Wavelength dependence of sub-laser-cycle few-electron dynamics in strong-field multiple ionization

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Abstract. Recoil-ion momentum distributions for double and triple ionization of Ne and Ar, as well as for double ionization of N\textsubscript{2} molecule by intense (0.3–0.5 PW cm\textsuperscript{-2}), short (\sim 35–40 fs) laser pulses have been recorded in a so far unexplored long laser-wavelength regime at 1300 nm. Compared to earlier results at 800 nm, the direct (e, ne) ionization pathway during recollision is strongly enhanced manifesting itself in a pronounced double-hump structure in the longitudinal ion momentum spectra not only for Ne, but also surprisingly distinct for Ar and, found for the first time, for molecules. Observed wavelength dependence of the sub-laser-cycle correlated few-electron dynamics might be of paramount importance for possible future applications in attosecond science, in particular, for imaging of ultrafast molecular processes via recollision-induced fragmentation.
1. Introduction

The possibility to control electronic motion in atoms and molecules with intense short-pulsed laser radiation has recently opened an entirely new domain of attosecond science [1], which covers such important applications as generation of attosecond light pulses [2] and ultrafast molecular imaging [3] or tomography (see e.g. [4]). The main physical concept behind this rapidly emerging area of research is a fascinatingly simple recollision scenario [5, 6], where an electron released from an atom via tunnelling, is driven back by the oscillating laser field and interacts with its parent ion, leading to the production of energetic photons and electrons (high-order harmonic generation and above-threshold ionization), or to the ejection of one or more further electrons. Whereas the former two classes of phenomena are essentially one electron processes, and can be reasonably well described within the so-called ‘single active electron’ (SAE) approximation, the latter, known as ‘non-sequential’ double or multiple ionization (NSDI, NSMI) [7, 8], necessarily involves dynamical correlation between individual electrons. Being the most simple and fundamental correlated strong-field reactions beyond SAE-dominated processes and thus representing the transition towards more complicated, driven correlated electron dynamics occurring in macroscopic systems where many-body interactions are crucially important, NSDI/NSMI attract continually increasing interest but, at the same time, remain a severe challenge for the theoretical description.

Signatures of NSMI were originally observed in the measurements of total ionization cross-sections, where it was discovered that in certain laser intensity regimes yields of doubly and multiply charged ions created by intense linearly polarized laser pulses exceed by many orders of magnitude those expected from SAE models assuming the sequential removal of two or more independent electrons [9]. This enhancement manifested itself in a characteristic ‘knee’ structure in the intensity-dependent ion yield curves, but its physical origin remained controversial until 2000, when the first differential data (recoil-ion momentum distributions [10, 11]) along with the suppression of the effect for elliptically polarized light [12, 13] provided convincing evidence in favour of the recollision model. Since then, due to the development of novel elaborate few-particle imaging techniques [14]–[16] in combination with the rapid advance of femtosecond laser technology [17], tremendous experimental progress has been achieved in studies on NSDI/NSMI, culminating in fully differential data for the prototype two-electron reaction, NSDI of He, that have become available recently [18].
The present work now aims at exploring the wavelength dependence of NSDI and NSMI using the latest detection technologies in order to draw conclusions on the sub-cycle, attosecond correlated electron dynamics underlying NSMI. For several reasons outlined in detail below we focus our interest on longer wavelengths where, up to now, NSMI has not been investigated to the best of our knowledge, with the goal to realize a cleaner recollision scenario which is the heart of attosecond science.

First measurements on NSMI \[9\] were performed with 1064 nm radiation for Xe and Kr targets, and this remains the longest wavelength at which this phenomenon has ever been studied\(^4\). Later experiments typically used shorter wavelength lying in the optical and near infrared domain. The vast majority of recent differential data mentioned above were obtained employing Ti:Sa lasers with a wavelength close to 800 nm (see \[8\] and references therein). Apart from possible resonant effects (see e.g. \[20, 21\]), probably the most important result obtained in the experiments on NSDI/NSMI performed with different wavelength, is the disappearance of this phenomena in the UV domain, as was concluded from the absence of the ‘knee’ structure in the He\(^{2+}\) yield measurement at 247 nm \[22\]. The latter observation can be easily interpreted in terms of the recollision model. In this scenario the maximum kinetic energy of the returning electron is given by \(3.17U_p\), where \(U_p = F^2_{\text{max}}/4\omega^2\) is the so-called ponderomotive potential, i.e. the cycle-averaged quiver energy of a free electron in an oscillating laser field of strength \(F\) and frequency \(\omega\), which is proportional to the squared wavelength. For 247 nm, where this quantity is about ten times smaller than that for 800 nm at a given intensity, a contribution from the recollision-based NS ionization mechanism is strongly suppressed. This, however, directly suggests that recollision-induced phenomena would be even more clearly observed at longer wavelengths.

