

Nondipole Coulomb sub-barrier ionization dynamics and photon momentum sharing

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The nondipole under-the-barrier dynamics of the electron during strong-field tunneling ionization is investigated, examining the role of the Coulomb field of the atomic core. The common analysis in the strong-field approximation is consequently generalized to include the leading light-front nondipole Coulomb corrections and demonstrates the counterintuitive impact of the sub-barrier Coulomb field. Despite its attractive nature, the sub-barrier Coulomb field increases the photoelectron nondipole momentum shift along the laser propagation direction, involving a strong dependence on the laser field. The scaling of the effect with respect to the principal quantum number and angular momentum of the bound state is found. We demonstrate that the signature of Coulomb induced sub-barrier effects can be identified in the asymptotic photoelectron momentum distribution via a comparative study of the field-dependent longitudinal momentum shift for different atomic species with state-of-the-art experimental techniques of midinfrared lasers.

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Exchanges of photon's energy, momentum, and angular momentum with charged particles are fundamental building blocks of light-matter interactions. In the presence of an intense laser field, photon energy absorption can trigger nonlinear processes like high-order harmonic generation [1,2], above-threshold ionization [3], and nonsequential double ionization [4]. Meanwhile, the angular momentum transfer from the laser beam is vital for generating vortex light [5,6], polarized electrons [7,8] and positrons [9], and nuclei vortices [10]. The absorption of the photon momentum has attracted considerable interest recently due to various breakthroughs in the experimental technique [11–18]. High precision measurements raised questions on how the absorbed photon momentum is partitioned between the photoelectron and the parent ion. The analysis relies on investigating the photoelectron momentum distribution (PMD) along the laser propagation direction. The coincidence measurement of the photoelectron and its parent ion provides a direct check of the momentum conservation law in the microscopic world [19].

It follows from energy and momentum conservation that in the simple-man model of tunneling ionization [2], when the electron appears in the continuum with a vanishing momentum due to a circularly polarized laser pulse, the total absorbed photon momentum is shared between the ion and electron as I_p/c and U_p/c , respectively [11,20], where I_p is the ionization energy, U_p the ponderomotive potential, and

c the speed of light. However, in deviation from the simple-man picture, there is a longitudinal momentum transfer to the electron during tunneling due to the laser Lorentz force $\delta p_z \sim (\kappa/c)B_0\tau \sim I_p/c$, where $\kappa = \sqrt{2I_p}$ is the atomic velocity, $\tau = \gamma/\omega$ the tunneling formation time, $\gamma = \omega\kappa/E_0$ the Keldysh parameter, E_0 and B_0 the laser electric and magnetic fields, respectively, and ω their frequency. Atomic units are used throughout. Then, the most probable electron longitudinal momentum at the tunnel exit becomes $I_p/(3c)$ [14,21]. This is the case for a short-range atomic potential, when the electron begins the sub-barrier motion at the bound state with a longitudinal momentum $v_{zs} = -2I_p/(3c)$ [21,22]. The Coulomb field of the atomic core reshapes the tunneling barrier and, consequently, the nondipole momentum shift due to the sub-barrier dynamics. The latter is also affected by the momentum distribution of the bound state, which contributes into the birth probability of the quantum orbit at v_{zs} .

The long-range character of the Coulomb field of the atomic core is known to play a key role in strong-field ionization. It enhances the tunneling probabilities [23–26]. In the continuum, it is responsible for the low-energy [27–33] and high-energy structures [34,35] in the PMD and the interplay between nondipole and Coulomb effects [12,13,36–39]. The sub-barrier dynamics can induce specific effects in tunneling ionization [7,8,21,40,41]. For instance, the sub-barrier nonadiabatic dynamics results in a transverse momentum shift and polarized photoelectrons [7,8]. The sub-barrier Coulomb effect can induce a phase shift of the quantum orbit [40], disturbing the holography pattern. However, there is yet no conclusion on how the under-the-barrier Coulomb action affects the nondipole momentum shift. While simulations via the time-dependent Schrödinger equation (TDSE) [42–44] and time-dependent Dirac equation [16] give accurately the final longitudinal momentum shift, they do not provide intuitive insight on the underlying sub-barrier dynamics.

