Carrier-envelope phase effects in above-threshold ionization of atomic hydrogen

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Abstract. Recent experiments in ultrafast physics have established the importance of above-threshold ionization (ATI) experiments in measuring and controlling the carrier-envelope phase (CEP) of few-cycle laser pulses. We have performed an investigation of atomic hydrogen subjected to intense CEP-stable few-cycle laser pulses. The experimental ATI spectra have been compared to predictions from an \textit{ab initio} numerical solution of the time-dependent Schrödinger equation in three dimensions. Good agreement between experiment

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and theory has been achieved without using any free fit parameters. Our results provide an important step towards obtaining calibrated reference data for a direct comparison of ATI electron yields for a range of gas species and experimental conditions.

Advancements in laser technologies have led to the generation of intense laser pulses with durations approaching the period of a single electric field oscillation [1]. The temporal evolution of the electric field of these pulses can be expressed as \( E(t) = E_0(t) \cos(\omega_0 t + \phi) \), where \( E_0(t) \), \( \omega_0 \) and \( \phi \) denote the pulse envelope, carrier frequency and carrier-envelope phase (CEP) respectively. The importance of the role that \( \phi \) plays in high-field physics is evidenced by its ability to alter above-threshold ionization (ATI) spectra [2], high-harmonic generation spectra [3], electron emission [4], and electron localization [5]; and to generate isolated attosecond pulses [6].

There has been great interest in the role of the CEP in ATI experiments, with a recent breakthrough being the ability to ‘phase-tag’ individual laser shots [7]. A number of theoretical models have been used to simulate the ATI process with varying degrees of success; strong-field approximation [8], single-active electron approximation (SAE) [7] and quantitative rescattering theory [9]. These models all incorporate some form of approximation that call into question the accuracy of the simulation. In contrast, numerical calculations based on the three-dimensional (3D) time-dependent Schrödinger equation (TDSE) avoid ambiguities owing to these approximations and hence are capable (subject to computational limitations) of yielding a highly accurate representation of the interactions under investigation. Admittedly, 3D TDSE calculations are computationally expensive for all but the simplest atomic species—atomic hydrogen (H), an element that has traditionally served as the primary test case for atomic physics. In this paper we present CEP-dependent experimental data from the interaction of atomic H with an intense few-cycle pulse and perform a fit-free comparison to 3D TDSE calculations. The combination of the undisputed accuracy of the 3D TDSE and our results serves as a key preliminary comparison between experiment and theory. The data sets a benchmark for experimental accuracy and provides the potential for absolute CEP calibration measurements of other atomic species to be made.

The experimental apparatus and data acquisition method have been detailed elsewhere [10] and hence are only described briefly here. The apparatus consists of an atomic H beam and an intense few-cycle laser beam intersecting within an electrostatic lens assembly. Photoelectrons ejected via the ATI process are detected by dual channel electron multiplier detectors located above and below the interaction region, coincident with the laser polarization vector. Each detector is operated as a retarding analyser via the application of a deflector voltage, \( V_D \), to the lens assembly. Only photoelectrons with a kinetic energy above some repeller energy \( R_E \) are detected.

The laser system is a commercial Femtolaser ‘Femtopower Compact Pro’, which outputs \( 5.5 \pm 0.3 \) fs, \( 150 \mu \text{J} \) energy pulses at a 1 kHz repetition rate with 800 nm central wavelength. Stabilization of the CEP to \(< 200 \) mrad rms phase noise is achieved using the \( f–2f \) interferometric technique [11] at the laser output. The laser traverses an air path of about 10 m from the laser output before being focused into the interaction region via a 750 mm focal length off-axis parabolic (OAP) mirror. The \( 1/e^2 \) radius, which is the width at which the intensity falls to 13.5% of the maximum value, of the focused beam is 47 \( \mu \text{m} \). Variation of the laser peak
intensity is achieved by the insertion of 2 μm thin pellicle beamsplitters, which impart negligible temporal broadening to the pulse. The error in the measured peak laser intensity is typically on the order of 10% [10]. Fine control of the CEP is achieved by insertion of a pair of fused silica wedges located in the beam path.