Another reason to study recollision processes further in the infrared domain is the possibility to enter the ‘true’ tunnelling regime. Indeed, the recollision scenario is based on a semiclassical three-step model, where an electron first tunnels from the atom, is then accelerated by the laser field, and comes back when the latter changes its sign, interacting with its parent ion \[5\]. The first step, here, assumes tunnelling ionization at a certain (well-defined) phase of the field. The criterion for ionization to be considered as tunnelling is given by the condition \(\gamma \ll 1\), where \(\gamma = \sqrt{I_p/2U_p}\) (\(I_p\): ionization potential) is the so-called Keldysh, or adiabaticity parameter \[23\]. However, for typical experimental conditions at optical frequencies (intensity of \(\sim 10^{14} - 10^{15}\) W cm\(^{-2}\), ionization potential \(\sim 10 - 20\) eV) this parameter is close to unity, and further increase of intensity leads to the onset of barrier-suppression ionization (BSI) \[24\]. Thus, the semiclassical recollision model should be better applicable at longer wavelength, where tunnelling conditions can be fulfilled before entering the BSI regime.

Another reason to investigate NSMI at smaller frequencies is connected with the intriguing issue of the timing of the recollision-induced scattering event. For a given ionization phase the time when a recolliding electron returns to the nucleus is defined by the laser period, being typically \(2/3\) of the latter (i.e. \(\sim 2\) fs at 800 nm) for the trajectories leading to the largest electron energies. The exciting possibility to modulate this time by changing the laser wavelength was recently exploited for ultrafast molecular imaging \[19\]. On the other hand, there might be a considerable time delay between the instant of recollision and the time when all the emitted electrons are finally set free, especially if excited states which are not immediately ionized by the

\(^4\) Some contribution of NSDI might have been observed in recent experiments \[19\] on D\(_2\) fragmentation at 1200–1850 nm though the authors claim that the fragments originate from recollision-induced dissociation.
The paper is organized as follows: in section 2, a brief overview of recollision kinematics and relevant multiple ionization mechanisms is provided, followed by a description of the experimental set-up in section 3. Results of this study are presented and discussed in section 4. Finally, conclusions and outlook are given in section 5. Atomic units are used throughout the paper if not stated otherwise.

