Nonsequential double ionization of N_2 in a near-single-cycle laser pulse

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We present a comparative study of nonsequential double ionization (NSDI) of N_2 and Ar exposed to near-single-cycle laser pulses. The NSDI process is investigated using carrier-envelope-phase-tagged electron-ion coincidence spectroscopy. The measured NSDI spectra of N_2 and Ar exhibit a striking resemblance. In particular, the correlated two-electron momentum distribution arising from NSDI of N_2 also displays a cross-shape very similar to that reported for Ar [Bergues *et al.*, Nat. Commun. **3**, 813 (2012)]. We interpret our results in terms of recollision-excitation with subcycle depletion and discuss how this mechanism accounts for the observed similarities and differences in the ionization behavior of the two species.

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I. INTRODUCTION

In nonsequential double ionization (NSDI) of an atom or molecule exposed to a strong laser field, the ionization acts of the first and second electrons are not independent from each other. Because it represents a prototypical example of multielectron dynamics driven, steered, and potentially controlled by an external field, NSDI of atoms and molecules has received considerable attention over the past four decades (for a recent review see [1]). Among different experimental techniques, including intensity-dependent ion-yield measurements [2,3], charge-state-resolved electron time-of-flight spectroscopy [4], and recoil-ion momentum spectroscopy [5], coincidence electron-ion recoil-momentum spectroscopy, where the momenta of electrons and ions are measured in coincidence [6], provides the most detailed information about the process. Besides NSDI in a strong field reported here, the study of NSDI has been extended to the extreme-ultraviolet-wavelength regime at free-electron lasers [7,8] and, recently, using time-resolved two-XUV-photon ionization spectroscopy with XUV radiation generated from a tabletop high-harmonic source [9].

While it is now well established that in NSDI the emission of the second electron is facilitated by the laser-driven recollision of the first electron with its parent ion, the exact mechanisms governing this recollision-induced ionization are not yet fully understood. One of the reasons is that in multicycle laser pulses, NSDI may involve multiple recollisions, which hamper the interpretation of experimental results and represent a great challenge for theoretical description. Owing to the advances in the production of ultrashort laser pulses [10–12], the problem of multiple recollisions can be largely avoided by driving NSDI with a carrier-envelope-phase (CEP) controlled near-single-cycle laser pulse to essentially confine the process to a single recollision event. While active CEP stabilization has allowed for recording the recoil-ion momentum spectrum of doubly charged Ar ions at various CEPs [13], measurements of the more instructive CEP-resolved electron correlation spectra were hindered in the past by the difficulty to keep the laser CEP stable over the entire acquisition time (on the order of 24 h) of such an experiment. The use of single-shot CEP tagging [14,15] has recently permitted overcoming those limitations and enabled the detailed investigation of NSDI of Ar in the near-single-cycle [16] and the few-cycle [17] regimes.

In the present study, we pioneer NSDI in a single laser cycle for diatomic molecules, where N_2 serves as a test case. The first and second ionization potentials of N_2 ($I_P^I = 15.58$ eV and $I_P^{II} = 27.12$ eV) are very close to those of Ar ($I_P^I = 15.76$ eV and $I_P^{II} = 27.63$ eV), and similarities in the ionization behavior, particularly NSDI, of the two species have been reported in numerous studies [18–22]. The fact that those similarities are not observed for all atom-molecule pairs with nearly equal ionization potentials (such as, for instance, Xe and O_2) [19–21] provides evidence, however, that this criterion is not sufficient to explain the resemblance in the ionization behavior.

Only a few studies so far have addressed this question for N_2 and Ar using highly differential experimental methods [23]. Coincidence spectra of Ar and N_2 have been recorded in the multicycle regime by Eremina *et al.* in Refs. [24,25], respectively. In both measurements, electrons were preferentially emitted with the same kinetic energy. While for most of the electron pairs the momenta point in the same direction, in a small part of the signal, the electrons are emitted with anticorrelated momenta. In a study with similar laser parameters, Zeidler *et al.* demonstrated that the ratio of anticorrelated to correlated electrons can be controlled to some extent via the molecular alignment [26].

