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Journal Article**Author(s):**

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Publication date:

2021-10

Permanent link:

<https://doi.org/10.3929/ethz-b-000494190>

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Originally published in:

Nature Photonics 15, <https://doi.org/10.1038/s41566-021-00842-7>

Funding acknowledgement:

801459 - Fellowship Program of the NCCR MUST (National Competence Center for Research in Molecular Ultrafast Science and Technology) and the Cluster of Excellence RESOLV (EC)

1 Complete characterization of sub-Coulomb-barrier 2 tunneling with phase-of-phase attoclock

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14 ABSTRACT

Laser-induced electron tunneling, triggering a broad range of ultrafast phenomena such as the generation of attosecond light pulses, photoelectron diffraction and holography, has laid the foundation of strong-field physics and attosecond science. Using the attoclock constructed by single-color elliptically polarized laser fields, previous experiments have measured the tunneling rates, exit positions, exit velocities and delay times for some specific electron trajectories, which are mostly born at the field peak instant where the laser electric field and the formed potential barrier are stationary in terms of the derivative versus time. From the view of the wave-particle dualism, the electron phase under a classically forbidden, tunneling barrier has not been measured, which is at the heart
15 of quantum tunneling physics. Here we present a robust measurement of tunneling dynamics including the electron sub-barrier phase and amplitude. We combine attoclock technique with two-color phase-of-the-phase spectroscopy to accurately calibrate the angular streaking relation and to probe the non-stationary tunneling dynamics by manipulating a rapidly changing potential barrier. This phase-of-phase attoclock (POP attoclock) directly links the measured phase of the two-color relative phase with the ionization instant for the photoelectron with any final momentum on the “detector”, which allows us to reconstruct the imaginary tunneling time and the accumulated phase under the barrier. The POP attoclock provides a general time-resolved approach to access the underlying quantum dynamics in intense-light-matter interactions.

16 The tunneling of a micro-particle through a barrier is one of the most fundamental and ubiquitous quantum
17 processes. In strong-field ionization the oscillating electric field of laser pulses creates a time-dependent potential
18 barrier in atoms or molecules allowing the bound electron to tunnel. At the instant of the field peak the potential
19 barrier is the thinnest and stationary as the derivative versus time is zero. After the peak instant, the potential barrier
20 changes quickly on the attosecond timescale, still triggering a series of tunneling bursts or electrons with different
21 probabilities at different positions. This sub-cycle electron dynamics greatly enriches strong-field phenomena¹,
22 ranging from strong-field ionization to high-order harmonic emission. Previous measurements mainly focus on the
23 most probable tunneling burst which is born at the instant of the field peak, such as the efforts on the tunneling
24 time delay²⁻¹¹, exit velocities¹²⁻¹⁴ and exit positions¹⁵⁻¹⁷. In contrast, the tunneling dynamics under a classically
25 forbidden, non-stationary barrier is less explored, especially for the electron sub-barrier phase, which is essential to
26 fully understand and characterize this fundamental quantum mechanical process.

27 The sub-cycle tunneling bursts could be mapped to different emitting angles within elliptically or circularly
28 polarized light fields, which is the principle of the attoclock^{2,3}. There, the mapping relation between the final
29 photoelectron momentum \mathbf{p} and its ionization instant t relies on an important, but unproved assumption that the
30 angle-time mapping is angularly uniform. For the photoelectron with the maximum yield in the final momentum
31 space, it is clear that it originates from the tunneling burst born at the instant of the electric-field peak. However, as
32 for the photoelectron with a general momentum (e.g., see the red box in Figure 1a), actually one cannot determine
33 its ionization instant without the assumption of the uniform mapping. Indeed, as we shown in this work, this
34 assumption is not valid (i.e., the rotation angle of the electric field vector β_ω is not identical to that of the electron
35 momentum vector α_p in Figure 1a). To realize the time-resolved ability in the full momentum space, determining
36 the ionization instant of each non-stationary tunneling burst and probing its sub-Coulomb barrier dynamics become
37 the key challenges in ultrafast science.

38 Recently, the phase-of-phase photoelectron spectroscopy was introduced to extract the scattering structural
39 information of atoms, molecules and nanoparticles¹⁸⁻²³, in which the momentum-resolved electron yield is Fourier
40 transformed with respect to the relative phase between two-color laser fields to obtain the phase of the yield
41 oscillation, which is the so-called phase of the phase. The phase-of-phase spectrum provides rich dynamical
42 information than the static photoelectron spectra measured in the one-color field. However, the physical meaning
43 of the obtained phase of the phase is not straightforward and thus the application of this elegant time-resolved
44 photoelectron spectroscopy is limited.