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In this Letter, we investigate the electron's sub-barrier nondipole dynamics in strong-field tunneling ionization within an enhanced analytical description including the role of the Coulomb field of the atomic core. The sub-barrier dynamics is described within the light-front [45] nondipole Coulomb-corrected strong-field approximation (SFA), while the continuum one via the light-front classical trajectory Monte Carlo (CTMC) simulation, disentangling the effect of the sub-barrier Coulomb action from that in the continuum. We found that the sub-barrier Coulomb effect induces an increase of the photoelectron nondipole longitudinal momentum shift and discuss its counterintuitive physical origin. The dependence of the Coulomb effect on the angular momentum of the bound state is analyzed. Finally, we discuss how the Coulomb effects in the sub-barrier nondipole momentum shift can be observed experimentally using midinfrared laser fields.

Our description of the tunneling process is based on TDSE in the Göppert-Mayer gauge [46]

$$i \frac{\partial}{\partial t} \psi(\mathbf{x}, t) = \left[\frac{1}{2} [\mathbf{p} + \mathbf{A}(\mathbf{x}, \eta)]^2 + V(\mathbf{x}) - \phi(\mathbf{x}, \eta) \right] \psi(\mathbf{x}, t), \quad (1)$$

where $V(\mathbf{x}) = -\frac{Z}{|\mathbf{x}|}$ is the Coulomb potential of the atomic core, $A^\mu = (\phi, \mathbf{A})$ the laser four-vector potential in the Göppert-Mayer gauge, with $\phi = -\mathbf{x} \cdot \mathbf{E}(\eta)$ and $\mathbf{A}(\mathbf{x}, \eta) = \hat{\mathbf{z}}\phi/c$, $\mathbf{E}(\eta) = E_0[\hat{\mathbf{x}} \cos(\omega\eta) - \hat{\mathbf{y}} \sin(\omega\eta)] \exp(-2 \ln 2 \frac{\eta^2}{T^2})$ the laser field of circular polarization with $L = 4T$ and T is the optical cycle, and with the light-front coordinate $\eta = t - z/c$. The Hamiltonian in Eq. (1) is accurate up to the $1/c$ order, suitable in the considered parameter regime. The deviation from the fully relativistic theory [47–50] is given by I_p/c^2 terms for the sub-barrier and U_p/c^2 terms for the continuum dynamics. We apply light-front nondipole Coulomb-corrected SFA [51], which corresponds to the nondipole expansion of the relativistic theory [50]. The nondipole transition amplitude in the Göppert-Mayer gauge reads [59]

$$M_{\mathbf{p}} = -i \int d\eta \frac{d^3\mathbf{x}}{(2\pi)^{3/2}} e^{-i \int_{\eta}^{\infty} d\eta' \frac{\frac{1}{2} \tilde{\mathbf{p}}(\eta')^2 + I_p}{1 - \frac{p_-}{c}}} \mathcal{F}_C^*(\eta, \mathbf{x}) \times (1 - \tilde{p}_-/c) e^{-i \tilde{\mathbf{p}}(\eta) \cdot \mathbf{x}} \cdot \mathbf{E}(\eta) \psi_{\kappa lm}(\mathbf{x}), \quad (2)$$

where $\tilde{\mathbf{p}}(\eta) = (\mathbf{p}_\perp + \mathbf{A}_\perp(\eta), p_- - \frac{I_p}{c})$, with $p_- = p_z - \frac{p_\perp^2}{2c}$ being the light-front momentum, $\mathbf{A}_\perp(\eta) \equiv -\int_{\eta}^{\infty} d\eta' \mathbf{E}(\eta')$, and $\psi_{\kappa lm}(\mathbf{x})$ is the bound-state wave function. Note that $p_z/c \approx p_-/c \approx \tilde{p}_-/c$ up to the $1/c$ order. The use of the Göppert-Mayer gauge in the employed SFA is justified as it provides the accurate Coulomb corrected Perelomov-Popov-Terent'ev (PPT) ionization rate [23–25], which is well confirmed experimentally. The influence of the Coulomb field on the sub-barrier continuum electron is described by the Coulomb correction (CC) factor:

$$\mathcal{F}_C(\eta, \mathbf{x}) = \exp \left\{ i \int_{\eta}^{\infty} d\eta' V(\mathbf{x}(\eta', \eta')) \right\} \left(1 - \frac{p_-}{c} \right). \quad (3)$$

With saddle-point integration of Eq. (2), the differential ionization rate in the adiabatic regime is obtained