2. ‘Attosecond streaking’ and mechanisms of strong-field ionization

Though NSMI was originally discovered via the observation of intensity-dependent ion yields, such measurements alone do not allow one to identify particular ionization pathways. Much more information can be extracted from differential data. In particular, recoil-ion momentum distributions along the (linear) laser polarization direction (which we will refer to as ‘longitudinal’) have proven to be extremely useful. The identification of different mechanisms of strong-field multiple ionization via analysis of the measured ion momenta is based on momentum conservation, and on the so-called attosecond-streaking principle, which provides information about the phase of the laser field at the instance of ionization, i.e. on the ionization time within the laser cycle [10, 11, 30]. If a particle of charge $n$ is set into an oscillating laser field of frequency $\omega$ and strength $F(t) = F_0(t)\sin(\omega t)$ at a time $t_0$, it acquires the drift momentum $P_\parallel(t_0) = (n/\omega)F_0(t_0)\cos(\omega t_0)$ [31]. Thus, if an electron is born near the maximum of the electric field cycle, which is the case for tunnelling ionization or sequential multiple ionization (SMI), its drift momentum is negligible. In contrast, the largest $P_\parallel^{\text{max}} = 2\sqrt{U_p}$ can be gained if an electron is emitted exactly at the zero crossing of the field, which is close to the time where the most energetic recollision occurs. [10, 11, 26–31]. Therefore, recollision-induced double and multiple ionization produces electrons and, thus, ions with large longitudinal momenta (since the momentum carried by the photons is negligible, recoil-ion momentum balances the momenta of all electrons) leading to characteristic double-hump structures in the ion momentum distributions. The maximum drift momentum which can be gained by an $n$-fold charged ion then is $P_\parallel^{\text{max}}(n) = 2n\sqrt{U_p}$ and, as shown in [31], due to the typical recollision kinematics this value gives an upper classical limit for the most probable ion momenta in the final state.
Correspondingly, for any combination of sequential and NS channels the width of the ion momentum distribution, as well as the positions of the classical cut-offs are defined mainly by the total number of electrons set free near the zero crossing of the field, i.e. participating in the recollision event [31]. However, the detailed shape of the spectrum depends on the sub-laser cycle dynamics induced by the recolliding electron. For instance, in the case of double ionization the final-state momentum distribution will depend on whether the second electron is emitted directly upon rescattering in an ‘instantaneous’ (e, 2e) event, or set free after a certain time delay. The latter situation can be realized if the recollision populates some excited states of the ion which are later ionized by the laser field [25]–[28]. Differences in the relative importance of this mechanism, known as recollision–excitation with subsequent field ionization (RESI), were shown to be responsible for strong atomic structure dependences of longitudinal momentum distributions observed in NSDI at 800 nm [26]–[28]. The RESI mechanism as it was originally suggested [25] implies that the second ionization step occurs close to the field maximum following the recollision, thus, producing ions with small longitudinal momenta and ‘filling the valley’ between two peaks from direct (e, 2e) ionization. However, it should be noted that depending on the particular excited states involved and, especially, on the intensity regime considered, excited states might be ionized earlier, varying the time delay between recollision and ionization within the laser cycle and, thus, modifying the structures in the momentum distributions.

3. Experiment

Measurements were performed using a reaction microscope [15, 16] described in detail in a previous publication [32]. A sketch of the whole experimental set-up is shown in figure 1. Linearly polarized 1300 nm radiation was generated by an optical parametric amplification (OPA) system (TOPAS-C, Light Conversion) pumped by 30 fs, 800 nm pulses from a Ti:Sa laser (KM-Labs system with 10 kHz repetition rate, amplified pulse energy up to 0.7 mJ). The estimated pulse duration at 1300 nm was \(\sim35\text{–}40\text{fs}\), and the pulse energy ranged up to 90 \(\mu\text{J}\). The OPA beam was focused to a spot size of \(\sim10\ \mu\text{m}\) onto the collimated supersonic gas jet in the ultra-high vacuum chamber (2 \(\times\) \(10^{-11}\) mbar) of the reaction microscope. Created ions and electrons were guided to two position-sensitive channel plate detectors by weak electric (2 V cm\(^{-1}\)) and magnetic (\(\sim10\) G) fields applied along the laser polarization axis.
time-of-flight and position on the detector the full momentum vectors of the recoil ions and electrons were calculated.

The momentum resolution along the laser polarization (longitudinal) axis was $\Delta P_\parallel < 0.02$ au for both ions and electrons. Along the transverse directions (i.e. in the plane perpendicular to the laser polarization) the resolution for ions differed from $\sim 0.7$ au along the gas jet direction to less than $0.1$ au in the direction perpendicular to the jet, being limited by the internal jet temperature in the former case, and by the resolution of the imaging system in the latter. Absolute calibration of the laser intensity has been performed using a clear kink in the measured longitudinal momentum distribution of the singly charged Ne ions (and, thus, in the photoelectron spectra, since for single ionization momentum distributions of ions and electrons are just mirror images of each other because of momentum conservation), which corresponds to the maximum drift momentum of $2\sqrt{U_p}$ that an electron can gain in the laser field ([26, 33], see also the discussion in section 2). An alternative calibration method with a cut-off for maximum electron kinetic energy at $10U_p$ [34] yielded results which agreed with those obtained by the first method within 20%.

4. Results and discussion

4.1. Double ionization of Ne and Ar

Figure 2 displays two-dimensional (2D) momentum distributions for double ionization of Ar and Ne at 1300 nm. The data are integrated over the momentum component parallel to the gas jet direction. The spectra exhibit two clear maxima at non-zero longitudinal (parallel to the laser polarization direction) momenta for both targets. The most noticeable feature, which can be seen in this figure, is a pronounced two-maxima structure in the Ar data, which has never been observed so clearly at 800 nm (compare figures 2(a)–(c) with, e.g., figure 2 of [35]). This is further illustrated in figure 3, where longitudinal momentum distributions, i.e. the projections of the data of figure 2 onto the horizontal axes, are shown in comparison with the data obtained at 800 nm at the same intensities. From now on, we will concentrate on the longitudinal momenta since, as discussed in section 2, they have proven to contain most valuable information on strong-field ionization dynamics. Transverse momentum components for the data of figure 2 exhibit structureless Gaussian-like shapes similar to those reported in [10, 11, 28, 35] for 800 nm.

