

Giant counter-rotating oscillations on the attosecond timescale

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We predict an unexplored type of ultrastrong coupling between atoms and intense ultraviolet light that leads to giant population oscillations on the attosecond timescale. These counter-rotating oscillations stem from the interference of energy-conserving and nonconserving photon absorption and emission processes via Rydberg states. They can be of similar amplitude as the elementary femtosecond Rabi oscillations between the two strongly coupled states. The effect, which is beyond the two-level atom, is nonreciprocal: It only affects the excited state, while the ground state is unaffected. We propose that two-photon Rabi oscillations ($1s^2-1s3d$) in helium are suitable for the generation of this type of ultrastrong coupling with realistic pulses. We use a combination of Floquet theory and effective Hamiltonian theory to test our predictions against *ab initio* simulations.

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The rotating wave approximation (RWA) is an important simplifying assumption in light-matter interaction because it separates slow and fast dynamics by enforcing energy conservation between atoms and photons [1]. Understanding the breakdown of the RWA is at the foundation of diverse research fields, such as ultrafast strong-field physics [2], cavity quantum electrodynamics (cQED) [3], and development of quantum technologies [4–6]. Historically, counter-rotating (CR) effects in two-level systems were explored in pioneering works by Bloch and Siegert [7], and Autler and Townes [8]. In further work, Shirley introduced Floquet theory as a limit of quantum optics, with infinite photon numbers and cavity volume [9], to derive phenomena beyond the RWA in the form of an induced energy shift (Bloch-Siegert shift) [7], as well as subcycle modulations of the atomic populations (CR oscillations). Due to limits of laser technology at the time, Shirley dismissed these CR oscillations as unobservable and calculated averaged transition probabilities that did not depend on the instantaneous electric field. In the same spirit, weak and rapid fluctuations of populations that occur in exact propagation of the time-dependent Schrödinger equation for atoms in strong fields have been noted, but passed over [10–12], with one recent exception [13].

In cQED, strong coupling effects are studied using the quantum Rabi model [14]. Here, “CR effects” imply a breakdown of the Jaynes-Cummings model [15], where, in the “ultrastrong coupling” regime, the number of excitations of the atom and field is no longer conserved [3,16–19]. Another breakdown of the RWA occurs in nonresonant interactions between atoms and intense low-frequency laser fields. Over the

last decades, semiclassical treatment of such interactions has revealed novel phenomena, such as high-order harmonic generation [20] and formation of attosecond pulses [21], which have found numerous applications in atoms [22], molecules [23], and solids [24–26]. Attosecond technology has also enabled the study of subcycle effects of bound states, and CR oscillations in the time domain, as recently shown by polarizing Rydberg states with infrared fields [27–29]. Recently, the research communities of quantum optics and strong-field physics have begun to intersect with researchers considering the generation of quantum states driven by strong fields [30–33].

Strong coupling implies first and foremost Rabi oscillations. In this work, we primarily focus on two-photon Rabi oscillations (2-PROs). 2-PRO is a counterintuitive quantum process that allows for strong coupling between two atomic states of equal parity via weakly coupled states of opposite parity [34]. As it is a nonlinear process, which requires high intensities sustained for long times, the process has been largely unexplored in the UV and XUV regimes. One notable exception is the work of Dörr *et al.*, where it was shown that population inversion through 2-PROs in hydrogen atoms between the $1s$ and $2s$ states is impossible due to unavoidable ionization losses induced by the laser field [11]. In cQED terminology, large losses compared to the Rabi period lie in the “weak coupling” regime, $\Omega < \Gamma$, with ionization rate Γ and Rabi frequency Ω . In Fig. 1(a), we reproduce this important and unexpected result by showing that the 2-PROs of the excited $2s$ state in hydrogen are overdamped in the entire resonant parameter space (spanned by pulse intensity I_0 and duration t). Physically, this effect arises due to the similar scaling of both Ω and Γ , $(\Omega, \Gamma) \propto E_0^2$, as can be understood from effective Hamiltonian theory [35,36]. While 2-PROs in the UV and XUV regimes have not yet been realized, intense XUV fields produced by seeded free-electron lasers (FELs) [37–39] have recently been used to demonstrate one-photon Rabi oscillations (1-PROs) [40] and strong-field quantum control [41] between the atomic ground state (GS) and excited states (ES) in helium atoms. In these experiments, $\Gamma < \Omega$,

