

Single-shot carrier-envelope-phase-tagged ion-momentum imaging of nonsequential double ionization of argon in intense 4-fs laser fields

Nora G. Johnson, O. Herrwerth, A. Wirth, S. De, I. Ben-Itzhak, M. Lezius, B. Bergues, and M. F. Kling^{*}
Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany and
J. R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506, USA

A. Senftleben, C. D. Schröter, R. Moshhammer, and J. Ullrich
Max-Planck-Institut für Kernphysik, D-69117 Heidelberg, Germany

K. J. Betsch and R. R. Jones
University of Virginia, Charlottesville, Virginia 22904, USA

A. M. Saylor, T. Rathje, K. Rühle, W. Müller, and G. G. Paulus
Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität, D-07743 Jena, Germany and
Helmholtz Institut Jena, D-07743 Jena, Germany

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Single-shot carrier-envelope-phase (CEP) tagging is combined with a reaction microscope (REMI) to investigate CEP-dependent processes in atoms. Excellent experimental stability and data acquisition longevity are achieved. Using this approach, we study the CEP effects for nonsequential double ionization of argon in 4-fs laser fields at 750 nm and an intensity of 1.6×10^{14} W/cm². The Ar²⁺ ionization yield shows a pronounced CEP dependence which compares well with recent theoretical predictions employing quantitative rescattering theory [S. Micheau *et al.*, *Phys. Rev. A* **79**, 013417 (2009)]. Furthermore, we find strong CEP influences on the Ar²⁺ momentum spectra along the laser polarization axis.

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I. INTRODUCTION

The electric field of a Fourier-limited laser pulse, $E(t) = E_0(t) \cos(\omega t + \varphi)$, is fully characterized by the pulse envelope, $E_0(t)$, the carrier frequency, ω , and the carrier-envelope phase (CEP), φ , which is freely adjustable and stabilized in modern laser systems [1]. By manipulating these variables, electron dynamics can be controlled on a subfemtosecond time scale [2]. A few prominent applications of CEP stable laser pulses are the reliable and reproducible generation of attosecond light pulses [3], the controlled electron emission from atoms [4–6], and the control of electron localization in the dissociative ionization of molecular hydrogen [7–9] as well as in the more complex multielectron CO molecule [10]. Most previous experiments employed CEP locking, which results in a signal-to-noise ratio and phase resolution limited by the duration of continuous locking [4,9]. In addition, a pair of fused silica wedges were used in most previous studies to change the CEP, inherently also modifying the pulse duration and peak intensity by introducing chirp [4,7,9,10]. Reaction microscope (REMI) measurements usually rely on single target particles interacting with individual laser pulses and, therefore, require rather long acquisition times when used with kHz repetition rate laser systems making CEP-dependent REMI studies scarce. The limited duration of continuous CEP locking has triggered studies toward the possibility to measure the CEP as an alternative to CEP locking [11]. This was recently achieved experimentally

for every single laser shot using stereographic detection of electrons from above-threshold ionization (Stereo-ATI) [12], which has also been investigated theoretically [13]. Here, we report on a combination of single-shot phase tagging with REMI measurements. Our measurements do not require CEP stabilization and allow for long data acquisition times. Furthermore, no dispersive material is required to vary the CEP during the experiment, such that the laser pulse envelope remains constant.

We study the nonsequential double ionization (NSDI) of argon in strong, few-cycle laser fields. NSDI accounts for an enhanced production of doubly charged ions over the yields expected for a purely sequential tunneling mechanism [14]. It is one of the most interesting phenomena in strong-field ionization as it involves the correlated motion of at least two electrons (see, e.g., [15]) and many studies have addressed the underlying mechanisms for different target species, pulse durations, intensities, and wavelengths (see, e.g., [4,16–24]). Because of the technical difficulties mentioned previously, however, there exists only one measurement with CEP stable few-cycle laser pulses [4]. Such data are highly desirable, as they might further elucidate the subcycle electron dynamics responsible for NSDI. Using our new approach, we reinvestigate the NSDI of argon and observe pronounced phase dependencies in the double ionization yield and the Ar²⁺ momentum spectra.

