PHYSICAL REVIEW A **93**, 021403(R) (2016)

Universal pulse dependence of the low-energy structure in strong-field ionization

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(Received 30 June 2015; published 17 February 2016; corrected 18 February 2016)

We determine quantitatively the laser pulse duration dependence of the low-energy structure (LES) in strong-field atomic ionization and establish its universal character. The electron energy measurement is performed on krypton and argon by varying the duration of a 1.8 μm midinfrared pulse from two to ten cycles. Comparing the experiment with analytical and numerical results, the soft-recollision mechanism leading to electron momentum bunching is confirmed as the origin of the LES. The universal behavior of the LES peak energy on pulse duration emerges from an analytical description as a product of two factors: one contains the influence of the laser parameters and the target, while the other one describes the pulse duration dependence in terms of optical cycles.

DOI: 10.1103/PhysRevA.93.021403

The absorption of a few photons beyond the minimum required to ionize an atom, first observed in 1979 [1], eventually led to high-energy above-threshold ionization (HATI) and high-harmonic generation (HHG). A lot of attention has been devoted to these phenomena since their understanding via a three-step model involving simple classical trajectories in the strong laser field [2,3], succeeded to describe such processes (for example, their high-energy cutoff [4,5]). More recently, the concept of strong-field "quasifree" motion of a high-energy rescattering electron was even used for laser-induced electron diffraction [6–9].

However, the observation of new structures [10,11] in the electron spectrum at low energies posed a puzzle: reproduced by numerical solutions of the time-dependent Schrödinger equation as well as by classical simulations [11–18], these now called "low" [10] and "very-low" [13] energy structures (LES and VLES) evade the well established three-step model. Early numerical studies [11–13] clearly hinted at the importance of the Coulomb potential from the parent ion during formation of the low-energy structures. Eventually their origin was identified by theory as soft recollisions which allow for a simple strong-field description [14], complementary but analogous to that for HATI and HHG. The HATI can be interpreted as a consequence of "hard" recollisions, where the electron is back-reflected with high momentum from the nucleus in an almost elastic recollision immune to influences from the binding potential. During soft recollisions, on the other hand, the electron revisits its parent ion with its momentum approaching zero. This makes the electron trajectory susceptible to external influences, leading to energy gain or loss from the binding potential which gives rise to a spectral bunching of electron momenta at the LES-VLES position [14,16].

The LES peak position is predicted to depend on the carrier envelope phase (CEP) [15,16] and the laser pulse duration [15]. To date, experimental verification was lacking since the required tunable pulse duration in the midinfrared spectral region, a prerequisite for achieving the strong-field limit, is technically quite demanding.

In this Rapid Communication, we report direct experimental evidence of the LES's soft-recollision mechanism by varying the laser's pulse duration from two to ten cycles. Our CEP-averaged experiments for different targets and laser parameters demonstrate that this dependence is universal. We introduce a simple analytical approach, based on classical trajectories and the soft-recollision mechanism, which reproduces the experimental pulse-duration dependence of the LES and reveals its universality: The peak LES energy position factorizes into a universal time-dependent function and another factor which contains the dependence on the laser and target properties. Moreover, the analytical treatment explains the surprising finding that in order to describe the CEP-averaged experiments one can simply ignore the sensitive CEP dependence of soft recollisions.

Using a setup similar to Schmidt *et al.* [19], and controlling the amount of dispersion in the laser system, we achieved a source with pulse duration tunable from two to ten cycles. A beta barium-borate-based commercial optical parametric amplifier (HE-TOPAS, Light Conversion Ltd.) is pumped by a Ti:sapphire-based chirped pulse amplification system with 60 fs FWHM pulse duration, 6 mJ pulse energy, and center wavelength of 790 nm. The 1 mJ, 75 fs (FWHM) idler from the TOPAS with a central wavelength of 1800 nm is focused into a 1-m-long, 400-μm-diameter, Ar-filled hollow core fiber (HCF). The 1800 nm pulses are spectrally broadened by self-phase modulation inside the fiber. By adjusting the gas pressure between 0 and 30 psi, the spectrum could be broadened from about 80 nm up to 400 nm, enough to support a laser pulse with FWHM duration down to two optical cycles at 1800 nm (12 fs). The spectrally broadened pulses then pass through fused silica windows with a thicknesses from 1.5 to 3 mm to compensate for the spectral dispersion introduced by the Ar gas in the HCF, the 1.5-mm-thick CaF_2 exit window of the HCF, the propagation in air, and the entrance CaF_2 window to the vacuum chamber [19]. A pair of GaAs plates at Brewster angle were used as polarizers after the beam collimation. Combining the GaAs plates with a half-wave plate before the HCF, the