The data for the longer wavelength display a clear target dependence, with the characteristic double-hump structure being much more pronounced for Ne, similar to earlier findings for 800 nm [26]. The 1300 nm spectra in figure 3 are considerably broader than those obtained at 800 nm, and exhibit a deeper minimum at zero momenta (this difference is especially clear for Ar). The discussion in section 2 suggests that the difference in width might originate not only from different ionization mechanisms being at work, but also from the difference in the ponderomotive potential, and thus, in the drift momentum acquired by the doubly-charged ion. Plotting the data in units of $\sqrt{U_p}$ in order to account for the latter effect (figure 4), we indeed observe very similar widths of the distributions for both wavelengths, with differences not exceeding the uncertainties caused by the precision of our intensity and, correspondingly, $U_p$ determination. The spectra are consistent with the classical cut-offs at $4\sqrt{U_p}$ expected for (e, 2e)-like processes lying within these limits for Ar and extending slightly beyond for Ne. This indicates that in general, similar double ionization mechanisms are active for both wavelengths.
Figure 2. 2D recoil-ion momentum distributions for double ionization of Ar (a)–(c) and Ne (d) at 1300 nm. $P_\parallel$ and $P_\perp$ denote the momentum components parallel to the laser polarization (‘longitudinal’) and to the laser propagation (‘transverse’) directions, respectively. The data are integrated over the third momentum component. Intensities in PW cm$^{-2}$ are given in the figure.

However, pronounced differences become obvious at small longitudinal momenta, close to zero, where the valley in between the two maxima is more significant for the longer wavelength. From an earlier analysis of the 800 nm data for different rare gases it was concluded that RESI is responsible for the formation of this part of the spectra [25]–[27]. Thus, it is natural to attribute the differences observed at very small momenta, to the reduction of the contribution from this mechanism at 1300 nm. In order to clarify this issue, we have extended...
Figure 3. Longitudinal momentum distributions for double ionization of Ar (a)–(c) and Ne (d) at 1300 nm (solid lines with shaded areas under the curves) and 800 nm (open circles). Intensities in PW cm\(^{-2}\) are given in the figure.

our earlier analysis of the relative importance of RESI versus the direct recollision-induced (e, 2e) channel to the conditions of the present experiment.

4.2. The semiclassical model of recollision-induced ionization and excitation

For this analysis, we used a slightly modified semiclassical model suggested in [26]. Briefly, in order to calculate the relative contributions of direct (e, 2e) ionization and RESI, we apply the standard stepwise scheme within the recollision scenario [5]. In a first step, one electron is field-ionized via tunnelling at a phase \(\omega t_0\) of the oscillating electric field \(F(t) = F_0 \sin(\omega t)\). Depending on the initial phase the electron may be driven back to its parent ion at a phase \(\omega t_1\) with a classical recollision energy \(E_{\text{rec}} = 0.5 F_0^2 \omega^2 \left[\cos(\omega t_0) - \cos(\omega t_1)\right]^2\). Here, in a second step, electron impact ionization or excitation takes place. In the first case, the second electron is directly ionized in an (e, 2e) process, whereas in the second the recollision-induced excitation is followed by subsequent tunnelling (field) ionization of the excited electron (RESI). In the intensity range considered here the latter ionization step occurs with almost 100% probability. Thus, for the calculation of the relative contribution of these two mechanisms we only need to calculate the ratio of the effective (phase averaged) probabilities for ionization, \(W_{\text{ion}}\), and
excitation, $W_{\text{exc}}$:

$$R = \frac{W_{\text{ion}}}{W_{\text{exc}}}, \quad W_{\text{ion,exc}} = \int_{\pi/2}^{\pi} W_{\text{ADK}}(\omega t_0) P_{\text{ion,exc}}(\omega t_0) \, d(\omega t_0). \quad (1)$$