45 Here we introduce phase-of-phase attoclock (POP attoclock) spectroscopy, i.e., combining the attoclock and

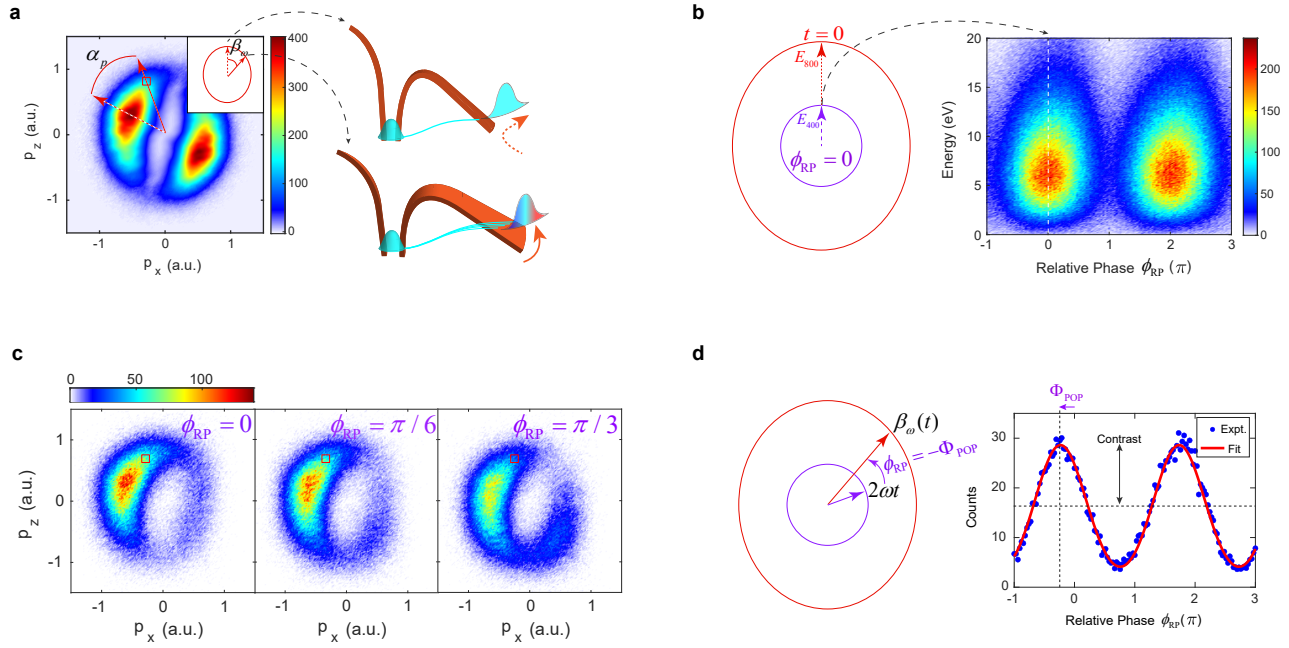


Figure 1: Phase-of-phase attoclock photoelectron spectroscopy. **a**, Measured photoelectron momentum distribution in a common attoclock configuration, in which a strong elliptically polarized 800-nm fundamental wave (FW, indicated by a red ellipse in the inset) is used to trigger a series of tunneling bursts following the rotating electric-field vector. The red dashed arrow and the red solid arrow identify the static tunneling at the field crest and the non-stationary tunneling in the falling edge, respectively. At the two ionization instants, the change tendency of the potential barrier is different, which is indicated by the crooked orange arrows near the barrier. Different sub-barrier dynamics gives rise to the different phase distribution on the emitting electron wave-packet (indicated by the color). **b**, Calibration of the relative phase in the POP attoclock configuration, in which a weak co-rotating circularly polarized second harmonic (SH) field is added into the common attoclock. The left panel shows the defined zero point for the relative phase, where the two fields both point to the vertical upward direction at the ionization instant of $t = 0$ so that the barrier will be bent to the maximum extent and its corresponding electron lobe in the final momentum plane (i.e., the left lobe) will reach the yield maximum. The right panel shows measured photoelectron energy spectrum for the left electron lobe as a function of the relative phase between two fields, from which we can accurately calibrate the relative phase between two fields. Here the emitting angle $\theta = \text{atan}(p_z/p_x)$ was integrated from 70° to 250° . **c**, Measured photoelectron momentum distributions at the relative phases of 0 , $\pi/6$, $\pi/3$. **d**, Measured phase-resolved electron yield (blue dots) for a given photoelectron momentum $[(p_x, p_z) = (-0.2, 0.74)$ a.u., indicated by the red boxes in **a** and **c**] and the fit curve by $A + P\cos(\phi_{\text{RP}} + \Phi_{\text{POP}})$ (red curve). Two characteristic parameters of the yield oscillation, the contrast P and the phase of the phase Φ_{POP} , are indicated in the plot. Since the direction of the SH clock hand is determined by $2\omega t + \phi_{\text{RP}}$, tuning the relative phase ϕ_{RP} (equivalent to rotating the clock hand) can build a mapping relation between the direction of the FW clock hand, $\beta_\omega(t)$, with the phase of the phase, Φ_{POP} , as illustrated in the left panel of **d**. When the ϕ_{RP} is tuned to be $-\Phi_{\text{POP}}$, the two clock hands are parallel and the ionization probability is the maximum.

46 the phase-of-phase spectroscopy, to resolve the ionization instant of photoelectrons with any momenta on the
 47 “detector” and further reconstruct the imaginary tunneling time and the sub-barrier phase acquired in the non-
 48 stationary tunneling process. A strong elliptically polarized fundamental wave (FW) is used to trigger the dynamical
 49 sub-cycle tunneling bursts, and a weak co-rotating circularly polarized second harmonic (SH) serves to perturb
 50 the potential barrier uniformly and to probe the time-dependent tunneling dynamics by adjusting the two-color
 51 relative phase. The synthesized electric field can be expressed as $E(t) = [E_\omega \cos(\omega t) + E_{2\omega} \cos(2\omega t + \phi_{RP})]\hat{z} +$
 52 $[\varepsilon E_\omega \sin(\omega t) + E_{2\omega} \sin(2\omega t + \phi_{RP})]\hat{x}$, where $\varepsilon = 0.80$ is the ellipticity of the FW. The relative phase between the
 53 two-color fields ϕ_{RP} was continuously controlled by a pair of fused silica wedges mounted on the piezo-driven
 54 motors. The peak intensity of the fundamental field is 1.0×10^{14} W/cm² ($E_\omega = 0.04$ a.u.; a.u. is atomic unit)
 55 and that of the second harmonic was controlled at a very low level about 1.0×10^{12} W/cm² ($E_{2\omega} = 0.004$ a.u.).
 56 We measured the photoelectron momentum distribution of single ionization from krypton atoms using cold-target
 57 recoil-ion reaction momentum spectroscopy (COLTRIMS)^{24,25}.

58 In the fundamental field solely, the photoelectron momentum distribution reveals two lobes, as illustrated in
 59 Fig. 1a, which can be understood by angular streaking by elliptically polarized fields (attoclock). The main lobes
 60 originate from strong-field tunneling ionization around the two field crests of one optical cycle. The emitting angle
 61 of the lobe center deviates from the direction of the laser vector potential at the peak instant, i.e., the x axis. The
 62 offset angle is caused by the Coulomb rotation in the continuum^{4,5}, while it could be related with the tunneling time
 63 delay and exit velocities⁶⁻¹¹. For all previous attoclock experiments, the offset angle was determined only for the
 64 momentum with the maximum yield, and then the angle-to-time mapping was assumed to be isotropic according to
 65 the semi-classical model. However, as discussed in this work, for the photoelectron with a general momentum in the
 66 final momentum plane, its offset angle is not simply identical to that with the maximum yield since its sub-barrier
 67 dynamics is different and its ionization instant will be modified accordingly. Therefore, the construction of the
 68 mapping relation between an arbitrary \mathbf{p} and t is the critical step to fully characterize the sub-barrier non-stationary
 69 tunneling dynamics.

70 In the geometry of POP attoclock, there are two temporal degrees of freedom, i.e., the ionization instant t and the
 71 two-color relative phase ϕ_{RP} . The direction of the FW clock hand is uniquely determined by the ionization instant
 72 and that of the SH clock hand depends on the two variables. The ionization instant is intrinsic, and we use the
 73 externally controllable relative phase to measure the ionization instant. Note that here the calibration of the relative
 74 phase does not rely on any theoretical models. At $\phi_{RP} = 0$, the two clock hands both point to the positive direction of
 75 z axis at $t = 0$ (see Figure 1b) and they will be oriented opposite along the z axis at the other peak instant $t = \pi/\omega$.

