

## Coherent strong-field control of multiple states by a single chirped femtosecond laser pulse

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**Abstract.** We present a joint experimental and theoretical study on strong-field photo-ionization of sodium atoms using chirped femtosecond laser pulses. By tuning the chirp parameter, selectivity among the population in the highly excited states 5p, 6p, 7p and 5f, 6f is achieved. Different excitation pathways enabling control are identified by simultaneous ionization and measurement of photoelectron angular distributions employing the velocity map imaging technique. Free electron wave packets at an energy of around 1 eV are observed. These photoelectrons originate from two channels. The predominant 2 + 1 + 1 resonance enhanced multi-photon ionization (REMPI) proceeds via the strongly driven two-photon transition  $4s \leftarrow \leftarrow 3s$ , and subsequent ionization from the states 5p, 6p and 7p whereas the second pathway involves 3 + 1 REMPI via the states 5f and 6f. In addition, electron wave packets from two-photon ionization of the non-resonant transiently populated state 3p are observed close to the ionization threshold. A mainly qualitative five-state model for the predominant excitation channel is studied theoretically to provide insights into the physical mechanisms at play. Our analysis shows that by tuning the chirp parameter the dynamics is effectively controlled by dynamic Stark shifts and level crossings. In particular, we show that under the experimental conditions the passage through

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an uncommon three-state ‘bow-tie’ level crossing allows the preparation of coherent superposition states.

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## 1. Introduction

Selective excitation of preselected target states making use of shaped femtosecond laser pulses is at the heart of coherent quantum control [1]–[10]. Closed-loop optimization strategies [4], [11]–[15] have proven to be enormously successful in controlling a huge variety of quantum systems; however, studies on model systems employing defined pulse shapes are the key to better understand the underlying physical mechanisms and to further develop quantum control concepts and techniques. This applies in particular to *strong-field* quantum control [16]–[20] characterized by non-perturbative interaction of a quantum system with intense-shaped laser pulses. Strong-field physical mechanisms involve—besides the interference of multiple excitation pathways—adiabatic and non-adiabatic time evolution accompanied by dynamic Stark shifts (DSSs) in the order of hundreds of meV. The latter is responsible for modification of the atomic states or molecular potential surfaces [21]–[23] such that new pathways become available and new target states—inaccessible in weak laser fields—open up. Recent studies of strong-field control on model systems devoted to the analysis of the basic physical mechanisms revealed that the concept of selective population of dressed states (SPODS) [24] provides a natural description of controlled dynamics in intense-shaped laser fields. For example, it was shown that ultrafast switching among different target channels by phase discontinuities within the pulse [16], [24]–[26], rapid adiabatic passage (RAP) by chirped pulses [27] and combinations thereof [28] are realizations of this general concept.

Chirped pulses are a well-established tool in quantum control because they usually serve as a prototype for shaped pulses with controllable envelope and time-varying instantaneous frequency. Therefore, they have played a prominent role in the development of quantum control concepts and techniques and are still the ‘workhorse’ to test novel strategies in quantum control. Examples of quantum control with chirped pulses comprise studies of selective excitation and ionization of a multilevel system in alkali atoms [19, 27], [29]–[34], control of molecular dynamics in diatomics and dyes [35]–[41], ([42] and references therein), measurement of coherent transients [43] and the development of adiabatic passage techniques [44].

In the present contribution, we employ chirped ultrashort laser pulses resulting from phase modulation of the laser spectrum to study resonance-enhanced multiphoton ionization (REMPI) of a multilevel system in sodium atoms. We demonstrate experimentally that different excitation pathways and, accordingly, different target channels can be addressed selectively by a single control parameter, i.e. the chirp. The nature of these pathways is unraveled by measurement of photoelectron angular distributions (PADs) from velocity map imaging (VMI) [45]–[50], yielding detailed information on the origin of the released photoelectron wave packets. Theoretical investigations of the light–atom interaction reveal an interplay of different physical mechanisms governing control. Analysis of the neutral excitation dynamics for a five-state model atom (including the most relevant states 3s, 4s, 5p, 6p and 7p) under the influence of a chirped ultrashort laser pulse highlights how physical mechanisms, such as RAP and DSS, act jointly to either address single states among the high-lying sodium states 5p, 6p and 7p (cf figure 1) or excite superpositions of any two neighboring states. We point out that the present paper extends two earlier techniques in several significant directions. The technique of Melinger *et al* [29] uses a single chirped picosecond laser pulse to selectively excite the two fine-structure components  $3p_{1/2}$  and  $3p_{3/2}$  in sodium atoms. The present technique adds a DSS to the control tools, which enables the population of a third state, and also the creation of coherent superposition states. The technique of Clow *et al* [34] makes use of a shaped femtosecond pulse to selectively populate a single highly excited state. The present technique is more flexible, since it allows to populate several different states by variation of a single parameter: the chirp.

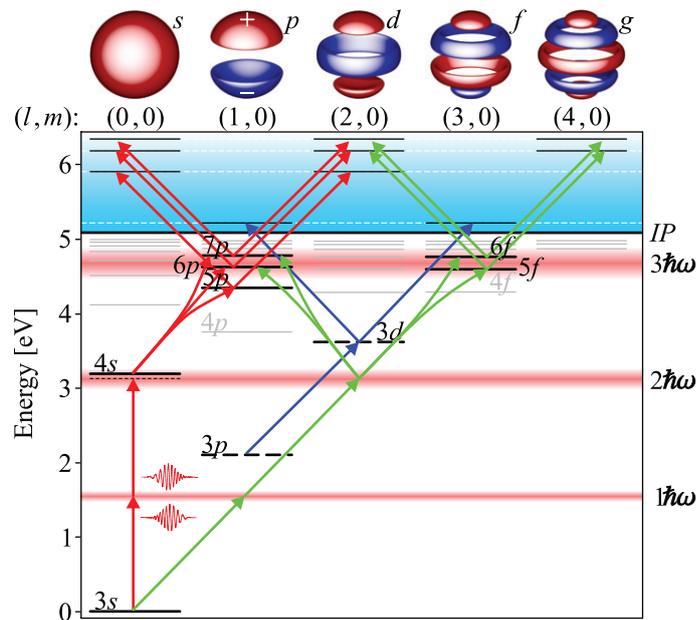
The paper is organized as follows. We start in section 2 by introducing the excitation and ionization scheme of sodium atoms exposed to ultrashort near-infrared laser pulses, and subsequently describe the details of our experimental setup. The experimental results are presented in section 3 along with a physical discussion of general features observed in the measured PADs supported by numerical simulations of the measurement results. Section 4 provides a detailed theoretical analysis of the strong-field-induced chirped excitation dynamics in terms of adiabatic states, highlighting different physical mechanisms that govern the light–atom interaction. We conclude the paper with a brief summary and conclusions.