Here, $W_{\text{ADK}}(\omega t_0)$ is the ADK [36] static-field tunnelling rate. The term $P_{\text{ion,exc}}$ in equation (1) denotes the probability of a direct (e, 2e) ionization or (e, e)$^*$ excitation with subsequent field ionization. This term can be written as

$$P_{\text{ion,exc}} = \sigma_{\text{ion,exc}}(E_{\text{rec}}(t_0)) \frac{1}{\pi[(R(t_1 - t_0))^2]}, \quad (2)$$

where $\sigma_{\text{ion,exc}}$ are the electron impact ionization/excitation cross-sections and $\pi[(R(t_1 - t_0))^2]$ is a rough estimate for an effective cross-section characterizing the transverse spread of the returning electron wave packet at the time $t_1$ of its recollision with the parent ion. This can be estimated using the mean value of $p_\perp$ given by the width of a Gaussian distribution (last term of equation (10) of [36]) as $\bar{p}_\perp = \frac{1}{2} \sqrt{F_0/8T_p}$. This yields the following expressions for the characteristic width (radius) of the electron wave packet as a function of the release time and its
cross-section:

\[ R(t_1 - t_0) = \frac{1}{m_e} \vec{p}_\perp(t_1 - t_0) = \frac{1}{2} \sqrt{\frac{F_0}{8I_p}} (t_1 - t_0), \]

(3)

\[ \pi [R(t_1 - t_0)]^2 = \frac{1}{4} \pi \frac{F_0}{\sqrt{8I_p}} (t_1 - t_0)^2. \]

(4)

Note that in the latter formula atomic units with \( m_e = 1 \) au have been used. Then, the integral (1) for \( W_{\text{ion,exc}} \) can be written as,

\[ W_{\text{ion,exc}} = \pi \int_{\pi/2}^{\pi} 2W_{\text{ADR}}(\omega t_0)\sigma_{\text{ion,exc}}(E_{\text{rec}}(\omega t_0)) \frac{8\omega^2 \sqrt{2I_p}}{\pi(\omega t_1 - \omega t_0)^2 F_0} d(\omega t_0). \]

(5)

The calculated ionization/excitation cross-sections \( \sigma_{\text{ion,exc}} \) were derived using general, simplified (field-free) expressions as described in detail in [26]. For the ionization we adopted the formula derived in [37], and for the excitation cross-sections the van Regemorter formula [38] was used considering the sum over all excited levels of the respective ion. In order to take into account the barrier suppression due to a finite field at the recollision time \( t_1 \) we introduce the suppressed ionization potential of the ion [39] \( I_p^\ast = I_p - 2\sqrt{2F(t_1)}. \)

The calculated ionization/excitation cross-sections as a function of the electron impact energy in units of \( I_p \) are shown in figures 5(a) for Ne and (b) for Ar, whereas the reconstructed intensity-dependent ratios of the contributions from both channels are presented in figure 5(c) for 1300 and 800 nm for both atomic species. From these results it is apparent that the RESI contribution is much weaker at 1300 nm, and thus, its suppression seems indeed to be responsible for the deeper minimum at zero longitudinal momentum observed in figures 3 and 4 for this wavelength. From figure 5 one can also see that the RESI contribution is expected to be weaker for Ne than for Ar at both 1300 and 800 nm, in good agreement with the data of figures 3 and 4.

Having calculated the ratios of the contributions from both channels as discussed above, we can now separate them in the total longitudinal momentum distributions applying a fitting procedure. In order to do this, we use the sum of two functions representing the contributions from direct (e, 2e) ionization and RESI, respectively. We assume that the distribution for the former channel is given by two Gaussians centred at some non-zero momenta (symmetric with respect to zero), and that for the latter it is a Gaussian peaked at zero momentum. Then we set the ratio of the areas under the respective curves to the ratio of ionization/excitation probabilities calculated in figure 5(c) and perform a fit with the widths of the Gaussians and the position of the maxima for the direct (e, 2e) contribution as free parameters. As shown in figure 6, this simple model allows one to reproduce the experimental data astonishingly well and to obtain separated curves for both considered channels. Even though this procedure is not unique in the sense that the data can be fitted using a similar pair of curves also for somewhat different ionization/excitation ratios resulting in different fit parameters, it provides important qualitative insight into the spectrum formation mechanisms, and allows one to estimate the maxima positions as well as classical cut-offs for the pure recollision-induced (e, 2e) reaction yielding results similar to those obtained in [25] by a combined analysis of ion momentum distributions and correlated two-electron momenta.
4.3. Double ionization of $N_2$ molecules at 1300 nm