76 Thus, at $\phi_{\text{RP}} = 0$ the left upper electron lobe in the final momentum plane, which corresponds to the peak instant
77 of $t = 0$, should reach the yield maximum and the right lower lobe should achieve the yield minimum. In Figure
78 1b, we illustrate the angle-integrated energy spectrum for the left upper electron lobe as a function of the relative
79 phase, from which the phase calibration can be performed accurately. After the phase calibration, we illustrate the
80 photoelectron momentum distributions in the two-color fields at the relative phase of zero, $\pi/6$ and $\pi/3$ in Figure
81 1c. With the increase of ϕ_{RP} , the centroid of the momentum distribution gradually rotates counterclockwise. In
82 Supplementary Section 3, we illustrate the complete phase-resolved movie of the photoelectron angular distribution.
83 For each momentum [see the example of $(p_x, p_z, p_y) = (-0.2, 0.74, 0)$ a.u. in Figure 1d], the yield oscillation with
84 respect to the relative phase is close to a standard cosine function $A + P\cos(\phi_{\text{RP}} + \Phi_{\text{POP}})$, where A is the background
85 count, P is the contrast, and Φ_{POP} is the so-called phase of the phase¹⁸ ranging from $-\pi$ to π . Mathematically, the
86 value of Φ_{POP} represents the waiting time ($\Phi_{\text{POP}} < 0$) or the passed time ($\Phi_{\text{POP}} > 0$) of the appearing instant of the
87 yield peak with respect to the instant of $\phi_{\text{RP}} = 0$ during the yield oscillation. However, a clear physical meaning of
88 Φ_{POP} is not identified in previous phase-of-phase experiments¹⁸⁻²³.

89 In the phase-of-phase spectroscopy, re-scattering and sub-cycle interference cause the difficulty to dig the deeper
90 physical meaning of the phase of the phase. Here we show it is directly corresponding to the photoelectron ionization
91 instant in this POP attoclock configuration. The photoelectron yield Y at the momentum \mathbf{p} exponentially depends
92 on the strength of the synthesized electric field $|E(t)|$ at its ionization instant t . In the POP attoclock, $|E(t)|$ can be
93 reduced to $F_\omega(t) + E_{2\omega}\cos(2\omega t - \beta_\omega + \phi_{\text{RP}})$, saving to the first order of the small quantity $E_{2\omega}/E_\omega$, where $F_\omega(t)$
94 is the strength of the FW clock hand and $\beta_\omega(t)$ is its orientation angle. Then, one can obtain the momentum- and
95 phase-resolved yield $Y(\mathbf{p}, \phi_{\text{RP}}) \sim \exp[|E(t)|] \sim Y_0(\mathbf{p})[1 + E_{2\omega}\cos(2\omega t - \beta_\omega + \phi_{\text{RP}})]$ (see Supplementary Section 2
96 for the mathematical derivation), where $Y_0(\mathbf{p}) \sim \exp[\beta_\omega(t)]$ is the yield without adding the perturbative field. Thus,
97 the phase of the yield oscillation with respect to ϕ_{RP} is $\Phi_{\text{POP}} = 2\omega t - \beta_\omega(t)$, while the contrast of the yield oscillation
98 is $Y_0(\mathbf{p})E_{2\omega}$. For the FW field with high ellipticity, there is $\beta_\omega \approx \omega t$. Finally, the phase of the phase is directly linked
99 with the ionization instant, i.e., $\Phi_{\text{POP}} = \omega t$.

100 The principle of the POP attoclock can be understood intuitively. As illustrated in Figure 1d, varying the relative
101 phase is equivalent to rotating the SH clock hand. When the SH clock hand points to the same direction of the FW
102 clock hand, the photoelectron yield will achieve the maximum and thus this instant is marked. Because the scale of
103 the SH clock has been calibrated, one can use it to measure the direction of the FW clock hand, i.e., the ionization
104 instant.

105 The experimental phase-of-phase spectrum and the contrast spectrum for krypton atoms are presented in Figure

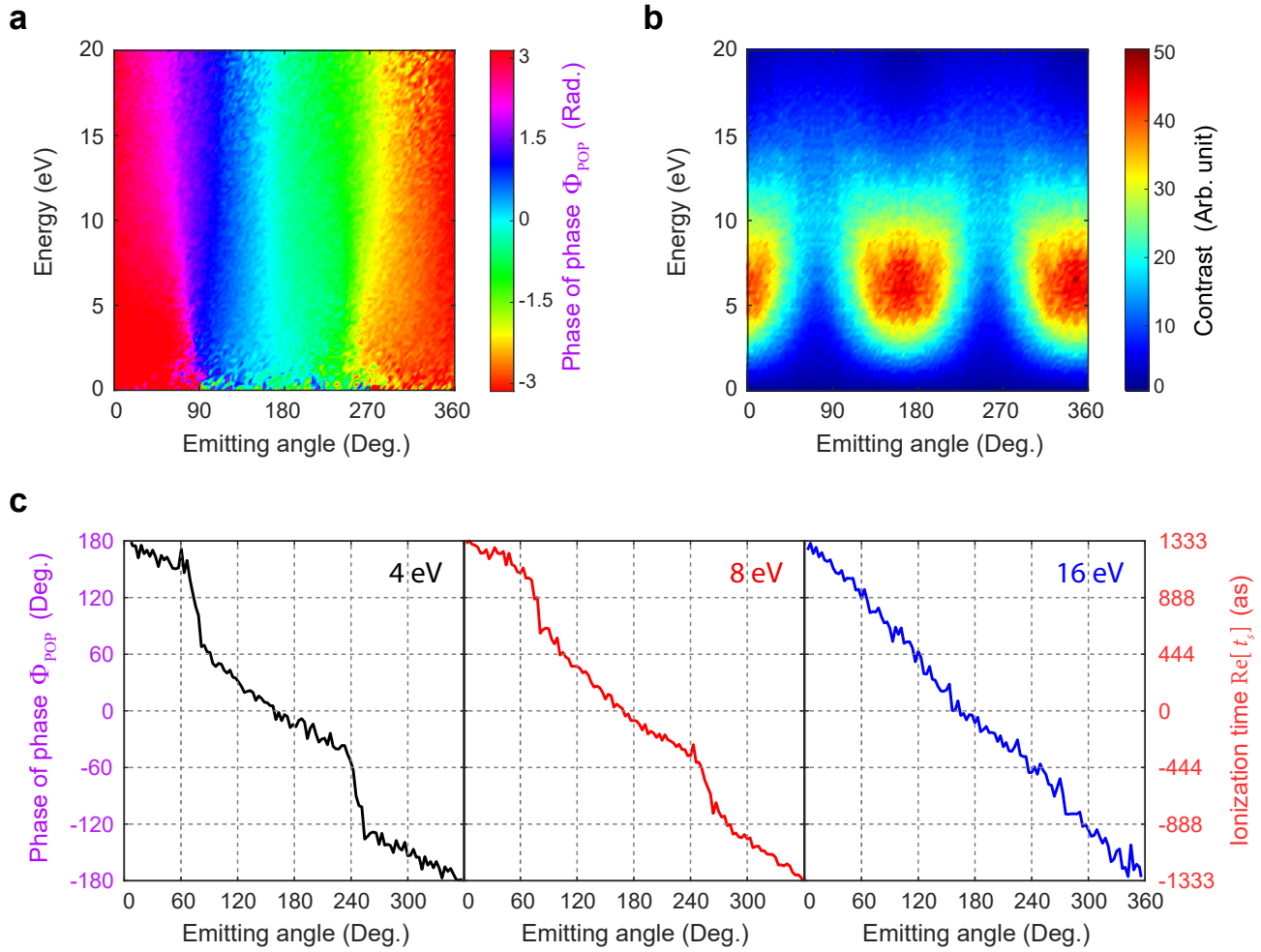


Figure 2: **Experimental results of the POP attoclock.** **a-b**, Measured phase-of-phase spectrum and contrast spectrum for krypton atoms in POP attoclock configuration, respectively. **c**, The cuts of Φ_{POP} along the energies of 4 eV, 8 eV and 16 eV. On the right vertical coordinate the phase of the phase is transformed to the ionization instant using $\Phi_{\text{POP}} = \omega t$.

