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PAPER

Chirp and carrier-envelope-phase effects in the multiphoton regime: measurements and analytical modeling of strong-field ionization of sodium

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Abstract

We investigate the influence of chirp on carrier-envelope-phase (CEP)-dependent strong-field few-cycle laser-induced photoelectron spectra of sodium, well within the multiphoton regime. Our measurements and analytical model of this process reveal a simple chirp-dependence, which has the potential to be utilized as an online monitor of laser chirp. Moreover, this effect could extend single-shot measurements of the CEP using above-threshold ionization to longer, chirped pulses, and significantly lower the required pulse energies. Specifically, at a wavelength of 775 nm and an intensity of 6.5×10^{12} W/cm² the CEP- and energy-dependent left-right asymmetries of emitted electrons are measured in a time-of-flight spectrometer. In these asymmetry maps, inclined stripe-like structures emerge, where the inclination is tunable with the chirp of the pulse. We report a simple analytical model, explaining the effect as the interference of electrons with even and odd angular momenta, located at energies *in between* adjacent above-threshold ionization peaks. As we demonstrate, the analytical model is in good agreement with the measurement, as well as with solutions of the three-dimensional time-dependent Schrödinger equation. Further, the analytical model, which can easily be extended to other atoms, allows us to derive an equation, describing the relation between the slope of the inclined stripes and the chirp of the laser.

1. Introduction

The ionization of atoms by strong laser fields can cause electrons to absorb more photons than are necessary for their liberation, a phenomenon known as above-threshold ionization (ATI) [1]. This process is the prerequisite for other important effects, such as high-harmonic generation (HHG) and non-sequential double ionization [2, 3]. The dynamics underlying ATI are commonly divided into two regimes, characterized by the Keldysh parameter, $\gamma = \omega_0 \sqrt{2I_p}/I$ [4] (atomic units are used throughout, unless otherwise stated), where ω_0 is the laser angular frequency, I_p is the ionization potential of the atom, and I is the laser intensity. In the multiphoton regime, $\gamma > 1$, the laser field is relatively weak and the ionization process is well described by the electron gaining energy, $E = \hbar\omega$, from each photon absorbed until it is above the ionization threshold and retains the excess energy. In contrast, in the tunneling regime, where $\gamma \ll 1$, the atom's potential well is sufficiently deformed by the laser field to allow the electron to tunnel through the barrier.

Many strong-field effects in the tunneling regime can be explained classically, employing the 'three-step' model [5, 6]. In this model, the electron ionized by the laser field (first step) may be driven back to the parent ion by the oscillating electric field (second step), where it can recombine with the ion (HHG) or be scattered at the core (third step). Elastic scattering can result in much higher electron kinetic energies than possible without this rescattering process [6], while inelastic scattering leads to excitation or further ionization of the parent ion [7].

For few-cycle pulses, the aforementioned rescattering process is highly sensitive to the offset between the maximum of the envelope and the peak of the carrier-electric field, i.e., the carrier-envelope-phase (CEP) of the

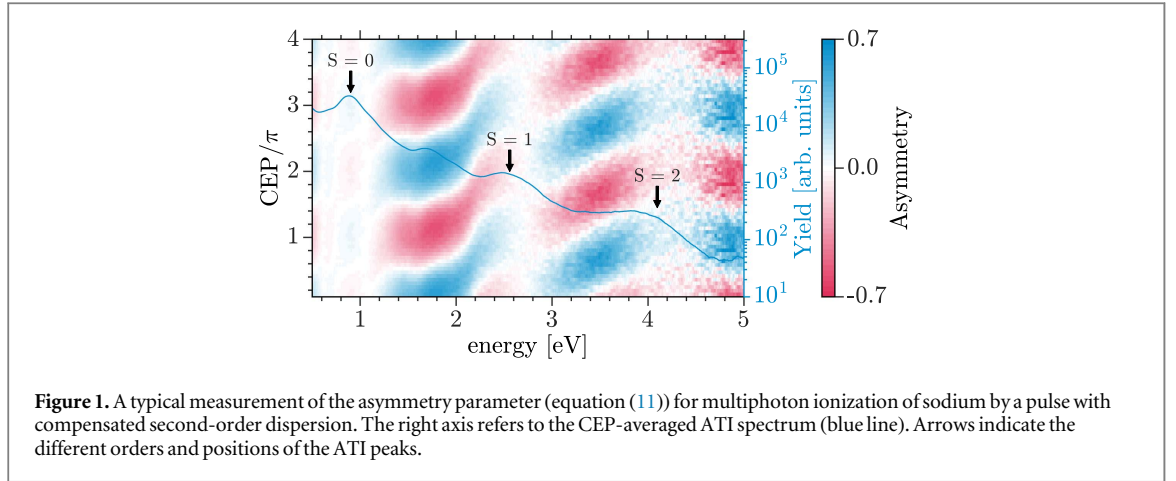
laser pulse [8], since the associated waveforms change significantly. This can be exploited to measure the CEP of single laser shots in real-time, using the so-called carrier-envelope phasemeter (CEPM) [9, 10]. This device typically operates using only rescattered electrons in the tunneling regime, i.e., at $\gamma \approx 1.1$, using $\lambda = 800$ nm, $I = 8 \times 10^{13}$ W/cm² pulses on Xe with $I_p = 12.13$ eV. The CEP effects under investigation in this paper are of a fundamentally different origin, namely interference phenomena located *in between* adjacent ATI peaks in the low-energy part of the spectrum for ATI well within the multiphoton regime. As a consequence, the CEP-dependence reported here can be observed even for relatively long pulse durations.

We move from the tunneling to the multiphoton regime by using an alkali target, which requires significantly less intensity to ionize and thus increases γ in comparison to more commonly used noble gases. Specifically, sodium atoms, with $I_p = 5.14$ eV, are used as the target gas, with laser intensities of $I \approx 6.5 \times 10^{12}$ W/cm², corresponding to $\gamma \approx 2.7$. Accordingly, the photoelectron spectra are largely free of contributions from rescattering and can be measured with significantly less pulse energy. This allows observation of chirp and CEP effects in a relatively unexplored regime.

