Polarisation response of delay dependent absorption modulation in strong field dressed helium atoms probed near threshold

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Abstract
We present the first measurement of the vectorial response of strongly dressed helium atoms probed by an attosecond pulse train (APT) polarised either parallel or perpendicular to the dressing field polarization. The transient absorption is probed as a function of delay between the APT and the linearly polarised 800 nm field of peak intensity $1.3 \times 10^{14}$ W cm$^{-2}$. The APT spans the photon energy range 16–42 eV, covering the first ionisation energy of helium (24.59 eV). With parallel polarised dressing and probing fields, we observe modulations with periods of one half and one quarter of the dressing field period. When the polarisation of the dressing field is altered from parallel to perpendicular with respect to the APT polarisation we observe a large suppression in the modulation depth of the above ionisation threshold absorption. In addition to this we present the intensity dependence of the harmonic modulation depth as a function of delay between the dressing and probe fields, with dressing field peak intensities ranging from $2 \times 10^{12}$ to $2 \times 10^{14}$ W cm$^{-2}$. We compare our experimental results with a full-dimensional solution of the single-atom time-dependent (TD) Schrödinger equation obtained using the recently developed \textit{abinitio} TD B-spline ADC method and find good qualitative agreement for the above threshold harmonics.

Introduction
Dressing an atom with a laser field changes its response to light in diverse ways. Weak resonant fields drive transitions between the field-free atomic states, enabling resonant atomic coherent phenomena such as electromagnetically induced transparency \cite{1, 2}, Autler–Townes splitting \cite{3, 4} and related phenomena such as slow light \cite{5} to be observed. Stronger non-resonant dressing fields can modify the optical response without inducing population transfer. By recording the absorption spectrum of an extreme ultraviolet (XUV) attosecond pulse or pulse train in the dressed medium, the response can be studied on timescales well within the period of the dressing field. This technique of attosecond transient absorption spectroscopy (ATAS) \cite{6–9} has recently been used to study phenomena ranging from electronic coherence in atoms, to ultrafast switching in dielectrics and the lifetime of autoionising states. In general this method holds great promise for monitoring the dynamics of the response of a laser dressed system on a sub- or few-femtosecond timescale in both gas and condensed phase media.

Despite the simplicity of its structure helium exhibits rich dynamics and the laser dressed response has received much attention. The double excitation around 60 eV serves as a model system for observing and manipulating a two-electron wavepacket \cite{10}. Even single excitations around the first ionisation threshold present a complex array of effects due to the various bound state resonances. The XUV pulses used in ATAS, produced by high-order harmonic generation (HHG), are typically too weak to induce multi-photon effects by themselves. Therefore, the diverse phenomena reported thus far can be broadly categorised by the order of the interaction induced by the dressing field. At the lowest intensity, new two photon XUV+IR transitions (to single-photon dipole forbidden states) have been observed at intensities as low as $0.5 \times 10^{11}$ W cm$^{-2}$ \cite{11–13}.
At slightly higher dressing field intensities three photon XUV + 2IR transitions occur, enhanced by bound-state resonances, and can undergo ‘which-way’ quantum interference with single photon XUV transitions [13–15] or optical interference with incident light [16, 17]. Additionally the sub-cycle AC Stark shift modulates the frequency of excited states [18] and reduces their lifetime. Both of these effects have been detected at \( \approx 3 \times 10^{13} \) W cm\(^{-2}\). More recently, processes involving four IR photons have been observed [13, 19, 22]. Earlier work [20, 21] has addressed the polarisation dependence of the field dressing in the perturbative regime, but here we investigate the polarisation dependence of the dressing field on the atom in the strong field regime.