The atomic H beam is created via collisional dissociation of molecular H2 in a radio frequency discharge powered by a helical resonator. The emergent beam is 80 ± 15% H atoms by number, with H2 comprising the remainder. At the interaction region, the H beam is uniform in density with a diameter of 0.5 mm and < 2 × 10−3 mrad divergence.

The photoelectron yield of atomic H at both detectors is obtained by taking the integrated photoelectron spectra as a function of the retarding voltage V_D (10–80 V), the laser CEP (0–2π) and the laser intensity (3.0–6.4 × 1014 W cm−2). Acquisition time for a data set at a single intensity is on the order of 45 min. Over this timeframe, CEP drifts can occur due to the 10 m air path to the interaction region. We employ a procedure which averages the CEP drifts over the entire data set, thereby avoiding systematic shifts in the data during the integration time. For a given intensity, an integrated electron spectrum (energy scan) is obtained by varying the deflector voltage V_D at a fixed laser CEP. The laser CEP is then set to a new value and the energy scan retaken, until data is obtained for CEP values spanning 2π. Three separate measurements are required to isolate the photoelectron yield from both the background and the residual H2 in the beamline [10]. This method is then repeated for each peak laser intensity. Technical difficulties precluded a common f−2f CEP lock across all intensities.

Utilizing a third f−2f interferometer placed just prior to the OAP, we observe both fast and slow fluctuations in the phase noise. Slow drifts contributions occur predominately from thermal gradients and air currents within the laboratory owing to the relatively long air path and acquisition time, whereas fast ‘jumps’ occur due to acoustic noise coupling into the optical components near the interaction region. Our independent measurement of these contributions yielded an ∼860 mrad rms CEP noise over a 10 min duration.

Our experimental data are compared to theoretical predictions of the photoelectron yield from the interaction of atomic H with a few-cycle pulse. The calculations were performed by direct integration of the 3D TDSE using the velocity-gauge form of the Hamiltonian [12], before any experimental data was taken. A sin⁴ pulse shape with 6.3 fs pulse duration at full-width half-maximum of the intensity envelope was used for the simulation. Figure 1 shows a comparison of CEP averaged integrated photoelectron yields of atomic hydrogen against the theoretical simulations as a function of R_E for several laser peak intensities. The uncertainty in each experimental data point is computed from the uncertainties in the measured dissociation efficiency, shot noise, and Allan deviation of the photoelectron yield. The experimental data have been fit to the theoretical simulations using the same method as in [10]. Due to the computationally expensive nature of the 3D TDSE calculations it was not feasible to obtain suitable theoretical predictions for 5.5 fs pulses. It is well known that CEP effects depend strongly on the pulse duration, but the difference between 5.5 and 6.3 fs pulses should not be resolvable in our experiment [13].

The raw theoretical predictions give the photoelectron yield as a differential rate (I_R) of the ionization probability (P) with respect to both the electron energy (E) and the angle of emission from the laser polarization direction (θ). The ionization probability is itself a function of E and θ; and of the laser peak intensity (I_0) and laser CEP (φ),

\[ I_R = \frac{dP(E, \theta, I_0, \phi)}{d\theta dE}. \] (1)
Electron repeller energy $R_E$ (eV) and laser peak intensity (W cm$^{-2}$). The experimental data (points) have been CEP averaged and fit to the theoretical TDSE predictions (dashed lines). Error bars are typically smaller than the data points.

Ionization rates are calculated for 181 values of $\theta$ from 0 to $180^\circ$, 14 values of $I_0$ between 0.5 and $8.0 \times 10^{14}$ W cm$^{-2}$, and 25 values of $\phi$ for $0 \leq \phi \leq 2\pi$. To simulate the experimental signal, a post-processing routine is applied to the theoretical ionization rates [14]. This routine integrates the theoretical ionization rates over the detector function, the laser intensity distribution and the atomic H density distribution in the interaction region. The detector function is derived from SIMION simulations of the photoelectron trajectories, taking into account the deflector voltage $V_D$ and the $0^\circ \leq \theta \leq 7^\circ$ detector acceptance angle. The SIMION simulations showed that $R_E$ had an approximately linear dependence on $V_D$, given by $R_E \approx 0.8(V_D - 5)$. Focal-volume averaging is performed by modeling the laser beam profile as a Gaussian of the form $I(r, z) \approx I_0 \exp\left(-2r^2/w(z)^2\right)$, where $r$ is the transverse direction and $w(z)$ is the waist along the axial direction of the laser. The interaction region is approximated by a cylinder of length 0.5 mm and radius 47 $\mu$m, representing the width of the atomic H beam and the laser beam waist, respectively. The resultant integrated electron yield $S_T$ after post-processing of $I_R$ for the top detector as a function of CEP is given as

$$S_T(V_D, I_0, \phi) \propto \int_0^\infty P(V_D, I_0 e^{-2r^2/w(z)^2}, \phi) r \, dr.$$  