II. EXPERIMENTAL TECHNIQUE

The experiments were performed at the AS-1 beamline at the Max Planck Institute of Quantum Optics [25] delivering

^{*}Corresponding author: matthias.kling@mpq.mpg.de

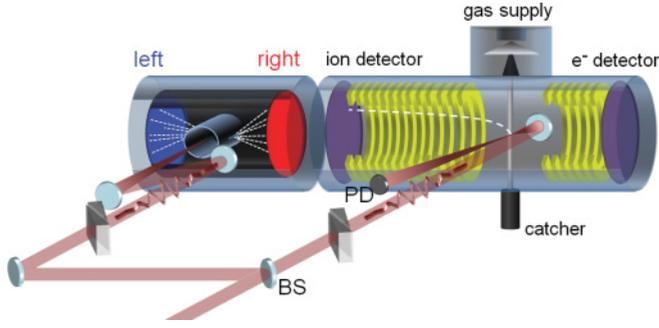


FIG. 1. (Color online) Schematic of the setup consisting of a single-shot Stereo-ATI phase meter (left chamber) and a REMI spectrometer (right chamber). A beamsplitter (BS) directs light into both chambers. Each arm is equipped with its own wedges for compensating residual chirp. The phase meter contains a layer of μ metal to shield from external electric and magnetic fields. A Xe gas pressure of 6.5×10^{-3} mbar fills the inner gas cell. In the Stereo-ATI, xenon atoms are ionized near the focus of the horizontally polarized laser beam and the electrons produced enter the high-vacuum drift tubes through slits. Electrons resulting from the ionization by a single laser shot are detected by MCP detectors to the left and to the right, enabling the determination of the CEP for each REMI event as described in the text. Simultaneously, Ar^{2+} ions created in the overlap of the laser focus and a skimmed supersonic jet of the REMI are detected by an MCP detector with a delay line anode. The jet stream is efficiently pumped by the catcher. The master trigger for the electronics is provided by a fast photodiode (PD). Note that the ion and electron optics are symmetric and a few REMI electrodes have been removed for visualization of the laser path. Furthermore, the electron detection in the REMI, which is facilitated by additional magnetic fields, was not utilized in these first proof-of-principle experiments, where only ions were detected.

4-fs pulses at 750 nm with an energy of 400 μJ per pulse at a 3-kHz repetition rate. The experimental setup is sketched in Fig. 1. Approximately 30 μJ of the beam are focused into a single-shot Stereo-ATI phase meter [12], while the main beam is directed into a vacuum chamber hosting the REMI. The latter is composed of time-of-flight spectrometers for electrons and ions combined with position-sensitive detectors to enable measuring the particles' three-dimensional momentum vectors [26]. The intensity in both devices can be varied by apertures and residual chirp is compensated by fused silica wedges (see Fig. 1). In the REMI chamber, the laser is focused by a spherical mirror ($f = 25$ cm) into a cold supersonic gas jet of neutral argon atoms. The jet crosses the laser beam slightly before the focus in order to minimize averaging over the CEP due to the Gouy phase shift [27].

The CEP for every single laser shot was determined via a Stereo-ATI measurement [12]. This apparatus utilizes the fact that the shape of the ATI spectrum for ultrashort pulses in Xe, especially for rescattered electrons with high energy, and correspondingly short time-of-flights (TOF), is sensitive to changes in the CEP of the laser pulse [5]. Here, the laser is focused into a xenon target and electrons that are emitted within an angle of approximately 2° around the laser polarization are detected as time-dependent electron currents on the microchannel plate (MCP) detectors with metal anodes to the left, L , and right, R as depicted in Fig. 1. Converting