Nevertheless, a few theoretical works exist that study the effects of chirped and unchirped pulses in multiphoton ionization, primarily by solving the time-dependent Schrödinger equation (TDSE). For caesium, asymmetries in the photoelectron angular distributions (PADs) of few-cycle chirped pulses have been calculated [11]. The PADs and ATI spectra for sodium have been shown to exhibit a strong chirp-dependence [12]. However, chirp-effects vanished, if intermediate bound states were eliminated in the calculation. Similarly, the chirp-dependence of ATI spectra for hydrogen has been predicted [13]. One interesting conclusion was that the varying instantaneous frequency of the laser can induce different resonant excitation dynamics of bound states, depending on the sign of the chirp. Specifically, the behavior of the ATI plateau [14] for different chirps has been studied in [15]. Additionally, the CEP- and chirp-dependence of the ionization of hydrogen by attosecond pulses has been theoretically explored [16]. The use of chirped attosecond pulses for measuring electron dynamics on the attosecond time scale has also been studied analytically [17].

In this paper, we present measurements of CEP- and chirp-dependent photoelectron spectra for few-cycle pulses at 775 nm. The chirp imprints an additional phase on the emitted electrons' wave function. In the corresponding ATI spectra this becomes observable as stripe-like structures in the CEP- and energy-dependent asymmetry maps. The slope of these stripes can be tuned depending on the chirp of the laser pulse. Expanding on the work of Cormier *et al* [18], we present an analytical model that predicts and explains these stripe-like structures as quantum interference of electrons from adjacent ATI peaks. To further confirm the validity of our model, the 3D TDSE is solved for sodium atoms [19]. Both theoretical models are then compared to measurements and good agreement is found between all three. Special emphasis is put on the connection between the slope of the emerging stripe-like structures and the chirp of the pulse. To our knowledge, the first observation of this kind of effect was made by Abel *et al* [20], who explained their measurements using a perturbative model based on a sum over indistinguishable quantum paths. They numerically modeled chirp by considering the N -fold autoconvolution of the laser spectrum to describe the N -photon absorption peak. This approach is similar to ours. However, they used xenon as a target gas and no systematic, chirp-dependent analysis of the stripe-like structures was performed.

The results reported here are of general interest for several reasons. Firstly, low- I_p targets, such as the alkali atoms, have rarely been studied in strong-field laser physics. In contrast to the noble gases, alkalis largely behave like one-electron systems and exhibit strong resonances, whose single photon transitions are within the bandwidth of typical few-cycle lasers. Secondly, as the intensity required for ionization of these atoms is significantly lower than for the noble gases, one can increase the wavelength while keeping a constant ponderomotive potential, $U_p = F_0^2 / (4\omega_0^2) \propto I\lambda^2$, where F_0 is the peak electric field amplitude of the laser. This counteracts the well-known drop-off in rescattered electron yield at mid-infrared wavelengths [21]. Thus, when moving to the tunneling regime, experimental techniques which rely on rescattered electrons can be advanced into a new wavelength regime, using alkali atoms. Thirdly, the results of this paper can be exploited for laser diagnostics. Namely, a real-time evaluation of asymmetry maps could be used to monitor the laser chirp online. The main advantage, as compared to other pulse characterization methods, is that the laser chirp can be evaluated in the interaction zone where the final experiment is performed. Finally, and perhaps most interestingly, the structures in CEP- and energy-dependent asymmetry maps can be used to measure the CEP of individual laser shots [8, 10]. The main advantages of the effect reported here, as compared to CEP effects in the tunneling regime, are the abilities to use lower-energy, multi-cycle, and chirped pulses.



2. Theoretical background

2.1. Analytical model

In order to model the chirp- and CEP-dependence of ATI spectra, we follow the approach of [18]. For M -photon ionization, the ATI spectrum shows peaks at energies

$$E_M = \frac{\mathbf{p}^2}{2} = M\omega_0 - U_p - |I_p|, \quad (1)$$

where $\mathbf{p}$ is the drift momentum measured at the detector. We work in the single active electron approximation. The only property of the initial ground state, which enters our model, is its ionization potential I_p . Thus, for a wavelength of $\lambda = 775$ nm and an intensity of $I = 6.5 \times 10^{12}$ W cm $^{-2}$ ($\gamma \approx 2.7$), electrons with energies around the first peak ($S = 0$) in figure 1 have absorbed four photons, whereas the $S = 1$ peak corresponds to 5-photon absorption from the ground state (3s in the present case). Since photons carry angular momentum $\ell = \pm 1$ and due to the dipole selection rules, the first ATI peak consists of electrons with even parity, i.e., their angular momenta fulfill $\ell = 0, 2, 4, \dots$. The second peak, in contrast, has odd parity, i.e., $\ell = 1, 3, 5, \dots$, and so on [18].

Due to the broad spectral bandwidth of few-cycle pulses, the ATI peaks will also be substantially broadened, as the peak width scales with the spectral bandwidth of the pulse. This leads to interference of odd and even parity electrons. The contrast of the interference will be particularly strong at photoelectron energies with equal contribution from neighboring ATI peaks, i.e., in the region *in between* the ATI peaks. Mathematically, the electronic wave function can be expanded using spherical harmonics, $Y_\ell^m(\theta, \phi)$. Their parity can be written as

$$Y_\ell^m(\pi - \theta, \pi + \phi) = (-1)^\ell Y_\ell^m(\theta, \phi). \quad (2)$$

This implies that even (odd) parity electrons are described by even (odd) functions. Since even (odd) functions have gerade (ungerade) geometry, the final wave function of even (odd) parity electrons will reflect this behavior. This, in turn, influences the left-right emission of the photoelectrons. Therefore, interference of electrons having different parity leads to observable left-right asymmetries.

To include the chirp-dependence, the model described in [18] needs to be expanded. To do this, we assume a linearly polarized, linearly chirped Gaussian laser pulse of the form $\tilde{\mathbf{F}}(t) = F(t)\mathbf{e}_z$ with

$$F(t) = \Theta W(t) e^{i(\omega_0 t + \varphi_{\text{CEP}})}, \quad (3)$$

where $W(t) := e^{-\Gamma t^2}$ is a function describing the pulse envelope, φ_{CEP} is the CEP of the laser pulse and $\Gamma = a - ib$ with

$$a = \frac{a_0}{1 + (2k'' d a_0)^2}, \quad b = \frac{2k'' d a_0^2}{1 + (2k'' d a_0)^2} \quad (4)$$

for $a_0 = 2 \ln 2 / \tau_0^2$ [22]. Here, τ_0 is the full width at half maximum (FWHM) pulse duration measured in the intensity envelope of the Fourier-transform-limited (FTL) pulse, d is the propagation length inside some dispersive medium (here, fused silica) and k'' is the group velocity dispersion of this medium. The maximal amplitude, F_0 , decreases with increasing chirp according to $\Theta := F_0 \sqrt{\tau_0 / \tau}$, where τ is the pulse duration of the chirped pulse, i.e., $\tau = \sqrt{2 \ln 2 / a}$.