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It was shown recently that for NSDI of Ar, the transition to the single-cycle regime leads to dramatically different correlation spectra. There, the two electrons exhibit totally asymmetric rather than totally symmetric energy sharing, resulting in a characteristic cross-shaped pattern in the correlation momentum spectrum [16]. The theoretical interpretation of this experiment involves electron impact excitation of the Ar^+ ion [16]. Since the excited-state spectra of Ar^+ and N_2^+ are significantly different from each other (the lowest excited energy of Ar^+ is 13.5 eV above the ground state, while N_2^+ has several excited states at much lower energy; see, e.g., [27,28]), there is no obvious reason to expect strong similarities between the two-electron momentum distributions of both species. In fact, recent predictions from quantum-mechanical [29,30] as well as classical-ensemble [31] calculations indicate that, in contrast to Ar [32,33], symmetric energy sharing between the electrons should be expected for NSDI of N_2 in a near-single-cycle laser pulse (for a recent review about theoretical approaches to NSDI see [34]). Here, we elucidate this question experimentally and show that, in spite of the significant differences in the excited-state spectra of the Ar^+ and N_2^+ ions, the CEP-resolved electron correlation spectra of Ar and N_2 exhibit striking similarities.

II. EXPERIMENTAL SETUP

A detailed description of the experimental setup has been given in earlier publications [16,35]. Briefly, near-single-cycle, linearly polarized laser pulses with a central wavelength of 750 nm are generated at a repetition rate of 3 kHz by a short-pulse laser system [36]. The laser beam is split into two parts. The first part is focused into a cold supersonic gas jet inside a reaction microscope (REMI) [37], where the momenta of ions and electrons arising from ionization of a target atom (or molecule) in the laser focus are measured in coincidence. The second part of the laser beam is sent to a single-shot stereo-ATI phase meter [14,15], facilitating the CEP tagging of the data recorded with the REMI as detailed in Ref. [38].

III. RESULTS AND DISCUSSION

In order to provide a solid basis for the comparison of the NSDI process in Ar and N_2, we alternately exposed both targets to the same laser focus and recorded the CEP-dependent recoil-ion momentum spectra of singly and doubly charged ions generated in the interaction region. The peak intensity in the focus was determined to be $I_0 = 3.6 \times 10^{14}$ W cm^{-2} by fitting the measured recoil-ion momentum distributions of Ar^+ with the semiclassical model described below. The yield ratios of Ar^{2+} to Ar^+ and N_2^{2+} to N_2^+ were 0.26% and 0.07%, respectively. This difference cannot be explained by double-ionization events occurring via dissociative channels alone since these represent less than 20% of the N_2^+ count, as estimated from the recorded N^+ fragments. The CEP-dependent modulations of the NSDI yield relative to the CEP-averaged yield (as defined in Ref. [38]) were 5.1% ± 0.5% and 4.2% ± 0.4% for Ar and N_2, respectively.

The CEP-dependent momentum spectrum of the Ar^{2+} and N_2^{2+} ions along the polarization axis are shown in Figs. 1(a) and 1(b), respectively. Both spectra exhibit a pronounced CEP-dependent asymmetry in the emission direction of the doubly charged ions and are hardly distinguishable from each other. Only a closer inspection shows that the islands in the N_2^{2+} spectrum extend slightly further towards smaller momentum values than the corresponding Ar^{2+} islands.