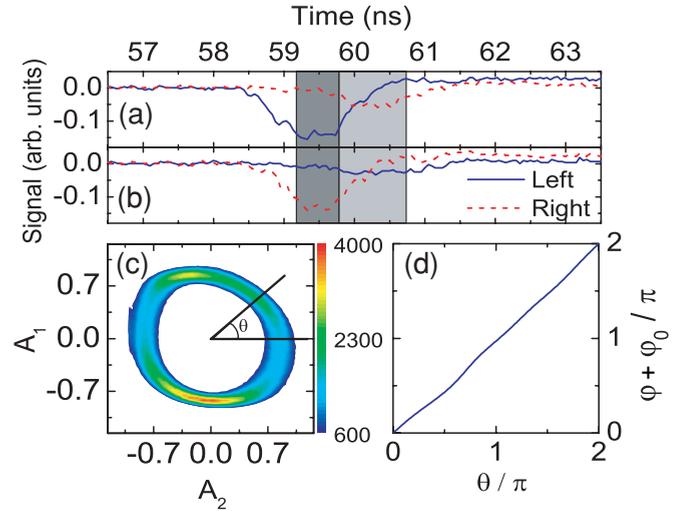


FIG. 2. (Color online) (a) and (b) Time-of-flight traces from the left and right detectors of the Stereo-ATI for two laser shots with different CEPs. Light and dark shaded areas indicate the gated regions for determining A_1 and A_2 , respectively. (c) PAP taken under the same conditions as the Ar^{2+} data (see text for further details). The color scale indicates the number of counts. (d) The relative CEP, $\varphi + \varphi_0$, as a function of the polar angle in the PAP, θ . The small deviation from the line $\varphi + \varphi_0 = \theta$ indicates only minor adjustments are needed for retrieving the correct relative phase.

the two currents into voltages yields the time-dependent ATI spectra in the form of two quickly varying voltage signals.

Figures 2(a) and 2(b) show TOF spectra for two laser shots with different CEPs. In order to process the ATI spectra and output a signal corresponding to the CEP in real-time, we utilize the fact that there are energy regions in which the contrast or asymmetry, $A = (L-R)/(L+R)$, with L and R being the number of counts on the left and right detector, respectively, in the ATI spectra of Xe varies like sine and cosine of the CEP, $A_1 \approx \sin(\varphi + \varphi_0)$ and $A_2 \approx \cos(\varphi + \varphi_0)$. These energy regions are chosen by appropriate gates in the TOF spectra as depicted in Figs. 2(a) and 2(b) (light and dark gray shaded areas). From the integral of the spectra within the gate windows, two asymmetry values, A_1 and A_2 , can be determined and a parametric asymmetry plot (PAP) of A_1 vs A_2 can be generated [see Fig. 2(c)], in which the polar angle, θ , corresponds to the CEP: $\theta \approx \varphi + \varphi_0$. A specially designed circuit facilitates the calculation of the asymmetry parameters, A_1 and A_2 , in $\sim 20 \mu\text{s}$ [28], thereby allowing the asymmetry parameters to be recorded simultaneously with the REMI data, therefore enabling the phase tagging of each recorded event.

Although the dependence of the asymmetry parameters on the CEP is only approximated by sine and cosine functions, the exact relationship between φ and θ can be determined based on the assumption that the laser produces a uniform and random distribution of CEP values. This constraint means that, given enough laser shots, every equally sized interval in φ must have the same number of counts, i.e., $d\varphi = \frac{\rho(\theta)d\theta}{\langle\rho\rangle} = \lambda(\theta)d\theta$, where $\rho(\theta)$ is the density of laser shots as a function of θ , $\langle\rho\rangle$ is the average density, and $\lambda(\theta) \equiv \rho(\theta)/\langle\rho\rangle$. Therefore, $\varphi(\theta) = \varphi_0 + \int_0^\theta \lambda(\theta')d\theta'$ [29]. The assumption of a uniform distribution is validated by shifting the CEP via tuning the

fused silica wedges just before the Stereo-ATI while observing no change in the PAP. Since the REMI data acquisition is triggered only when an event occurs (and therefore the PAP may be biased by recording only these events), we separately recorded unbiased PAP data for the transformation of θ to φ for approximately 10 min at the full repetition rate. The transformation from the raw phase, θ , to the actual CEP value, $\varphi + \varphi_0$, for the PAP data displayed in Fig. 2(c) is shown in Fig. 2(d). As can be seen in Fig. 2(d), the necessary correction to θ is small.