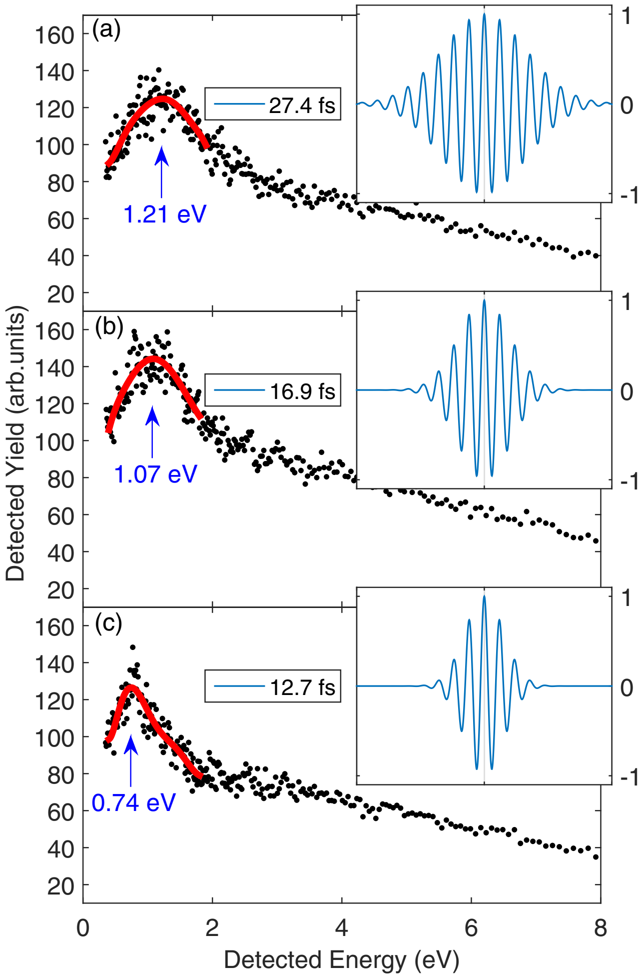

FIG. 1. Photoelectron spectra of krypton (black dots) for three different pulse durations at constant intensity. The curve over the data points is a polynomial fitting to smooth the data and localize the LES peak position. The line in the inset of each figure illustrates the electric field at different pulse durations. Note, the signal below 0.3 eV falls off rapidly due to the transfer function of the electron spectrometer.

laser intensity is controlled precisely to maintain a constant field strength at different pulse durations by adjusting the pulse energy while monitoring the $2U_{\mathrm{p}}$ and $10U_{\mathrm{p}}$ classical cutoffs in the photoelectron spectrum, where U_{p} is the pondermotive energy.

Pulse durations are determined using a home-built all-reflective second harmonic generation, frequency resolved optical gating apparatus [20,21]). Then, for each pulse duration, angle-resolved photoelectron spectra within $\pm 1^{\circ}$ to polarization direction were measured with a time-of-flight (TOF) spectrometer [10]. Argon and krypton gases are fed into the TOF chamber through a leak valve. The peak intensity of the field is kept constant (within $\pm 10\%$) at different pulse durations. We extract the LES peak positions as illustrated in Fig. 1 for krypton illuminated with a constant peak intensity of 8.3×10^{13} W/cm^2, i.e., constant U_{p}, and different pulse durations. The corresponding LES positions for krypton and argon and for different laser intensities normalized to the