A similar expression for the yield $S_B$ of the bottom detector is found by repeating the post-processing routine for $\theta \approx 173–180^\circ$. The validity of this post-processing routine has been demonstrated in its accuracy [10]. It is well known that variation of the CEP induces an asymmetry between the number of electrons detected at the top and bottom detectors. This asymmetry between detectors is quantified by the asymmetry parameter

$$\alpha(\phi) \equiv \frac{S_T(\phi) - S_B(\phi)}{S_T(\phi) + S_B(\phi)}.$$  

Figure 1. Integrated photoelectron yield of atomic H as a function of repeller energy $R_E$ (eV) and laser peak intensity (W cm$^{-2}$). The experimental data (points) have been CEP averaged and fit to the theoretical TDSE predictions (dashed lines). Error bars are typically smaller than the data points.

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The periodicity of the asymmetry observed in our data is well modelled by a sinusoid in all cases. A sinusoidal least-squares fit to $\alpha$ is performed using only two fit parameters,

$$\alpha(\phi) = A \sin(\phi - \phi_0).$$

Here $A$ is the modulation depth coefficient that quantifies the maximum possible asymmetry, while $\phi_0$ is the phase offset that quantifies the position of the maximum possible asymmetry relative to the CEP value $\phi$. Figure 2 shows an example of experimental asymmetry data obtained at a laser intensity of $5.6 \times 10^{14}$ W cm$^{-2}$ and repeller energy of 52 eV.

Theory–experiment comparison is facilitated by calculating the modulation depths and phase offsets for both the experimental and theoretical results. However, further modification of the theory predictions is required in order to account for the added CEP fluctuations incurred between the laser output and the interaction region, as any form of phase noise serves to both decrease the modulation depth and increase the error in the phase offset. Our data acquisition method has been chosen to be resistant to slow drifts in the phase and is the least sensitive to systematic errors arising from fast ‘jumps’ in the CEP. By taking an energy scan at a fixed CEP, any fast phase fluctuations manifest in the results as an increase in the error bars of both the modulation depth and phase offset without affecting the accuracy of the data points themselves. In contrast, if we were to perform a CEP scan at a fixed $V_D$, fast CEP fluctuations would result in a systematic and more pronounced decrease in the modulation depth as well as a random shift in the value of the phase offset between adjacent data points.

We can determine the reduction in theoretical modulation depth without losing accuracy by using our independent measurements of the phase noise. The required modification of the theory predictions is simply a universal constant rescaling factor applied to the modulation depth. This scaling factor can be calculated by assuming a sinusoidal form of the data, as shown in equation (4), and applying a fluctuating phase noise. The phase noise $P(\phi')$ is approximated
by a Gaussian of the form

\[ P(\phi') = e^{-\phi'^2/(2\sigma_0^2)}, \]

(5)

where \( \sigma_0 \) is the rms phase noise. A histogram of the measured rms phase noise was plotted and found to obey a normal distribution, hence justifying the use of a Gaussian as an approximation to the noise function. The universal scaling factor \( \eta \) is then calculated by taking the integral over the product of the phase noise and a sine curve,

\[ \eta = \int_{-\infty}^{\infty} P(\phi')\sin(\phi + \phi') \, d\phi' = e^{-\sigma_0^2/2}. \]

(6)

Substituting our independently measured phase noise of \( \sim 860 \) mrad into equation (6) yields a scaling factor of 0.69, which was subsequently applied to the theoretical modulation depth.