For an electron starting in the ground state and absorbing M photons to reach the continuum we estimate the total probability amplitude of the M -photon absorption to behave as

$$\mathcal{S}_M \propto K_M \Theta^M. \quad (5)$$

The term K_M denotes the multiphoton coupling between the ground state and the final continuum state at kinetic energy E_M (see equation (1)). The values of K_M are adjusted to reproduce the relative height of the ATI peaks from figure 1.

The probability amplitude to find an electrons with energies off the M -photon peak can be estimated as

$$\tilde{\mathcal{S}}_M = K_M \Theta^M g(E_M) = \alpha_M(E_M) e^{iM\varphi_{\text{CEP}}}, \quad (6)$$

where $g(E_M)$ is the Fourier transform of the field $W(t)^M e^{i(E_M t + M\varphi_{\text{CEP}})}$. The result is a function depending on ω , i.e., energy in atomic units:

$$[g(E_M)](\omega) = e^{-(\omega - E_M)^2 / (4M\Gamma)} e^{iM\varphi_{\text{CEP}}} \sqrt{\pi / (M\Gamma)}. \quad (7)$$

The function $g(E_M)$ describes the shape of the M^{th} ATI peak. In addition, we introduced

$$\alpha_M(E_M) := K_M \Theta^M e^{-(\omega - E_M)^2 / (4M\Gamma)} \sqrt{\pi / (M\Gamma)}. \quad (8)$$

Performing the same consideration for the $(M + 1)^{\text{th}}$ peak yields

$$\begin{aligned} \tilde{\mathcal{S}}_{M+1}(E_{M+1}) &= K_{M+1} \Theta^{M+1} g(E_{M+1}) \\ &= \alpha_{M+1}(E_{M+1}) e^{i(M+1)\varphi_{\text{CEP}}}. \end{aligned} \quad (9)$$

Possible phase contributions resulting from the atomic potential during the ionization process are not included. Note that by restricting ourselves to $\omega \geq 0$ in the above equations, we confine our one dimensional model to a single emission direction, which we will label ‘right’. It points along the direction of the electric field, $\mathbf{e}_z$.

We are interested in interference effects in between two adjacent ATI peaks. The probability of an electron having an energy between the M^{th} and $(M + 1)^{\text{th}}$ ATI peak is calculated as a *coherent* sum, i.e.,

$$\begin{aligned} P_M(E = E_M + \Delta E^{(1)}, \varphi_{\text{CEP}}) &= |\tilde{\mathcal{S}}_M(E_M + \Delta E^{(1)}) + \tilde{\mathcal{S}}_{M+1}(E_{M+1} - \Delta E^{(2)})|^2 \\ &= |\alpha_M(E_M + \Delta E^{(1)}) e^{iM\varphi_{\text{CEP}}} \\ &\quad + \alpha_{M+1}(E_{M+1} - \Delta E^{(2)}) e^{i(M+1)\varphi_{\text{CEP}}}|^2 \\ &= |\alpha_M|^2 + |\alpha_{M+1}|^2 + 2\text{Re}(\alpha_M \alpha_{M+1} e^{-i\varphi_{\text{CEP}}}). \end{aligned} \quad (10)$$

Here, $\Delta E^{(1)}$ and $\Delta E^{(2)}$ are arbitrary energy intervals, such that $\Delta E^{(1)} + \Delta E^{(2)} = \omega$, which reflects the fact that two adjacent ATI peaks are separated by the photon energy. Note that we omit the energy-dependences in the final formulation for better readability, as they are identical to the previous line.

In section 4 we will show that the numerical evaluation of equation (10) leads to chirp-dependent, inclined stripe-like structures in the CEP asymmetry maps. One of our goals will be to quantitatively analyze the slope of these stripe-like asymmetry structures, which can be seen in figure 1 around 1.8 and 3.4 eV. For this purpose, we define the asymmetry parameter, plotted in figure 1, as

$$A(E, \varphi_{\text{CEP}}) := \frac{Y_{\text{left}}(E, \varphi_{\text{CEP}}) - Y_{\text{right}}(E, \varphi_{\text{CEP}})}{Y_{\text{left}}(E, \varphi_{\text{CEP}}) + Y_{\text{right}}(E, \varphi_{\text{CEP}})}, \quad (11)$$

where $Y(E, \varphi_{\text{CEP}})$ is the yield, recorded at the left or right detector, respectively. These types of false color plots are referred to as asymmetry maps.

As the stripes in the TDSE and experiment will show irregular structures, as compared to the perfect stripes in the analytical model (see figure 4), the determination of their slopes is not as straightforward. We discuss a fitting procedure, which was employed to quantitatively determine the slope of the structures for the TDSE and measurements.

Firstly, different orders of the ATI peaks behave differently, therefore we focus on the first asymmetry region between $S = 0$ and $S = 1$, i.e., we set the relevant energy interval to be $D := [1.0 \text{ eV}, 3.0 \text{ eV}]$. Next, a function of the form

$$f(E, \Omega) = B(E) \sin(\Omega - \Phi(E)) \quad (12)$$

is fitted to $A(E, \varphi_{\text{CEP}})$ at all energies $E \in D$ for $\Omega \in [0, 4\pi]$, yielding the two energy-dependent fitting parameters $B(E)$ and $\Phi(E)$. The final fitting region is determined by the condition $|B(E) / \max(B(E))| \geq B_0$, where B_0 was chosen to be 0.65. In this region, a linear function, weighted by the error $1/\Delta\Phi(E)$, is fitted to the phase-offset angle $\Phi(E)$, yielding the slope, s , of the asymmetry structure. Finally, intermediate slopes, s_i , are fitted between the start and end point of each energy interval in the fitting region. We define the error of s as

$$\Delta s = \sqrt{\frac{1}{N} \sum_{i=1}^N (s - s_i)^2}. \quad (13)$$

Remarkably, this fitting procedure is not necessary for the analytical model, as we will now derive an analytical expression for the slope of the inclined structures in this model. Using equation (10) we can write $P_M(E, \varphi_{\text{CEP}}) = Y_{\text{right}}(E, \varphi_{\text{CEP}})$. Here, M is uniquely determined by the condition $E_M \leq E \leq E_{M+1}$. As our model only considers one emission direction, we use the following identity to calculate the asymmetry: $Y_{\text{left}}(E, \varphi_{\text{CEP}}) = Y_{\text{right}}(E, \varphi_{\text{CEP}} + \pi)$ for a fixed CEP. This assertion, which is independent of the symmetry of the initial state, has also been confirmed by 1D TDSE calculations. Then, in equation (11), we can write the asymmetry parameter as

$$A(E, \varphi_{\text{CEP}}) = \frac{4\text{Re}(\alpha_M \alpha_{M+1})}{|\alpha_M|^2 + |\alpha_{M+1}|^2}. \quad (14)$$