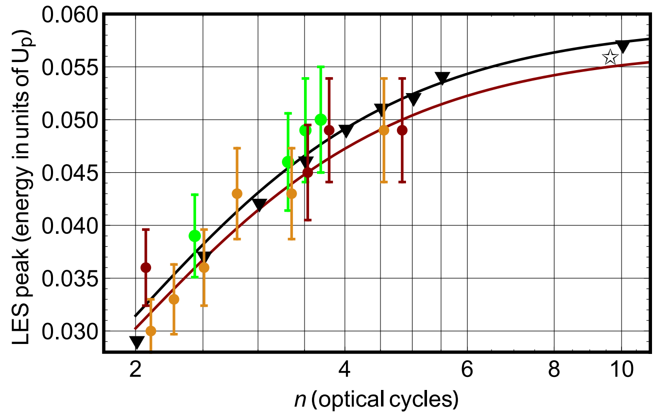

FIG. 2. LES peak energy positions for a frequency of $\omega = 0.0253$ a.u. (corresponding to a wavelength of 1800 nm) normalized to the peak pondermotive energy $U_{\mathrm{p}} = F_0^2/(2\omega)^2$ as a function of laser pulse duration in terms of optical cycles $n = T\omega/(2\pi)$. Data is for krypton [$U_{\mathrm{p}} = 20$ eV, green (light gray) and 25 eV, orange (gray)] and argon [31 eV, black triangle and 55 eV, red (black circle)]. Circles are from experiment, triangles are from a numerical classical calculation as in [15], and lines are analytical results (see text). Theoretical results are focus averaged and shifted down by 15% to match the experiment. The star is adapted from the experiment by Blaga *et al.* [10] for argon with $U_{\mathrm{p}} = 35.6$ eV.

respective pondermotive energy U_{p} are plotted in Fig. 2 versus the pulse duration expressed in optical cycles.

The results indicate that (i) the LES position depends sensitively on the pulse duration, but (ii) only weakly on the target and other laser parameters, once the trivial dependence on U_{p} is factored out, and (iii) that the CEP-averaged experimental results agree quite well with our classical numerical calculation, laser focus averaged over a Gaussian pulse profile but at fixed zero CEP (triangles). The fact that the CEP-fixed numerical results agree with the experimental ones, which can be interpreted as an average over the CEP due to the Gouy phase shift along the Gaussian laser focus is not obvious, especially considering the pronounced CEP dependence of the LES for short pulses [15,16].

The expression for our analytical results (solid lines in Fig. 2) directly explain and illustrate points (i)–(iii). Although easily generalizable to higher order LES recollisions and the VLES, we restrict ourselves here for the sake of clarity to the principal LES peak. To this end we consider a free electron moving in one dimension in a time-dependent vector potential $A(t)$ with a Hamiltonian

$$H = [p + A(t)]^2/2 \tag{1}$$

whereby atomic units are used unless stated otherwise. Hamilton's equations produce a constant drift momentum which yields the value $p_{\mathrm{d}} = -A(t_0)$ if we assume that the velocity $\dot{x}(t_0) = 0$ (tunneling regime). Integration of $\dot{x}(t) = p_{\mathrm{d}} + A(t)$ in time from t_0 to the recollision time $t^* = t_0 + \Delta t$ gives

$$x(t^*) = p_{\mathrm{d}}\, \Delta t + \int_0^{\Delta t} dt\, A(t_0 + t). \tag{2}$$

For a soft recollision leading to the LES observed in the experiment, we get $\Delta t = 3\pi/\omega$. This follows from the fact that both tunneling and recollision occur when the vector potential $A(t)$ is near zero [14]. According to (2) the recollision condition $x(t^*) = 0$ requires a particular drift momentum which can be interpreted as the average of the time-dependent vector potential over Δt, the time it takes the electron from the tunnel exit to the soft recollision,

$$p = -\frac{1}{\Delta t}\int_0^{\Delta t} dt\, A(t_0 + t)\,. \tag{3}$$

We will demonstrate that the position of the LES maps out directly via Eq. (3) the time dependence of the vector potential which underscores the universal dynamical validity of the very simple soft-recollision picture. We obtain an analytical expression for (3) by taking the vector potential as the derivative of the quiver amplitude with Gaussian envelope [22]:

$$A(t) = -\frac{A_0}{\omega}\frac{d}{dt}[e^{-2\ln 2(t/T)^2}\cos(\omega t - \phi)], \tag{4}$$

where A_0 is the peak amplitude of the vector potential, ω is the laser frequency, and ϕ the CEP. We have checked that the results are insensitive to the actual pulse shape (Gauss and $\cos^2$ pulses) with (4) giving particularly simple analytical expressions. The pulse duration T will be expressed below by the number of cycles n in the pulse, i. e., $T = n\,2\pi/\omega$. For the vector potential (4) the field is maximal at $t_0 \approx \phi/\omega$. Consequently, the pulse-duration-dependent drift momenta according to (3) is simply

$$p(n,\phi) = \zeta(n,\phi)p_\infty, \tag{5a}$$

$$\zeta(n,\phi) = \frac{1}{2}(e^{-c\phi^2/(n\pi)^2} + e^{-c(3+\phi/\pi)^2/n^2}), \tag{5b}$$