106 2a-b, respectively, which were obtained by Fourier transforming 72 frames of the angle-resolved photoelectron energy
107 spectra with the resolution of $\Delta\phi_{\text{RP}} = \pi/12$. Here we define the photoelectron emitting angle as $\theta = \arctan(p_z/p_x)$,
108 and the momentum p_y along the laser propagation direction was integrated within $[-0.1, 0.1]$ a.u. in the off-line data
109 analysis. When the emitting angle varies 360 degrees, the phase of phase changes 2π radians or 360 degrees. This is
110 equivalent to that the photoelectron emitting angle will rotate 360 degrees when varying the relative phase by 2π .
111 More importantly, the slope of POP with respect to the photoelectron emitting angle reflects the electron rotation
112 speed in the momentum plane. In Fig. 2c, we illustrate three cuts of the phase-of-phase spectrum along the energies
113 of 4 eV, 8 eV and 16 eV, respectively, and convert Φ_{POP} to the ionization instant (see the right vertical coordinate of
114 the plots). The results directly show the mapping relation between the emitting angle and the ionization instant is
115 not angularly uniform, especially for the low-energy photoelectrons. The absolute value of the curve slope is less
116 than 1 when θ ranges from 120° to 240° and from -60° to 60° , which indicates the electron rotates slowly when it
117 arrives in the regions of the two main lobes (see Supplementary Section 3 for the animated photoelectron angular
118 distribution). The mapping curve jumps quickly in the low-yield transitional zone between the two main lobes,
119 where the FW clock hand points around the minor axis of the ellipse. For the high-energy electrons, their rotation
120 speed is close to be isotropic. The POP spectrum intuitively shows a dynamical phenomenon that photoelectrons
121 have a angle- and energy-dependent rotation speed even in a circular streaking field, which originates from the
122 non uniform mapping relationship between the emitting angle and the ionization instant. The contrast spectrum is
123 the extent of the response to the weak SH perturbation. Due to the perturbation is isotropic in POP attoclock, the
124 contrast spectrum faithfully reflects the static photoelectron angle distribution in the elliptical FW.

125 To validate our observation and explanation, we first resort to two different tunneling models, the classical
126 trajectory Monte-Carlo (CTMC) simulation^{26–28} that is widely used for tunneling from a static barrier, and the
127 strong-field approximation (SFA) model within the saddle-point approach²⁹ for the non-stationary tunneling. The
128 previous attoclock experiments are mostly explained using the CTMC model^{2,3,15}. In the CTMC model, there
129 is neither electron sub-barrier dynamics nor the sub-barrier phase. The potential barrier shape is dictated by the
130 instantaneous strength of the electric field adiabatically. At each ionization instant the tunneled electron burst has a
131 Gaussian distribution centered at zero on the transverse momentum and has no any longitudinal momentum. Thus,
132 the final momentum of the electron burst follows the opposite direction of the laser potential vector at the ionization
133 instant with an additional Coulomb rotation angle. Finally, the photoelectron emission angle is linearly changed
134 with respect to the ionization instant. In the CTMC simulation, we calculated a series of photoelectron momentum
135 distributions at different relative phases and then extracted the phase of the phase with the same procedure for

136 experimental data. The obtained POP spectrum shows that the degree of linearity between the ionization instant
 137 and the emitting angle is pretty good (i.e., the slopes of the mapping curves in Fig. 3b are all close to -1). The
 138 left-right shift between these mapping curves can be attributed to the Coulomb rotation. In SI, we present the CTMC
 139 result without including the Coulomb potential and the obtained mapping curves are all overlapped. Thus, previous
 140 attoclock experiments that were interpreted with the CTMC model is not very accurate because the sub-barrier effect
 141 is not seriously included.

142 By contrast, the non-stationary tunneling model (SFA) includes a time-dependent tunneling process on the
 143 imaginary time axis (see Fig. 4a). For the quantum path of each final momentum \mathbf{p} , its ionization time, i.e., the saddle
 144 point $t_s(\mathbf{p}) = t_r(\mathbf{p}) + t_i(\mathbf{p})i$, is a complex number governed by the saddle-point equation $[\mathbf{p} + \mathbf{A}(t_s)]^2/2 + I_p = 0$. The
 145 real part of the ionization time, t_r , represents the emitting instant from the barrier, which is the measured phase of
 146 the phase in this work. The imaginary part of the ionization time, t_i , is the so-called Keldysh tunneling time³⁰, which
 147 decides the duration time of the tunneling along the imaginary time axis. During the tunneling, i.e., progressing from
 148 t_s to t_r , the electron accumulates a time-dependent complex action $S[t_r(\mathbf{p})] = \int_{t_s}^{t_r} [\mathbf{p} + \mathbf{A}(t)]^2/2 + I_p dt$. The imaginary
 149 part of the action at the tunnel exit is associated with the probability of the quantum path by the exponential
 150 factor $\exp(-2\text{Im}[S])$, which corresponds to the measured contrast in POP attoclock. The real part of the action is
 151 the sub-barrier phase, which governs the initial phase of electron wave function in the continuum (see Fig. 4b).
 152 The laser field will excite the tunneling electron under the barrier and thus the quantum path model predicts a
 153 center-shifted Gaussian-like distribution for the transverse momentum and a nonzero time-dependent distribution
 154 for the longitudinal momentum at the exit^{13,31,32}. These non-stationary tunneling features will destroy the uniform
 155 $\theta - t_r$ mapping relation in the CTMC model. In SI, we illustrate the time-resolved distributions of the exit velocities
 156 from SFA and use them as the initial condition for the CTMC simulation. The obtained POP spectrum emerges the
 157 non-uniform mapping. More discussions about the relation and difference between the static and the non-stationary
 158 tunneling models are presented in the theoretical part of Methods.