## 2. Experiment

In our experiment, we combine spectral phase shaping to produce chirped ultrashort laser pulses with the measurement of PADs resulting from REMPI of sodium atoms, employing the VMI technique. In this section, we first introduce the sodium excitation scheme with emphasis on the different accessible excitation and ionization pathways. Then we describe the experimental setup and layout of our photoelectron imaging spectrometer.

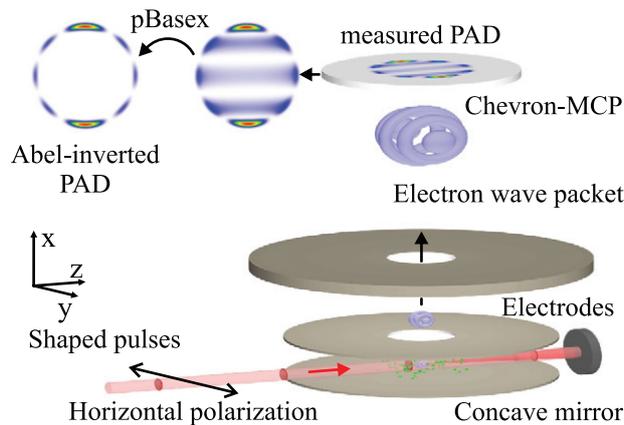
### 2.1. Excitation scheme

Figure 1 shows the excitation and ionization scheme of sodium atoms based on energy-level information taken from the NIST database [51]. Different multi-photon excitation pathways are accessible during the interaction of sodium atoms with intense ultrashort laser pulses (laser specifications are given in section 2.2). The predominant excitation pathway is a 2+1+1 REMPI process via the two-photon transition  $4s \leftarrow \leftarrow 3s$  (red arrows in figure 1), which is nearly resonant with our laser spectrum [52]. Consequential population of states 5p, 6p and 7p gives rise to photoelectron wave packets in the ionization continuum having s- or d-symmetry.



**Figure 1.** Excitation and ionization scheme of sodium atoms illustrating the excitation pathways that arise during the interaction with an intense 795 nm, 30 fs full width at half maximum (FWHM) laser pulse. These pathways comprise a 2 + 1 + 1 REMPI (red arrows) and a 3 + 1 REMPI (green arrows) process from the 3s ground state as well as a two-photon ionization process from state 3p (blue arrows). Blurred red bars represent the one-, two- and three-photon spectra of our laser, respectively. Since state 4s lies within the bandwidth of the two-photon spectrum, the laser strongly drives the transition  $4s \leftarrow \leftarrow 3s$ . Once state 4s is populated, population flows to states 5p, 6p and 7p, giving rise to photoelectron wave packets with combined s- and d-symmetry at characteristic kinetic energies 0.76, 1.04 and 1.20 eV in the ionization continuum. A competing excitation pathway is opened up by three-photon absorption leading to population of states 5f and 6f in addition. Photoelectrons from this excitation channel are characterized by a combined d- and g-symmetry of the measured PADs at kinetic energies 1.02 and 1.18 eV, respectively. Two-photon ionization from the non-resonant, transiently populated state 3p results in photoelectron wave packets at about 0.2 eV, having combined p- and f-symmetry. For illustrative purposes, the relevant symmetries of the released photoelectron wave packets are visualized on top of the figure in red and blue, encoding the positive and negative signs of the electron wavefunction, respectively.

The recorded PADs therefore exhibit a combined s- and d-symmetry and are measured at the distinct kinetic energies 0.76, 1.04 and 1.20 eV, corresponding to states 5p, 6p and 7p, respectively. Alternatively, a 3 + 1 REMPI process (green arrows in figure 1) based on three-photon absorption from the 3s ground state with no intermediate resonances is taken into account, contributing also to the population of states 5p, 6p and 7p but, in addition, transferring population to states 5f and 6f. One-photon ionization of the latter results in photoelectron wave packets with d- and g-symmetry at kinetic energies 1.02 and 1.18 eV, respectively.



**Figure 2.** Experimental setup. Horizontally polarized femtosecond laser pulses are sent into a vacuum chamber and refocused by a 50 mm on-axis concave mirror into sodium vapor provided by an alkali metal dispenser source (not shown). Photoelectrons emitted by the light–atom interaction are projected toward a position-sensitive MCP detector using the VMI method. The amplified signal is recorded by a 1.4 million pixels camera system and sent to a computer. An Abel inversion is performed using the pBasex algorithm.

These photoelectrons are distinguished from the p-state contributions (at 1.04 and 1.20 eV) by the symmetry of their angular distributions. In the following, we will refer to the different photoelectron contributions as different *energy channels* at nominal kinetic energies of 0.8, 1.0 and 1.2 eV, and infer their origin, i.e. the excitation pathway, from the *angular* distribution. Both multi-photon excitation pathways proceed via the intermediate, non-resonant state 3p, which is only transiently populated. However, since ionization takes place *during* the excitation also photoelectrons from this state are detected at low kinetic energies around 0.2 eV (blue arrows in figure 1). For more details see caption of figure 1.