There have been few experimental studies of intensities above \( 5 \times 10^{12} \) W cm\(^{-2}\). Due to the potential for higher order processes, one might expect the physics to be even richer. One possibility is for XUV initiated HHG (XiHHG), in which a continuum electron is ionised by the XUV pulse, and undergoes acceleration in the laser field before recombining with the ion [23, 24]. It has also been shown that HHG from an excited atomic state may lead to efficient control over the rescattering efficiency and harmonic polarisation [25]. XiHHG has been reported at \( 5 \times 10^{13} \) W cm\(^{-2}\) [26], and has also been proposed as a probe of core dynamics such as Auger decay [27]. At these intensities, resonant enhancements [22] are expected to be weakened by appreciable broadening of the bound states [28]. At sufficiently high intensities, a trajectory-based view is likely to become applicable, in which the excited state dynamics are dominated by acceleration in the laser field in a similar fashion to the widely used strong-field approximation [29–31] for conventional HHG.

In this article we explore the response of helium atoms probed by transient absorption around the ionisation threshold in this high intensity regime (up to intensities of \( 2 \times 10^{14} \) W cm\(^{-2}\)). We investigate the polarisation sensitivity of the response by measuring the delay dependent absorption modulation for dressing fields parallel and perpendicular to that of the probe field. By using an attosecond pulse train (APT), our measurements are sensitive to the components of the response with the same half-cycle periodicity as the driving laser [22].

Alongside the vectorial dependence we also present the intensity response of the delay dependent modulation depth amplitude over a broad intensity range extending from \( 2 \times 10^{13} \) W cm\(^{-2}\), in the perturbative regime, to \( 2 \times 10^{14} \) W cm\(^{-2}\), just below the onset of strong-field ionisation. Our experimental results are compared with a full-dimensional (3D) time-dependent (TD) Schrödinger equation calculation performed using the recently developed \textit{ab initio} TD B-spline ADC method [32].

**Experimental methods**

Figure 1 shows a schematic of the experimental setup used to measure the transient absorption. The input pulses, of central wavelength 800 nm, full-width at half maximum duration (FWHM) 30 fs and energy \( \approx 3 \) mJ, were supplied by a 1 kHz, Ti:Sapphire CPA laser (KMLabs Red Dragon). An annular mirror (AM1) split the incoming beam. The transmitted portion was used to produce the XUV APT using HHG in an effusive gas jet (GJ), with a 100 \( \mu \)m diameter nozzle backed by 1.5 bar krypton. The residual IR was blocked by an aluminium filter (AF). The beam reflected from (AM1) was used to dress the helium atoms. The intensity was controlled using a half-wave plate (HWP1) and polariser (P), and the polarisation was switched between vertical and horizontal by a further half-wave plate (HWP2). An insertable beam block (BB) was used to block the dressing field beam for reference (field-free absorption) spectra. The time delay between the XUV and dressing arms was controlled with either a delay translation stage (TS) or piezo-driven mirror (PM). An auxiliary interferometer (AI) provided high-resolution tagging for the delay between the two arms. The arms were recombined using a further annular mirror (AM2) and refocused with a toroidal mirror (TM) into a 2.6 mm diameter helium-filled tube target (T). The transmitted XUV spectrum was dispersed by a 1200 lines mm\(^{-1}\) flat-field grating (FFG) and detected on a micro-channel plate (MCP) with phosphor coating, monitored by a CCD camera. The measured spectral range was 16–42 eV, corresponding to harmonic orders 11–27. In this range, the individual harmonics were well resolved. The signal contribution for each individual harmonic was obtained by integrating over the brightest region around each harmonic peak. The regions of integration were kept fixed when analysing the data for each figure.

The peak intensity of the dressing field in the target plane was inferred from power and beam profile measurements, onto which the beam was directed by an insertable pick-off (PO). The FWHM of the dressing beam focus was \( \approx 100 \) \( \mu \)m from focal spot imaging. The spot size of the XUV beam was \( \approx 70 \) \( \mu \)m FWHM, measured using a knife edge method. The uncertainty in the absolute intensity was \( \pm 50\% \), however, the uncertainty in the relative intensity between measurements was an order of magnitude less. The experimental pulse duration of the dressing field was estimated to be \( \approx 50 \) fs, where increasing positive delay corresponds to a later arrival of the XUV pulse. The absolute pump-probe delay was not measured. Typical backing pressures used for the helium tube target were around 50 mbar.