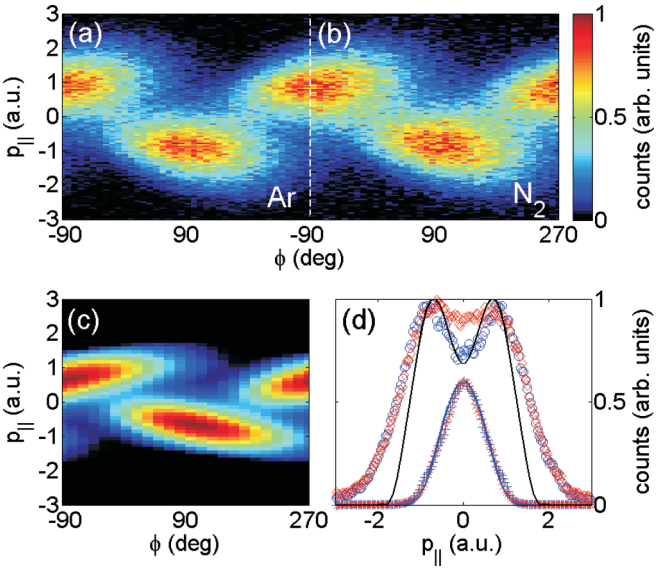

FIG. 1. (Color online) Measured CEP dependence of the recoil-ion momentum distribution along the laser polarization axis: (a) for Ar^{2+} and (b) for N_2^{2+}. The ions were generated in the same laser focus at a peak intensity of $I_0 = 3.6 \times 10^{14}$ W cm^{-2}. Invariance under the symmetry transformation $(p,\phi) \rightarrow (-p,\phi+\pi)$ was used to symmetrize the experimental images. (c) Calculated CEP-dependent $p_{||}$ distributions of Ar^{2+} using the semiclassical model described in the text. The undetermined CEP offset in the experimental data was chosen such that measured and calculated doubly charged ion spectra oscillate in phase. (d) Measured momentum spectra of Ar^+ (blue pluses), N_2^+ (red crosses), Ar^{2+} (blue circles), and N_2^{2+} (red diamonds), averaged over CEP, shown together with predictions for the Ar^+ spectrum [blue (gray) solid line] and the Ar^{2+} spectrum (black solid line). The maxima of the doubly charged ion spectra are normalized to 1, and the spectra of the singly charged ions are normalized to 0.6 for visual convenience. The statistical uncertainty of the measured data is of the order of the marker size.

For the discussion of the mechanisms that govern NSDI of N_2 and Ar, it is instructive to compare the experimental results to predictions of a semiclassical NSDI model for Ar. The model, which was described in detail in Ref. [16], builds on recollisional excitation with subsequent ionization (RESI) [39,40]. In RESI, the recolliding electron excites the parent ion, which is then further ionized by the laser field. In the calculation we assume that the main contribution comes from RESI via the lowest-lying excited state of Ar. Just after the recollision, the magnitude of the momentum of the recolliding electron is determined by energy conservation, while its direction, i.e., the angle between the momentum vectors just before and just after the recollision, is described by a free parameter, β, in our model. The dependence of the simulation results on β was discussed in Ref. [16]. The best agreement is found for $\beta = 25°$. Depletion of the bound-state population as well as the intensity distribution over the focal volume are taken into account in the calculations.

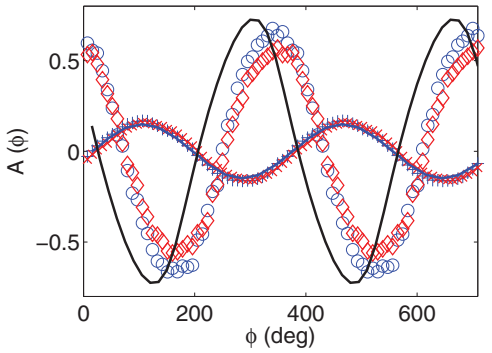

FIG. 2. (Color online) CEP-dependent asymmetries for Ar^+ (blue pluses), N_2^+ (red crosses), Ar^{2+} (blue circles), and N_2^{2+} (red diamonds), shown together with predictions for the Ar^+ asymmetry [blue (gray) solid line] and the Ar^{2+} asymmetry (black solid line). The undetermined CEP offset in the experimental data was chosen such that the asymmetries of the measured and the calculated Ar^+ spectra oscillate in phase. The statistical uncertainty of the measured data is of the order of the marker size.

The calculated CEP-resolved momentum spectrum of Ar^{2+} is displayed in Fig. 1(c). While the simulation qualitatively reproduces the experimental result, the signal in the measured Ar^{2+} spectrum extends to somewhat higher momenta than in its calculated counterpart. As discussed in Ref. [16], this is probably due to small contributions from higher excited states of Ar^+ that are neglected in the present model. To compare the spectra more quantitatively, in Fig. 1(d) we plot the CEP-averaged Ar^{2+} and N_2^{2+} spectra together with the CEP-averaged spectra of the simultaneously recorded Ar^+ and N_2^+ ions and the predictions of the model for Ar. Within the resolution of the measurement (of about 0.1 a.u.), the Ar^+ and N_2^+ spectra lie essentially on top of each other. The Ar^{2+} and N_2^{2+} momentum distributions, in contrast, differ by about 10%, around $p_{||} = 0$. The measured and calculated Ar^{2+} spectra have a pronounced dip, while the central part of the N_2^{2+} spectrum is rather flat.