## 2.2. Setup

In this section, the experimental setup comprising the laser system and the photoelectron imaging spectrometer is described. Intense 795 nm, 30 fs FWHM laser pulses provided by an amplified 1 kHz Ti:sapphire laser system (*Femtolasers Femtopower Pro*) were phase modulated in frequency domain by a home-built pulse shaper [53], applying quadratic phase masks of the form  $\varphi_{\text{mod}}(\omega) = \varphi_2/2(\omega - \omega_0)^2$ , where  $\omega_0$  is the central frequency of our laser spectrum [27]. The chirp parameter  $\varphi_2$  was varied in the range from  $-2000 \text{ fs}^2$  to  $+2000 \text{ fs}^2$  in steps of  $\Delta\varphi_2 = 100 \text{ fs}^2$ . The chirped output pulses of  $12 \mu\text{J}$  energy were sent into a vacuum chamber and refocussed by a concave mirror (5 cm focal length; we estimated a *peak* intensity of about  $10^{13} \text{ W cm}^{-2}$  for the bandwidth-limited pulse) into sodium vapor supplied by an alkali metal dispenser source, as shown in figure 2. Photoelectrons released during the strong-field interaction of the shaped pulses with single atoms were detected by a photoelectron imaging spectrometer using the VMI method. In order to compensate the residual chirp of the unmodulated pulse, we performed an *in situ* adaptive optimization of the multi-photon ionization of water vapor background (about  $4 \times 10^{-7} \text{ mbar}$ ) in the interaction region of

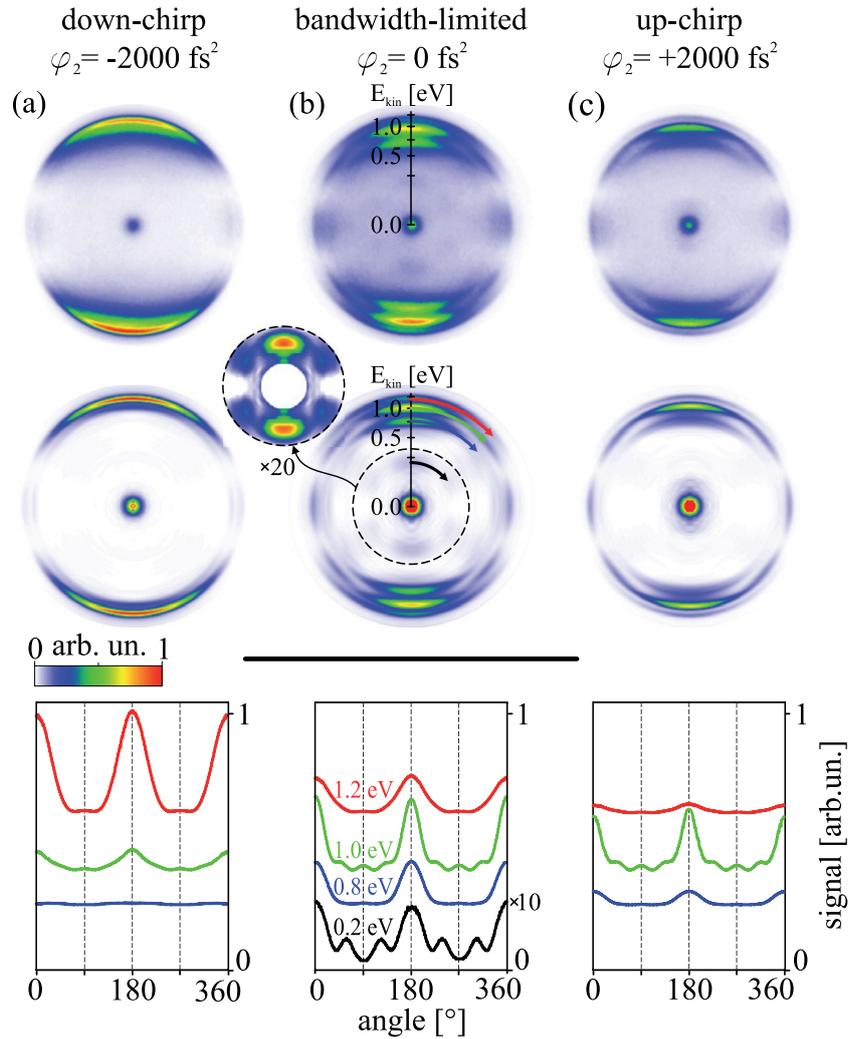
the spectrometer. The resulting optimal compensation phase was additionally applied to the pulse shaper during the experiments, ensuring an error in the chirp parameter  $\varphi_2$  of less than  $150 \text{ fs}^2$ . The energy calibration of the imaging spectrometer was performed using a  $3+1$  REMPI of xenon atoms excited by a Nd:YAG ns laser system at 355 nm, achieving a spectrometer resolution of 60 meV at 0.5 eV. Employing the energy-calibrated photoelectron imaging spectrometer, we studied angular and energy-resolved photoelectron spectra as a function of the chirp parameter  $\varphi_2$ .

### 3. Experimental results and discussion

Figure 3 (upper row) shows measured PADs from REMPI of sodium atoms with chirped fs laser pulses for three exemplary values of the chirp parameter  $\varphi_2$ . The middle row displays the corresponding Abel-inverted (retrieved) PADs obtained by employing the pBasex algorithm [50, 54]. When PADs arise from ionization with polarization-shaped pulses [55], direct tomography methods have been developed for three-dimensional reconstruction of ultrashort free photoelectron wave packets [56]. Angular sections through the retrieved PADs at kinetic energies 0.8, 1.0 and 1.2 eV, as plotted in the lower row, serve to identify the symmetry of the different energy channels observed in the PADs. The PAD measured for the unmodulated, i.e., bandwidth-limited pulse is depicted in the central column. Three major contributions are observed at kinetic energies 0.8, 1.0 and 1.2 eV, related to the energy channels discussed above (cf section 2.1). The angular section taken at 1.2 eV exhibits two minor nodes between  $0^\circ$  and  $180^\circ$ , i.e. d-symmetry. This channel is attributed mainly to ionization via state 7p (red excitation pathway in figure 1), though our numerical simulations (inset of figure 4) indicate that also ionization via state 6f (green excitation pathway in figure 1) delivers a minor contribution. The contribution of an s-wave to this channel, as expected from the excitation scheme figure 1, is reflected in the *weak* equatorial signal. At an angle of  $90^\circ$  s- and d-waves have opposite sign and, thus, interfere destructively, whereas at the poles, i.e. at  $0^\circ$  and  $180^\circ$ , both waves add up constructively. The section taken at 1.0 eV exhibits four nodes between  $0^\circ$  and  $180^\circ$ , corresponding to g-symmetry. This contribution originates predominantly from ionization via state 5f. The observation that the lobe at  $90^\circ$  (and  $270^\circ$ , respectively) is slightly lowered with respect to its two neighbors indicates a weak d-wave contribution interfering destructively with the g-wave in this angular segment. The contribution measured at 0.8 eV shows again combined s- and d-symmetry and is ascribed to ionization via state 5p.