The temporal resolution was taken as the root-mean-square fluctuation of the delay between the XUV and dressing arms as measured in the AI. Over the duration of one exposure of the MCP camera this was \( \approx 150 \) as...
placing an upper limit on the observable modulation frequency of $\omega_1$, where $\omega_1 = 2.35 \text{ rad fs}^{-1}$ is the dressing field frequency.

To convert our delay-dependent spectra into absolute absorption cross sections, we first determined the density-length product $\eta$ of the target from its field-free absorption and its known absorption cross section $\sigma_{FF}$ [33], using the Beer–Lambert law. The determined density-length products for harmonics 17 and 19 were averaged. With the dressing field applied, the measured delay-dependent absorption was converted into a delay-dependent cross section $\sigma(t)$ via the Beer–Lambert law. This was decomposed into the field-free absorption and the additional absorption resulting from the dressing field $\sigma_D(t)$

$$\sigma(t) = \sigma_D(t) + \sigma_{FF} = \eta^{-1} \log \frac{I_{EF}}{I(t)} + \sigma_{FF}$$

where $I_{EF}$ and $I(t)$ are the measured field-free and dressed-atom harmonic intensities respectively.

**Theoretical methods**

We calculated the single-atom delay-dependent absorption using B-spline time-dependent (TD) algebraic-diagrammatic construction (ADC) method [32]. The basis set consists of spherical harmonics for the angular part and B-splines $B_i(r)$ for the radial coordinate. The single particle basis functions $\psi_{lin}$ used in this calculation are therefore expressed as

$$\psi_{lin} = \frac{1}{r} B_i(r) Y_{lm}(\theta, \phi).$$

The TD problem is solved within TD-ADC making the following ansatz for the TD many-electron wavefunction $|\Psi(t)\rangle$

$$|\Psi(t)\rangle = C_0(t)|\Psi_0(t)\rangle + \sum_n C_n(t)|\Psi_n(t)\rangle,$$

where the coefficients $C_0(t)$ and $C_n(t)$ refer to the ground-state $|\Psi_0\rangle$ and to the correlated excited states (CES) $|\Psi_n\rangle$ of the ADC theory respectively. These configuration basis states include single, double, etc. excitations with respect to the ground state of the system; the maximum number of electrons which are allowed to be excited at

**Figure 1.** (a) Schematic diagram of the experimental setup. The incident angle for zero-degree optics was minimised with additional mirrors not shown in this schematic. The parity of the arms was matched to minimise effects from drifts in beam pointing. The label definitions are as follows: annular mirror (AM), insertable beam block (BB), half-wave plate (HWP), polariser (P), piezo-driven mirror (PM), translation stage (TS), focusing optic (F), (TE1) and (TE2) form an expanding telescope, effusive krypton gas jet (GJ), aluminium filter (AF), toroidal mirror (TM), insertable pick-off (PO), differentially pumped helium tube-target (T), flat-field grating (FFG), micro-channel plate (MCP), auxiliary interferometer (AI), D-shaped mirror (DM), beam splitter (BS), spectrometer (SPEC).

(b) Example comparison of the source harmonics (blue) generated in the effusive gas jet (GJ) with no gas in the target (T) compared with the field-free absorption (red) of the source harmonics in the undressed helium target (T). The harmonic transmission of the laser dressed helium is also shown (black) for a given delay and peak intensity $1.3 \times 10^{14} \text{ W cm}^{-2}$. The above ionisation energy harmonics (17–25) are shown in the zoomed insert.
the same time, i.e. the point at which the expansion of equation (3) is truncated, defines the order $n$ of the ADC ($n$) hierarchy. In the following calculation we have used the first-order method of the ADC-hierarchy, namely ADC(1), in which only single excitations are included in the expansion of the wavefunction. This is a good approximation to the current application as in He the threshold for double excitation is above 60 eV and the photon energies of the harmonics investigated here are much lower. The presented results have been calculated making explicit use of the atomic spherical symmetry and they are in principle exact as long as double excitations do not play an important role in the dynamical process of interest. The TD Schrödinger equation (TDSE) for the unknown coefficients $C_{ni}$, $C_{ni}$ is solved via the Arnoldi–Lanczos algorithm. A complex absorbing potential (CAP) in $W(\text{cap})$, has been employed in order to eliminate wave-packet reflection effects from the grid boundaries. The form of the CAP used is this work reads as