In order to discuss the CEP dependence in more detail, the CEP-dependent asymmetries in the singly and doubly charged ion spectra of Ar and N_2 are compared in Fig. 2. The asymmetry $A(\phi)$ is defined as $A(\phi) = [N_+(\phi) - N_-(\phi)]/[N_+(\phi) + N_-(\phi)]$, where, for a given CEP ϕ, $N_-(\phi)$ $[N_+(\phi)]$ are the number of ions with a negative (positive) momentum component along the polarization axis. The CEP-dependent asymmetry can be approximated by $A(\phi) = A_0 \sin(\phi + \phi_0)$, where A_0 is the asymmetry amplitude and ϕ_0 is an offset phase. Since all the ions were recorded simultaneously, the phase shifts between the different asymmetry curves are meaningful quantities. Note that this is not the case for data recorded in separate experiments since the CEP is measured up to a constant but unknown offset, which depends on the specific conditions of the experiment. One can see that the asymmetry curves of Ar^+ and N_2^+ coincide perfectly in amplitude and phase. While those for Ar^{2+} and N_2^{2+} oscillate in phase with each other, the amplitude of the N_2^{2+} asymmetry curve is slightly smaller than for the case of Ar^{2+}. While the measured asymmetry amplitude for Ar^+ is well reproduced by the semiclassical model, the asymmetry for Ar^{2+} is overestimated by 10%. The predicted phase shift of 165° between the Ar^{2+} and Ar^+ asymmetry curves differs from the measured value of 125°. As discussed in Ref. [16], quantitative agreement would require a more sophisticated theory.

In order to measure electron correlation spectra of NSDI of N_2, data were collected over 48 hours at a focal peak intensity

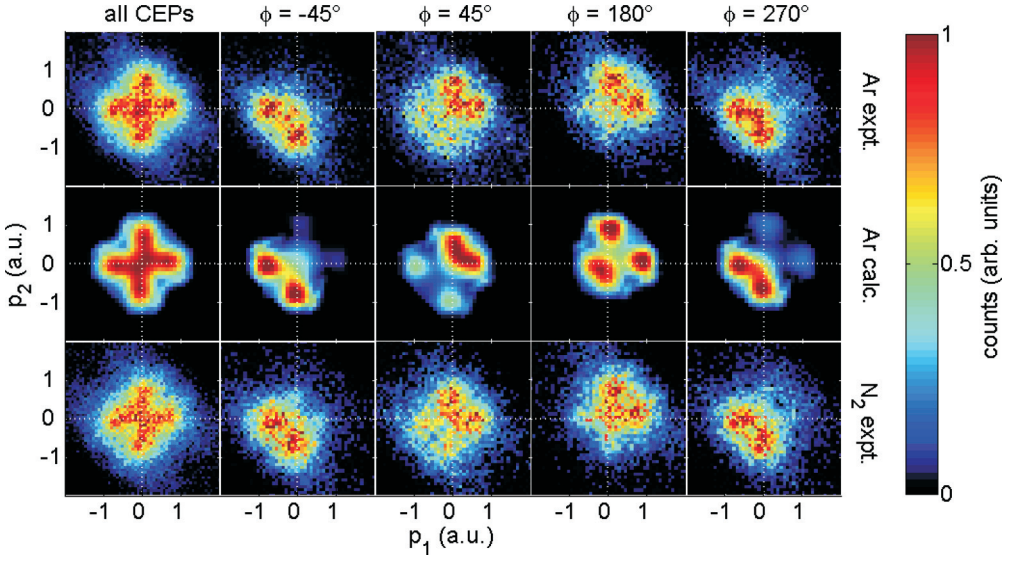

FIG. 3. (Color online) Experimental two-electron momentum distributions showing the correlated momenta along the laser polarization axis of the first (p_1) and second (p_2) electrons emitted from Ar^{2+} (top row) or N_2^{2+} (bottom row) and the corresponding predictions for Ar (middle row) at the peak intensity $I_0 = 3.2 \times 10^{14}$ W cm^{-2}. The CEP-averaged results are displayed in the left column. In the second through fifth columns, the correlation spectra are shown for CEP values of −45°, 45°, 180°, and 270°, respectively. The signal in each image is averaged over a CEP range of ±22.5° and normalized to its maximum value. The undetermined CEP offset in the experimental data was chosen such that measured and calculated doubly charged ion spectra oscillate in phase. Invariance under the symmetry transformation $(p,\phi) \rightarrow (-p,\phi + \pi)$ and the symmetry with respect to the $p_2 = p_1$ diagonal were used to symmetrize the experimental images.