159 In Fig. 3c, we show the momentum-resolved distribution of t_r , which is obtained directly by solving the saddle-
 160 point equation *in the fundamental field*. The saddle-point result agrees with the experiment and the energy-dependent
 161 mapping relations between θ and t_r are well reproduced. Therefore, this POP-attoclock could reveal the features of
 162 the non-stationary tunneling, which are usually hidden in static photoelectron spectra of attoclock with one-color
 163 elliptically or circularly polarized fields. In Supplementary Figure 5, we present the extracted phase of phase
 164 spectrum using POP attoclock from calculated photoelectron momentum distributions by numerically integrating
 165 the transition matrix element of SFA, following the experimental and CTMC procedure. The extracted result

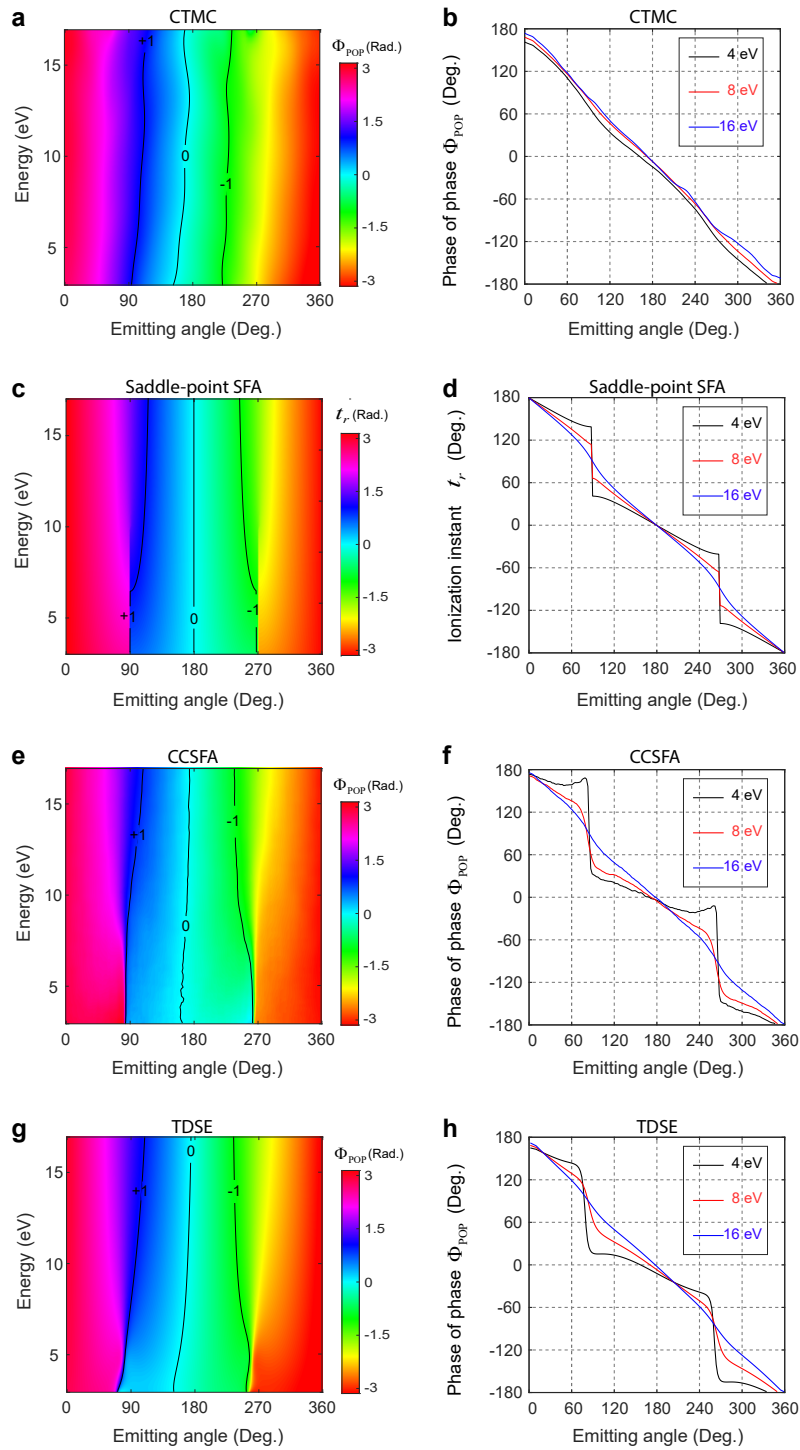


Figure 3: **Calculated phase-of-phase spectra.** **a,b,** The spectra of Φ_{POP} and the corresponding cuts along different energies from the CTMC simulation. **c,d,** The distribution of the real part of the saddle point and its corresponding cuts based on the SFA model. **e,f,** The results of the CCSFA simulation. **g,h,** The results of the TDSE simulation. The isolines of Φ_{POP} or $t_r = 1.0, 0,$ and -1.0 radian are plotted in **a,b,e,** and **g.**

166 agrees very well with the saddle-point result, which self-consistently proves that the POP attoclock can measure the
167 ionization instant. In circular or elliptical light fields, one needs to consider the influence of the magnetic quantum
168 number of the ground state of atoms, since the p_+ and p_- orbitals will display the ionization dichroism in static
169 photoelectron energy spectra^{33,34}. The magnetic quantum number will affect the probability of electron trajectories
170 via the pre-exponential factor of the transition matrix element but has no effect on the saddle point. Therefore, the
171 POP spectrum is less sensitive to the magnetic quantum number, and on the contrary the contrast spectrum will
172 show the same ionization dichroism features as the static photoelectron energy spectrum (see Supplementary Figure
173 4). To reveal the dependence of the POP spectrum on the nonadiabaticity of tunneling, in Supplementary Figure
174 6 we present the calculated POP spectra at other two wavelengths (1200 nm and 1800 nm) for the FW from the
175 SFA model. The result demonstrates that the non-uniform mapping gradually disappears with the increase of laser
176 wavelength or the nonadiabaticity of tunneling.

177 In all attoclock experiments, the rotation of the long-range Coulomb potential by elliptical or circular fields will
178 modify the electron momentum after tunneling, and mainly causes the offset angle in the final momentum plane. To
179 investigate the effect of the Coulomb potential on the POP spectrum, we have further performed the calculation
180 with the Coulomb-corrected strong-field model³⁵⁻³⁷ (CCSFA) and the numerical solution of the time-dependent
181 Schrödinger equation³⁸ (TDSE) (see Methods for the details). The CCSFA model is based on SFA and the effect of
182 the long-range Coulomb potential has been included. The TDSE results can be viewed as the benchmark for all the
183 models. From the results of these two models, the POP spectrum will be shift by the offset angle, as indicated by the
184 ioslines in Fig. 3e-g. Both models show the low-energy electrons will be more affected by the Coulomb rotation.
185 Although the Coulomb rotation will shift the POP spectrum left to some extent, basically it does not alter the slope of
186 the POP curves (see the CTMC result in Fig. 3a-b), which reflects the sub-barrier non-stationary tunneling dynamics.