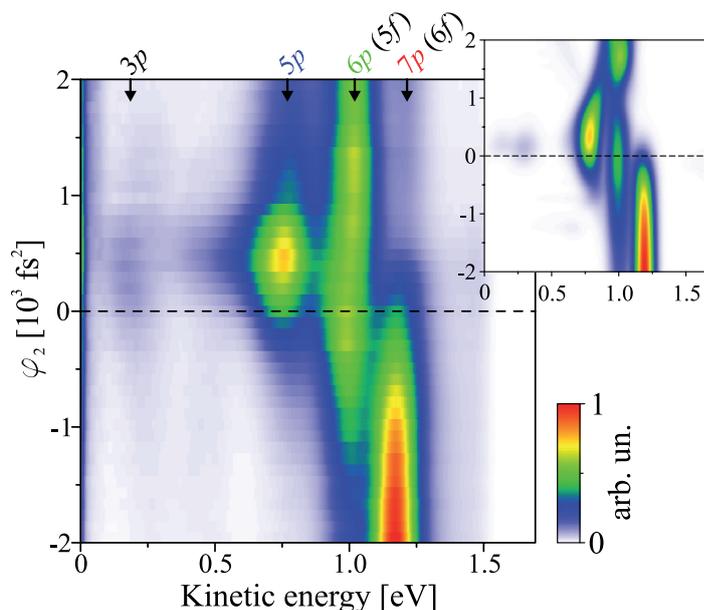
Moreover, a weak contribution is observed at about 0.2 eV, a magnification of which is shown in the inset of figure 3(b). The nodal structure of this signal exhibits distinct f-symmetry. However, the pronounced poles of the PAD as well as the fact that the nodes at  $45^\circ$  and  $135^\circ$  in the angular section are raised with respect to the node at  $90^\circ$  give a hint on a p-wave contribution to the photoelectron signal. Observation of photoelectron wave packets with combined p and f-symmetry close to the ionization threshold is consistent with two-photon ionization from state 3p (blue pathway in figure 1). Note that state 3p is—although non-resonant—transiently populated during the interaction, mediating the multi-photon processes to the state 4s and the high-lying f states.

For large negative values of  $\varphi_2$  (left column in figure 3), i.e. strongly *down*-chirped laser pulses, the outer channel at kinetic energy 1.2 eV is considerably enhanced in comparison to the bandwidth-limited case, whereas the intermediate channel at 1.0 eV is strongly reduced and the two innermost contributions have essentially vanished. Note the change in symmetry of



**Figure 3.** Measured PADs from excitation and ionization of sodium atoms using both chirped and bandwidth-limited fs laser pulses. In the upper row, measured PADs for different values of the chirp parameter  $\varphi_2$  are shown. (a)  $\varphi_2 = -2000 \text{ fs}^2$  (down-chirp). (b)  $\varphi_2 = 0$  (bandwidth-limited). (c)  $\varphi_2 = +2000 \text{ fs}^2$  (up-chirp). All images are scaled to the same maximum value. The middle row contains the corresponding Abel-inverted PADs obtained using the pBasex algorithm. Angular sections through the retrieved PADs at kinetic energies of about 0.2, 0.8, 1.0 and 1.2 eV (lower row) reveal the symmetries of the observed contributions and shed light on the underlying ionization pathways. The signal offsets are introduced for better visibility.

the intermediate channel which exhibits combined s- and d-symmetry in this case, indicating more efficient ionization from state 6p, while the 5f contribution is very small. Changing the sign of  $\varphi_2$ , i.e. using strongly *up*-chirped laser pulses (right column in figure 3), suppresses the high-energy channel in favor of the intermediate channel at 1.0 eV, which dominates the PAD in this case. From its angular section at 1.0 eV, we find a combined d- and g-symmetry, as in the bandwidth-limited case. This contribution is therefore traced back mainly to state



**Figure 4.** Measured photoelectron kinetic energy distributions as a function of the chirp parameter  $\varphi_2$ . The data were obtained by angular integration of the retrieved PADs. Three main energy channels are observed at 0.8, 1.0 and 1.2 eV, each of which can be activated by appropriate choice of the chirp parameter. For  $\varphi_2 \ll 0$ , i.e. strongly down-chirped laser pulses, photoelectrons with high kinetic energies related to the high-lying state 7p (and minor 6f contribution) are produced. The intermediate channel at 1.0 eV, related to states 6p and 5f, is addressed by strongly up-chirped laser pulses with  $\varphi_2 \gg 0$ . Photoelectrons with kinetic energies around 0.8 eV, corresponding to state 5p, are favored at small positive values of  $\varphi_2$ , i.e. high laser pulse peak intensities. The weak contribution at 0.2 eV in the same  $\varphi_2$  region stems from ionization of the non-resonant state 3p. The inset shows results from a numerical simulation of the multi-photon excitation and ionization process.

5f. The finding that the symmetry of photoelectrons from the intermediate channel alters from d to g is rationalized by the change of the ordering of red and blue frequency components within the chirped pulse. For a down-chirped pulse, i.e. when the blue components arrive first, initially, the system is in resonance with the two-photon transition  $4s \leftarrow \leftarrow 3s$  implying efficient ionization via the p states (red pathway in figure 1). On the other hand, up-chirped pulses favor ionization via state 5f since at early times the system is in resonance with the three-photon transition  $5f \leftarrow \leftarrow \leftarrow 3s$  (green pathway in figure 1). Such processes have also been observed in [39] under different excitation conditions.