of $I_0 = 3.2 \times 10^{14}$ W cm^{-2}. The total count rate was kept low (0.2 ions per laser shot) in order to decrease the contribution of false coincidences to less than 15% of the measured signal. Under the experimental conditions of this measurement, the ratio of the measured yields of N_2^{2+} and N_2^+ ions was 0.05%. The data are compared to correlation spectra of NSDI of Ar recorded in a separate experiment under similar conditions and at essentially the same peak intensity. The ratio of the Ar^{2+} and Ar^+ yields in this measurement was 0.13%.

The measured two-electron momentum distributions from NSDI of Ar and N_2 are presented in Fig. 3 together with the predictions of the NSDI model for Ar. The experimental correlation spectra are generated by considering the laser shots where at least one electron and one Ar^{2+} (or N_2^{2+}) ion were detected. Since one of the two NSDI electrons is frequently missing because of the limited single-particle detection efficiency of about 50%, the momentum of the missing electron is calculated as the negative momentum sum of the dication and the detected electron. All the measured and the simulated CEP-averaged correlation spectra shown in Figs. 3(a), 3(b), and 3(c) exhibit a fairly similar crosslike structure. The dependence of the two-electron momentum distributions on the CEP is shown in the second through fifth columns for CEP values of 135°, 225°, 0°, and 90°, respectively. Here, again, it can be seen that all three series of spectra exhibit qualitatively the same features.

The comparison of the measured and simulated correlation spectra for Ar confirms the results of Ref. [16] that were obtained at a slightly lower intensity. The pronounced maxima along the momentum axes at ±0.7 a.u. are well reproduced by the calculation. The essentially asymmetric energy sharing observed in the measured correlation spectra of N_2 [Fig. 3(c)] strongly contrasts with the results reported in previous experimental studies using longer pulses [25,26]. Although it is very similar to the measured correlation spectrum of Ar [Fig. 3(a)], the correlation spectrum of N_2 slightly differs by contributions of low-energy anticorrelated electrons, leading to the flat top of the N_2^{2+} spectrum. Together with the disparity in the measured double- to single-ionization yield ratios, this constitutes the main difference observed between NSDI of Ar and N_2.

The reason for the asymmetric energy sharing of NSDI electrons in Ar was discussed in detail in Ref. [16]. Subcycle depletion of the excited-state population in the course of the RESI process was shown to govern the moment of the second-electron release on a subcycle time scale. The instant of emission, in turn, determines the momentum that the electron gains in the laser field. The strong resemblance between the correlation spectra of Ar and N_2 suggests that the same mechanism should also apply for NSDI of N_2. A crucial difference between recollision excitation of Ar and N_2, however, is the existence of excited states in N_2^+ with energies ranging well below that of the lowest excited state of Ar^+.

In order to investigate the role of such lower-lying excited states, we considered in our model for Ar the contribution