187 Importantly, the tunneling process not only modifies the amplitude of the electron wavepacket (i.e., initial
188 momentum distributions), but also imprints a remarkable phase on it. The POP attoclock enables us to obtain the
189 electron sub-barrier phase information. This approach does not need the interference pattern on photoelectron
190 momentum distribution³⁹. With the measured phase of the phase and the contrast, we can further reconstruct the
191 other two quantities of the imaginary-time tunneling picture, t_i and $\text{Re}[S]$, according to the sub-barrier integration
192 equation of the action. Our reconstruction approach is very straightforward and robust, which has been justified
193 using the input of t_r and $\text{Im}[S]$ extracted from the photoelectron momentum spectra of SFA (see Supplementary
194 Figure 5). To isolate the sub-barrier dynamics from the classical dynamics in the continuum, one needs to subtract
195 the Coulomb rotation angle from the measured photoelectron emission angle, being consistent with the SFA model.

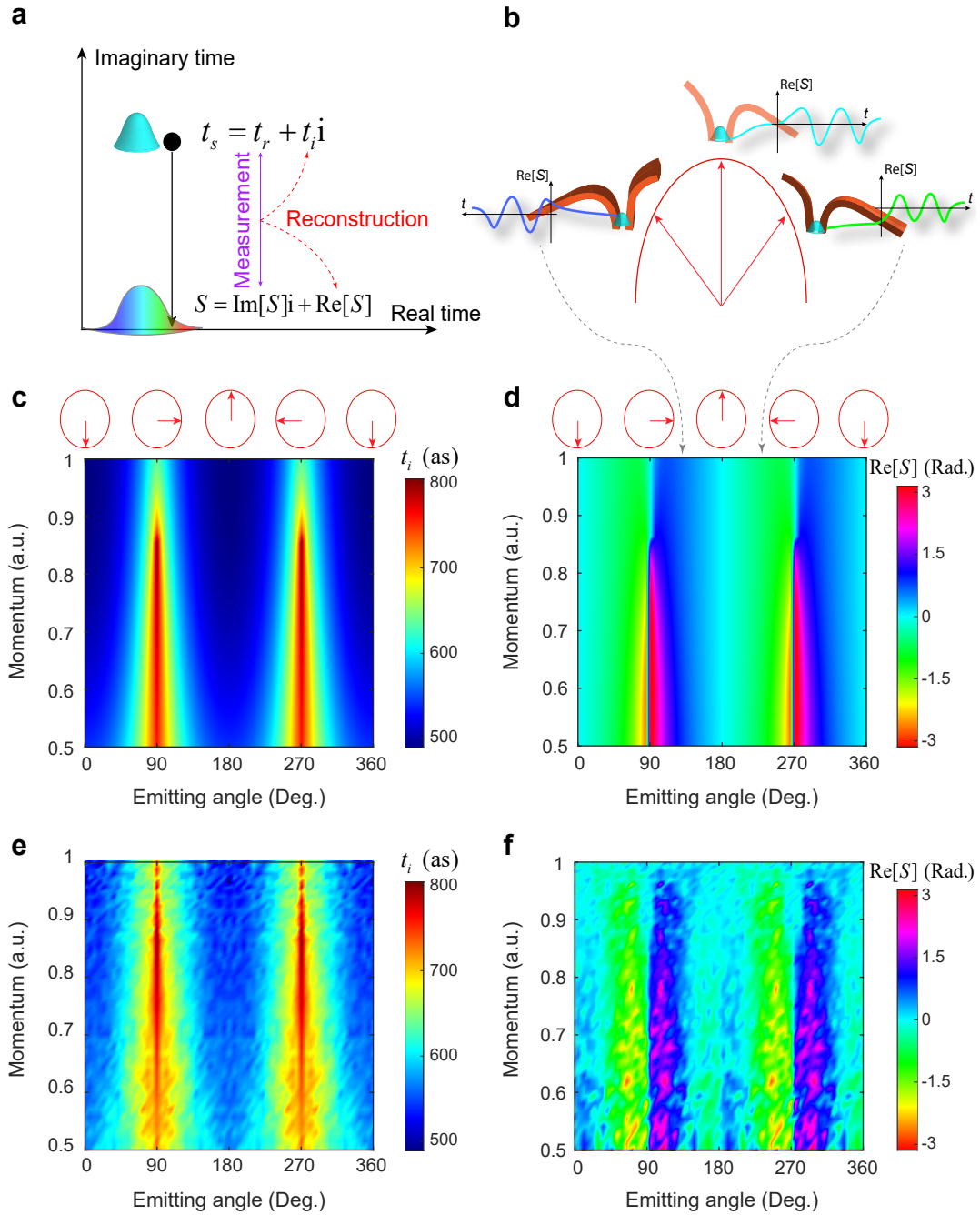


Figure 4: **Sub-barrier tunneling dynamics.** **a**, Tunneling picture and the reconstruction of t_i and $\text{Re}[S]$. **b**, Illustrations for the phase evolution at three ionization instants. The sub-barrier phase indicated by $\text{Re}[S]$ at the tunnel exit, is dependent on the dynamical change of the barrier. **c-d**, The final-momentum-resolved distribution of the imaginary tunneling time t_i and the sub-barrier phase $\text{Re}[S]$, respectively, which are obtained by solving the saddle-point equation and the integration equation of the action. **e-f**, The reconstructed distributions of t_i and $\text{Re}[S]$ using the measured phase-of-phase spectrum and the contrast spectrum, respectively.

196 The reconstructed results from experiments (Fig. 4e-f) agree well with the theoretical results of the quantum path
197 approach (Fig. 4c-d), revealing a clear time- and energy-dependent sub-barrier feature on the distributions.

198 For the imaginary tunneling time t_i , it reflects the static property of the potential barrier, i.e., the barrier thickness
199 decided by the instantaneous strength of the electric field. The imaginary tunneling time will reach the minimum (\sim
200 500 attoseconds) when the electric field vector points to the major axis of the ellipse (i.e., the emitting angle $\theta = 180^\circ$)
201 and it will increase to 800 attoseconds when the polarization vector points to the minor axis. The energy dependence
202 of t_i is also important, which controls the radial spread of the photoelectron momentum distribution. Under the
203 adiabatic approximation [$E(t) = E$, $A(t) = -Et$], the imaginary tunneling time can be obtained analytically as
204 $t_i = \sqrt{(2I_p)/E}$, and it has no any energy dependence. The corresponding ionization rate $\exp(-2\text{Im}S)$ becomes
205 back to the ADK rate⁴⁰ used in CTMC (i.e., $\exp[-2(2I_p)^{3/2}/(3E)]$), which shows a narrower radial spread of the
206 photoelectron momentum distribution compared with the non-adiabatic case^{13,14}.

