Ion-charge-state chronoscopy of cascaded atomic Auger decay

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New Journal of Physics 10 (2008) 025009 (12pp)
Received 29 September 2007
Published 29 February 2008
Online at http://www.njp.org/
doi:10.1088/1367-2630/10/2/025009

Abstract. It has recently been demonstrated that apart from the electron detection realized in the attosecond streak camera, also ion detection can be used for establishing extreme-ultraviolet pump/visible probe experiments, temporally resolving the dynamics of atomic inner-shell relaxation processes. We utilize this method for studying the Auger decay of krypton atoms following the creation of vacancy states in the 3d shell. It is shown that the electronic relaxation occurs through different pathways, each involving cascades of sequential steps which are followed in their native temporal succession.

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1. Introduction

Of the many electrons building the shell of an atom, infrared to ultraviolet radiation readily available from lasers will excite only the weakest bound ones. The majority of electrons remains unaffected and will contribute to the dynamics of the ‘active’ electron only indirectly in terms of a structureless cloud screening the charge of the atomic nucleus. Excitation with shorter wavelength radiation dramatically changes this situation, as deeply bound electrons are also reached and can be ejected from the atom. The created vacancies represent highly unstable states that give rise to a relaxation by rearrangement of a larger number of less bound electrons. The decay may take place in a single step, but usually occurs as a cascade of successive radiative or non-radiative processes with corresponding transient intermediate states. Atomic inner-shell relaxation therefore represents a show-case example of complex multi-particle dynamics and is studied vigorously—mostly utilizing synchrotron radiation for excitation [1]–[6]. While this source reliably delivers pulses of tunable, narrow-band ionizing radiation, the pulse duration of typically a few tens of picoseconds is usually too large for directly following the relevant temporal evolution. Generally, the lifetime decreases with increasing binding energy of an intermediate state. While the final steps in a relaxation cascade imply radiative decay via fluorescence [5] that may take pico-, nano-, or even microseconds to occur, the early steps often involve radiationless Auger decay which takes place in a few femtoseconds or even on an attosecond timescale [7, 8]. In spite of the long duration of synchrotron pulses, temporal information on the femtosecond scale has been extracted by thorough analysis of spectral linewidths [9]. However, apart from practical limitations connected with spectral resolution, spectral complexity, or line-asymmetries introduced by post-collision interaction [10], energy-domain approaches do not provide information on the order of successive processes. In contrast, the correct order of events is usually directly observable in time-domain experiments. Utilizing the pump-probe technique, femtochemistry demonstrates how transient intermediate states of a photochemical reaction can be interrogated with an ionizing probe pulse [11]. Extension of this concept to studies of extremely short-lived intermediates in an inner-shell relaxation cascade calls for adequately short pump and probe pulses that introduce sufficient energy into the system for initiating and probing such dynamics, respectively. In the work described in this paper, sub-femtosecond extreme-ultraviolet pulses from a laser-driven high-harmonic source are used for creating highly excited vacancy states in noble gas atoms. The delivered high-energy photons would also represent suitable probes for sampling the population of the resulting transient states, but unfortunately the outstanding temporal properties of high-harmonic radiation still come...
with a lack of intensity. The corresponding small probability of creating an intermediate state in a sample of gaseous neutral atoms therefore must be compensated by a high probability for detecting it. To this end, we use the intense 800 nm laser fundamental as the probe beam, which ionizes essentially all populated weakly bound intermediate states through multiphoton absorption or tunnelling. This delayed ionizing probe changes the charge state of the ion that is finally detected, thus establishing ion-charge-state chronoscopy as a time-domain method for following the evolution of complex relaxation pathways in the electron shell of atoms. In section 2, we will introduce a few relaxation mechanisms that are of significance for this work. In the following sections, we will briefly introduce the method before presenting new results on the Auger cascade of krypton atoms after excitation of the 3d shell. The results will be discussed in section 4 and compared with work performed in the energy domain. Finally, we compare the capabilities of ion-charge-state chronoscopy with our previous results on the krypton Auger decay obtained with electron spectroscopy utilized in the attosecond streak camera [12].