$$p_\infty = \frac{2A_0}{3\pi} \tag{5c}$$

with the LES peak position p_∞ for a cw ($n\to\infty$) pulse [14] and the abbreviation $c \equiv \ln 2/2$. Equation (5) predicts a reduction of the LES peak momenta for finite pulse duration by the factor $\zeta(n,\phi)$ compared to a cw pulse ($\zeta = 1$) and a pronounced dependence on the CEP ϕ as illustrated in Fig. 3. However, the $\zeta(n,\phi)$ for $\phi = \pm\pi/2$ sit almost symmetrically next to the central curve for zero CEP which is easily seen from a Taylor expansion of Eq. (5b) about $\phi = 0$, since the quadratic term is already suppressed by a factor $(n\pi)^2$. This suggests that up to a small correction proportional to n^{-2}, CEP averaging $\bar{\zeta}(n) = \frac{1}{\pi}\int_{-\pi/2}^{+\pi/2} d\phi\,\zeta(n,\phi)$ will give the zero CEP result $\bar{\zeta}(n) \approx \zeta(n,0)$,

$$\bar{p}(n) \approx p_\infty\bar{\zeta}(n) \equiv \frac{p_\infty}{2}(1 + e^{-c(3/n)^2}) \tag{6}$$

for the CEP-averaged LES momenta, indistinguishable from exact CEP averaging (gray line in Fig. 3) for $n \geqslant 2$.

The laser properties enter in Eq. (6) only as an overall scale. Hence, the LES dependence $\bar{\zeta}(n)$ on the pulse duration is universal in the sense that the ratio of the LES position for two different pulse durations $\bar{p}(n)/\bar{p}(n') = \bar{\zeta}(n)/\bar{\zeta}(n')$ depends neither on properties of the light or of the target. Remarkably, this property of the LES time dependence is largely preserved even under modifications of the target (Coulomb) potential

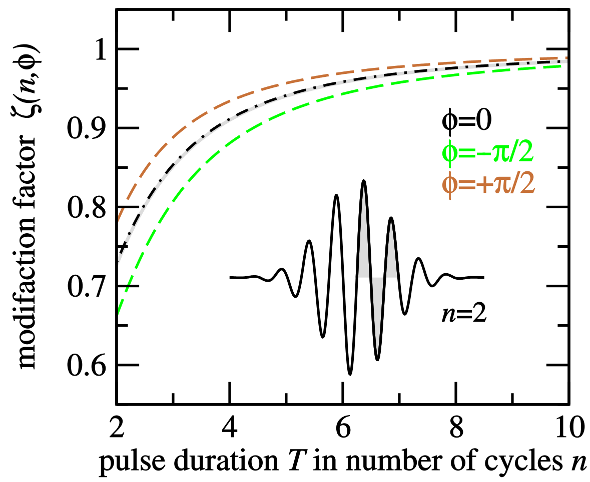

FIG. 3. Time dependence of $\zeta(n,\phi)$ from (5b) for the carrier-envelope phases $\phi = 0, \pm\pi/2$. The trained eye sees a gray line, which shows the exact CEP average given by $\bar{\zeta}_{\rm ex}(n) = n/4\sqrt{\pi/c}\,(2\,C_1 + C_7 - C_5)$ with $C_\alpha = \mathrm{erf}[\alpha\sqrt{c}/2n]$. Remember that $c = \ln 2/2$.

and the experimentally necessary laser focus averaging which we will introduce below.

First, in the numerical calculations all LES peaks appear slightly redshifted compared to Eq. (6), since the electron suffers a small momentum reduction before finally leaving the (attractive) potential. This momentum change along the laser polarization can be expressed as

$$\delta p = -\int_\infty^\infty dt \frac{d}{dz}V(\mathbf{r})\Big|_{\mathbf{r}_*(t)} = -C_{3\mathrm{D}}\left(\frac{\omega^3}{F}\right)^{1/5} \tag{7}$$

with $\mathbf{r}_*(t) = \{\rho,z\} = \{\rho_*, z_* + \frac{F}{2}[t - t_*]^2\}$, the soft-recollision trajectory without potential, and the soft collision at $\{\rho_*,z_*\} = (F\omega^2)^{1/5}\{2.723, -1.521\}$ for a 3D Coulomb potential with the value $C_{3\mathrm{D}} = 0.073$ of the integral (7) [14]. The resulting LES momentum redshift $\delta p/p_\infty$ amounts to about 3% for the laser parameters considered here and agrees very well with the classical numerical result (not shown).

