Nonsequential Double Ionization with Few-Cycle Laser Pulses

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We investigate differential electron momentum distributions in nonsequential double ionization with linearly polarized, few-cycle pulses, using a classical model based on a laser-assisted inelastic ($e^+, 2e^-$) rescattering mechanism. These yields, as functions of the momentum components parallel to the laser polarization, are highly asymmetric and strongly influenced by the phase difference between the pulse envelope and its carrier oscillation, radically changing their sign around a critical phase. This behavior provides a powerful tool for absolute-phase measurements.

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Recently, few-cycle laser pulses of intensities around or higher than $10^{14}$ W/cm$^2$ have proven to be extremely important. A particular characteristic of such pulses is that they may have very high intensities and, still, carry much less energy than their longer counterparts, such that, effectively, ionization is reduced. This has extended the damage threshold of solid-state materials up to the intensities in question [1], and has made the generation of high-order harmonic radiation up to astonishingly high frequencies possible [2]. Furthermore, their length, of the order of a few fs, permits controlling processes such as molecular motion or chemical reactions [3], as well as the production of isolated, x-ray attosecond pulses [4].

In this pulse-length regime, the so-called “absolute phase,” i.e., the phase difference between the pulse envelope and its carrier frequency, radically influences strong-field phenomena, such as high-order harmonic generation (HHG) [2] and above-threshold ionization (ATI) [5, 6]. Indeed, this phase determines, for instance, the maximal harmonic or photoelectron energies, the intensities in the spectra, and the time profiles of both phenomena.

This is not surprising, since the physics of HHG and ATI is directly related to the instantaneous, time dependent field. HHG, for instance, is described by a three-step mechanism in which an electron leaves an atom at an instant $t_0$ through tunneling ionization, propagates in the continuum under the influence of the external laser field, and, at a later time $t_1$, recombines with a bound state of the parent ion, generating harmonics [7]. Slightly different processes, either in which elastic rescattering with the parent ion is taken as the third step or in which the electron reaches the detector without recolliding explain the high-order and low-order ATI peaks, respectively [8].

In order to interpret the experimental data obtained in such cases, the precise knowledge of the absolute phase $\phi$ is required. This poses a serious practical problem, since this phase is difficult to stabilize, to control, or to measure [9]. For this reason, schemes for measuring $\phi$ have been suggested and realized, as, for instance, using the asymmetry in ATI photoelectron counts [6].

In this Letter, we propose laser-assisted nonsequential double ionization (NSDI) as a tool for absolute-phase diagnosis. This phenomenon is the subject of very active discussion, which was triggered by differential measurements of electron momentum distributions performed with the cold target recoil ion momentum spectroscopy technique, for linearly polarized fields of intensities of the order of $I \sim 10^{14}-10^{15}$ W/cm$^2$ incident in rare-gas samples [10]. Such measurements revealed very peculiar features, namely, two symmetric peaks at $p_{1||} = p_{2||} = \pm 2\sqrt{U_p}$, in the $(p_{1||}, p_{2||})$ plane, where $p_{j||}$ ($j = 1, 2$) and $U_p$ denote the momentum components parallel to the laser field polarization and the ponderomotive energy [11], respectively.

These features are explained by a physical mechanism very similar to those in HHG and high-order ATI. The main difference lies in the rescattering process at $t_1$, which is now inelastic: the first electron gives part of its kinetic energy upon return to the second electron, so that it can overcome the ionization potential of the singly ionized atom and reach the continuum [7]. This process has been considered by several groups, using classical [12], semiclassical [13, 14], or quantum-mechanical [15, 16] approaches, using different types of electron-electron interaction [13, 14], and neglecting or including electron-electron repulsion in the final states [14, 15]. So far, since the pulses involved were relatively long, they have been mostly approximated by monochromatic fields.