2. Ion creation by inner-shell relaxation

Generally, the creation of an inner-shell vacancy leaves an atom in an unstable state which will decay through radiative or radiationless transitions. Auger decay is a radiationless de-excitation process resulting in the ejection of one or more electrons. In the case of photon impact with attosecond XUV bursts, the creation of the inner-shell vacancy is treated as much faster than the lifetime of the inner-shell hole. Therefore, it is reasonable to describe the Auger electron ejection in two independent steps. In the first step, a photoelectron from an inner shell is emitted and in the second step the inner-shell vacancy decays upon ejection of one or more electrons, according to equation (1).

\[ \hbar \omega + A \rightarrow A^{++} + e_{\text{photo}}^{-} \rightarrow A^{++} + e_{\text{photo}}^{-} + e_{\text{Auger}}^{-}. \]  

This decay path requires that the binding energy of the electron from the vacancy exceeds the double ionization threshold to initiate the second step. The Auger decay is not necessarily connected with the emission of a photoelectron in the first step. Another possible channel is described by equation (2). The incident photon produces an inner-shell vacancy and lifts the electron to a highly excited Rydberg state without ejection of the electron. The photon energy matches a transition in such a way that the excited neutral atom decays by emission of an Auger electron—and often also of a Rydberg electron. This process is known as ‘resonant’ Auger decay.

\[ \hbar \omega + A \rightarrow A^{+} \rightarrow A^{++} + e_{\text{Rydberg}}^{-} + e_{\text{Auger}}^{-}. \]  

Beyond normal Auger decay, the ionization of deep inner shells can lead to the emission of two or more Auger electrons, if the total energy of the ionized or excited atom exceeds the threshold of higher ionic charge states. The emission of two Auger electrons is either characterized by sequential ((3a), cascade Auger decay) or simultaneous ((3b), double Auger decay) ejection.

\[ A^{+} \rightarrow A^{2+} + e_{\text{Auger1}}^{-} \rightarrow A^{3+} + e_{\text{Auger1}}^{-} + e_{\text{Auger2}}^{-}. \] (3a)

\[ A^{+} \rightarrow A^{3+} + e_{\text{Auger1}}^{-} + e_{\text{Auger2}}^{-}. \] (3b)

In the case of a sequential Auger decay the stepwise model described above is extended by a further step for the emission of the second Auger electron (see equation (3a)). The stepwise decay with ejection of two Auger electrons requires a sufficiently long lived intermediate level as
compared to the lifetime of the inner-shell vacancy. If this level does not exist, the two electrons may be emitted simultaneously and they share the total energy released in filling the vacancy. For the latter process, the measured electrons cover a range from zero kinetic energy to the maximum energy released. Consequently, extraction of dynamical information from spectral linewidths is precluded for double Auger decay by the fact that the kinetic energy of the emitted electron is not clearly determined as discussed in the case of the xenon 4d double Auger decay by Penent et al [8]. Our new technique is sensitive to the creation of the corresponding ions and thus able to resolve Auger lifetimes also in such cases.

3. Ion-charge-state chronoscopy

The method was stimulated by calculations of carrier-envelope-phase (CEP) dependences in the ionization yield of helium [13] and xenon [14]. Similar to electron streaking measurements [15, 16], the ion yield should be sensitive to the CEP of a few-cycle laser pulse. This behaviour should also strongly depend on the intensity of the IR-laser field. Those calculations considered the laser-excitation from atomic ground states and did not consider pumping of the atomic gas target with a synchronized XUV pulse. In this case, the processes taking place become different—usually much more complex—and the detection of the ionization yield with respect to the different ionic states opens up a new way to resolve valence as well as inner-shell dynamics, not directly accessible in the energy domain.

It was demonstrated by Uiberacker et al [7] that measuring the ionization yield of XUV-excited neon in its different ionic states resolves a field-strength dependent behaviour. It was also shown that this new technique provides access to the observation of a cascaded decay of a xenon 4d core-hole and allows determination of the time constant of the double Auger decay in good agreement with measurements performed in the energy domain by Penent et al [8].