We apply the definition of α_M and, for the sake of simplicity, assume $K_M = 1$ for all M . After a series of cumbersome but straightforward algebraic operations, equation (14) can be brought to the same form as equation (12), yielding

$$\Phi_M(E) = \frac{bZ(E)}{4M(M+1)(a^2 + b^2)} + \frac{\pi}{2}, \quad (15)$$

where $Z(E) = -(E + I_p + U_p)^2 + M(M+1)\omega_0^2$. The subscript M was introduced to indicate that equation (15) refers to the phase-offset angle, as defined in equation (12), between the M^{th} and $(M+1)^{\text{th}}$ ATI peak, as this angle changes depending on the peak order. Finally, the slope can be calculated by taking the derivative of equation (15) with respect to E , resulting in

$$m_M(E) = \frac{-2b(E + I_p + U_p)}{4M(M+1)(a^2 + b^2)}. \quad (16)$$

Since $\Phi_M(E)$ is a quadratic function, $m_M(E)$ is energy-dependent. However, in the region where the M^{th} and $(M+1)^{\text{th}}$ ATI peak overlap, i.e., the only region in which $\Phi_M(E)$ is of interest, the offset angle is approximately linear and the slope becomes comparably weakly energy-dependent. Specifically, the asymmetry regions in the analytical model will be between 1.4 and 2.1 eV (see figure 4). For example, for $d = -500 \mu\text{m}$ we obtain $m_4(1.4 \text{ eV}) = 15.2 \text{ rad/eV}$ and $m_4(2.1 \text{ eV}) = 16.7 \text{ rad/eV}$.

Furthermore, we have confirmed that the simplification $K_M = 1$ does not significantly alter the inclination, thus removing any experimental fitting parameters. Using the values of K_M derived from the measurement and applying the fitting procedure used for the TDSE and experimental results to the analytical model, yields a slope of $s = 20.58 \text{ rad/eV}$, for $d = -636 \mu\text{m}$, whereas equation (16) using the $K_M = 1$ simplification yields $m_4(1.8 \text{ eV}) = 20.46 \text{ rad/eV}$. Indeed, for all glass positions considered, the deviation never surpasses 1%. However, a consequence of setting $K_M = 1$ is a shift in the energy region, at which the highest asymmetry appears. In the example above, it shifts from 1.8 eV, using the experimentally derived values, to 2.1 eV, using $K_M = 1$.

2.2. Time-dependent Schrödinger equation

For comparison of the experimental results with a more complete theoretical model, we solved the three-dimensional TDSE, using the freely available code Qprop, to obtain CEP-dependent photoelectron spectra in the emission direction considered in the analytical model. The algorithm is described in detail elsewhere [19, 23]. Here, we briefly summarize the main concepts. The TDSE is solved for an atom in a classical laser field, whereby the interaction is described in dipole approximation. The single active electron (SAE) is bound by a spherically symmetric potential $V(r)$. The vector potential $\mathbf{A}(t)$ is used. It is connected to the electric field defined in equation (3) by $\mathbf{F}(t) = -\partial\mathbf{A}(t)/\partial t$.

Owing to its single valence electron, sodium, or alkali metal atoms in general, are the perfect candidates for the commonly employed SAE approximation. For modeling the emission of sodium's outermost electron, we use the spherically symmetric Hellmann pseudopotential [24, 25]

$$V(r) = -\frac{1}{r} + \frac{21}{r}e^{-2.5492r}. \quad (17)$$

The resulting TDSE is solved by expanding the wave function in spherical harmonics. Imaginary-time propagation is used to obtain the desired ground state of the valence electron. Afterwards, the wave function is propagated on a uniformly discretized radial grid using a Crank–Nicolson propagator. The final photoelectron spectra are calculated using the window-operator method [26]. Convergence was reached for a radial step size $dr = 0.2$, time step $dt = 0.05$, and a maximum angular momentum quantum number $\ell_{\text{max}} = 12$. For better comparison with experimental data, the spectra were intensity averaged over the laser focal volume using the method outlined in [27, 28].

3. Experimental setup

The experimental setup, shown in figure 2, consists of a few-cycle Ti:Sapphire laser system (FemtopowerTM CompactTM Pro). The laser pulses are spectrally broadened in a neon-filled, hollow-core fiber and subsequently compressed using chirped mirrors to compensate the dispersion of the fiber and the following optical elements. After a thin, broadband beamsplitter, one beam is coupled into an ATI time-of-flight spectrometer (TOF) in which an atomic oven dispenses a jet of sodium vapor. The other part of the beam is steered into a CE phasemeter (CEPM) [9], which measures relative CEPs of each individual laser shot of the pulse train with a single-shot uncertainty of 230 mrad. The CE phases of the laser pulses vary randomly. This is done by evaluating the left-right asymmetry of photoelectron TOF spectra of ionized xenon in real-time. The information is used to tag the data from the TOF spectrometer with the CEP. Subsequent sorting yields a CEP- and energy-dependent ATI spectrum for sodium. Note that even though the TOF measures only electrons emitted to one side, left-right asymmetries can still be determined using the symmetry of the laser pulse, i.e., the yield in the left direction for CEP φ is identical to the yield in the right direction at phase $\varphi + \pi$. In addition, the CEPM was used to measure the pulse duration as ~ 6 fs, for the pulse with compensated second-order dispersion.

In the experiment, the atomic oven is heated to 291 °C. At this temperature, the Na₂ dimer to Na monomer ratio is 1% [29] and thus the influence of Na₂ can be safely neglected. The spectral phase of the laser pulses is independently controlled by sets of fused silica wedges. For both beam paths, the intensity is controlled by constraining the beam diameter via adjustable iris apertures. The ionization of sodium is performed at a peak intensity of $6.5 \times 10^{12} \text{ W cm}^{-2}$, which is determined from pulse duration, energy and focal spot size measurements.

