimailo, HM Quiney, IV Litvinyuk, RT Sang, and D Kielpinski. New J Phys 15, 033002 (2013).

[P3] Measurement of laser intensities approaching 10^{15} W/cm^2 with an accuracy of 1%. MG Pullen, WC Wallace, DE Laban, AJ Palmer, GF Hanne, AN Grum-Grzhimailo, K Bartschat, D Wells, HM Quiney, XM Tong, IV Litvinyuk, RT Sang, and D Kielpinski. Phys Rev A 87, 053411 (2013). Featured as a "Research Highlight," Nature Photonics 7, 585 (2013).

b) papers published in non-peer-reviewed journals or in conference proceedings

[C1] Percent-level accuracy in strong-field measurements of photoionisation and laser intensity. WC Wallace, O Ghafur, JE Calvert, C Khurmi, DE Laban, MG Pullen, AJ Palmer, K Bartschat, AN Grum-Grzhimailo, D Wells, HM Quiney, XM Tong, IV Litvinyuk, RT Sang, and D Kielpinski. Australian Institute of Physics Congress 2014, Canberra, Australia, 7 – 11 Dec 2014

[C2] Accurate measurement of photoionization yield of H-atom using intense few cycle laser pulses. C Khurmi, WC Wallace, O Ghafur, DE Laban, IV Litvinyuk, RT Sang and D Kielpinski. Australian and New Zealand Conference on Optics and Photonics 2013, Fremantle, WA, Australia, 8 – 11 Dec 2013

[C3] Photoionization yield of atomic hydrogen using intense few-cycle pulses. O Ghafur, WC Wallace, JE Calvert, DE Laban, MG Pullen, AN Grum-Grzhimailo, K Bartschat, IV Litvinyuk, RT Sang, and D Kielpinski. 28th International Conference on Photonic, Electronic and Atomic Collisions, Lanzhou, China, 24 – 30 Jul 2013; J Phys: Conf Ser 488, 032045 (2014).

[C4] Photoionization yield of atomic hydrogen using intense few-cycle pulses. O Ghafur, WC Wallace, J Calvert, DE Laban, MG Pullen, AN Grum-Grzhimailo, K Bartschat, IV Litvinyuk, RT Sang, and D Kielpinski. Conference on Lasers and Electro-Optics – Pacific Rim, Kyoto, Japan, 30 Jun – 4 Jul 2013; DOI: 10.1109/CLEOPR.2013.6600024

[C5] Above-threshold ionization in atomic hydrogen using intense, few-cycle laser pulses. WC Wallace, MG Pullen, DE Laban, AJ Palmer, GF Hanne, AN Grum-Grzhimailo, B Abeln, K Bartschat, D Weflen, I Ivanov, A Kheifets, HM Quiney, IV Litvinyuk, RT Sang, and D Kielpinski 27th International Conference on Photonic, Electronic and Atomic Collisions, Belfast, UK, 27 Jul – 2 Aug 2012; J Phys: Conf Ser 388, 032055 (2012).

c) conference presentations

[C6] Precision attosecond physics with atomic hydrogen. D Kielpinski. International Workshop for Attosecond Science, Beijing, China, 27 – 28 Jun 2013.

[C7] Carrier-envelope phase effects in few-cycle ionisation of atomic hydrogen. D Kielpinski, WC Wallace, MG Pullen, DE Laban, AJ Palmer, GF Hanne, AN Grum-Grzhimailo, B Abeln, K Bartschat, D Weflen, I Ivanov, A Kheifets, HM Quiney, IV Litvinyuk, and RT Sang. 23rd International Conference on Atomic Physics, Palaiseau, France, 23 – 27 Jul 2012.

[C8] Carrier-envelope phase effects in few-cycle ionisation of atomic hydrogen. D Kielpinski, WC Wallace, MG Pullen, DE Laban, AJ Palmer, GF Hanne, AN Grum-Grzhimailo, B Abeln, K Bartschat, D Weflen, I Ivanov, A Kheifets, HM Quiney, IV Litvinyuk, and RT Sang. 43rd Annual Meeting of the American Physical Society Division of AMO Physics, Orange County, CA, 4 – 8 Jun 2012.

d) manuscripts submitted but not yet published

[S1] Percent-level accuracy in measuring strong-field photoionization and laser intensity. W.C. Wallace, O. Ghafur, S. Undurti, J.E. Calvert, C. Khurmi, D.E. Laban, M.G. Pullen, K. Bartschat, A.N. Grum-Grzhimailo, D. Wells, H.M. Quiney, X.M. Tong, I.V. Litvinyuk, R.T. Sang, and D.

Kielpinski. Submitted to Phys Rev Lett. Reviewers' questions about consistency of data currently being resolved (see above).

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e) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

None

Additional references.

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