Figure 1 shows a schematic drawing of the experimental set-up. A 5 fs laser pulse is focused into a neon filled tube (diameter \(\sim 3\) mm) and interacts with neon atoms inside the tube to produce high-harmonic radiation. The laser pulse and the highly collimated XUV beam emerging from the interaction region within the tube co-propagate collinearly towards a double structured focusing mirror. Differential pumping stages reduce the pressure from \(\sim 1 \times 10^{-2}\) mbar in the high-harmonic generation (HHG) chamber to the range of \(\sim 5 \times 10^{-7}\) mbar in the experimental chamber. In the beamline, the laser and harmonic beams pass through a 150 nm thick, 5 mm diameter zirconium foil placed on a 2 \(\mu\)m thick nitrocellulose pellicle in order to suppress the laser radiation at the central part and the XUV radiation in the annular part of the beam. The zirconium foil acts as a high-pass filter for the high harmonic radiation and suppresses photons with energies below 60 eV. The energy transported by the resulting annular laser beam can be adjusted with a motorized iris not shown in this diagram. The focussing mirror consists of an inner part coated with a Mo/Si multilayer. This mirror acts as a bandpass filter on the harmonic spectrum to pick an isolated pulse of attosecond duration from the cut-off range of the harmonic spectrum as shown in the inset of figure 1 at the lower right with the dashed curve representing the reflectivity of the XUV mirror. The outer part of the double mirror assembly reflects the IR laser pulse. A concentric hole of 5 mm diameter hosts the miniature Mo/Si mirror of slightly smaller diameter. The miniature XUV mirror is mounted on a closed-loop-controlled ultra high precision piezo translation stage (PI P-621.1 CD) allowing alignment and translation with respect to the external part with a reproducibility of better than 15 attoseconds. The total achievable delay range is about 660 fs. Both pulses are
focused into the experimental target with a focal length of 120.5 mm. The ions created in the target volume are measured with a reflectron type time-of-flight ion spectrometer. The inset at the upper left of figure 1 demonstrates the resolution achieved with this arrangement for the example of the XUV ionization of xenon. All ionic charge states are measured simultaneously with isotopic resolution.

The principle of ion-charge-state chronoscopy is shown in figure 2 assuming a laser pulse with an intensity sufficiently low so as not to ionize the bound electron with binding potential \( W_2 \) for \( \tau_D > 0 \) (when the laser pulse arrives before the XUV pulse), thus leaving the atom neutral. For \( \tau_D = 0 \) the XUV pulse meets the neutral atom at the peak intensity of the laser pulse. As in the first case, the laser pulse can be neglected for unexcited electrons because the

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**Figure 1.** Schematic diagram of the experimental set-up. The inset at the lower right shows the generated XUV spectrum and the XUV-mirror reflectivity. The mirror acts as a bandpass filter on the harmonic spectrum and selects an isolated attosecond pulse from the cut-off range of the spectrum. The inset at the upper left shows the charge-state resolution achieved with this arrangement for the case of xenon. The distribution of the ions is in good agreement with the cross-sections for the xenon ion production at 90 eV known from synchrotron radiation studies.
intensity is sufficiently low, but the XUV pulse can act on the atom in different ways, two of which are:

a. A valence electron is photoionized and appears in the continuum with a kinetic energy \( W_{\text{kin}} = \hbar \omega - W_{\text{bind}} \). This leads to a singly ionized atom.

b. An inner-shell electron is ionized, but the photoelectron appears in the continuum with a slightly lower kinetic energy with the deficit \( \Delta W_e = W_{\text{kin}} - \hbar \omega - W_{\text{bind}} \) being used for ‘shaking-up’ an additional electron into a highly excited state. Additionally, the rearrangement of the electron shell is accompanied by a core-level shift \( \Delta W \). The resulting atom is singly ionized and excited. This highly excited state may decay by an Auger transition.

For \( \tau_D > 0 \) the laser pulse meets atoms in an excited ionic state that the XUV pulse has created before.

a. For the laser intensity chosen sufficiently low, so that the bound electrons at \( W_2 \) cannot be ionized by the laser field, no further transition occurs.

b. Once there is an excited electron present at \( W_{\text{ms}} \) the laser intensity is sufficient to ionize it through multiphoton absorption or tunnelling \[7\]. In this case, the ionic charge state is increased by one and the atom is doubly ionized.

c. For sufficiently high XUV photon energies ionization of a core level may be involved, followed by inner-shell relaxation processes like Auger decay. In the considered case an Auger decay is initiated, while the excited electron is ionized due to the laser pulse. A triply charged ion is created.

Figure 2. Illustration of the ionization sequence related to attosecond excitation and IR probing. The intensity of the IR laser pulse is assumed to have an intensity that is too low to act on the bound electron at \( W_2 \). The ionization and excitation of the former neutral atom by the attosecond XUV pulse enables the IR laser pulse to ionize the excited electron \( W_{\text{ms}}^* \). Furthermore, inner-shell processes may be induced as shown in case c for \( \tau_D > 0 \).
For case c, two competing time constants for the process are active. The ionization of the Rydberg electron by the laser pulse results in the growth of the doubly charged ion yield. In a competing effect, an Auger decay inhibits the formation of doubly charged ions and gives rise to triply charged ions. The branching between these two processes depends not only on the laser pulse duration and intensity, but also on the time constant of the observed Auger decay.