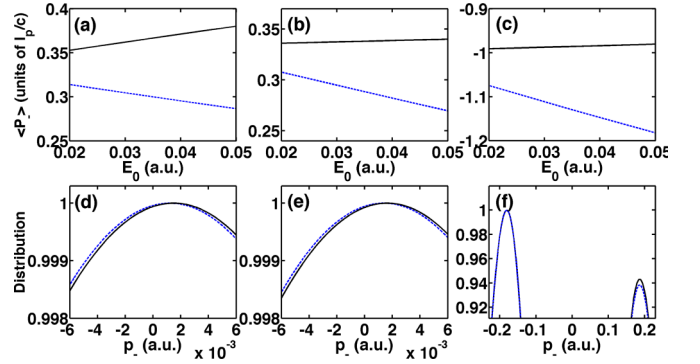


FIG. 1. Sub-barrier Coulomb effect, neglecting the continuum influence, in the quasistatic regime: (a)–(c) The expectation value $\langle p_- \rangle$ vs E_0 ; (d)–(f) PMDs at the laser intensity $I = 10^{14}$ W/cm². The initial states are (a),(d) hydrogen atom with quantum numbers (1,0,0) and argon atom (b),(e) with (3,1,1), and (c),(f) with (3,1,0). The solid black line includes the sub-barrier Coulomb effect, while the dashed blue line is the case of a short-range potential.

[51]:

$$\begin{aligned} \Gamma^{(n,l,m)}(\eta, v_\perp, p_-) &= \frac{\kappa}{\tilde{\kappa}} \frac{2^{2v+2} C_{\kappa l}^2 \kappa^{6v}}{(1 - \frac{p_-}{c})^{2v} E(\eta)^{2v}} \\ &\times |Y_{lm}(\tilde{\mathbf{p}}(\eta_s))|^2 \exp \left[-\frac{2}{3} \frac{\tilde{\kappa}^3}{E(\eta)(1 - \frac{p_-}{c})} \right], \end{aligned} \quad (4)$$

where (n, l, m) are the atomic quantum numbers (the angular momentum projection is determined with respect to the laser propagation axis), $E(\eta) = |\mathbf{E}(\eta)|$, $\tilde{\mathbf{p}}(\eta_s)$ is the saddle-point momentum, $v = Z/\kappa$ the effective principal quantum number, $v_\perp$ the initial velocity component in the polarization plane, $C_{\kappa l}^2 = \frac{2^{2v-2}}{v(v+l)!(v-l-1)!}$, and $\tilde{\kappa} = \sqrt{\kappa^2 + \tilde{p}_z^2 + v_\perp^2}$.

To implement the light-front CTMC simulations, we sample over all possible ionization instants, tunneling exits, and initial momenta as initial conditions for the classic equation of motion

$$\frac{dQ}{d\eta} = \{Q, H\}, \quad \frac{dP}{d\eta} = \{P, H\}, \quad (5)$$

where $\{A, B\}$ is the Poisson bracket, with the canonical coordinates $Q = (x, y, z)$ and $P = (p_x, p_y, p_-)$. Trajectories are weighted by $\Gamma^{(n,l,m)}$ of Eq. (4). Accurate to the $1/c$ order, we have the classical light-front Hamiltonian

$$H = \frac{(\mathbf{p}_\perp + \mathbf{A}_\perp)^2 + (p_- + V/c)^2}{2(1 - \frac{p_- + V/c}{c})} + V. \quad (6)$$

The tunneling exit $\mathbf{x}^e = -\frac{I_p}{1 - p_-/c} \frac{\mathbf{E}(\eta)}{E(\eta)^2}$ is obtained by taking the real part of the complex trajectory [51].

Let us first discuss the sub-barrier Coulomb effect. Neglecting the Coulomb field effect in the continuum leads to the conservation of $P = (p_x, p_y, p_-)$. We plot the expectation value of p_- in Fig. 1. From Eq. (4), we derive $\langle p_- \rangle$ for an s state in the leading-order correction of E_0/E_a [51]:

$$\langle p_- \rangle = \frac{I_p}{c} \left[\frac{1}{3} + (2v - 1) \frac{E_0}{E_a} \right], \quad (7)$$

with the atomic field $E_a = \kappa^3$. Note that the leading $I_p/(3c)$ term can be derived only employing the binding energy I_p and classical Lorentz sub-barrier dynamics in imaginary time without any extra information about the binding potential and any time or spatial dependencies of the laser field; see Sec. IB.2 in [51]. Apart from the constant $I_p/(3c)$ [14,21,42,59], the sub-barrier Coulomb effect yields an increase in the momentum shift, despite the attractive nature of the parent ion, and induces a strong dependence on the laser field and the principal quantum number of the bound state (Fig. 1).