In particular, classical models reproduce the main features either observed experimentally or obtained by more refined, quantum-mechanical methods, astonishingly well. Indeed, recently, we have computed NSDI yields considering rescattering in its simplest form, i.e., electron-impact ionization, both classically and within a quantum-mechanical $S$-matrix framework, with practically identical results [14].
In this work, we use a similar classical model as in [14], in which a laser ensemble is subject to a few-cycle pulse \( \mathbf{E}(t) = -d\mathbf{A}(t)/dt \), with the vector potential \( \mathbf{A}(t) = A_0 \sin^2(\omega t/(2n)) \sin(\omega t + \phi) \hat{\mathbf{x}} \). (1)

Thereby, \( \omega \), \( A_0 \), \( \phi \), and \( n \) denote the frequency, amplitude, absolute phase, and number of cycles, respectively. The electrons reach the continuum at a time \( t_0 \) with vanishing drift velocities and from the origin of the coordinate system. The start times are uniformly distributed, and the ejection probability per unit time, unless stated otherwise, is given by the quasistatic [17] tunneling rate

\[
R(t_0) \sim \frac{1}{|E(t_0)|} \exp \left[ -\frac{2(|E_{01}|)^{3/2}}{3|E(t_0)|} \right].
\]

where \( |E_{01}| \) is the ionization potential of the atom in question. Subsequently, these electrons propagate under the influence of only the laser field. Finally, some of them return to the site of their release at \( t_1 > t_0 \) and free a second electron ensemble through inelastic collisions.

Each pair in such ensembles obeys

\[
[k + \mathbf{A}(t_0)]^2 = 0,
\]

\[
\int_{t_0}^{t_1} [k + \mathbf{A}(t)]^2 dt = 0,
\]

and

\[
\sum_{j=1}^{\infty} [p_j + \mathbf{A}(t_1)]^2 = [k + \mathbf{A}(t_1)]^2 - 2|E_{02}|,
\]

given in atomic units. Equation (3) expresses the energy conservation at the ionization time. Equation (4) imposes restrictions upon the intermediate electron momentum \( k \) such that the electron returns to its parent ion. Finally, Eq. (5) yields the energy conservation at the recollision time \( t_1 \). Thereby, the first electron gives part of its kinetic energy \( E_{\text{rel}}(t_1) = [k + \mathbf{A}(t_1)]^2/2 \) upon return to the second electron, so that it is able to overcome the ionization potential \( |E_{02}| \) of the singly ionized atom. In terms of the momenta components parallel and perpendicular to the electric-field polarization, denoted \( p_{\parallel} \) and \( p_{\perp} \) \((j = 1, 2)\), respectively. Eq. (5) is written as

\[
\sum_{j=1}^{\infty} [p_{\parallel, j} + A(t_1)]^2 = [k + \mathbf{A}(t_1)]^2 - 2|E_{02}| - \sum_{j=1}^{\infty} p_{\perp, j}^2.
\]

For constant \( p_{\perp, j} \), Eq. (6) describes a circle in the \((p_{\parallel, 1}, p_{\parallel, 2})\) plane, centered at \( A(t_1) \) and whose radius depends on \( E_{\text{rel}}, |E_{02}| \), and on \( p_{\perp, 1} \). Such momenta effectively shift the second ionization potential so that rescattering may become classically forbidden.

The electron momentum distributions then read

\[
\Gamma \sim \int dt_0 R(t_0) \delta \left( E_{\text{rel}}(t_1) - \frac{1}{2} \sum_{j=1}^{\infty} [p_j + \mathbf{A}(t_1)]^2 - |E_{02}| \right).
\]

where the argument of the \( \delta \) function gives the energy conservation upon return and \( p_{\perp, j} \) are integrated over. Details about this model are given in [14].

These distributions are displayed in the upper panels of Fig. 1, for neon [18], as density plots in the \((p_{\parallel, 1}, p_{\parallel, 2})\) plane. Their circular shapes and the maxima along \( p_{\parallel, 1} = p_{\parallel, 2} \) are features also present for monochromatic driving fields [19], and mean, physically, that both electrons are leaving the parent ion most probably with equal parallel momenta. However, the fact that the yields are mainly concentrated in only one quadrant of the \((p_{\parallel, 1}, p_{\parallel, 2})\) plane makes them strikingly different from the former distributions, which are symmetric in \((p_{\parallel, 1}, p_{\parallel, 2}) \leftrightarrow (-p_{\parallel, 1}, -p_{\parallel, 2})\). Furthermore, around a critical phase \( \phi_c \) [cf. Fig. 1(b)], the distributions start to change, until the whole yield is shifted from the first to the third quadrant. For increasing pulse length, these effects gradually disappear, so that the distributions become symmetric and phase-independent [Figs. 1(e)–1(h)].