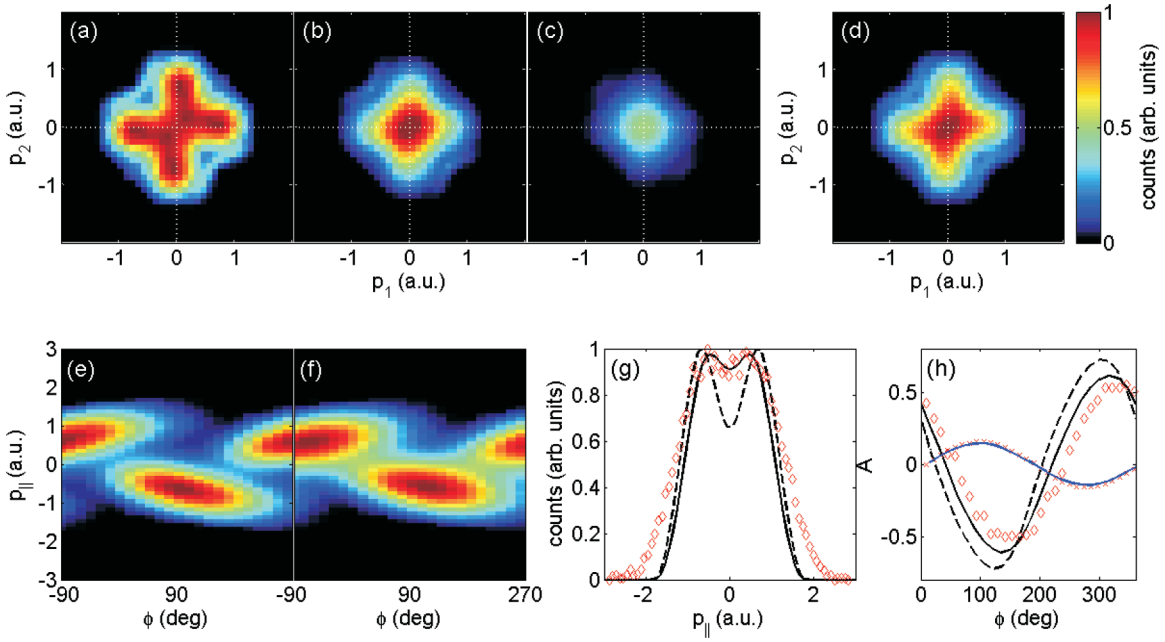

FIG. 4. (Color online) Contribution of (a) the lowest excited state of Ar^+ with an excitation energy of 13.5 eV and two additional fictitious states with excitation energies of (b) 11.5 and (c) 9.5 eV to the correlation spectrum. (d) Sum of the contributions in (a), (b) and (c) normalized to 1. The excitation probability by an electron with a kinetic energy greater than the excitation energy is assumed to be equal for all three states. Comparison of the calculated CEP-resolved momentum spectra of the doubly charged ions (e) without and (f) with the contribution of the two lower-lying states. The corresponding CEP-averaged spectra and the asymmetry curves of singly and doubly charged ions are compared in (g) and (h) to the measured N_2^{2+} data (red diamonds). The black solid (dashed) lines in (g) and (h) are the calculation results obtained with (without) inclusion of the lower-lying states. The blue (gray) solid line and the red crosses in (h) denote, respectively, the calculated and measured asymmetry curves of the singly charged ions. Here, the undetermined CEP offset in the experimental data was chosen such that the asymmetries of the measured and the calculated Ar^+ spectra oscillate in phase.

of two additional fictitious states with excitation energies of 9.5 and 11.5 eV. The calculation results are summarized in Fig. 4. Due to the decreasing tunneling probability, the NSDI yields from states with energies below the first excited state of Ar rapidly decrease with decreasing excitation energy and thus constitute just a small correction. In particular, it can be seen in Figs. 4(a)–4(d) that these lower-lying states add a small contribution of low-energy anticorrelated electrons in the correlation spectrum that leads to a slight merging of the islands in the CEP-resolved momentum spectrum of the doubly charged ions, as can be seen in Figs. 4(e) and 4(f). This feature, which is consistent with the experimental observations, flattens the dip in the corresponding CEP-averaged spectrum presented in Fig. 4(g). Finally, one can see from Fig. 4(h) that the agreement between the measured and calculated asymmetry curves is also improved. In spite of the good agreement, we would like to point out that the contribution of lower-lying excited states is considered only qualitatively in the present analysis. Rather than a quantitative description, our approach may provide a basis for a more detailed treatment taking into account exact electron-impact excitation cross sections for the relevant molecular states.

IV. CONCLUSIONS

We have performed a direct comparison of NSDI of Ar and N_2 triggered by a single recollision event. A remarkably high degree of similarity was observed in the CEP-resolved correlation spectra, which both exhibit totally asymmetric energy sharing between the electrons. We have shown that not only the striking similarities but also the differences in NSDI of the two studied species can be understood in terms of RESI with subcycle depletion of the excited-state population. The development of quantitative models for NSDI in molecules should benefit from both the many constraints imposed by our highly differential experimental data and the fact that a rigorous theoretical treatment of NSDI is significantly easier for an isolated recollision event than for the more complex long-pulse dynamics involving multiple recollisions.

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