In the current measurements our uncertainty in the single-shot CEP detection is less than 210 mrad, which was determined from the PAP data as outlined in Ref. [29]. Note that this uncertainty is comparable to the rms phase variation obtained by employing a Stereo-ATI as feedback for phase locking [30]. However, our technique requires neither feedback nor the precise stabilization necessary for locking. Moreover, removing the need for locking and stabilization allows for the collection of data over much longer periods, which, in turn, facilitates a significantly more precise determination of the CEP dependence. Additionally, unlike common locking schemes [1,31], we measured the CEP after amplification, hollow-core fiber compression, and transport, such that our phase-tagging technique accounts for any CEP fluctuations up to the interaction chamber.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Using the described technique, we explored the CEP dependence of NSDI of argon. The prevailing interpretation of NSDI is based on laser-driven rescattering: at a peak of the laser electric field tunnel ionization produces an electron, which is then driven by the laser field and may recollide with the singly charged ion. Depending on the kinetic energy of the returning electron and the target species, the collision can result in either the direct emission of two electrons in an $(e,2e)$ process (recollision-induced direct ionization, RIDI) or the excitation of the singly charged ion with subsequent field ionization by the laser field (recollision-induced excitation plus tunneling, RIET). The electron as well as the doubly charged ion recoil momentum spectra along the laser polarization axis (parallel momentum) exhibit characteristic signatures of the NSDI process: within a classical description the two emitted electrons in an $(e,2e)$ process can acquire up to $p_{\parallel}^{\max} = 4\sqrt{U_p}$ (a.u.) [23], where U_p is the ponderomotive potential, defined as $U_p = I/4\omega^2$ (a.u.) for laser intensity I . This simple expression can be derived under the assumption that the two electrons are liberated with zero initial momentum and that the excess energy of the first recolliding electron is consumed completely in the liberation of the second electron. The release of the second electron at recollision energies sufficiently above the threshold for ionization of the ion typically occurs in RIDI when the first electron returns close to its highest kinetic energy near a zero crossing of the oscillating electric field [corresponding to a maximum in the vector potential $A(t_i)$ at time t_i given by $A(t_i) = -\int_{t_i}^{\infty} E(t) dt$]. In this case both electrons are emitted into the same hemisphere along the polarization direction and the momentum distribution of the doubly charged ion exhibits maxima at nonzero momentum [23]. In RIDI, a strong CEP-dependent asymmetry

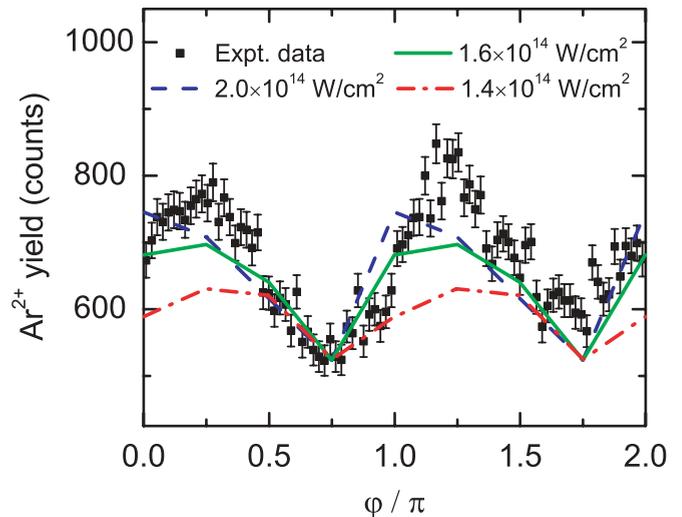


FIG. 3. (Color online) Comparison of the experimental CEP-dependent Ar^{2+} yield (solid black squares) with theoretical predictions (colored lines) for three different intensities from Micheau *et al.* [16]. The error bars indicate the statistical error of the experimental data. The offset phase, $\varphi_0 = 0.43\pi$, for the experimental data has been chosen such that the minimum in the experimental data coincides with the theory.

in the electron and ion-momentum spectra is expected [22]. In the RIET mechanism, the returning first electron leads to excitation of the singly charged ion. Double ionization occurs in RIET by tunnel ionization at one of the subsequent peaks of the electric field (near zeros of the vector potential). Depending on the sign of the electric field causing tunneling of the second electron it can leave in either the same or the opposite direction as the first, resulting in an ion-momentum spectrum with a maximum at zero according to Ref. [23].