In ion-charge-state chronoscopy, we detect the yields of all occurring charges as a function of the delay between an exciting XUV and a probing laser pulse. The competitive depopulation of an intermediate state through an inner atomic Auger decay or laser ionization can be interpreted as probing of the current intermediate state population by the laser field. The branching ratios of the different ion yields depend on the relative timing between both light pulses. An application of this method for following complex relaxation paths in an excited atomic system is discussed in the following section for the case of krypton 3d Auger cascades.

4. Results on the evolution of Auger cascades in krypton and discussion

For the experiment presented here, XUV pulses with a carrier frequency of $\hbar \omega = 91$ eV and a bandwidth of 9 eV corresponding to 250 as duration were used for creating vacancies in the 3d electron shell of atomic krypton. The same pulses were used in a previous ion-charge-state chronoscopy experiment on xenon [7]. In contrast to xenon being excited non-resonantly far above the triple ionization threshold of 64.09 ± 0.04 eV [8] at these photon energies, krypton exhibits both non-resonant as well as resonant Auger decay.

Figure 3 displays the levels and transitions relevant for the interpretation of the pump-probe measurement presented below. For excitation energies within the mirror bandwidth the most probable transitions are indicated by the purple arrows in the diagram. The de-exciting resonant as well as non-resonant Auger transitions are indicated by green arrows. Singly-charged krypton originating from XUV photoionization with a 3d vacancy decays predominantly to doubly charged krypton, but with a fraction of 30% also to triply charged krypton [17]. Highly excited neutral atoms in $3d^{-1}np$ states, produced by photoabsorption, decay via Auger transitions to the states of singly charged ions with configuration $4p^{-2}np$ and via Auger cascades through intermediate states of $4s^{-1}4p^{-1}np$, $4s^24p^{-3}4dnp$ and $4s^{-2}4p^{4}nl$ configurations to doubly charged krypton ions. A fraction of 10% will directly decay to triply charged krypton [22]. For the laser intensity of $4 \times 10^{13}$ W cm$^{-2}$ chosen here (see the discussion at the end of this section), corresponding to Keldysh parameters $\gamma$ [23] of 1–2 (see scale in figure 3), the NIR laser pulse can probe bound levels down to approximately 15 eV below the $4p^{-3}$ ionization threshold. At this intensity, the laser field alone produces only singly ionized krypton. The probing transitions are indicated by red arrows in figure 3.

For the enhanced production of triply charged ions with participation of the IR pulse two competing mechanisms corresponding to resonant and non-resonant Auger decay are included in the diagram. The simpler dynamics is connected with normal Auger decay of the $3d^{-1}$ vacancy to the doubly charged ion. It may populate the intermediate $4s^24p^{-4}d^3$ and $4s^24p^{-3}d^4$ states with the decay constant of 7.9 fs, related to the lifetime of the 3d vacancy, determined previously with the attosecond streak camera [12]. These states of Kr$^{2+}$ are below the triple ionization threshold and comparatively long lived, thus the laser induced depletion of those levels samples the population of the levels caused by the Auger transition.

In the case of the resonant Auger decay an increase of the yield for production of Kr$^{3+}$ ions due to the IR pulse can originate from three different channels. These channels are not
independent, making a complete interpretation difficult. In the first case, a resonant Auger decay populates $4s^{-2}4p^6nl$ intermediate states with approximately the same time constant as for the decay of the $3d^{-1}$ vacancy, given above. These states are depopulated by two competing processes. One is a second step Auger decay to the $4p^{-2}$ ground state, while the other is the depopulation caused by the probe laser pulse. The second channel is related to the probe laser pulse in the beginning of the decay cascade. The resonantly excited $3d^{-1}np$ states may be depleted by the probing laser pulse populating the $3d^{-1}$ states and thus the transition probability of 10% for the direct channel increases to 30% as for the decay of a singly charged ion with a genuine $3d^{-1}$ vacancy. Hence, the transition is followed by an increase of the ionization yield in comparison to the direct transition by 20% and the initiation of the Auger transition is delayed by the interaction with the laser pulse. For the third channel the laser needs to act on the atom twice, initiating the channel for the normal Auger decay described above due to depletion of resonantly excited states $3d^{-1}np$. The depletion of the excited $np$ states in this case will happen in the wings of the laser pulse. Therefore, it is expected that this channel becomes indistinguishable from the normal Auger decay to the intermediate states $4s^24p^{-4}4d^2$ and $4s^24p^{-3}4d$.