The active electrons of most rare-gas atoms are in the p state. For instance, the ionization of the argon atom is dominated by signals from $(3, 1, \pm 1)$ orbits, while the nondipole shift for $(3,1,0)$ is more distinctive:

$$\langle p_- \rangle = \frac{I_p}{c} \left[-1 + (6v - 5) \frac{E_0}{E_a} \right]. \quad (8)$$

The physical origin of the Coulomb field effects for the nondipole sub-barrier dynamics is encoded in the exponent and the prefactor of the ionization amplitude of Eq. (2). The action contributed by the kinetic part, i.e., $\frac{1}{2} \tilde{\mathbf{p}}(\eta')^2 + I_p$ of Eq. (2), gives the tunneling exponent in Eq. (4). Due to the laser magnetic field, the tunneling electron obtains a momentum in the laser propagation z direction, which decreases the longitudinal tunneling energy and the tunneling probability, the latter being maximal at $p_-^m = I_p/(3c)$ [21]. We can estimate the contribution of the Coulomb action by evaluating Eq. (3) with quantum orbits starting at the saddle point of the SFA-matrix element $\mathbf{x}_s$ [60]. The momentum shift due to the sub-barrier Coulomb action $\mathcal{F}_C$ is evaluated by the η integration from the saddle-time η_s to the exit time $\eta_r = \text{Re}\eta_s$. The sub-barrier Coulomb action $\mathcal{F}_C$ contributes to the momentum shift, yielding a peak at $p_-^m > I_p/(3c)$; see Fig. 2(a). An essential sub-barrier CC for the nondipole momentum shift stems from matching the bound-state wave function in the Coulomb field to the tunneling wave packet. The prefactor accounts for the probability of the quantum orbit, which originates at the bound state with the momentum v_{zs} . The width of the momentum space wave function $\psi_{\kappa lm}(x_s, y_s, v_{zs})$ (in the mixed representation, at the starting point of the ionization x_s) in the Coulomb field is narrower than that in the short-range potential; see Fig. 2(b). The most probable v_{zs}^m is the peak of the function: $|\psi(x_s, y_s, v_{zs})|^2 \mathcal{W}(v_{zs})$, which is indicated by the grid lines in Figs. 2(b) and 2(c). The solid orange grid line in (b) includes the sub-barrier CC and is closer to the origin (larger v_{zs}), which yields an increase of the longitudinal momentum at the tunnel exit, as the total longitudinal momentum transfer from $\mathbf{x}_s$ to $\mathbf{x}^e$ is I_p/c .

The momentum shift and the corresponding CC depend essentially on the angular momentum of the bound state. We compare in Fig. 1 the expectation value of $\langle p_- \rangle$ with and without CC for quantum numbers $(1, 0, 0)$ (hydrogen) and $(3, 1, 0)$, $(3, 1, 1)$ (argon). There is a significant difference between s and p states. While the PMD of $(1,0,0)$ and $(3, 1, \pm 1)$ has only one peak, it has two peaks around $\pm \sqrt{E_0/\kappa}$ for $(3,1,0)$, which originate from the momentum distribution of the initial bound state; see Fig. 2(c). In the dipole theory, the two peaks are equally probable and $\langle p_- \rangle = 0$. The center of the window $\mathcal{W}$ is at $v_{zs} = -2I_p/(3c)$ in the nondipole