Good agreement between experimental modulation depths and theory is illustrated by figure 3. The modulation depths were obtained by applying equation (3) to both the experimental data and the theoretical predictions, then subsequently performing the fit in equation (4). Errors in the data points are propagated through from uncertainties in the yield measurements. We note that the experimental data lie systematically below the theoretical predictions. This is predicated by a combination of the error in the measured laser intensity; as well as an underestimation of the rms phase noise, owing to the difference in measurement times of the independently measured phase noise (\( \sim 10 \) min) versus that of a typical data set (\( \sim 45 \) min).

As expected from the literature, a negligible modulation depth is observed at the lowest repeller energies for photoelectrons resulting from direct ionization. However, as the repeller energy approaches the \( 2U_P \) cutoff (where \( U_P \) is the ponderomotive energy), significant asymmetries can occur even for direct ionization if the laser pulse is sufficiently short [15]. We typically obtain data approaching \( \sim 1.8U_P \). At energies \( \sim 2U_P \), both directly ionized and re-scattered electrons contribute to measured asymmetries, and for energies \( \gg 2U_P \) electron rescattering dominates, with asymmetries orders of magnitude larger than from directly ionized electrons. In this experiment, we observe the onset of CEP effects at laser intensities and electron energies consistent with this theoretical picture.

We emphasize that figure 3 displays a fit-free comparison between experiment and theory, using a completely independent measurement of the phase noise to rescale the theoretical modulation depths. This good agreement was also achieved without the need to adjust the pulse duration in the theoretical simulations. Statistically speaking, comparison without free parameters is a stricter test than any algorithm used to fit experimental modulation depths to theory and, indeed, is more stringent than any test model that uses potentials, such as most SAE, or other approximations.

Figure 4 once again exhibits good agreement between experimentally measured phase offsets and their corresponding theoretical predictions. The phase offsets were obtained in identical fashion as the modulation depths in figure 3. Due to the nature of the \( f-2f \) locking technique, the absolute laser CEP is initially unknown. To enable comparison, the experimental data must be shifted by an arbitrary but constant offset in order to overlap with the theory. This is done by calculating the weighted average of the experimentally measured phase offsets (where the weighting is given by the inverse of the error bars) at a fixed intensity. The experimental data is then shifted such that its weighted average matches the theoretical predictions. For viewing clarity, an arbitrary phase shift has been introduced to separate each pair of theory and experiment curves. Data points at the lowest repeller energies have been excluded as the modulation depths were very small, thus resulting in large errors in the phase offset.

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Figure 3. The experimental data (points) and phase-noise modified theoretical predictions (dashed lines) for the modulation depth plotted as a function of repeller energy $R_E$ (eV) for intensities of 3.0, 4.6 and 6.4 (a); and 4.2 and 5.6 (b) in units of $10^{14}$ W cm$^{-2}$. Data typically approaches energies of $\sim 1.8U_p$. For clarity, the data have been split across two graphs.

At our level of precision, there is no statistically significant variation between individual data points within any one intensity data set, supporting the claim that our data acquisition method is resistant to fast phase fluctuations. This compares well with the theoretical predictions for the phase offset which are relatively flat over comparable energy ranges. We note that we observe an intensity-dependent phase shift in the theoretical predictions which was not observed experimentally due to the lack of common \( f-2f \) lock across data sets. Further work is required in order to increase the precision in the measurement of the phase offset in order to observe both the energy and intensity-dependent phase shifts. Regardless, by employing our data acquisition method, good agreement within the data set for each individual intensity has again been achieved.

The experimental data presented here have important implications to the future of stereo-ATI as an experimental means of both measuring and stabilizing the CEP of a few-cycle laser.
Figure 4. The experimental data (points) and theoretical predictions (dashed lines) for the phase offset plotted as a function of both electron repeller energy $R_E$ (eV) for intensities of 3.0, 4.6 and 6.4 (a); and 4.2 and 5.6 (b) in units of $10^{14}$ W cm$^{-2}$. The relative CEP between different intensities was not determined. For clarity, each theory and experiment pair are separated by an arbitrary phase.

pulse. By comparing ATI data of atomic H with other gases, one can accurately calibrate the phase offsets obtained in these gas species to the true value of the laser CEP. A set of reference values constructed for different gas species across a range of laser intensities and electron energies can then serve as a comparison tool between completely separate ATI apparatus. Precise stereo-ATI of atomic H will serve as the ultimate test case for both precise and accurate measurement of the laser CEP, as H is the only species for which highly accurate calculations can be carried out.

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