In order to provide the full picture of the chirp-dependent population flow to the different energy channels, we performed an angular integration of all 41 measured PADs and present the resulting energy-resolved photoelectron spectra in terms of a two-dimensional map as a function of the kinetic energy and the chirp parameter  $\varphi_2$ . The result obtained upon variation of  $\varphi_2$  in the range from  $-2000$  to  $+2000$   $\text{fs}^2$  is displayed in figure 4. The three major channels at 0.8, 1.0 and 1.2 eV are clearly visible. Note that for e.g. rare gas atoms under our experimental

conditions ponderomotive shifts of more than 0.5 eV are calculated. No such shifts are observed in the experiment, since the high-frequency approximation [57, 58] (necessary condition for the application of the ponderomotive energy concept) is not valid for alkalis excited by near-infrared laser radiation. An analysis of the neutral excitation dynamics behind the observed contributions will be given in section 4. The map illustrates the above statements that for large negative values of  $\varphi_2$  the high-energy channel at 1.2 eV is addressed with high efficiency, i.e. a down-chirped pulse steers the population predominantly towards the high-lying state 7p. For large positive chirp values the intermediate channel is selectively addressed, corresponding to predominant population of states 6p and 5f. The low-energy channel is accessed most efficiently in the vicinity of  $\varphi_2 = 500 \text{ fs}^2$ . In fact, in the regime  $0 \leq \varphi_2 \leq 1000 \text{ fs}^2$  the photoelectron spectrum is made up of contributions from states 5p, 6p and 5f. Because the excitation (and simultaneous ionization) takes place on an ultrashort timescale precluding decoherence processes, a *coherent* superposition of states 5p, 6p and 5f is excited in this chirp regime. Upon changing the sign of  $\varphi_2$ , i.e. for  $-1000 \text{ fs}^2 \leq \varphi_2 \leq 0$ , the laser pulse induces a coherent superposition of states 6p, 5f and 7p. Photoelectrons observed at about 0.2 eV for moderate positive chirps are attributed to two-photon ionization from state 3p.

The inset to figure 4 shows results from a numerical simulation of the simultaneous multi-photon excitation and ionization process. The calculations are based on numerical integration of the time-dependent Schrödinger equation for a neutral 20-state system (comprising those states labeled in figure 1 and taking the fine structure splitting into account) interacting with an intense chirped 795 nm, 30 fs FWHM Gaussian input pulse. One-photon ionization from the high-lying p and f states is treated within a simplified model employing the first-order perturbation theory. We assume a flat continuum and unit coupling elements with no additional phases for all bound-free transitions. A more rigorous treatment of the ionization step involving the determination of radial coupling matrix elements also for the bound-free transitions is provided by, e.g. single-channel quantum defect theory [59] as reported for instance in [60, 61]. In order to model the two-photon ionization from state 3p proceeding, for example, via state 3d as indicated by the blue pathway in figure 1, we employed second-order perturbation theory. For a more detailed description of our method see [25, 27, 62]. The simulation of photoelectron spectra reproduces the main features of the experimental results very well. This allows us to look into the underlying *neutral* excitation dynamics and follow the population flow within the bound atomic system. We find that for *large negative chirp*  $\varphi_2$  state 7p is addressed almost selectively, while for large positive  $\varphi_2$  values both states 6p and 5f are populated efficiently in equal measure. The latter is in accordance with the experimental observation of the PAD with pronounced g-symmetry in the intermediate channel at 1.0 eV for *large positive chirp* (see figure 3(c)). The most efficient excitation of state 5p occurs for *moderate positive chirp*. However, in this chirp regime states 6p and 5f receive comparable population confirming the observation of a PAD with a contribution of g-symmetry at 1.0 eV and *zero chirp*. At *moderate negative chirp*, we obtain a coherent superposition of states 6p, 5f and 7p. Note that the weak contribution around 0.2 eV and small positive values of  $\varphi_2$  observed in the experiment (shown in the inset to figure 3(b)) is also reproduced in the simulation. Within the framework of our simulation, these photoelectrons are ascribed to two-photon ionization from state 3p which receives non-perturbative *transient* population. We note that in a perturbative regime, ionization from this transiently populated state could be interpreted as a transition from a *virtual* state.

In the next section, we will further investigate the neutral population dynamics by means of a reduced atomic system in order to rationalize the general features observed in the experiment in terms of physical mechanisms governing the excitation process.

#### 4. Theoretical model

In this section, we provide mainly a qualitative description of the system at hand. To this end, we assume that the photoelectron signal arises most significantly through the 2 + 1 + 1 REMPI channel (red pathway in figure 1), involving the five states 3s, 4s, 5p, 6p and 7p. The idea of this reduction is to demonstrate the basic principles influencing the dynamics of the whole system, which become more transparent in this simplified model, involving the most significant states for our experiment. In this approach, we adiabatically eliminated state 3p [44, 63, 64] because it is off resonance and receives smaller transient population than the other coupled states. Its presence, however, affects the population dynamics significantly for it induces strong DSSs in the energies of states 3s and 4s, which substantially modify the energy diagram.

The quantum dynamics of this five-state system obeys the time-dependent Schrödinger equation

$$i\hbar \frac{d}{dt} \mathbf{c}(t) = \mathbf{H}(t) \mathbf{c}(t). \quad (1)$$

The Hamiltonian  $\mathbf{H}(t)$  in the rotating-wave approximation, rotating with the instantaneous laser frequency  $\omega(t) = \omega_0 + 2at$  (see equation (A.5) in the appendix), is given by [44, 64]

$$\mathbf{H}(t) = \hbar \begin{bmatrix} \Delta_1 - S_1 & \frac{1}{2}\Omega_{12} & 0 & 0 & 0 \\ \frac{1}{2}\Omega_{12} & \Delta_2 - S_2 & \frac{1}{2}\Omega_{23} & \frac{1}{2}\Omega_{24} & \frac{1}{2}\Omega_{25} \\ 0 & \frac{1}{2}\Omega_{23} & \Delta_3 & 0 & 0 \\ 0 & \frac{1}{2}\Omega_{24} & 0 & \Delta_4 & 0 \\ 0 & \frac{1}{2}\Omega_{25} & 0 & 0 & \Delta_5 \end{bmatrix}. \quad (2)$$

Here the explicit time dependence is dropped for ease of notation. The vector  $\mathbf{c}(t) = [c_1(t), c_2(t), \dots, c_5(t)]^T$  consists of the amplitudes of the five states, ordered as shown above, which are obtained by numerical integration of the Schrödinger equation (1), the respective populations are  $P_n(t) = |c_n(t)|^2$ ,  $\Delta_n(t) = \omega_n - k\omega(t)$  are the generally time-dependent atom–laser detunings, where  $\omega_n$  are the atomic state eigenfrequencies, with  $\omega_{3s}$  taken as zero,  $k$  is the transition order,  $\Omega_{2n} = d_{2n}\Omega_0 f(t)$  represent the one-photon couplings of state 2 to state  $n$  ( $n = 3, 4, 5$ ),  $\Omega_{12} = q_{12}\Omega_0^2 f^2(t)$  is the two-photon coupling between states 1 and 2, with  $f(t)$  being the chirped laser electric field envelope,  $d_{mn}$  are the relevant transition dipole moments in atomic units,  $q_{12}$  is the effective two-photon transition moment (cf equation (A.1)) and  $S_1$  and  $S_2$  represent the DSS of states 1 and 2, respectively,

$$S_1 = \frac{\Omega_{3s3p}^2}{4\Delta_{3p}}, \quad S_2 = \frac{\Omega_{3p4s}^2}{4\Delta_{3p}}. \quad (3)$$