Previous studies have shown that both RIDI and RIET typically contribute to NSDI of argon and their importance was studied as a function of the laser pulse duration, intensity, and wavelength [4,15,17,18,22–24]. It was found that RIDI dominates NSDI for longer wavelengths and higher intensities due to the higher ponderomotive potential [17]. Liu *et al.* [4] have employed phase-stabilized 5-fs laser pulses at 760 nm and $3.5 \times 10^{14} \text{ W/cm}^2$ and have observed a CEP dependence in the parallel Ar^{2+} momentum spectra. Interestingly, a double-hump structure in the momentum spectra was obtained for all phases, which indicated a predominant role of RIDI for their experimental parameters [4]. The results were interpreted in terms of a semiclassical model by Rottke *et al.* [22], the validity of which was confirmed by time-dependent Schrödinger equation (TDSE) calculations by Liao *et al.* [20]. Most recently, Micheau *et al.* [16] adapted quantitative rescattering theory to NSDI. They predicted the Ar^{2+} ion yield to be CEP dependent. To date this quantity has not been measured, most likely due to (i) the lack of long-term stability of experiments employing CEP stabilization and (ii) the fact that changing the CEP in previous experiments led to small changes in pulse duration and intensity, thus prohibiting a measurement of the Ar^{2+} yield as a function of CEP alone.

Employing the phase-tagging approach reported here, we have obtained the CEP dependence of the Ar^{2+} ion yield

for constant pulse duration and intensity (see Fig. 3). The peak intensity in our measurement $1.6(\pm 0.4) \times 10^{14}$ W/cm² is determined from the $2U_p$ cutoff in the longitudinal momentum (along the laser polarization axis) of H₂O⁺, which was present as a small background in the jet in our experiments on argon and, in contrast to Ar⁺, did not saturate the detector. The experimental Ar²⁺ ion yield data in Fig. 3 are compared to recently published theoretical results by Micheau *et al.* [16]. They calculated the Ar²⁺ yield for a five-cycle pulse centered at 800 nm at three intensities of $(1.4, 1.6, 2.0) \times 10^{14}$ W/cm². The theoretical curve for 1.6×10^{14} W/cm² agrees best with the data presented here, consistent with the experimentally determined intensity. The calculations predict minima in the Ar²⁺ yield at approximately $3/4\pi$ and $7/4\pi$, which are independent of intensity in the range considered [16]. In Fig. 3, we choose $\varphi_0 = 0.43\pi$ in the experimental data such that the dip in the total Ar²⁺ yield occurs at the same phase as in the theoretical curves. Although the yield is clearly CEP dependent, it has $\pm\pi$ inversion symmetry. However, this ambiguity can be eliminated by taking the phase-dependent Ar²⁺ momentum spectra into account, which are shown in Fig. 4.

Figure 4 shows the longitudinal momentum, $p_{||}$, of the Ar²⁺ ions parallel to the laser polarization axis, integrated over the perpendicular momenta. Considering a pulse with $\varphi = 0$, an electron that tunnels out just after the maximum of the laser electric field and recollides about three-quarters of a cycle later (near a zero of the electric field and a maximum in the vector potential) gains maximum kinetic energy. In this case, the two electrons emitted in an ($e, 2e$) process have positive momentum. Due to momentum conservation, the recoiling