$$W = 0 \quad \text{for} \quad r < r_{\text{CAP}}, \quad \tilde{W} = \eta (r - r_{\text{CAP}})^2 \quad \text{for} \quad r \geq r_{\text{CAP}},$$

where the absorbing radius $r_{\text{CAP}}$ defines the size of the inner region and the strength coefficient $\eta$ regulates the smoothness and steepness of the CAP profile. The CAP used in the calculations starts at a radius $r_{\text{CAP}} = 120$ a.u. and has a strength $\eta = 0.0005$. With the addition of the CAP term the form of the total TD Hamiltonian of the system reads

$$\hat{H} = \hat{H}_0 + 2E(t) - i \hat{W},$$

where $\hat{H}_0$ is the field-free Hamiltonian and $2E(t)$ is the laser–atom interaction in length form and within the dipole approximation.

This calculation has been performed using a radial box size of $R_{\text{max}} = 200$ a.u. and 360 radial B-splines. The maximum angular momentum employed was $l_{\text{max}} = 60$ and in the case of perpendicular polarisations of the XUV and IR electric fields the maximum value of the magnetic quantum number used in the calculations has been set $|m_{\text{max}}| = 2$. Convergence of the results with respect to the basis set parameters has been checked. With this choice the typical number of singly excited configurations included in the simulation is of the order of a few tens of thousands depending on the polarisation of the dressing field relative to the APT. Since the IR induced couplings are sensitive to the APT spectral amplitudes and phases of the contributing harmonics, the calculated results have a dependency on the XUV pulse shape used. For this reason the pulse shapes of the XUV and IR dressing field used in the ADC(1) calculations were generated to approximate those in the experiment. The calculation of the XUV spectrum was required to access the harmonic phases that could not be measured directly in the experiment. The XUV field was calculated numerically by simulating the dominant processes in the experimental XUV generation process. This included a 2D axisymmetric simulation of the macroscopic HHG process within the krypton GJ, and then vacuum propagation, filtering and focussing of the generated XUV field. The XUV field at the point of maximum on-axis fluence was used in the single atom calculations. A bandwidth limited $\cos^2$ envelope was used for the spectrum of the IR dressing field, with a FWHM pulse duration set to be 50 fs.

The frequency dependent absorption $S(\omega; \tau_d)$ is calculated from the expectation value of the electric dipole moment of the atom $\mathbf{\sigma}(t)$ and the incident XUV field $E_X(t)$ as

$$S(\omega; \tau_d) = - \text{Im} \left[ \tilde{E}_X^* (\omega) \langle \tilde{\mathbf{\sigma}}(\omega; \tau_d) \rangle \right],$$

where tilde denotes the Fourier transform from the time domain to XUV frequency $\omega$, $^*$ denotes complex conjugation, and $\tau_d$ is the dressing IR-XUV delay. This quantity is then Fourier transformed with respect to $\tau_d$ to even multiplets of the dressing field frequency $\omega_d$. The generalised cross-section, $\sigma (\omega; \tau_d)$ was calculated using the following equation [34]

$$\sigma (\omega; \tau_d) = \frac{4 \pi \alpha \omega S(\omega; \tau_d)}{|\tilde{E}_X|^2},$$

where $\alpha$ is the fine structure constant.