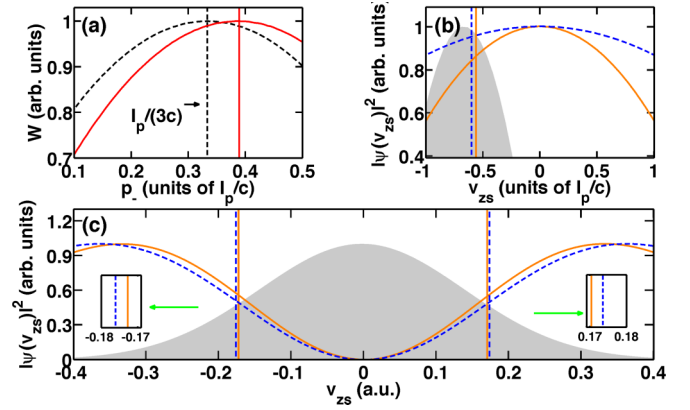


FIG. 2. (a) Nondipole CC ionization window $\mathcal{W}(p_-) \equiv |\mathcal{F}_C|^2 \exp[-\frac{2}{3} \frac{\tilde{\kappa}^3}{E(\eta)(1-p_-/c)}]$: (solid red) including the sub-barrier action, the peak is at $p_-^m > I_p/(3c)$ (solid red grid line); (dashed black) neglecting the sub-barrier Coulomb correction, $p_-^m \approx I_p/(3c)$ (dashed black grid line). The bound state wave functions at the starting point of the tunneling trajectory x_s : (b) for $(1,0,0)$ state and (c) for $(3,1,0)$ state; the wave function for the zero-range potential (dashed blue) and for the Coulomb case (solid orange); the tunneling window $\mathcal{W}$ is shadowed; $E_0 = 0.04$ a.u. The grid lines in (b) and (c) indicate the peaks of $\mathcal{W}(v_{zs})|\psi(x_s, y_s, v_{zs})|^2$ with (solid orange) and without the sub-barrier CC (dashed blue).

theory. The asymmetry of the window with respect to the origin results in suppression of the probability of the positive peak with respect to the negative one, yielding a negative longitudinal momentum $\langle p_- \rangle < 0$. After accounting for the atomic Coulomb field, the peaks of the wave function become closer, which results in an increase of the PMD peaks. The sub-barrier CC for the right peak is larger [Fig. 1(f)], which leads to a decrease of $\langle p_- \rangle$ by absolute value. Hence, for the state $(3,1,0)$, the exponent of Eq. (4) prefers the peak $p_- \approx -\sqrt{E_0/\kappa}$ due to the nondipole momentum shift I_p/c , while the prefactor prefers the peak $p_- \approx \sqrt{E_0/\kappa}$ due to the sub-barrier Coulomb field effect.

Next let us discuss the role of the Coulomb field in the continuum and the nonadiabatic effect; see Fig. 3. $P = (p_x, p_y, p_-)$ is no longer conserved and the momentum is reduced by absolute value as a result of the Coulomb momentum transfer mostly at the tunnel exit. For the states $(3, 1, \pm 1)$ (argon) the continuum CC decreases the momentum shift, however, approximately maintaining the slope of the field dependence [51]. For a smaller electric-field strength, the tunneling exit is farther away from the atomic core, while the photoelectron can stay a longer time around the nuclei. As a consequence, $\langle p_- \rangle$ decreases in the absolute value, while the slope of the curve remains less affected. For the state $(3,1,0)$, the continuum CC reduces the momentum shift for the peak at $p_- \approx \sqrt{E_0/\kappa}$ while increasing it for the peak at $p_- \approx -\sqrt{E_0/\kappa}$. The net result increases the total momentum shift. The qualitative behavior of the nonadiabatic corrections is similar (those without sub-barrier CC were considered in [61]). While the nonadiabatic effect decreases the absolute value of $\langle p_- \rangle$, it increases the slope of the field dependence. The contributions of the nonadiabatic and the continuum CC effects are calibrated via the difference of the momentum

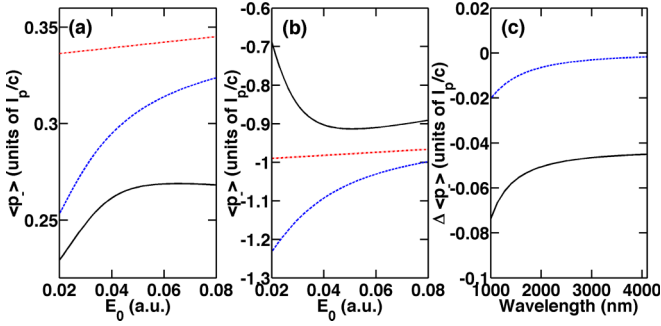


FIG. 3. Asymptotic light-front momentum $\langle p_- \rangle$ vs E_0 when the initial states are an argon atom with quantum numbers (a) (3, 1, 1) and (b) (3, 1, 0). The solid black lines include the continuum and the sub-barrier CC, the dashed-dotted red lines only the sub-barrier CC, and the dashed blue lines only the sub-barrier CC and nonadiabatic effects. (c) The contributions $\Delta \langle p_- \rangle$ of the nonadiabatic effect (dashed blue) and the continuum CC (solid black) to $\langle p_- \rangle$ vs the laser wavelength, for the laser intensity of 10^{14} W/cm 2 .

shift $\Delta \langle p_- \rangle$, calculated with and without these effects; see Fig. 3(c). Both the continuum CC and the nonadiabatic effects are suppressed at longer laser wavelengths.