Figure 4 shows the experimental result for the delay-dependent Kr$^{3+}$ yield. The shape of the curve is explained in terms of the different channels described above assuming early depletion of the resonantly excited $3d^{-1}np$ states. The rising edge is determined by the decay of the initial vacancy, gradually populating states that are susceptible to the NIR probe pulse. Accordingly, the routes to a triply charged ion can be attributed to an enhancement of the ionization yield by
Figure 4. Kr$^{3+}$ signal versus laser/XUV delay. The indicated time constants result from a fitting procedure using the model described in the supplementary information of [7]. The laser intensity is in the range of $4 \times 10^{13}$ W cm$^{-2}$. $	au_{A1}$ indicates the time constant of the first Auger decay taken from [12] while $	au_{A2}$ gives the time constant determined for the second step Auger cascade.

the following processes, where an $L$ above the arrow indicates the action of the probing NIR pulse:

$$3d^{-1}np \xrightarrow{L} 3d^{-1} \rightarrow 4p^{-3},$$  \hspace{1cm} (4)  

$$3d^{-1} \rightarrow 4s^24p^{-4}4d^2, 4s^24p^{-3}4d \xrightarrow{L} 4p^{-3},$$  \hspace{1cm} (5)  

$$3d^{-1}np \rightarrow 4s^{-2}4p^6nl \xrightarrow{L} 4p^{-3}.$$  \hspace{1cm} (6)  

Referring to region I in figure 4, for negative delays, i.e. the laser pulse arriving prior to an inner-shell excitation, the relaxation paths of the 3d$^{-1}$ and 3d$^{-1}np$ states are not affected by the probe laser pulse and the ion yield remains unaffected as compared to purely XUV-generated Kr$^{3+}$ ions. This indicates at the same time that the laser-induced generation of singly charged krypton is not saturated—otherwise XUV ionization of singly ionized krypton would become favourable for negative delays and an enhancement of the ionization yield would be observed in this delay range, as well.

For large positive delays (region IV) the enhancement of the ionization yield follows the transition described by equation (5) since the 4s$^24p^{-4}4d^2$ state is long lived compared to the maximum delay range of the experimental set-up.

Region II of figure 4 corresponds to the time-dependent population of the intermediate states 4s$^24p^{-4}4d^2, 4s^24p^{-3}4d$ following the decay of the 3d vacancy given by equation (5). With increasing delay the decay of the resonantly excited 3d$^{-1}np$ states into 4s$^24p^6nl$ states becomes favourable. The early depletion of the 3d$^{-1}np$ to 3d$^{-1}$ indicated by the red arrow in
Figure 5. Intensity-dependent yield of ionization to Kr$^{3+}$ ions for two laser/XUV delays: red circles indicate the intensity-dependent behaviour at +20 fs delay; black squares the intensity-dependent behaviour at −20 fs delay; blue triangles depict the laser–induced formation of Kr$^{3+}$ ions with the dashed vertical line indicating the threshold for their formation. In the upper graph the ratio Kr$^{3+}$ (+20 fs)/Kr$^{3+}$ (−20 fs) is plotted. From the ratio of ion counts from Kr$^{+}$ (not shown here) to Kr$^{2+}$ the laser intensity was deduced. The grey box indicates the intensity range utilized for the analysis of the Kr MNN Auger decay cascades.

Figure 3 is only dominant for delays less than the time constant $\tau_{A1} = 7.9$ fs for the Auger decay of the 3d vacancy.

Furthermore the population of 4s$^{-2}$4p$^6$nl states is characterized by a higher transition probability from the 3d$^{-1}$np than a transition to 4s$^2$4p$^{-4}$4d$^2$ and 4s$^2$4p$^{-3}$4d states from the 3d$^{-1}$ state. This explains the pronounced peak in the measured ion population compared to region IV of figure 4.