Second, in the experiment the photoelectrons emerge from the full Gaussian focus, so volume averaging is also required. Since we are interested here only in the position of the LES peak, it is sufficient to consider ionization from the center half-cycle. The ionization rate is given by the static field expression $P(\tilde{F}) = 4/|\tilde{F}|\exp(-2/3|\tilde{F}|)$ [23]. $\tilde{F}$ is the reduced field strength $\tilde{F} = F/[2E_{\rm ip}]^{3/2}$ with the instantaneous field F and the ionization potential $E_{\rm ip}$. Using the volume intensity distribution in a Gaussian focus [24], with $I_0 = F_0^2$ the maximal intensity at the center, $\eta \equiv F/F_0$ and $\tilde{F}_0 = F_0/[2E_{\rm ip}]^{3/2}$, the ionization rate reads

$$P_{\tilde{F}_0}(\eta) \propto \exp\left[-\frac{2}{3\tilde{F}_0\eta}\right]\frac{4\eta^2 + 2}{\eta^5}\sqrt{1-\eta^2}. \tag{8}$$

The focal-averaged LES-peak positions are obtained by integration $\bar{p} \propto \eta$ from Eq. (6) with the weight Eq. (8) between 0 and 1. Realizing that for relevant parameters $\tilde{F}_0 < 1/10$ one can expand Eq. (8) about $\eta = 1$, the averaging can be done analytically [25] to give $P(n) = \bar{p}(n)/C_{\mathrm{av}}$ where

$$C_{\mathrm{av}} = 1 + \frac{9}{4}\frac{F_0}{(2E_{\mathrm{ip}})^{3/2}}, \quad (9)$$

which gives another reduction $1/C_{\mathrm{av}}$ of the LES momentum of about 10% for the laser parameters and targets considered here.

The $F^{1/5}$ dependence of δp is too weak for a significant averaging effect. At first sight, δp and the focal averaging seem to destroy the universality of the pulse duration dependence of the LES with additional dependence on laser and target (E_{ip}) properties, respectively. However, taking into account the fast convergence of Eq. (6) with n to p_∞ and the fact that δp is a small correction to $p(n)$, it turns out that the universality of the LES pulse dependence is preserved, since to a good approximation, the δp-shifted and focus-averaged LES momentum assumes the original form (6) with p_∞ replaced by p_∞^{eff},

$$p_{\mathrm{LES}}(n) = \left(\frac{p_\infty}{C_{\mathrm{av}}} + \delta p\right)\bar{\zeta}(n) \equiv p_\infty^{\mathrm{eff}}\,\bar{\zeta}(n). \quad (10)$$

This is the final expression for the LES peak momentum position. In Fig. 2 we plotted LES-energy positions scaled by the ponderomotive energy, $p_{\mathrm{LES}}^2(n)/2U_{\mathrm{p}}$, with solid lines to compare with experiment. Note, that the corrections due to the potential δp and focus averaging C_{av} reduce the LES position in energy by about 25% compared to the "pure" result (6). The product form (10) restores the universal pulse duration dependence $\bar{\zeta}(n)$ of the LES and shows how the laser parameters and the binding energy of the electron influence the LES. Expanding (10) in ω for fixed Keldysh parameter $\gamma = \sqrt{E_{\mathrm{ip}}/2U_p}$,

$$p_{\mathrm{LES}}(n) = p_\infty\bar{\zeta}(n)\left(1 - \frac{9}{16\gamma^3}\frac{\omega}{U_{\mathrm{p}}} - \frac{3\pi C_{3\mathrm{D}}}{2^{11/5}}\frac{\omega^{2/5}}{U_{\mathrm{p}}^{3/5}}\right), \quad (11)$$

shows that in addition to the strong-field nature of the LES, it is also a phenomenon unique with the long-wavelength laser field.

In summary, we have presented experiments which reveal the dependence of the LES peak energy on the laser pulse duration. A completely analytical formulation in good agreement with the numerical and experimental results shows that the pulse duration dependence is universal since all residual dependencies on the laser properties and the target can be cast into a factor of proportionality which describes the position of the LES for long pulses with analytical target dependence and laser focus averaging also included. Utilizing this universal behavior, the pulse durations of MIR lasers could be calibrated by measuring the LES peak positions.

K.Z. acknowledges the support from the Presidential Fellowship of the Ohio State University. Y.H.L. and J.L.X. acknowledge the support of the Ohio Supercomputer Center. This work was supported by the National Science Foundation under Grant No. PHY-1304218.

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