The effect of the DSS due to state 3d is neglected for it is very weakly coupled to the states whose energies it might influence: the p states are coupled about 10 times stronger to state 4s

as compared with state 3d; state 3d is not directly coupled to state 3s, but rather through a two-photon transition. In the first two diagonal elements of the Hamiltonian (associated with the energies of states 3s and 4s) the atom–laser detuning and the DSS add up to a time-dependent *effective* chirp: the former resulting from the time-dependent instantaneous laser frequency  $\omega(t)$ , and the latter deriving from the time-dependent shift of the level energies due to DSS.

#### 4.1. Excitation regimes

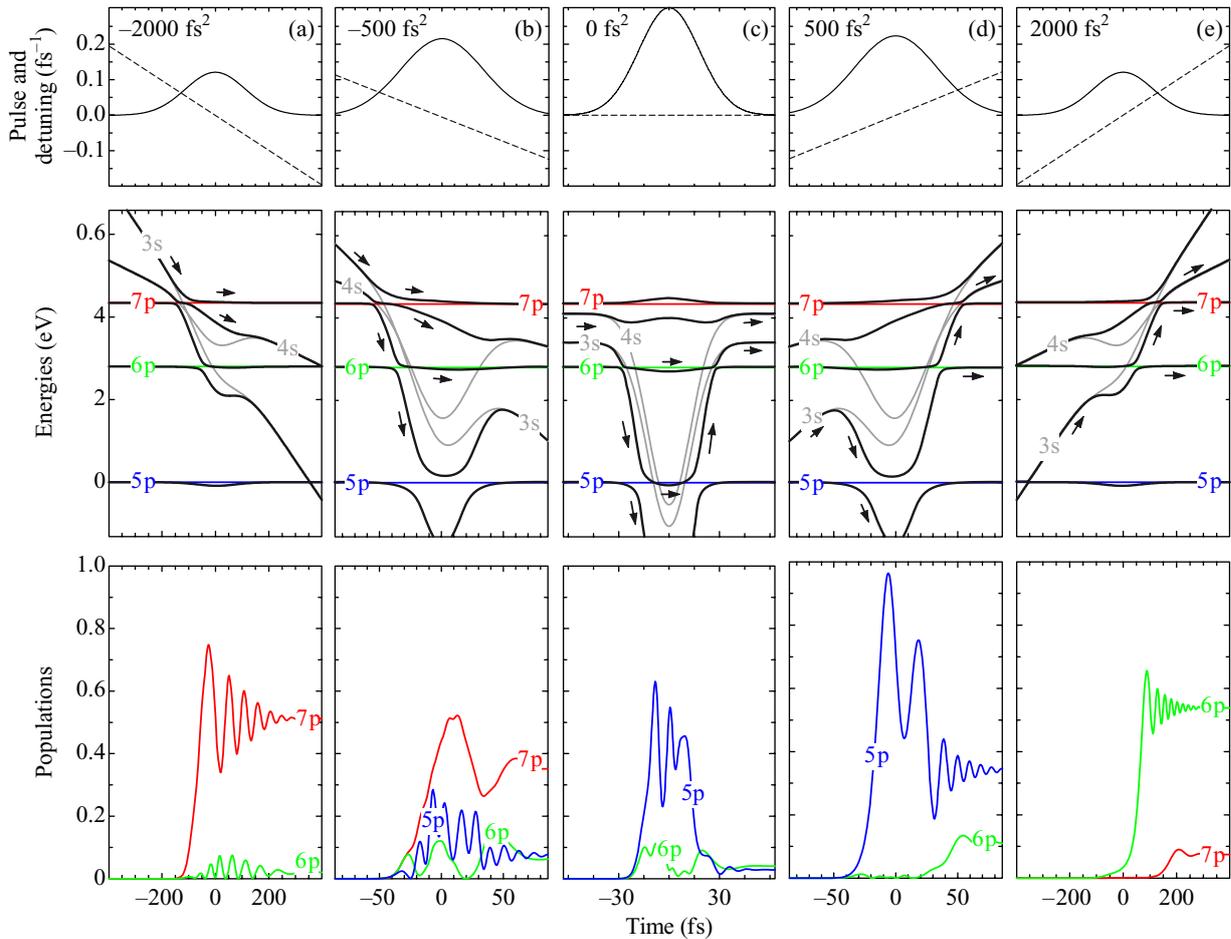
In figure 5, we distinguish five different regimes in regard to the value of the chirp  $\varphi_2$ . In all cases, we plot the bare-state energies and analyze the dynamics by accounting for the presence of level crossings. Because it is the ionization signal that is observed in the experiment it is also important *when* a particular level crossing occurs: a level crossing at *early* time, and the ensuing adiabatic passage transition, would translate into a larger ionization signal than a *late* crossing, where even a significant population transfer to a certain discrete state would not be reflected in the ionization signal.

Below we examine the dynamics of our system with particular interest in states 5p, 6p and 7p. In figure 5, we show the populations and the energies of the five bare states for the chirp  $\varphi_2$  varied between  $-2000$  and  $2000$  fs<sup>2</sup> (from left to right) with the system initiated in state 3s. For illustrative purposes we pick  $\Omega_0 = 0.3$  fs<sup>-1</sup>, corresponding to an intensity of  $3.7 \times 10^{12}$  W cm<sup>-2</sup> [63] and  $\Delta t = 30$  fs.

**4.1.1. Large negative chirp.** For large negative chirp ( $\varphi_2 = -2000$  fs<sup>2</sup>, figure 5(a)) the laser field reaches resonances relative to the  $7p \leftarrow 4s$  (one-photon) transition and the  $4s \leftarrow \leftarrow 3s$  (two-photon) transition in nearly the same instant, thus creating a ‘bow-tie’ level crossing pattern [44], [65]–[69], which is of particular significance because it involves three rather than two states. This crossing results in efficient population transfer to states 4s and 7p and depopulation of state 3s. Because state 7p is populated at such early times, it is exposed to ionization for most of the interaction dynamics and hence has a dominant contribution in the photoelectron signal (see figure 4 at 1.2 eV and  $-2000$  fs<sup>2</sup>).