4. Results and discussion

To determine if the simple model described in section 2.1 predicts the correct behavior for the CEP-, energy-, and chirp-dependent ATI left-right asymmetry in sodium, we measure the spectra with the aforementioned setup, calculate them with the TDSE and model them analytically. All three methods are displayed side-by-side in figure 4 for three different chirps.

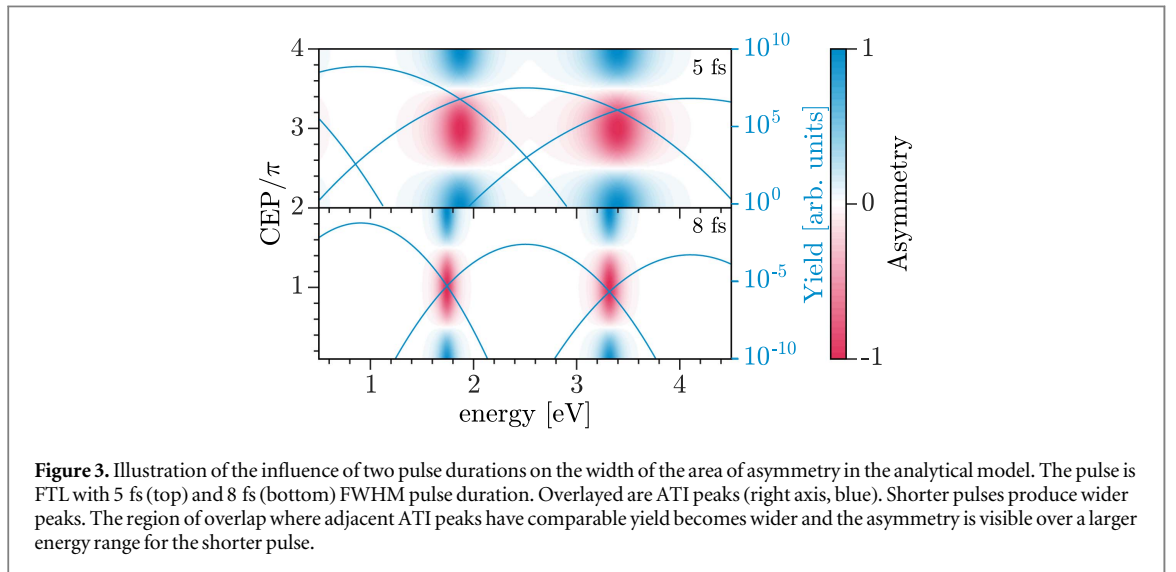
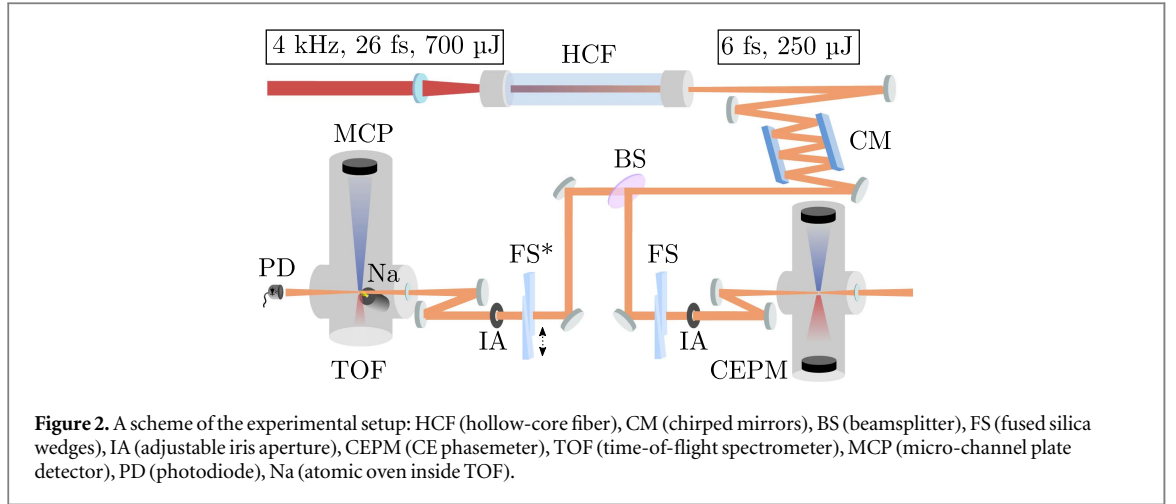
Using motorized fused silica wedges, asymmetry maps were recorded for different chirps by scanning the amount of fused silica in the beam path over a range of 1.60 mm. The results of the scan were evaluated and the position producing slope $s = 0$ was assumed to belong to a pulse whose second-order dispersion is compensated. For this position, we defined the propagation distance inside the glass to be $d = 0 \text{ }\mu\text{m}$.

In figure 4 asymmetry maps obtained from the analytical model, experimental data and TDSE calculations are compared for $d = -636 \text{ }\mu\text{m}$ (left), $0 \text{ }\mu\text{m}$ (middle), and $+636 \text{ }\mu\text{m}$ (right). Calculations for both theoretical models were performed employing a central wavelength of $\lambda = 775 \text{ nm}$, an FTL pulse duration $\tau_0 = 6 \text{ fs}$, a peak intensity $I_0 = 6.5 \times 10^{12} \text{ W/cm}^2$ and a group velocity dispersion $k'' = 38.22 \text{ fs}^2/\text{mm}$. This results in a chirp, pulse duration and intensity of -24.3 fs^2 , 12.7 fs and $I_0 = 3.1 \times 10^{12} \text{ W/cm}^2$ (left), 0.0 fs^2 and 6 fs (middle), as well as $+24.3 \text{ fs}^2$ and 12.7 fs (right) in figure 4.