It is possible to detect the sub-barrier CC signal with the current experimental capabilities similar to Refs. [14,15]. The momentum shift as well as the slopes of the field dependence are different after including the sub-barrier effect. In Fig. 4(a), we sample the laser intensity range 10^{14} – 2×10^{14} W/cm 2 for the linear fitting. We have carried out TDSE simulations based on the split-operator method, from which we obtain the scaling of the momentum shift as $\langle p_- \rangle \sim 1.44 \pm 0.22 (E_0/E_a)(I_p/c)$. After including both the nonadiabatic effect and Coulomb corrections, the light-front CTMC simulations yield $\langle p_- \rangle \sim 1.42 \pm 0.02 (E_0/E_a)(I_p/c)$. Without the sub-barrier CC, the CTMC simulation gives only $\langle p_- \rangle \sim 0.96 \pm 0.02 (E_0/E_a)(I_p/c)$. Thus the sub-barrier Coulomb dynamics induces a quite strong-field dependence in the nondipole longitudinal momentum shift (a large slope vs the laser field) partly distinguishable in an experiment.

Comparing atoms with different effective principal quantum numbers ν provides another possibility to observe the sub-barrier CC; see Fig. 4(b). The qualitatively robust signature of the sub-barrier CC is the increase of $\langle p_- \rangle \sim 2\nu \frac{E_0}{E_a} \frac{I_p}{c}$ with a higher effective principal quantum number of the active electron. For the ionization of Xe and Xe $^+$ with

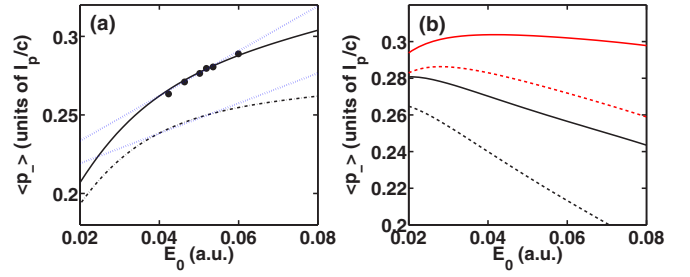


FIG. 4. (a) $\langle p_- \rangle$ vs E_0 for the ground state hydrogen atom when the laser wavelength is 800 nm. Circles are results of TDSE simulations. Dotted blue lines are linear fittings of the curves. (b) $\langle p_- \rangle$ vs E_0 for Xe and Xe $^+$ when the laser wavelength is 3000 nm. The solid lines include the full CC, while the dashed lines do not include the sub-barrier CC. The black lines are for Xe and the red lines are for Xe $^+$.

quantum numbers (3,1,1), we have $\nu = 1.06$ and $\nu = 1.60$, respectively. The sub-barrier CC signal is robust against the average over the magnetic quantum numbers. Thus the comparison of the longitudinal momentum shift and its dependence on the laser field for different atoms can unambiguously confirm the signature of the sub-barrier CC in an experiment. The longer laser wavelengths are more beneficial for the observation of the considered effects, as the masking effect of nonadiabaticity can be avoided at $\gamma < 1$; see Fig. 3(c), which shows an increase of the continuum CC due to nonadiabaticity at shorter wavelengths.

Concluding, after introducing the light-front nondipole Coulomb-corrected theory, we reveal a counterintuitive role of the Coulomb field of the atomic core in nondipole sub-barrier dynamics, which increases the photoelectron nondipole momentum shift despite its attractive nature, with consequences for the momentum partition of the absorbed laser photons. We demonstrate, disentangling the sub-barrier and continuum effects, that the Coulomb sub-barrier signatures can be observed in the asymptotic momentum distribution with long-wavelength laser fields by measuring the characteristic slope of the field dependence of the average longitudinal momentum in different gas targets.

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