**Results and discussion**

Figure 2(a) shows the results of one delay scan at a dressing field intensity of $2.4 \times 10^{14}$ W cm$^{-2}$ (~3.8 mJ input pulses) with parallel polarised XUV and dressing fields. Clear modulation is observed for harmonics 13–21, (20–32.5 eV), spanning the ionisation potential of helium. The Fourier transform along the delay axis, shown in figure 2(b), reveals that while the modulation is predominately at twice the dressing field frequency, harmonics 17 and 19 also have significant modulation at $4\omega_d$. Since the modulation frequency is determined by the difference in the frequencies of the harmonics coupled by the dressing field [22], we infer that there is strong coupling between all adjacent harmonics. Given H11 was blocked by the AF, the entire $2\omega_d$ component of H13 must be due to coupling with H15, whereas the $2\omega_d$ components of the other harmonics are coherent.
combinations of the couplings between both adjacent harmonics. The $4\omega_1$ component implies weaker but significant coupling between $H_{17}$ and $H_{13}$ or $H_{21}$ (or both) and also between $H_{19}$ and $H_{15}$ or $H_{23}$ (or both).

Presented in figure 3 is the experimentally measured absorption cross section for the above ionisation threshold harmonics (17 and 19) as a function of delay for dressing field and APT (a) parallel and (b) perpendicularly polarised. Our experimental dressing-field intensity was estimated to be $1.3 \times 10^{14}$ W cm$^{-2}$. Changing the relative polarisation of the dressing and XUV fields from parallel ($\Theta = 0^\circ$) to perpendicular ($\Theta = 90^\circ$) causes a significant reduction in the modulation amplitude. In addition to the total dynamic cross section, we also plot the field-free cross sections (FF) [33] as dashed lines for each harmonic. The decreased modulation amplitude of the above threshold harmonics as the relative polarisation is changed from parallel to
perpendicular can be understood by considering the direction of the dipole induced by the XUV (figure 3(c)). If the two fields are aligned parallel, the XUV-induced dipole will experience maximum modulation from the IR-induced distortion to the bound potential. When the two fields are aligned perpendicularly, the XUV-induced dipole will experience minimum modulation of the bound potential resulting in a reduction in the absorption modulation amplitude.

Figure 4 again shows the comparison of the delay dependent absorption cross section modulation for different polarisation alignments however this figure also includes the below ionisation threshold harmonics (panels (e) and (f)) as well as a comparison to theory (panels (c), (d), (g) and (h)). The theoretical and the experimental data assume a similar peak intensity of the dressing field \(1.2 \times 10^{14} \text{ W cm}^{-2}\) and \(1.3 \times 10^{14} \text{ W cm}^{-2}\) respectively. The significant reduction in modulation amplitude for the above ionisation threshold harmonics H17 and H19 (a) and (b) when the polarisations of the dressing and XUV fields are varied from parallel (left column) to perpendicular (right column) is reproduced qualitatively by the theoretical model. In addition, the approximate phase difference between the modulation peaks for each pair of harmonics in the experiment and the theory is similar. (The absolute phase cannot be compared between experiment and theory.) By contrast, the experimentally measured modulation amplitude of the below threshold harmonics is not significantly altered by changing the relative polarisation alignment (panels (e) and (f)). This behaviour is partly reproduced by the model (panels (g) and (h)). The modulation amplitude for harmonic 13 is qualitatively reproduced. However, the behaviour of harmonic 15 is less well captured. The partial disagreement for the below threshold numerical results may be attributed to the use of the single electron excitation approximation. Truncating the ADC hierarchy to \(n = 1\) causes a small (~0.4–0.5 eV) discrepancy between the calculated and actual energy levels. Harmonic 15 lies just below the ionisation energy of helium, in the vicinity of many bound states. Discrepancy between these energy values may therefore significantly change the calculated absorption cross section. Other disagreements between theory and experiment are consistent with this hypothesis. The offset of the modulation relative to the field-free cross section is reproduced best by the theory for harmonics 13 and 19. These harmonic energies are situated respectively well below and well above the ionisation threshold of helium. For the 13th and 19th harmonics we would expect minimal effects from any inaccuracies in the calculation of the ionisation potential and problems capturing the diffuse Rydberg states for these harmonics. As for the difference in modulation depth between the experiment and theory, the theoretical calculation is a single atom treatment so does not capture any of the field propagation modifications other than...
the attenuation of the APT. As such it is to be expected that the visibility of the experimental data is by comparison reduced.