Considering the few fundamental assumptions that went into building the analytical model, the agreement with the measured data is remarkable. The two main areas of visible asymmetry, namely around 1.9 eV and 3.4 eV, coincide between analytical model and experiment. As discussed above and illustrated in figure 3, the highest asymmetry is predicted at the intersection of adjacent peaks and the width of these regions increases with the spectral bandwidth of the pulse. Thus, the analytical model reveals the origin of the asymmetry structures: Both, CEP and the laser spectral phase, i.e., the chirp, are imprinted on the electronic wave function. This manifests itself in the form of asymmetries, due to interference of even and odd parity electrons, in between the peaks. Moreover, the agreement between the measurement, TDSE, and simple model confirms that a multiphoton interpretation of the dynamics is appropriate as the analytical model completely neglects contributions from tunneling ionization or rescattering. Moreover, the measurement and modeling is in excellent agreement with the results in [20]. Since xenon starts in a p -state, whereas sodium is originally in an s -state, this shows that the nature of the stripe-like structures is independent of the symmetry of the initial state.

It is noticeable that the measurements show additional structures, deviating from the perfectly striped nature predicted by the analytical model. An example of these structures are the inclined ‘tails’, departing from the predicted horizontal structure, to the bottom left and top right side of the main horizontal stripes in figure 4(e). Unsurprisingly, the TDSE asymmetry maps (third row in figure 4) reproduce more of these effects.

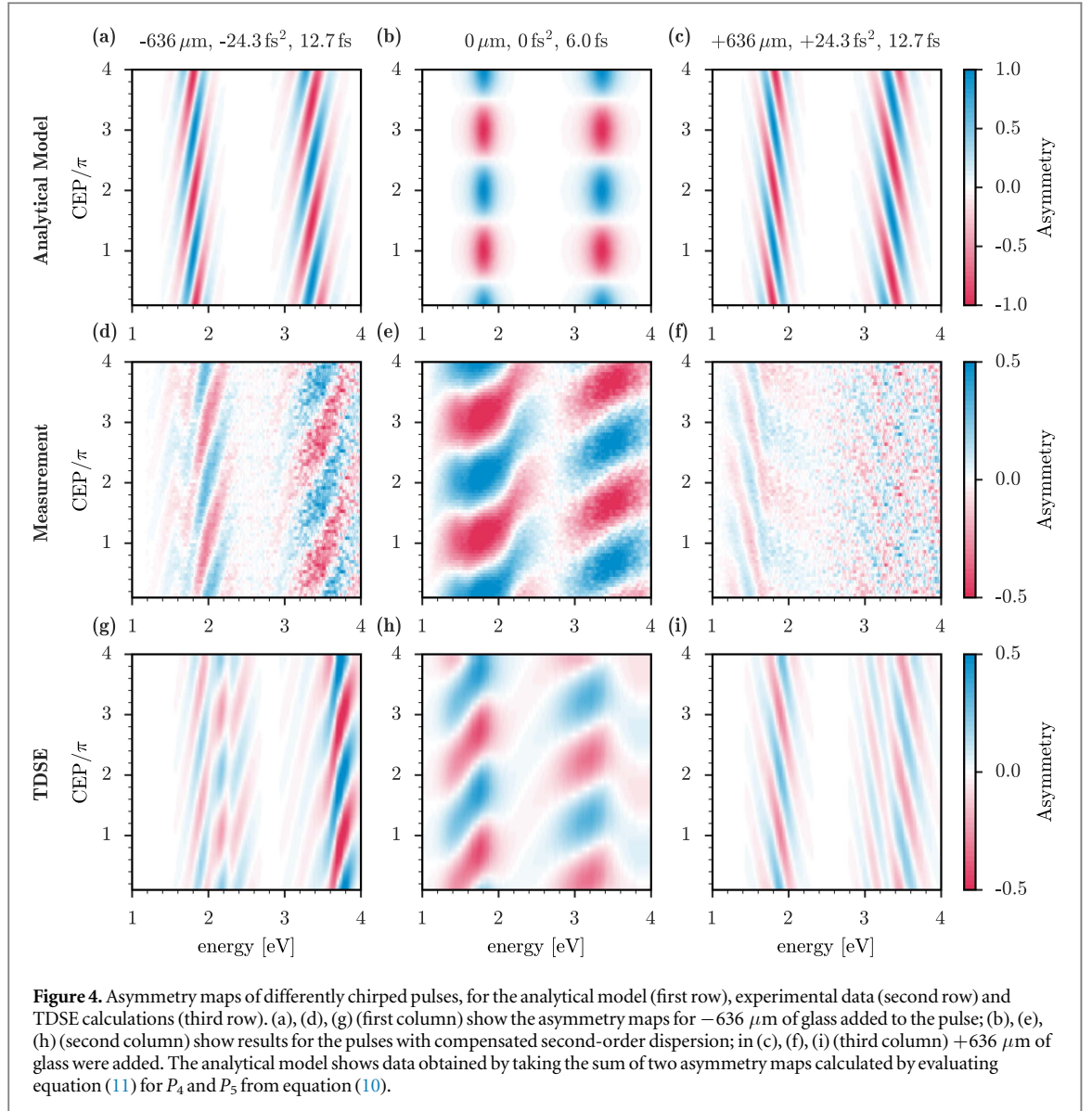
The emergence of these irregular structures is mainly attributed to three factors. Firstly, the influence of the atomic binding potential during the ionization process imprints an additional phase on the electron. The TDSE captures much of this influence, but relies on a pseudopotential, which may deviate from the realistic atom. These effects of the atomic binding potential are neglected in the analytical model.



Secondly, the experimental pulse contains higher-order dispersion not accounted for in the calculations. By numerically extending the analytical model to Gaussian pulses containing solely third- or fourth-order dispersion, we confirmed that these higher-orders cause characteristic structures of their own. However, for a Gaussian pulse chirped in the range -830 to $+830$ μm of glass, second-order dispersion dominates the behavior of the asymmetry structures.

Thirdly, and most importantly, sodium has several atomic resonances within the frequency bandwidth of the pulse, which become especially important for ionization by chirped pulses. Of particular importance here is the ground state to first excited state, $3s \rightarrow 3p$, transition, which corresponds to 589 nm. We can observe this resonance as a side peak around 1.7 eV in figure 1. As this is in the high energy part of the broadband laser spectrum used here, it plays a strong role in laser-induced electron dynamics for negatively chirped pulses, for which the leading edge is shifted to the short-wavelength high-energy portion of the spectrum. This allows resonant excitation to an excited state followed by ionization, which significantly increases the ionization yield [13], i.e., resonance-enhanced multiphoton ionization (REMPI) [30]. In contrast, for a positively chirped pulse, the resonance can only be slowly accessed and, thus, ionization is reduced. This effect is identified by inspecting the evolution of the population of the $3s$ and $3p$ bound states and the total bound state population in the TDSE, shown in figures 5(a), (c). A similar observation was previously reported in [13] by analyzing TDSE calculations of hydrogen. Depending on the chirp, different bound state dynamics were accessed, leading to differing final photoelectron spectra. Although these factors are not considered in the analytical model, there is good agreement between it, TDSE and measurement.