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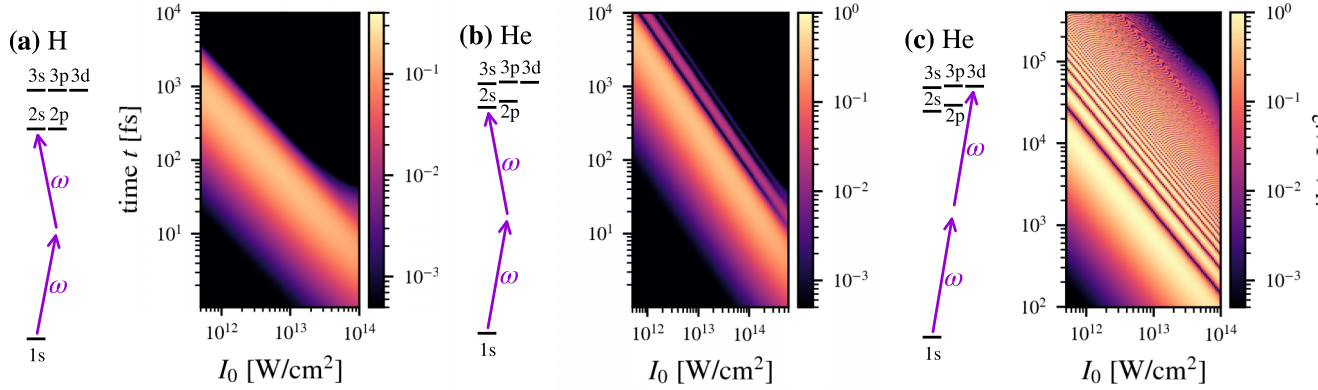


FIG. 1. Excited state (ES) population $|b(t)|^2$ for the resonant transition (i.e., adjusted for AC Stark shift) from the ground state (GS) to (a) the $2s$ state in hydrogen, (b) the $1s2s$ state in helium, and (c) the $1s3d$ state in helium, parametrically for different intensities. In analogy to classical mechanics, we can define a damping ratio ζ in order to classify the overdamped ($\zeta > 1$) and underdamped ($\zeta < 1$) regimes. In panel (a) $\zeta \sim 1.5$, in panel (b) $\zeta \sim 0.4$, and in panel (c) $\zeta \sim 6 \times 10^{-3}$.

since $\Omega \propto E_0$ and $\Gamma \propto E_0^2$ [42]. Therefore, we propose using the “strong coupling” nomenclature from cQED for this regime.

In this letter, we show that two-photon Rabi cycling helium atoms, driven by intense XUV light, are a promising system to explore light-matter coupling in novel domains. The *overdamped* nature of the $1s$ - $2s$ hydrogen UV transition [Fig. 1(a)] does not generalize to the experimentally relevant helium atom because the $1s^2$ - $1s2s$ XUV transition is *underdamped*, albeit with only few weak Rabi oscillations surviving, as can be observed in Fig. 1(b). Most importantly, the two-photon coupling between $1s^2$ and $1s3d$ states is underdamped, with strong Rabi cycles accessible in the parameter space, as shown in Fig. 1(c). Therefore, the $1s^2$ - $1s3d$ transition allows for “strong coupling” conditions and population inversion over a broad range of intensities and pulse lengths, which enables us to increase the field intensity to explore physics beyond the RWA [43]. In analogy with cQED, we will refer to this regime as “perturbative ultrastrong coupling.” In traditional cQED, which is concerned with ideal two-level systems, the ultrastrong coupling regime is reached when the coupling is comparable to the optical frequency: $\Omega \approx \omega$. In our work, which concerns real multilevel atoms, we find that giant CR oscillations take place already for couplings much smaller than the optical frequency, $\Omega \ll \omega$, and can coexist with the Rabi oscillations. We find that the giant CR effects can break the reciprocity of ultrastrong coupled states, by which we mean that CR oscillations may appear solely on one of the strongly coupled states. Providing a quantitative explanation of giant, nonreciprocal CR oscillations in a strongly coupled atom is the main point of this letter. Atomic units are used unless otherwise stated: $e = \hbar = m = 1/4\pi\epsilon_0 = 1$.