NSDI has been observed not only for atomic but also for some molecular systems like e.g. $H_2(D_2)$ [19], [40]–[43], $N_2$ [44]–[47] and $O_2$ [45, 46]. Typical features known from atomic targets have been observed as well for molecules, in particular, enhanced ion yields of doubly charged molecular ions in certain intensity regimes [44, 45], a strong polarization dependence [40, 43, 45], signatures of electron–electron correlation in coincident electron spectra [46, 47] etc. However, a characteristic double-hump structure in the longitudinal ion momentum distributions has never been seen for molecules. In experiments on $N_2$ and $O_2$ (performed at 800 nm) distributions of doubly-charged molecular ions were found to have a maximum at zero momentum [46], and this was also the case for the centre-of-mass distribution of two protons emerging from recollision-induced double ionization of $H_2$ reported in [43]. Even though in the later work it was argued that the absence of the double-hump structure might be the signature of the dominance of RESI, attention was mainly concentrated on specific molecular effects like the role of orbital symmetries [46], influence of the alignment [47] etc, which make an identification of different recollision mechanisms much more complicated.
Figure 6. Separation of the contributions from different recollision-induced pathways of double ionization at 0.4 PW cm$^{-2}$ in the longitudinal momentum distributions. Open triangles: experimental data for Ar ((a) and (c)) and Ne ((b) and (d)) at 1300 nm ((a) and (b)) and 800 nm ((c) and (d)). Red solid lines: total fits of the experimental data. Blue lines: contributions from the direct (e, 2e) ionization. Green lines: RESI contributions. Calculated ratios $R$ of both contributions are given in the figure.

In figure 7, we present the longitudinal momentum distribution of $N_2^{2+}$ molecular ions measured with 1300 nm light. Here, a characteristic double-hump shape of the spectrum can be observed, to some extent resembling the Ar data in figures 3 and 4. Taking into consideration an overall similarity between NSDI results on $N_2$ and Ar observed at 800 nm (for these two systems with essentially identical ionization potentials, similar ion yields [45] as well as two-electron correlation patterns [46] have been measured), one is tempted to attribute the appearance of the minimum at zero $P_\parallel$ to the suppression of the RESI contribution at longer wavelength. Even though one certainly needs more quantitative analysis (e.g. similar to that presented above for Ar and Ne) in order to support this statement, having in mind numerous possible effects of the molecular structure, the enhancement of direct recollision-induced ionization might have important consequences for molecular imaging [3, 19]. It should be noted that in our measurement the molecules were randomly oriented, and it was shown in [47] that for $N_2$ NSDI dynamics strongly depend on the molecular orientation.

4.4. Triple ionization of Ar and Ne

After obtaining a consistent qualitative picture of NSDI at 1300 nm, it would be interesting to extend this study beyond two-electron ejection. Detailed studies on higher charge states often
Figure 7. Longitudinal momentum distribution for $\text{N}_2^{2+}$ ions at 1300 nm, 0.4 PW cm$^{-2}$. Arrows show the classical cut-offs of $4\sqrt{U_p}$.

Figure 8. Longitudinal momentum distributions of triply (solid lines) and doubly (open circles) charged Ar (a) and Ne (b) ions for 1300 nm, 0.4 PW cm$^{-2}$. The data are plotted in units of $\sqrt{U_p}$. Dashed vertical lines indicate the classical cut-offs of 2, 4 and $6\sqrt{U_p}$.

represent an experimental challenge due to very low reaction rates, especially at low intensities. Even though the maximum peak intensity which we could achieve with our OPA system did not exceed 0.5 PW cm$^{-2}$, its high repetition rate of 10 kHz allowed us to perform differential measurements on triple ionization of both Ar and Ne.