To further confirm the predicted behavior of the slope of the asymmetry structure described in equation (16), we perform a slope analysis, using the algorithm outlined at the end of section 2.1, and plot the slope of the structure as a function of fused silica in the beam path for the analytical model, TDSE and measurement in figure 5(b). Here, one sees that there is excellent agreement between the measurement and



TDSE and that, despite its simplicity, the analytical model quantitatively predicts the behavior quite well. As expected, the analytical model works better for positively chirped pulses, as the aforementioned resonance does not play an important role there. The influence of the resonance can be seen in the jump in slope around 2 eV in figures 4(d) and (g), which corresponds to the deviation between measurement and analytical model around $-600 \mu\text{m}$ of fused silica in figure 5(b). The differences between measurement and TDSE are most likely due to the higher-order chirp in the measurements. They become the more pronounced, the more glass is added.

Although this measurement can be extended to even larger chirps, the experiment becomes more challenging. As the absolute value of the chirp increases, the pulse length increases, which decreases the peak intensity of the pulse and decreases the yield. However, this might be circumvented by increasing the target density, or adjusting the pulse energy. Nevertheless, here we display measurements from the near FTL 6 fs pulse to heavily positively and negatively chirped 15.7 fs pulses and even larger chirps could be measured given sufficient count rate.

Note that as one moves further into the tunneling regime; by some combination of increasing intensity, increasing wavelength or decreasing ionization potential; the qualitative effect of stripes in the asymmetry map remains. However, in this regime electron rescattering is much more prevalent and problematic than in the multiphoton regime. This rescattering process, i.e., the ionized electron returning to and scattering off of the parent ion, is highly sensitive to the CEP and thus creates additional structures in the asymmetry map, which makes the determination of the chirp much more difficult if not impossible.

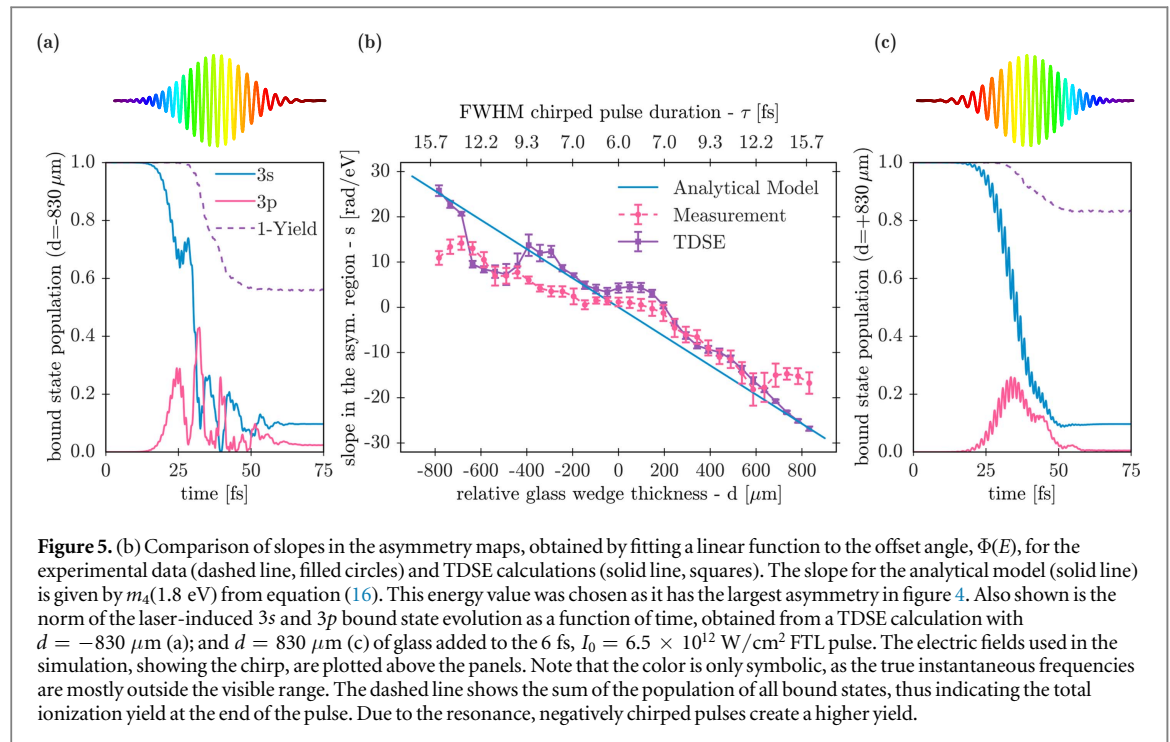


Figure 5. (b) Comparison of slopes in the asymmetry maps, obtained by fitting a linear function to the offset angle, $\Phi(E)$, for the experimental data (dashed line, filled circles) and TDSE calculations (solid line, squares). The slope for the analytical model (solid line) is given by $m_4(1.8 \text{ eV})$ from equation (16). This energy value was chosen as it has the largest asymmetry in figure 4. Also shown is the norm of the laser-induced 3s and 3p bound state evolution as a function of time, obtained from a TDSE calculation with $d = -830 \mu\text{m}$ (a); and $d = 830 \mu\text{m}$ (c) of glass added to the 6 fs, $I_0 = 6.5 \times 10^{12} \text{ W/cm}^2$ FTL pulse. The electric fields used in the simulation, showing the chirp, are plotted above the panels. Note that the color is only symbolic, as the true instantaneous frequencies are mostly outside the visible range. The dashed line shows the sum of the population of all bound states, thus indicating the total ionization yield at the end of the pulse. Due to the resonance, negatively chirped pulses create a higher yield.

5. Summary

The carrier-envelope-phase- and chirp-dependent multiphoton ionization of sodium by few-cycle laser pulses at 775 nm and $6.5 \times 10^{12} \text{ W/cm}^2$ has been measured. We observed stripe-like structures in the CEP- and energy-dependent left-right asymmetry maps, whose inclination can be controlled by applying different chirps to the laser pulse. Using a perturbative multiphoton model, we explained this observation as the interference of electrons with different parities *in between* adjacent above-threshold ionization peaks. The experimental findings were compared with solutions of the three-dimensional time-dependent Schrödinger equation, as well as an analytical model. This allows us to derive a simple equation, which relates the slope of the stripe-like structures to the chirp of the laser. Good agreement between the theoretically predicted and experimentally measured slope is found, which suggests that the effect reported here could be used in online laser diagnostics in parallel to an experiment to monitor the chirp of the laser. Finally, measurement of the carrier-envelope-phase in the multiphoton regime, even for longer pulse durations, is possible.