Theory. In the following, we provide the outline for the derivation of the CR population dynamics for the two-photon Rabi cycling case. The theoretical modeling of 2-PROs in atoms is enabled through an essential states formalism. The essential states span a sub-Hilbert space $\mathcal{P}$, and they are coupled effectively via the orthogonal Hilbert space of nonessential states, called $\mathcal{Q} = \mathcal{P}^\perp$. Let us refer to the essential states as $|a, 0\rangle$ (GS) and $|b, -2\rangle$ (ES), where a and b refer to the ground and excited atomic states, respectively. The

second index labels “photon numbers” of the mode. Unlike quantum optics, the absolute photon number is simply an inconsequential energy offset in Floquet theory. Therefore, the photon numbers may take on negative values. Parametrizing the effective coupling of the essential states to and via the $\mathcal{Q}$ space leads to an effective two-level Hamiltonian H_{eff} . If the $\mathcal{Q}$ space contains CR states, H_{eff} incorporates CR effects, such as the Bloch-Siegert shift and CR contributions to effective couplings. Since ionization in strong high-frequency fields cannot be neglected, the effective Hamiltonian must be non-Hermitian, and photoionization can occur in a range of different nonlinear processes [35,36,42]. However, the two-level H_{eff} generates only a *cycle-averaged* time evolution with populations $|a(t)|^2$ (GS) and $|b(t)|^2$ (ES). It can thus not be used to model rapid CR oscillations. In Floquet theory, CR oscillations of an atomic state $|b\rangle$ stem from interference of the essential uncoupled atom-photon state $|b, -2\rangle \in \mathcal{P}$, with Floquet states comprised of the same atomic state, but a different number of photons, $|b, k\rangle \in \mathcal{Q}$, where $k \neq -2$, so that the CR oscillations have frequencies $2\omega, 4\omega$, etc. (the odd harmonics vanish due to the inversion symmetry of the atom).

Method. The non-Hermitian effective Hamiltonians employed in this work are obtained through nonperturbative time-independent calculations, enabled through Floquet theory [44]. We construct a complex-scaled Floquet Hamiltonian H_F^θ , whose continuous spectrum has been rotated into the complex plane [45,46]. The complex scaling renders H_F^θ non-Hermitian, with complex eigenvalues $E_n - i\Gamma_n$ and complex-scaled eigenfunctions $|n\rangle$ that are not orthonormal under the usual scalar product [47]. However, the eigenfunctions still fulfill biorthonormality $\langle \tilde{m} | n \rangle = \delta_{nm}$, where $\langle \tilde{m} |$ solves the left eigenvalue problem $\langle \tilde{m} | H = (E_m - i\Gamma_m) \langle \tilde{m} |$. The closure relation then reads $1 = \sum_n |n\rangle \langle \tilde{n}|$.

From H_F^θ , we parametrize the influence of the $\mathcal{Q}$ -space states in an all-order approach [48]. Specifics on effective Hamiltonian theory can be found in the Supplemental Material [49] and Refs. [50–56] therein. H_F^θ is constructed in the length gauge from complex-scaled energies and transition dipole moments, both obtained using the configuration-interaction singles (CIS) method [57,58], with exterior complex scaling [59].

To validate the results, we perform *ab initio* simulations using the time-dependent configuration-interaction singles (TDCIS) method in the length gauge [60]. The simulations are done with a CIS basis that is constructed using a B-spline basis with a complex-absorbing potential [61,62]. The use of length gauge is required: As shown by Kobe and Smirl, the complex amplitudes of the states are probability amplitudes only if the length gauge is employed [63]. This means that their squares correspond to the probabilities to find electrons in the respective field-free states.