Figures 8(a) and (b) display longitudinal momentum distributions of triply charged Ar and Ne ions (solid lines), respectively, measured with 1300 nm light at 0.4 PW cm$^{-2}$. In both cases, the distribution of doubly charged ions (open circles) is shown for comparison. If plotted in units of $\sqrt{U_p}$, the Ne$^{3+}$ spectrum is essentially identical to the distributions reported in

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[11, 27, 28] for Ne triple ionization at 800 nm up to 2 PW cm\(^{-2}\). The spectrum extends slightly beyond the classical cut-off of \(6\sqrt{U_p}\), with the maxima at \(4\sqrt{U_p}\) and almost no ions produced with zero longitudinal momentum are observed, indicating that the dominating triple ionization pathway is a recollision-induced (e, 3e) reaction.

The Ar\(^{3+}\) distribution instead is considerably narrower, with the maxima close to \(2\sqrt{U_p}\), and is very different from those reported for 800 nm [27, 28]. As in the case of double ionization, the difference can be observed mainly in the region of low \(P_{\parallel}\): whereas 800 nm data exhibit either a maximum or a very shallow dip at zero, the results for 1300 nm show a pronounced minimum with a clear double-hump structure. Again, the most likely reason for this is a suppression of the RESI-like mechanism, which, however, becomes much more complicated for triple ionization due to the increased number of available reaction pathways and the possibility of multiple excitations. However, there is another important change with respect to the results obtained at shorter wavelength. Comparing momentum distributions from triple and double ionization (open circles in figure 8), we can see for both Ar and Ne, that the spectra for the respective higher charge state are clearly broader, and the maxima are shifted towards larger momenta. At 800 nm this was the case only for Ne, whereas for Ar double and triple ionization very similar distributions were produced (compare figure 8(a) with figure 3 of [27]). The most likely interpretation for this behaviour is that one electron less is involved in the recollision at 800 nm, being emitted independently via tunnelling. In contrast, at 1300 nm at 0.4 PW cm\(^{-2}\) all three electrons seem to participate in the scattering event within the dominant triple ionization channel. In order to trace the reasons for this behaviour, one needs to estimate the probabilities for impact ionization and excitation involving three electrons in the energy range relevant for each wavelength which is beyond the scope of this work.

5. Conclusions and outlook

In this paper, we have presented the first experimental data on NS double and triple ionization of Ne and Ar at 1300 nm. Analysing recoil-ion momentum distributions, we have extracted information about dominant ionization pathways. We observe significant changes in the reaction dynamics compared to the NSDI/NSMI at 800 nm, which has been extensively studied in the recent past. The main difference is due to the suppression of recollision-induced excitation, which for the case of double ionization can be quantitatively explained within our simple semiclassical model. The latter can be of special interest for longer wavelengths since here, the most powerful theoretical method of strong-field physics, numerical solutions of the time-dependent Schrödinger equation, will experience severe computational difficulties due to the large excursions of the ionized electrons, and, thus, the large spatial numerical grid required [48]. We have also presented experimental data for NSDI of nitrogen molecules, where the double-hump structure in the ion momentum distribution characteristic for recollision-induced ionization is, for the first time, observed for a molecular target. Though not discussed in this paper, coincident electron spectra have been recorded as well, being the subject of further analysis.

The results of the present work directly show that the investigated wavelength regime would be beneficial for such applications as ultrafast molecular imaging via recollision-induced fragmentation [3, 19, 41, 42]. A demonstrated decrease of the contributions from confusing excitation channels in double or multiple ionization would considerably facilitate the separation of different fragmentation pathways, and would allow one to enhance a direct (e, ne)-like...
process with its well-defined timing, thus, improving time resolution of the imaging procedure and facilitating the interpretation of the data.

In general, the mid-infrared wavelength range represents a very promising regime for further investigations and applications of all phenomena involving recollision, and thus, for the upcoming field of attosecond physics. This is essentially due to the fact that the recollision energy scales with $\lambda^2$, and by using larger wavelengths high scattering energies can be achieved already at moderate laser intensities [1]–[3]. Even though experiments on above-threshold ionization or harmonic generation beyond 1 $\mu$m are still scarce (see, e.g., [49]–[51]), the recent development of intense high-repetition rate laser sources delivering few-cycle pulses for this frequency domain [52, 53] are expected to lead to an experimental breakthrough in the nearest future. Correspondingly, much more detailed studies of correlated few-electron laser-induced dynamics in this regime will become possible, including kinematically complete experiments feasible with the reaction microscope used in this work. Especially interesting would be the possibility to scan over a broad range of wavelengths, considerably changing both energy and timescale of the strong-field reactions.

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