The decaying part of the peak in region III is best explained by competition of the following cascades:

\[ 3d^{-1}np \rightarrow 4s^{-2}4p^6nl \xrightarrow{L} 4p^{-3}, \]  
\[ 3d^{-1}np \rightarrow 4s^{-2}4p^6nl \rightarrow 4p^{-2}. \]  

With increasing delay, the depopulation of the intermediate state 4s$^{-2}$4p$^6$nl to Kr$^{2+}$ dominates the laser-induced depopulation to Kr$^{3+}$. This requires that the 3d$^{-1}$np is not depleted by the laser for delays > 15 fs because a substantial fraction needs to decay to the intermediate state 4s$^{-2}$4p$^6$nl to yield the observed depopulation.
In order to extract the relevant time constants from the observed data, we used the fitting procedure already applied for the analysis of the time-dependent ion-charge states of xenon (for details see supplementary information of [7]). According to the same conditions for the generation of the laser pulse as in [7], an identical 4.3 fs duration for the laser probe enters in the fit. For the time constant of the MNN Auger decay of the Kr 3d−1 core hole we used the value of $\tau_{A1} = 7.9$ fs from [12]. With these parameters as input, the fit to the trailing edge of the measured curve yields a decay constant of $\tau_{A2} = 20 \pm 4$ fs for the second step Auger decay $4s^{-2}4p^{6}nl \rightarrow 4p^{-2}$, corresponding to the lifetime of the intermediate state involved in this decay cascade.

The nonlinear nature of laser probing by multiphoton or tunnel ionization generally complicates the analysis and interpretation of the data, but also qualifies the laser intensity as a variable parameter for testing hypotheses on the relevant decay mechanisms. Such intensity dependence shall be discussed in the following. For laser intensities higher than $4 \times 10^{13}$ W cm$^{-2}$, used in the above study, an additional channel is expected to open for a population transfer from the $4s^{-1}4p^{-1}np$ states to the $4p^{-3}$ ground state. For large positive delays the increased ionization yield should change in magnitude depending on whether the $4s^{-1}4p^{-1}np$ states are depleted or not; the behaviour of the graphs in figure 5 supports this statement. An intensity-dependent change in the ionization yield due to the accessibility of more strongly bound excited states for increased intensities can be observed. At laser intensities that already create a substantial amount of Kr$^{2+}$ ions even without pumping with XUV photons, the ratio Kr$^{3+}(+20$ fs)/Kr$^{3+}(-20$ fs) becomes larger which could be an indication of the $4s^{-1}4p^{-1}np$ states getting depleted by the laser pulse as well.

5. Conclusion

Information on the dynamics of inner-shell relaxation processes, as obtained in this work and in [7], is also delivered by the attosecond streak camera concept, based on the spectral analysis of photoemitted electrons energetically streaked by a probing laser field [15, 16], [24]–[29]. The case of the Kr MNN Auger decay discussed in this paper has previously been studied with electron streaking [12], thus suggesting an assessment of the peculiarities and capabilities of both methods. Electron streaking probes electrons at the instant of their transition into the continuum. Probe field intensities are usually kept well below $10^{13}$ W cm$^{-2}$ in order to avoid an excessive background of electrons emitted by above-threshold ionization (ATI). Consequently, it is assumed that the laser field does not influence the dynamics and one observes intrinsic inner-shell electron dynamics terminated by an autonomous ionization process. Electron detection facilitates analysis of their kinetic energy as well as their angular distribution [30, 31], thus providing high state specificity and information depth. Ion-charge-state chronoscopy, on the other hand, does not deliver direct information about the involved electronic transitions; the interpretation of the observed temporal evolution therefore must be made with an advanced model of the studied system’s electronic structure in mind. As the considered higher charge states are not easily produced by ATI, their chronoscopy can usually be performed at light intensities exceeding $10^{13}$ W cm$^{-2}$. The laser field probes the transient population of bound excited states, thus allowing the observation of intermediate steps of a relaxation cascade and providing access to relaxation pathways that do not result in ionization. The involved high light intensities require some care in the interpretation, because the probe field may also actively participate in the electron dynamics or at least modify the properties of some relevant states,
especially loosely bound Rydberg states. Rigorous studies of the intensity dependence of the observed dynamics will aid in detecting such effects.

Acknowledgments

We thank Marc Vrakking and Ferenc Krausz for fruitful discussions. We are grateful for financial support from the Volkswagenstiftung (Germany), the Marie Curie Research Training Network XTRA, a Marie Curie Intra-European Fellowship, and by the cluster of excellence Munich Centre for Advanced Photonics. MFK acknowledges support by the Emmy-Noether program of the German Science Foundation. NMK acknowledges support by the Russian Foundation for Fundamental Researches via grant 06-02-16289.

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New Journal of Physics 10 (2008) 025009 (http://www.njp.org/)