At certain time delays and for a small range of intensities, the absorption of harmonic 13 was negative, meaning that transmitted light was stronger than if the target were absent. This is examined in greater detail in figure 5. Note the relative phases of modulations between different intensity data sets cannot be reliably established in these measurements, but the relative phases of harmonics at each intensity is well defined. We focus on harmonics 13 and its neighbour 15 (harmonic 11 was blocked by the AF). At $2.3 \times 10^{12}$ W cm$^{-2}$ (dark grey), the absorption is negligible for harmonic 13 as it does not overlap with any weakly dressed bound states. At this dressing-field intensity harmonic 15 has a small absorption offset as it lies resonant with the 1s3p electronic state [13, 28]. Increasing the intensity to $6.8 \times 10^{12}$ W cm$^{-2}$ introduces a two-IR-photon coupling between these harmonics, causing both to modulate with a half-cycle period of the driving laser. Harmonic 13 continues to oscillate around zero indicating a continued absence of available absorption channels via either single-photon or multi-photon pathways. Harmonic 15 however acquires an increased cycle-averaged offset totalising approximately 3.7 Mb. This is attributed to an increase in the absorbance of the 1s3p state with increasing IR field intensity harmonic 15 as it lies resonant with the 1s3p electronic state [13, 28]. Increasing the intensity further ($2.0 \times 10^{13}$ W cm$^{-2}$) strengthens the coupling between the two harmonics and hence the modulation amplitude, but also opens up an absorption channel for harmonic 13 which offsets the modulation so that the absorption cross section becomes positive for all delays. This absorption channel could be attributed to the XUV + 1IR multi-photon transition to the previously dipole-forbidden 1s3s state [13].

Further insight is obtained from the dependence of the modulation amplitudes on the dressing field intensity. Figures 6(a) and (c) show the measured amplitude of the $2\omega_1$ and $4\omega_1$ components respectively for harmonics 13–19. Across the full intensity range of $2.3 \times 10^{12}$–$2.3 \times 10^{14}$ W cm$^{-2}$ we detected $2\omega_1$ modulations. We also detected $4\omega_1$ modulations above $10^{13}$ W cm$^{-2}$ in harmonics 13 and 17, with a spike of $4\omega_1$ modulation in harmonic 15 and possibly harmonic 19 around $1.5 \times 10^{13}$ W cm$^{-2}$. The results of the corresponding theoretical calculations are shown in figures 6(b) and (d). The theory, which covers up to $1.2 \times 10^{14}$ W cm$^{-2}$, reproduces qualitatively several aspects of the experiment: the double peak structure of the harmonic 15 $2\omega_1$ modulation, the minima of the harmonic 13 $2\omega_1$ modulation depth towards the highest intensity dressing fields and the general flat response of harmonic 19 $2\omega_1$ modulation depth. Considering the $4\omega_1$ response we again see many of the general shapes from the experimental data reproduced by the theory. The modulation depth of harmonic 17 rises and falls throughout this intensity range, and the sharp peak in the $4\omega_1$ response for harmonic 15 and possibly harmonic 19 is reproduced around a similar dressing field intensity by the theory. More generally, the theory also predicts the higher intensity onset of the $4\omega_1$ modulation contributions compared with the $2\omega_1$ response. In general, the theoretical amplitudes are much larger than those observed. Possible reasons for the discrepancy include the spatial variation of the dressing field intensity over the XUV beam, and the propagation in the target medium which can reduce or eliminate modulations [22].