Later on we observe almost adiabatic evolution and the population is shared mainly between states 4s and 7p in the form of Rabi oscillations with fading amplitude [70]. State 6p acquires only marginal population mainly due to its crossing with state 3s (which is, however, already depleted due to the preceding ‘bow-tie’ crossing) via a three-photon excitation through state 4s. The late crossings between states 3s and 5p, and also between states 4s and 6p are of no importance because they occur after the pulse intensity has essentially vanished. State 5p remains unpopulated since it is far off-resonant throughout the entire dynamics.

**4.1.2. Large positive chirp.** For large positive chirps ( $\varphi_2 = 2000$  fs<sup>2</sup>, figure 5(e)) the energy diagram is mirrored compared with the one for large negative chirps  $\varphi_2$  (figure 5(a)). Then initially the system evolves adiabatically, with minor (off-resonant) population transfer from state 3s to state 4s due to their strong mutual couplings. Around the time of the peak laser intensity, as state 3s sweeps across resonance with 6p, the latter starts to effectively populate through the three-photon 3s–6p crossing. Because this crossing occurs approximately in the middle of the laser pulse the population of state 6p is exposed to ionization for a considerable



**Figure 5.** Populations (lower frames) and energies (middle frames) of the states of interest 5p, 6p and 7p versus time for  $\varphi_2$  varied (from left to right) between  $-2000 \text{ fs}^2$  (down-chirp) and  $2000 \text{ fs}^2$  (up-chirp),  $\Omega_0 = 0.3 \text{ fs}^{-1}$  and  $\Delta t = 30 \text{ fs}$ . In the middle frames, colored and gray lines depict the bare state energies. The latter are related to states 3s and 4s and include the effective chirp, i.e. the chirp of the laser as well as the chirp due to ac Stark shifts. The black lines represent the dressed state energies and the arrows show the population flow. The populations in the lower frames are consistent with the asymmetry in the experimental results presented in figure 4: for large chirps states 6p (positive chirp) and 7p (negative chirp) are predominantly populated, whereas around zero chirp the contribution comes mostly from state 5p. The envelopes (straight lines) and detunings (dashed lines) of the modulated pulses are shown in the uppermost frames. Note that the energies are mirrored when changing the sign of the chirp  $\varphi_2$ .

time interval, which results in significant photoelectron signal from 6p (see figure 4 at 1.0 eV and  $+2000 \text{ fs}^2$ ). For the same reason—the 3s–6p crossing occurring near the laser pulse maximum—the population transfer from state 3s to state 6p is relatively efficient and only about half of the population is left in states 3s and 4s thereafter; then only a part of this already reduced population is transferred to state 7p at the subsequent ‘bow-tie’ crossing 3s–4s–7p. Moreover,

this crossing occurs at late times and hence state 7p is not visible in the photoelectron spectrum. State 5p remains unpopulated once again as it stays far off any resonance.

We now turn our attention to the regimes of a moderately large chirp  $\varphi_2$ , where the photoelectron spectrum changes from a single-state feature to one displaying double features.

*4.1.3. Moderate negative chirp.* For a moderate negative chirp ( $\varphi_2 = -500 \text{ fs}^2$ , figure 5(b)) an early crossing occurs between states 3s, 4s and 7p in the rising edge of the pulse, which leads to a partial population transfer from state 3s to states 4s and 7p, because the laser intensity is not strong enough to enforce adiabatic evolution. The population in state 7p is exposed to ionization for the rest of the pulse, whereas the population in state 4s proceeds until the subsequent 4s–6p crossing where it is partially transferred to state 6p. The leftover temporary flows into state 5p, which starts to emerge in the photoelectron spectrum, and is finally driven back into state 3s. In result, all states 5p, 6p and 7p are visible in the photoelectron signal, which is an indication for the creation of a coherent superposition of these (see figure 4 at about  $-500 \text{ fs}^2$ ).

*4.1.4. Moderate positive chirp.* For moderate positive chirps ( $\varphi_2 = 500 \text{ fs}^2$ , figure 5(d)) state 3s first comes very close to state 5p at times of the laser pulse maximum; during this proximity the population undergoes Rabi-type oscillations between states 3s and 5p and is exposed to ionization from state 5p. The signature of state 5p is clearly visible and indeed this is the regime where this state indisputably dominates in the photoelectron signal (see figure 4 at 0.8 eV and  $+500 \text{ fs}^2$ ). In other words, it is the DSS induced by the two-photon transition  $4s \leftarrow \leftarrow 3s$  which makes the population of the far-off-resonant state 5p possible [71]. If this Stark shift were absent (e.g. if the two-photon transition  $4s \leftarrow \leftarrow 3s$  were instead a single-photon one in a gedanken scenario) state 5p would never receive sizeable population. As we proceed beyond the pulse maximum state 3s crosses state 6p and the population is partially transferred to the latter. Hence state 6p emerges in the photoelectron signal due to the ensuing ionization, whereas state 7p is invisible in this regime because all population left flows into state 4s.

*4.1.5. Zero chirp.* In this regime, the laser pulse is unchirped,  $\varphi_2 = 0$ . Therefore, the *effective* chirp is entirely due to ac Stark shift. The latter is symmetric to the pulse because it is induced by the same pulse. Moreover, because state 3s crosses states 6p and 5p (figure 5(c)), sizeable population will visit these two states through the respective first crossings 3s–5p and 3s–6p. A second pair of crossings in the falling edge of the pulse will induce additional transitions  $5p \leftarrow \leftarrow \leftarrow 3s$  and  $6p \leftarrow \leftarrow \leftarrow 3s$ . The implication is that states 5p and 6p will contribute significantly to the photoelectron signal (see figure 4 around  $\varphi_2 = 0$ ). State 7p, on the other hand, remains well off resonance throughout and receives only a small population due to (weak) non-resonant interaction. Its contribution to the photoelectron signal should be therefore more muted than those from states 5p and 6p.