Important questions concern the physical origin of both the asymmetry and the critical phase: are they caused by the phase space or by the tunneling rate \( R(t_0) \)? Depending on the parameters, a whole phase-space region may become classically inaccessible, such that the radius of the circle described by Eq. (6) would collapse and the corresponding yields would vanish. The quasistatic tunneling rate, on the other hand, favors the start times \( t_0 \) for which the instantaneous field amplitude \( |E(t_0)| \) is large, as compared to those for which \( |E(t_0)| \) is small. Thus, the

\[\text{FIG. 1. Electron momentum distributions along the laser polarization, for neon (}|E_{01}| = 0.79 \text{ a.u. and } |E_{02}| = 1.51 \text{ a.u.}|, subject to pulses of intensity } I = 4.7 \times 10^{14} \text{ W/cm}^2 \text{ and carrier frequency } \omega = 0.057 \text{ a.u., respectively. In panels (a) to (d) and (i) to (l) we consider a four-cycle pulse } (n = 4), \text{ whereas in panels (e) to (h) the pulse length is varied. Panels (a) to (h) and (i) to (l) were computed with the quasistatic and a constant tunneling rate, respectively. Panels (a) and (i) } \phi = 0.5 \pi; \text{ panels (b) and (j) } \phi = 0.8 \pi; \text{ panels (c) and (k) } \phi = 1.0 \pi; \text{ panels (d) and (l) } \phi = 1.2 \pi. \text{ Panels (e), (f), and (g) } n = 6, 8, \text{ and } 10, \text{ respectively, and } \phi = 0.5 \pi. \text{ In panel (h) } n \text{ is the same as in (g) and } \phi = 1.2 \pi.\]

133006-2
contributions from the former or from the latter case would be enhanced or suppressed, respectively. In order to single out the influence of the phase space, we assume that the electrons belonging to the first ensemble reach the continuum at a constant rate. Such results are shown in Figs. 1(i)–1(l) and are radically different from those obtained with the more realistic, quasistatic tunneling rate. Indeed, the momentum distributions, though asymmetric, exhibit peaks in both first and third quadrants of the \((p_{\parallel}, p_{\perp})\) plane, vaguely resembling those obtained with monochromatic driving fields. The asymmetry is expected, since, for such pulses, the relative tunneling rate is large and, therefore, whose relevant contributions to the yield in the first and third quadrants.

In conclusion, we perform a theoretical investigation of NSDI with few-cycle pulses, using a classical model based on electron-impact ionization. Both electrons have, most probably, equal final momentum components parallel to the field polarization which are mainly positive or negative. The sign of such momenta and their most probable value depend on the absolute phase \(\phi\). In particular, around a critical phase, this sign changes. Such features are explained as the interplay between the tunneling rate for the first electron and the phase space, and agree even quantitatively with quantum-mechanical computations [23].

FIG. 2. Parallel electron momenta \(p_{\parallel}\) along \(p_{\parallel} = p_{2\parallel}\), for \(p_{\perp} = 0\) (\(j = 1, 2\)), as functions of the start times \(t_0\) of the electrons belonging to the first ensemble, in units of the field cycle, together with the quasistatic tunneling rates. The remaining parameters are the same as in Figs. 1(a)–1(d).
The modifications in the NSDI yields upon a critical phase are far more extreme effects than those observed for HHG or ATI. In fact, the advantage of NSDI over the phase are far more extreme effects than those observed for financial support.

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[5] In ATI, an atom absorbs more photons than is necessary for it to ionize, releasing high-energy electrons.
[11] The ponderomotive energy is the temporal average $U_p = (At^2)/2$, where $A(t)$ is the vector potential. In order to compare our results more directly with the existing literature, we approximate it by the expression for the monochromatic case, i.e., $U_p = A_0^2/4$.
[18] For neon, computations based on electron-impact ionization agree very well with the experiments. For other species, such as argon, excitation tunneling should also be considered [V. L. B. de Jesus (unpublished)].
[19] This holds only if the interaction by which the first electrons dislodge the second is of contact type, i.e., $V_{12} = \delta(r_1)\delta(r_2 - r_1)$. This interaction is implicit in Eq. (7). For other types of interactions see, e.g., Ref. [14].
[21] Longer excursion times are not relevant due to wave-packet spreading, which makes the overlap between the wave packets of the first and of the second electrons negligible. In our classical model, this implies leaving the corresponding electrons out of the ensemble.
[22] Other sets of electrons practically do not contribute to the yield, since the corresponding tunneling rate is very small. Their influence is seen if this rate is artificially taken to be constant (cf. lower panels of Fig. 1).
[23] C. Figueira de Morisson Faria et al. (to be published).