207 In contrast to the imaginary tunneling time, the sub-barrier phase reflects the dynamical properties of the quickly
208 changing Coulomb barrier, i.e, the derivative versus time. When the electric field vector points to the major or
209 minor axis of the ellipse, the barrier is stationary and thus the electron doesn't acquire any phase in agreement with
210 the Wentzel–Kramers–Brillouin approximation solution (real number) under the barrier⁴¹. When the electric field
211 vector deviates the stationary instants, the electron acquires a sub-barrier phase whose sign is dependent on the
212 change direction of the barrier, i.e., the barrier thickness is increasing or decreasing. In Fig. 4b, we illustrate the
213 phase evolution at three ionization instants at the rising edge, stationary point, and falling edge of the light electric
214 field, respectively. This clearly proves the sub-barrier phase is dependent on the dynamical change of the barrier.
215 Moreover, both experiment and theory indicate that the low-energy component of the tunneling wave function has
216 a more prominent sub-barrier phase, as there the sub-barrier interaction time and length are larger. Note that the
217 POP attoclock can provide the time- and energy-resolved dynamical information about tunneling compared with the
218 common static attoclock scheme.

219 In conclusion, we have calibrated the attoclock with the two-color phase-of-phase photoelectron spectroscopy,
220 and employed this POP attoclock to build the accurate angular streaking relationship between an arbitrary final
221 momentum and its ionization instant in the full momentum space. The results demonstrate the widely-used angular
222 streaking capability of light fields is not angularly homogeneous and the ionization instants along the same emitting
223 angle have a strong energy dependence, which both contradict with the static tunneling model. Using the measured
224 ionization instant and ionization probability, we have reconstructed the imaginary tunneling time and the electron
225 sub-barrier phase, which contains the static and dynamical information of the tunneling process, serving as a

226 complete characterization of sub-barrier tunneling dynamics in strong-field ionization. Based on this work, the POP
227 attoclock has the appealing potential application in the attosecond metrology of the oriented molecules, crystal solids
228 and liquids⁴².

229 **Methods**

230 **Experimental details.** Supplementary figure 1 schematically shows the experimental setup for phase-of-phase
231 attoclock. The p -polarization fundamental light field pulses were delivered from a multipass Ti:sapphire amplifier at
232 a central wavelength of 800 nm with the repetition rate of 3 kHz. The pulse duration is 25 fs with full width of half
233 maximum in intensity. The second harmonic pulses at the central wavelength of 400 nm were produced by frequency
234 doubling with a 200- μm -thick type-I beta-barium borate (BBO) crystal (29.2° cut). Then, the two light fields were
235 synchronized in a Mach-Zehnder interferometer scheme with 5-attosecond precision and their polarizations and
236 intensities were controlled, independently. In each arm of the interferometer, the intensity and polarization of the
237 light field were monitored by a wire grid polarizer and half-wave and quarter-wave retardation plates, respectively.
238 The fundamental field was adjusted to be the elliptical polarization whose major axis was fixed along the z axis and
239 the second harmonic field was tuned to a co-rotating circular polarization in the same polarization plane (x - z plane).
240 The light beam was focused by a silver-coated concave mirror with focal length $f = 75$ mm, which was placed inside
241 the high-vacuum chamber ($< 10^{-10}$ mbar) of the COLTRIMS set-up. The supersonic gas jet of krypton atoms (2
242 Bar) was delivered along the x direction by a small nozzle with a opening hole diameter of 30 μm . In COLTRIMS,
243 static electric (~ 3.2 V/cm) and magnetic (~ 5.4 G) fields were applied along the z axis to collect the charged
244 fragments in coincidence. Only the single ionization (one electron is coincident with one kr^+) events are presented
245 in this work.

246 **CTMC simulation.** In the CTMC model, the tunnelled electrons have a Gaussian distribution on the transverse mo-
247 mentum (v_{\perp}) perpendicular to the instantaneous laser field and zero longitudinal momentum along the instantaneous
248 laser field direction. Each electron trajectory is weighted by the ADK ionization rate $W(t, v_{\perp}^i) = W_0(t)W_1(v_{\perp}^i)$ ^{27,28},
249 in which $W_0(t) = |(2I_p)^2/E(t)|^{2/\sqrt{2I_p}-1} \exp[-2(2I_p)^{3/2}/|3E(t)|]$ determines the ionization rate with respect to the
250 ionization instant, and $W_1(v_{\perp}^i) = \sqrt{2I_p}/E(t) \exp[-\sqrt{2I_p}(v_{\perp}^i)^2/|E(t)|]$ determines the initial transverse momentum
251 distribution, where $E(t)$ is the instantaneous strength of laser field and I_p is the ionization potential. The electron
252 tunneling exit is along the instantaneous direction of the synthesized light field and its value is given by $I_p/|E(t)|$.
253 After sampling all the electrons, their classical motion outside the barrier is governed by the Newtonian equation
254 $\ddot{\mathbf{r}} = -E(t) - \mathbf{r}/r^3$ until the laser is turned off, where r is the distance from the electron to the nucleus. In the
255 simulation, we use a \sin^2 -shape pulse envelope with the duration of 12 cycles for the SH field. At the end of the

256 laser pulse, we select the electrons with positive energy and record their positions and their momenta. The electron
 257 asymptotic momenta on the virtual detector are transformed according to the Kepler's laws. Finally, we statistic the
 258 electron trajectories using the 200×200 bins in the momentum regime of $p_x \times p_z = [-1.5, 1.5] \text{a.u.} \times [-1.5, 1.5] \text{a.u.}$. We
 259 totally calculate 24 frames of the photoelectron momentum distributions for the two-color relative phase distributing
 260 from 0 to 2π uniformly. The calculated phase-resolved photoelectron momentum distributions are presented in SI,
 261 from which we use Fourier transform to obtain the phase-of-phase spectrum shown in Fig. 3a.