Another perspective on these measurements comes from considering electron trajectories and XiHHG, which can be recast in a multi-photon picture. In our experiment, the cutoff of the incident APT extended above the highest observed harmonic. The harmonics emitted by recombining electrons were therefore already present.
in the incident field. In this situation, the emitted and incident light interfere producing delay-dependent absorption with modulation frequency determined by the frequency difference between the initiating and emitting harmonics. The highest order modulation observed is therefore equal to the maximum kinetic energy gained by an electron between ionisation and recombination. In our case, this was $\omega_4$. Our wavelength and typical laser intensity were such that the available kinetic energy gain using the three-step model was $U_3 \approx 2 \omega_1^2$, and our best estimates of the temporal resolution yielded a maximum resolvable modulation frequency of $\omega_8$. Likely reasons why we did not observe higher order modulation emerge through signal to noise considerations. Modulation components are detected as peaks after Fourier transforming along the delay axis. In our case, for the harmonics under study ($13-19$), the noise in the Fourier domain is caused by both detector noise and fluctuations in the XUV intensity (driven by laser fluctuations) between steps in our delay scans. Together, these obscure weak modulation components. By contrast, Gademann et al. observed XfHHG at photon energies not present in the incident APT, so their measurements were observed on a zero background. For these harmonic orders, interference between incident and generated harmonics therefore did not occur, and XUV fluctuations would not have affected the detection threshold. At similar intensities to our experiment, the IR-induced coupling they observed was equivalent to a photon energy gain of at least 6 IR photons.

The agreement between experiment and theory will be influenced by the reshaping of the pulse in the medium. At maximum IR intensity, the absorption of the harmonics was $\sim 80\%$ or more. Such a degree of absorption is accompanied by significant phase slip between the dressing field IR and the harmonics, and order dependent dispersion of the harmonics. This has been shown to be capable of modifying or even removing certain modulation components and changing absorption line shapes of bound state resonances. Physically, different dynamics are initiated and probed throughout the target length. Another source of discrepancy was the spatial intensity variation of the dressing field in the transverse plane. This is caused by the XUV focal spot size being a significant fraction of the dressing field focal spot size, non-uniformity in the input beam profile as well as uncertainty in the spatial and temporal overlap between the XUV and IR beams.

Figure 6. Dependence of absorption cross section modulation amplitudes on intensity; (a) and (c) show experimental results, (b) and (d) show theory. (a) and (b): half-cycle period component. (c) and (d): quarter-cycle period component. Representative error bars are shown. The theoretical error bars were obtained from frequency components of the Fourier transform other than those at even integer multiples of the driving field frequency. For each theory harmonic the maximum error bar is plotted. This imperfect periodicity reflects the finite envelopes of the dressing and probing fields.
intensity. Scans of this duration would take significantly longer to run, and as such we would need to ensure that the laser and other experimental parameters were sufficiently stable across this time-frame.

## Conclusion

We have presented the first measurements of the vectorial response of the transient absorption of singly excited helium dressed by a laser field with polarisation oriented either parallel or perpendicular to the polarisation of the probing APT, and intensity approaching the strong-field ionisation threshold of the ground state. We observed delay-dependent absorption with half and quarter-cycle periodicity with a strong dependence on the relative polarisations of the dressing and probing fields. Several aspects of our results were reproduced by single-atom TDSE calculations. We have studied the dependence of field induced absorption modulation on the relative polarisation of dressing and probe fields. This shows a strong modulation for the case of parallel polarisation, but the modulation is greatly suppressed in the case of perpendicular polarisations for the above threshold harmonics. The behaviour can be deduced qualitatively from a heuristic strong field picture of the laser dressed atom and the geometric dependence of the distortion as a function of angle with respect to the strong field polarisation axis. The full-dimensional calculation reproduces the main features of these results.

Little difference in the modulation amplitude was observed between the parallel and perpendicular polarisations for the below threshold harmonics. This behaviour is reproduced by the theory for harmonic 13, but is not so well captured for the 15th harmonic. We understand this to be due to small offsets in the calculated bound energy states in helium arising from the use of a truncated ADC hierarchy, i.e. $n = 1$.

The observations we have made extend our knowledge of strongly laser-dressed helium. We have observed the dependence of the XUV absorption modulations on the dressing field polarisation state and intensity, bridging the gap between the perturbative and strong-field intensity regimes. Our results are a step towards understanding the strongly driven dynamics in more complex systems.