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References

- [1] Agostini P, Fabre F, Mainfray G, Petite G and Rahman N K 1979 Free-free transitions following six-photon ionization of xenon atoms *Phys. Rev. Lett.* **42** 1127–30
- [2] Ferray M, L'Huillier A, Li X F, Lompre L A, Mainfray G and Manus C 1988 Multiple-harmonic conversion of 1064nm radiation in rare gases *J. Phys. B: At. Mol. Opt. Phys.* **21** L31
- [3] L'Huillier A, Lompre L A, Mainfray G and Manus C 1983 Multiply charged ions induced by multiphoton absorption in rare gases at $0.53 \mu\text{m}$ *Phys. Rev. A* **27** 2503–12
- [4] Keldysh L V 1965 Ionization in the field of a strong electromagnetic wave *Sov. Phys. JETP* **20** 1307
- [5] van Linden van den Heuvell H B and Muller H G 1987 Limiting cases of excess-photon ionization *Multiphoton Processes* ed S J Smith and P L Knight (Cambridge: Cambridge University Press) pp 25–34
- [6] Paulus G G, Becker W, Nicklich W and Walther H 1994 Rescattering effects in above-threshold ionization: a classical model *J. Phys. B: At. Mol. Opt. Phys.* **27** L703
- [7] Becker W, Grasbon F, Kopold R, Milošević D B, Paulus G G and Walther H 2002 Above-threshold ionization: from classical features to quantum effects *Adv. At., Mol., Opt. Phys.* **48** 35–98
- [8] Paulus G G, Lindner F, Walther H, Baltuška E, Lezius M and Krausz F 2003 Measurement of the phase of few-cycle laser pulses *Phys. Rev. Lett.* **91** 253004

- [9] Wittmann T, Horvath B, Helml W, Schatzel M G, Gu X, Cavalieri A L, Paulus G G and Kienberger R 2009 Single-shot carrier-envelope phase measurement of few-cycle laser pulses *Nat. Phys.* **5** 357–62
- [10] Zhang Y, Kellner P, Adolph D, Zille D, Wustelt P, Würzler D, Skruszewicz S, Möller M, Sayler A M and Paulus G G 2017 Single-shot, real-time carrier-envelope phase measurement and tagging based on stereographic above-threshold ionization at short-wave infrared wavelengths *Opt. Lett.* **42** 5150–3
- [11] Nakajima T and Cormier E 2007 Effects of the carrier-envelope phase of chirped laser pulses in the multiphoton ionization regime *Opt. Lett.* **32** 2879–81
- [12] Nakajima T 2007 Above-threshold ionization by chirped laser pulses *Phys. Rev. A* **75** 053409
- [13] Laulan S, Haché J, Ba H and Barmaki S 2013 Ionization process of atoms by intense femtosecond chirped laser pulses *J. Mod. Phys.* **4** 20–6
- [14] Paulus G G, Nicklich W, Xu H, Lambropoulos P and Walther H 1994 Plateau in above threshold ionization spectra *Phys. Rev. Lett.* **72** 2851–4
- [15] Xiang Y, Niu Y and Gong S 2009 Above-threshold ionization by few-cycle nonlinear chirped pulses *Phys. Rev. A* **80** 023423
- [16] Peng L-Y, Tan F, Gong Q, Pronin E A and Starace A F 2009 Few-cycle attosecond pulse chirp effects on asymmetries in ionized electron momentum distributions *Phys. Rev. A* **80** 013407
- [17] Yudin G L, Bandrauk A D and Corkum P B 2006 Chirped attosecond photoelectron spectroscopy *Phys. Rev. Lett.* **96** 063002
- [18] Cormier E and Lambropoulos P 1998 Effect of the initial phase of the field in ionization by ultrashort laser pulses *Eur. Phys. J. D* **2** 15–20
- [19] Mosert V and Bauer D 2016 Photoelectron spectra with Qprop and t-SURFF *Comput. Phys. Commun.* **207** 452–63
- [20] Abel M J, Pfeifer T, Jullien A, Nagel P M, Bell M J, Neumark D M and Leone S R 2009 Carrier-envelope phase-dependent quantum interferences in multiphoton ionization *J. Phys. B: At. Mol. Opt. Phys.* **42** 075601
- [21] Colosimo P et al 2008 Scaling strong-field interactions towards the classical limit *Nat. Phys.* **4** 386–9
- [22] Chang Z 2011 *Fundamentals of Attosecond Optics* (Boca Raton, FL: CRC Press)
- [23] Bauer D and Koval P 2006 Qprop: a Schrödinger-solver for intense laser–atom interaction *Comput. Phys. Commun.* **174** 396–421
- [24] Hellmann H 1935 A new approximation method in the problem of many electrons *J. Chem. Phys.* **3** 61–61
- [25] Milošević M Z and Simonović N S 2015 Calculations of rates for strong-field ionization of alkali-metal atoms in the quasistatic regime *Phys. Rev. A* **91** 023424
- [26] Schafer K J and Kulander K C 1990 Energy analysis of time-dependent wave functions: application to above-threshold ionization *Phys. Rev. A* **42** 5794–7
- [27] Wang P, Sayler A M, Carnes K D, Esry B D and Ben-Itzhak I 2005 Disentangling the volume effect through intensity-difference spectra: application to laser-induced dissociation of H_2^+ *Opt. Lett.* **30** 664–6
- [28] Sayler A M, Wang P Q, Carnes K D and Ben-Itzhak I 2007 Determining intensity dependence of ultrashort laser processes through focus z -scanning intensity-difference spectra: application to laser-induced dissociation of $\text{h} + 2\text{J}$ *J. Phys. B: At. Mol. Opt. Phys.* **40** 4367
- [29] Nesmeyanov A N 1963 *Vapor Pressure of the Elements* ed J S Carasso (New York: Academic)
- [30] Ashfold M N R and Howe J D 1994 Multiphoton spectroscopy of molecular species *Annu. Rev. Phys. Chem.* **45** 57–82