262 **Strong-field approximation model.** In the strong-field approximation, the final-momentum-resolved ionization
 263 matrix element is given by $M_p = -i \int_0^{t_f} \langle \mathbf{p} + \mathbf{A}(\tau) | \mathbf{r} \cdot \mathbf{E}(\tau) | \psi_{\text{initial}} \rangle \exp(iS(\tau)) d\tau$, where $S(\tau) = \int_0^\tau [\mathbf{p} + \mathbf{A}(t)]^2 / 2 +$
 264 $I_p dt$, t_f is the laser turn-off time, and ψ_{initial} is the initial wavefunction of electrons. In the calculation, we adopt
 265 the asymptotic form of the hydrogenlike atomic wavefunction $\psi_{\text{initial}} = \exp(-\sqrt{(2I_p)}r) Y_{1m}(\theta, \phi) / r$ as the initial
 266 wavefunction of Kr atoms, where $m = 1$ and -1 represent the p_+ and p_- orbitals, respectively. By the numerically
 267 integrating the ionization matrix element, we obtained the phase-resolved photoelectron momentum distributions
 268 from p_+ and p_- orbitals, respectively. From the photoelectron momentum spectra, we extracted the phase-of-phase
 269 spectrum. Note that using the saddle-point method one can avoid the numerical integration and introduce the
 270 concept of the quantum path. The saddle point $t_s(\mathbf{p})$ is determined by the equation $[\mathbf{p} + \mathbf{A}(t_s)]^2 / 2 + I_p = 0$, which
 271 is solved by a complex root finding routine. In the elliptical light field, one final momentum can correspond to
 272 two saddle points and only one of them is physical. By judging the ionization probability of the quantum path,
 273 one can select the physical one. During tunneling (i.e., from t_s to t_r), the electron acquires a complex action
 274 $S(t_r) = \int_{t_s}^{t_r} [\mathbf{p} + \mathbf{A}(\tau)]^2 / 2 + I_p d\tau$. The real part of the action is the sub-barrier phase and the imaginary part governs
 275 the ionization rate by $W = \exp(-2\text{Im}[S(t_r)])$. In the main text, we show t_r , t_i and $\text{Re}[S]$, which are obtained by
 276 directly solving the saddle-point equation and propagates the quantum path under the barrier. On the other hand, we
 277 present the corresponding results in SI, which are obtained by the extraction method of POP attoclock. They agree
 278 very well with each other, which strongly verified the reliability of POP attoclock. Note that in the adiabatic limit
 279 $[E(t) = E, A(t) = -Et]$, there is $t_i = \sqrt{(v_\perp + 2I_p)/E}$ and thus the exponential factor of the ionization rate can be
 280 evaluated as $W = \exp[-2(2I_p)^{3/2}/(3E)] \exp[-\sqrt{2I_p}(v_\perp)^2/E]$, which is exactly the ADK rate used in the adiabatic
 281 CTMC model.

282 **Coulomb-corrected strong-field approximation simulation.** The CCSFA model is based on the saddle-point
 283 approach of the SFA. We randomly sample 10^7 points in the final momentum plane ($p_x, p_z = [-1.5, 1.5] \text{a.u.}$). After
 284 determining the ionization time of each quantum path by the saddle-point equation in the two-color field, we then
 285 propagate these electron trajectories with real variables using Newton's equation $\ddot{\mathbf{r}} = -E(t) - \mathbf{r}/r^3$ to simulate the

286 electron dynamics outside the barrier. The weight of the electron trajectories is given by $W = \exp(-2\text{Im}[S(t_r)])$
 287 without considering the pre-exponential factor. The electron tunnel exit position and exit momentum are determined
 288 by the imaginary-time propagation under the barrier, i.e., $\mathbf{r}_{exit} = \int_{t_s}^{t_r} [\mathbf{p} + \mathbf{A}(t)] dt$ and $\mathbf{p}_{exit} = \mathbf{p} + \mathbf{A}(t_r)$. After the laser
 289 pulse turns off, we obtain the electron asymptotic momenta on the virtual detector according to the Kepler's laws.
 290 Then we obtain the photoelectron momentum distribution on the "detector" using the same method as CTMC.

291 **TDSE simulation.** Details of the algorithm and the source code are in the publication³⁸. Briefly, we first obtain the
 292 atomic initial-state wave-function through the imaginary time propagation method. In the simulation we use the
 293 effective model potential $V_{eff} = -[Z + (Z_{full} - Z)\exp(-r_s \cdot r)]/r$, where $Z = 1$ and $Z_{full} = 36$ are the asymptotic
 294 ion charges as $r \rightarrow \infty$ and $r \rightarrow 0$, respectively. The screening length $r_s = 7.7631$ is used to match the ionization
 295 potential $I_p = -14.0$ eV for krypton atom. The initial magnetic quantum number m can be tuned to 1, 0 and -1
 296 without changing the ionization potential. The laser parameters are identical in all simulations shown in the main
 297 text. Secondly, we perform the real-time evolution of the wave-function in the laser and the Coulomb combined
 298 field within the dipole approximation. The wave-function is expanded in spherical harmonics, and time-propagation
 299 is performed using a split-operator approach. Finally, when the laser turns off, the photoelectron momentum
 300 distribution is evaluated with a time-dependent surface flux method. The calculated phase-resolved movie of
 301 photoelectron momentum distributions is shown in SI. From these photoelectron momentum distributions we extract
 302 the POP spectrum shown in Fig. 3g.

Reconstruction of t_i and $\text{Im}[S]$. Using the expression of the laser field, one can derive the real part and imaginary
 part of the electron sub-barrier action analytically, as given by,

$$\begin{aligned} \text{Re}[S] = & -p_z E_\omega / \omega^2 \cos \omega t_r (1 - \cosh \omega t_i) - p_x \varepsilon E_\omega / \omega^2 \sin \omega t_r (1 - \cosh \omega t_i) \\ & + (1 - \varepsilon^2) E_\omega^2 / (8\omega^3) \sin 2\omega t_r (1 - \cosh 2\omega t_i). \end{aligned} \quad (1)$$

$$\begin{aligned} \text{Im}[S] = & (p^2/2 + I_p + U_p) t_i - p_z E_\omega / \omega^2 \sin \omega t_r \sinh \omega t_i \\ & + p_x \varepsilon E_\omega / \omega^2 \cos \omega t_r \sinh \omega t_i - (1 - \varepsilon^2) E_\omega^2 / (8\omega^3) \cos 2\omega t_r (\sinh 2\omega t_i). \end{aligned} \quad (2)$$

303 The phase-of-phase spectrum directly reflects the distribution of t_r , and the contrast spectrum is corresponding to
 304 $\exp(-2\text{Im}[S])$, which is normalized to the maximum of the theoretical distribution in the reconstruction process.
 305 Using the measured spectra of t_r and $\exp(-2\text{Im}[S])$, we start from Equation 2 to evaluate t_i by the numerical method
 306 of solving the non-linear equation for each final momentum. Then using the measured t_r and the reconstructed t_i ,

307 through Equation 1 we can obtain $\text{Re}[S]$, i.e., the sub-barrier phase. Note that we have reduced the classical Coulomb
308 rotating effect in experiment. The electron emitting angle θ was shifted to cancel the offset angle in reconstruction
309 process, which is in consistence with the SFA model.

310 Data availability

311 The data that support the plots within this paper and other findings of this study are available from the corresponding
312 author upon reasonable request.

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400 **Acknowledgements**

401 Y. L. acknowledges the finance support by the National Science Foundation of China (grant numbers: 92050201,
402 918850111, 11774013). M. H. acknowledges funding from the European Union’s Horizon 2020 research and
403 innovation programme under the Marie Skłodowska-Curie grant agreement No 801459 - FP-RESOMUS - and the
404 Swiss National Science Foundation through the NCCR MUST.

405 **Author contributions statement**

406 M. H., P. G., and Z. G. performed the experiments. M. H., Y. F. and Y. L. analyzed and interpreted the data.
407 Simulations were implemented by M. H.. This project was coordinated by Y. L.. All authors discussed the results
408 and wrote the paper.

409 **Competing interests**

410 The authors declare no competing interests

411 **Additional information**

412 Supplementary information is available for this paper. Correspondence and requests for materials should be addressed
413 to Y. L..