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## References

[1] Boller K-J, Imamoglu A and Harris S E 1991 Phys. Rev. Lett. 66 2593
[2] Fleischhauer M, Imamoglu A and Marangos J P 2005 Rev. Mod. Phys. 77 633
[3] Wu M, Chen S, Gaarde M B and Schafer K J 2013 Phys. Rev. A 88 043416
[4] Pfeiffer A N and Leone S R 2012 Phys. Rev. A 85 033422
[5] Hau L V, Harris S E, Dutton Z and Behroozi C H 1999 Nature 397 594
[6] Gouliebkakis E et al 2010 Nature 466 739
[7] Wang H, Chini M, Chen S, Zhang C-H, Hu S X and Chang Z 2010 Phys. Rev. Lett. 105 143002
[8] Santra R, Yakovlev V S, Pfeiffer T and Loh Z-H 2011 Phys. Rev. A 83 033405
[9] Schultz M et al 2013 Nature 493 75
[10] Ott C et al 2014 Nature 516 374
[11] Chen S, Beck M J, Beck A R, Mashiko H, Wu M, Pfeiffer A N, Gaarde M B, Neumark D M, Leone S R and Schafer K J 2012 Phys. Rev. A 86 063408
[12] Bell M J, Beck A R, Mashiko H, Neumark D M and Leone S R 2013 J. Mod. Opt. 60 1506
[13] Chini M, Wang X, Cheng Y, Wu Y, Zhao D, Telnov D A, Chu S-I and Chang Z 2013 Sci. Rep. 3 1105
[14] Chen S, Wu M, Gaarde M B and Schafer K J 2013 Phys. Rev. A 87 033408
[15] Wang X, Chini M, Cheng Y, Wu Y, Tong X-M and Chang Z 2013 Phys. Rev. A 87 063413
[16] Chini M, Wang X, Cheng Y and Chang Z 2014 J. Phys. B: At. Mol. Opt. Phys. 47 124009
[17] Holler M, Schper F, Gallmann L and Keller U 2011 Phys. Rev. Lett. 106 123601
[18] Chini M, Zhao B, Wang H, Cheng Y, Hu S X and Chang Z 2012 Phys. Rev. Lett. 109 073601
[19] Herrmann J, Lucchini M, Chen S, Wu M, Ludwig A, Kasimi L, Schafer K J, Gallmann L, Gaarde M B and Keller U 2015 New J. Phys. 17 013007
[20] Reduzzi M et al 2015 Phys. Rev. A 92 033408
[21] Lucchini M, Herrmann J, Ludwig A, Locher R, Sabbar M, Gallmann L and Keller U 2013 New J. Phys. 15 103010
[22] Chen S, Schafer K J and Gaarde M B 2012 Opt. Lett. 37 2211
[23] Schafer K J, Gaarde M B, Heinrich A, Biegert J and Keller U 2004 Phys. Rev. Lett. 92 023003
[24] Gaarde M B, Schafer K J, Heinrich A, Biegert J and Keller U 2005 Phys. Rev. A 72 013411
[25] Averbukh Y 2004 Phys. Rev. A 69 043406
[26] Gademann G, Kelkensberg F, Siu W K, Johnsson P, Gaarde M B, Schafer K J and Vrakking M J J 2011 New J. Phys. 13 033002
[27] Leeuwenburgh J, Cooper B, Averbukh V, Marangos J and Ivanov M 2013 Phys. Rev. Lett. 111 123002
[28] Shivaram N, Timmers H, Tong X-M and Sandhu A 2012 Phys. Rev. Lett. 108 193002
[29] Corkum P B 1993 Phys. Rev. Lett. 71 1994
[30] Krause J L, Schafer K J and Kulander K C 1992 Phys. Rev. Lett. 68 3535
[31] Lewenstein M, Balcou P, Ivanov M Y, L’Huillier A and Corkum P B 1994 Phys. Rev. A 49 2117
[32] Ruberti M, Averbukh V and Decleva P 2014 J. Chem. Phys. 141 164126
[33] Baker D J, Bedo D E and Tomboulian D H 1961 Phys. Rev. 124 1471
[34] Gaarde M B, Buth C, Tate J L and Schafer K J 2011 Phys. Rev. A 83 013419
[35] Pfeiffer A N, Bell M J, Beck A R, Mashiko H, Neumark D M and Leone S R 2013 Phys. Rev. A 88 051402