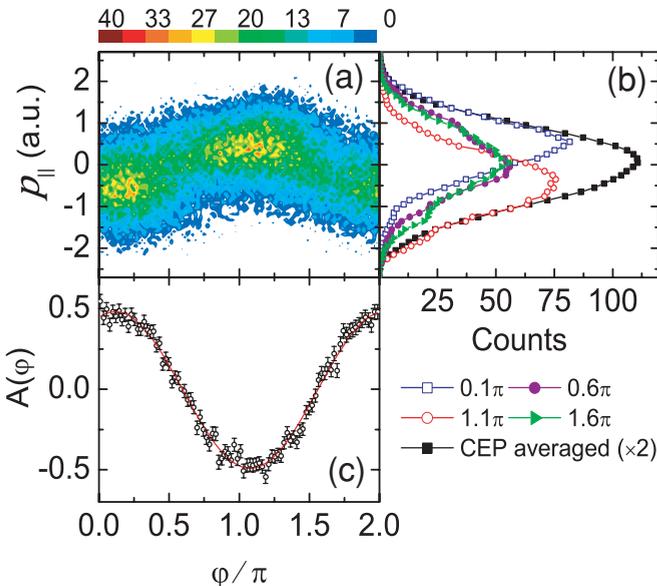


FIG. 4. (Color online) Experimental results for NSDI of argon showing (a) the CEP dependence of the Ar²⁺ longitudinal momentum along the laser polarization axis ($p_{||}$), (b) the longitudinal Ar²⁺ momentum spectra for the CEP averaged (solid black squares) and four phases φ as indicated in the legend (integrated within $\pm 0.04\pi$ and averaged over five adjacent $p_{||}$ values), and (c) the asymmetry parameter, $A(\varphi)$, as defined in the text. The error bars denote the statistical error.

Ar²⁺ ion has negative momentum for $\varphi = 0$. Figure 4(a) shows a two-dimensional plot of momentum distributions for the full 2π CEP range. The $\pm\pi$ ambiguity present in the CEP dependence of the Ar²⁺ yield is resolved by choosing $\varphi = 0$ for negative Ar²⁺ momentum.

Momentum spectra at four values of φ (integrated over a range of $\pm 0.04\pi$) are extracted from Fig. 4(a) and shown in Fig. 4(b). At $\varphi = 0.1$ and 1.1π , the momentum spectra are clearly asymmetric and peak at nonzero momenta, while the spectra at $\varphi = 0.6\pi$ and 1.6π are nearly identical and centered around zero. Comparison of the data to the earlier work by Liu *et al.* [4] shows that under the current conditions we observe one, not two, peak in the longitudinal Ar²⁺ momenta for all CEP values. We attribute this difference to the lower intensity used in the present study. Detailed theoretical modeling may give further insight into the corresponding correlated electron dynamics. This however, is beyond the scope of the current paper.

To explore the degree of control reached in our study, Fig. 4(c) shows the asymmetry in the Ar²⁺ momentum spectra as defined by the parameter $A(\varphi) = [N_-(\varphi) - N_+(\varphi)]/[N_-(\varphi) + N_+(\varphi)]$, where N_- and N_+ represent the number of ions with negative and positive $p_{||}$ momenta, respectively. We observe a very high degree of asymmetry of ± 0.5 which interestingly fits near perfectly (adjusted $R^2 = 0.98$) to a sine function, as indicated in Fig. 4(c).

IV. CONCLUSIONS

In conclusion, we combine single-shot CEP tagging with the REMI technique to allow studies on phase-dependent processes with high statistics and a correspondingly high signal-to-noise ratio as compared to earlier work based on the stabilization of the CEP. A particular advantage of this approach arises from the fact that the laser pulse envelope remains constant over the entire 2π phase-space variation as no dispersive material needs to be used to change the CEP. This experimental approach has been applied to study the NSDI of argon with 4-fs laser pulses at 750 nm and an intensity of 1.6×10^{14} W/cm². With these very short laser pulses, we observed a large (± 0.5) CEP-dependent asymmetry in the recoil momentum spectra of Ar²⁺ and obtained good agreement with the theoretically predicted phase dependence of the Ar²⁺ yield. The phase-tagging approach can be utilized far beyond the studies on NSDI shown here and facilitates the detailed exploration of the waveform control of atoms, molecules, and nanoscopic objects in few-cycle laser fields.

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