## 4.2. Discussion

Below we discuss the five excitation regimes in the dressed state (adiabatic) context. When adiabatic, which demands large couplings and low chirp rates for the avoided crossings in question, starting in state 3s we end up in state 7p for  $\varphi_2 < 0$  or in state 6p for  $\varphi_2 > 0$  (figure 5, middle frames; in the latter case a fully non-adiabatic passage across state 5p occurs, since

the pulse intensity is negligible for the 3s–5p resonance). Therefore, clearly from figure 5, our system exhibits a somewhat adiabatic behavior for chirp  $\varphi_2$  away from the origin. As we get closer, the crossings shift toward the pulse wings, whereas the pulse gets narrower in time, which in combination results in breaking adiabaticity. The latter is further hindered by the increased DSS, which effectively enhances the chirp rate.

We expect adiabaticity to remain almost unaffected for large negative values of the chirp  $\varphi_2$ , since the chirp rate  $a \propto 1/\varphi_2$  and  $\Omega \propto 1/\sqrt{\varphi_2}$ , and to break down for large positive values, for it relies on the three-photon transition  $6p \leftarrow \leftarrow \leftarrow 3s$ , which gets weaker, as the resonances relative to 3s–4s and 4s–6p further separate in time. Larger peak intensities  $\Omega_0$  strengthen adiabaticity for the transition  $7p \leftarrow \leftarrow \leftarrow 3s$  and make complete population transfer possible, as also indicated in [34], whereas for the transition  $6p \leftarrow \leftarrow \leftarrow 3s$  due to the unfavorable influence of the increased DSS we predict the contrary.

## 5. Summary and conclusion

In this contribution, we presented a joint experimental and theoretical study on strong-field REMPI of sodium atoms using chirped femtosecond laser pulses. Experimentally, PADs have proven to be the essential tool to identify the different excitation and ionization pathways.

We observed three distinct ionization pathways contributing to the measured PADs. The predominant contribution with combined s- and d-symmetry is due to  $2 + 1 + 1$  REMPI processes involving the strongly driven two-photon transition  $4s \leftarrow \leftarrow 3s$ , and subsequent ionization from the states 5p, 6p and 7p. Photoelectrons with combined d- and g-symmetry originated from  $3 + 1$  REMPI via states 5f and 6f. A weak contribution with combined p- and f-symmetry close to the ionization threshold is attributed to the third channel, that is two-photon ionization of the non-resonant transiently populated state 3p.

Selective population of the highly excited states 5p, 6p, 7p and 5f, 6f was achieved by controlling a single pulse parameter, i.e. the chirp parameter  $\varphi_2$ . In particular, we observed highly selective population of state 7p using strongly down-chirped laser pulses. For strongly up-chirped laser pulses states 6p and 5f were populated with high efficiency and a dominant signal from state 5p was obtained for moderately up-chirped laser pulses. Moreover, in the intermediate chirp regions coherent superpositions of neighboring states have been excited.

Simulations based on numerical integration of the time-dependent Schrödinger equation for a neutral 20-state system are in agreement with our experimental findings. In addition, a five-state model was developed in order to provide insights into the physical mechanisms at play. Our analysis of the time-dependent populations showed that by tuning the chirp parameter distinct physical mechanisms have been addressed, involving adiabatic and non-adiabatic time evolution along with DSSs and (multiple) level crossings. It was pointed out that the occurrence of an uncommon ‘bow-tie’ level crossing is responsible for the excitation of coherent superposition states as observed in the experiment. The strong DSS of the two-photon transition  $4s \leftarrow \leftarrow 3s$  turned out to be of particular significance for populating state 5p being inaccessible in weak laser fields.

Our results highlight the importance of studying model systems experimentally and theoretically to better understand the physical mechanisms of strong-field coherent control. Our findings demonstrate that, in general, in strong-field control multiple pathways involving different physical mechanisms are at play simultaneously.

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## Appendix. Details of calculations

Each p state consists of  $p_{1/2}$  and  $p_{3/2}$  substates, coupled by  $\Omega_{1/2}$  and  $\Omega_{3/2}$ , respectively, to a relevant s state. Therefore initially our system comprises overall 10 states (prior to eliminating state 3p). To simplify our approach we perform a transformation to a dark-bright basis for each of the p states and thus eliminate half of the p substates as dark (uncoupled) states, and keep the rest, which become coupled by the root mean square of the relevant  $\Omega_{1/2}$  and  $\Omega_{3/2}$  and are the ones to be referred to as p states throughout the theoretical part of the paper.

The effective two-photon transition moment between states 3s and 4s is

$$q_{12} = -\frac{d_a d_b + d_c d_d}{2\Delta_{3p}}, \quad (\text{A.1})$$

where  $d_{a,c}$  and  $d_{b,d}$  are the dipole moments for the transitions  $3p_{1/2,3/2} \leftarrow 3s_{1/2}$  and  $4s_{1/2} \leftarrow 3p_{1/2,3/2}$ , respectively.

The effect of a quadratic phase modulation in frequency domain of the form

$$\varphi(\omega) = \frac{\varphi_2}{2}(\omega - \omega_0)^2 \quad (\text{A.2})$$

is described in time domain by a modulated linearly polarized laser electric field  $E(t)$  given as [72]

$$E(t) = 2\text{Re} \{ E^+(t) \}, \quad (\text{A.3})$$

where for the positive-frequency part we have

$$E^+(t) = \frac{E_0}{2\gamma^{1/4}} e^{-(t^2/4\beta\gamma)} e^{i\omega_0 t} e^{i(at^2 - \varepsilon)} \quad (\text{A.4})$$

with

$$\varepsilon = \frac{1}{2} \arctan \frac{\varphi_2}{2\beta},$$

$$\beta = \frac{\Delta t^2}{8 \ln 2},$$

$$\gamma = 1 + \left( \frac{\varphi_2}{2\beta} \right)^2,$$

$$a = \frac{\varphi_2}{8\beta^2\gamma}$$

resulting in the time-dependent instantaneous laser frequency

$$\omega(t) = \omega_0 + 2at. \quad (\text{A.5})$$

Here  $\Delta t$  denotes the FWHM of the intensity  $I(t)$  of the unmodulated pulse,  $\omega_0$  is the laser carrier frequency and  $\varphi_2$  is the chirp parameter to be varied.

We define a reference Rabi-frequency  $\Omega(t) = \Omega_0 f(t)$ , where  $f(t)$  is the laser electric field envelope

$$f(t) = \frac{\exp\left(-\frac{t^2}{4\beta\gamma}\right)}{\gamma^{1/4}}. \quad (\text{A.6})$$

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