Analytical formulation. In Floquet theory, the transition amplitude between the atomic states $|a\rangle$ and $|b\rangle$ from $t = 0$,

$$U_{ba}(t) = \sum_k \langle b, k | e^{-iH_F^\theta t} | a, 0 \rangle e^{ik\omega t}, \quad (1)$$

is given as the sum over the transition amplitudes between the atom-photon states $|a, 0\rangle$ and $|b, k\rangle$, determined by H_F^θ , and weighted with the Fourier components $\exp(ik\omega t)$ [9,44]. In our essential states approach, only one photon block ($k = -2$) is included. Expanding in the dressed state basis of $H_{\text{eff}}|\pm\rangle = \lambda_\pm|\pm\rangle$, we obtain

$$U_{ba}(t) \approx \sum_{j \in \pm} \langle b, -2 | j \rangle e^{-i\lambda_j t} \langle \tilde{j} | a, 0 \rangle e^{-2i\omega t}. \quad (2)$$

The transition probability is the optical cycle-averaged ES population, $|U_{ba}(t)|^2 = |b(t)|^2$. Since H_{eff} is non-Hermitian, we require the biorthonormal $\langle \tilde{\pm} |$, which fulfill the left eigenvalue equation for H_{eff} , so that $1 = |+\rangle\langle \tilde{+}| + |-\rangle\langle \tilde{-}|$ [47]. For a brief introduction of Floquet theory, its application to the semiclassical two-level system, and expressions for the damped, effective two-level dynamics, we refer to the Supplemental Material [49] and Refs. [64,65] therein.

According to Eq. (1), CR oscillations are induced when several $|b, k\rangle$ —in our case $|b, 0\rangle$, $|b, -2\rangle$, and $|b, -4\rangle$ —have non-negligible overlap with the full eigenstates of H_F^θ , i.e., $H_F^\theta|\phi_\pm\rangle = \lambda_\pm|\phi_\pm\rangle$. However, diagonalization of H_{eff} only yields the eigenstates $|\pm\rangle \in \mathcal{P}$, while $|b, k\rangle \in \mathcal{Q}$, $k \neq -2$, thus requiring the knowledge of $|\phi_\pm\rangle$. The operator that produces the $\mathcal{Q}$ -space components of a given $\mathcal{P}$ -space state is known as the *reduced wave operator* χ [66,67]. With χ , we can obtain $|\phi_\pm\rangle \in \mathcal{P} \oplus \mathcal{Q}$ from $|\pm\rangle \in \mathcal{P}$ simply via

$$|\phi_\pm\rangle = (P + \chi)|\pm\rangle, \quad (3)$$

where $P := |a, 0\rangle\langle a, 0| + |b, -2\rangle\langle b, -2|$ projects on the essential states. In practice, we employ the pole approximation [1] to calculate the reduced wave operator, leading to

$$\chi \approx \frac{Q}{E_0 - QH_F^\theta Q} QH_F^\theta P, \quad (4)$$

where $Q = 1 - P$ projects onto $\mathcal{Q}$ and E_0 is the resonance energy. Equation (4) is obtained by partitioning the time-independent Schrödinger equation (see the Supplemental Material [49]).

Through Eq. (3), the transition amplitude in Eq. (1) can be expressed in a form that only involves quantities accessible through effective Hamiltonian theory,

$$U_{ba}(t) = \sum_{k \in \{0, -2, -4\}} \sum_{j \in \pm} \langle b, k | P + \chi | j \rangle e^{-i\lambda_j t} \langle \tilde{j} | a, 0 \rangle e^{ik\omega t}, \quad (5)$$

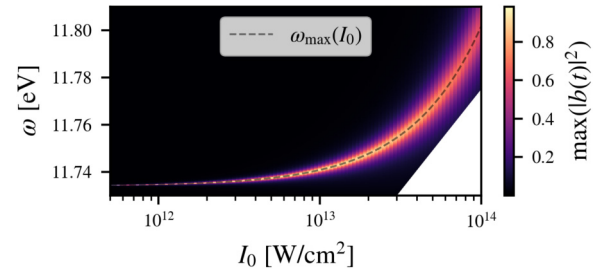


FIG. 2. Maximum of the excited state population, $\max(|b(t)|^2)$, in the parameter space of ω and I_0 ($1s^2$ - $1s3d$ system). The white space signifies an off-resonant high-intensity region of the parameter space, in which the effective two-level Hamiltonian does not apply.

but retains subcycle dynamics. Interference between the CR states and coupling between the GS and the $\mathcal{Q}$ space can be neglected ($\chi|a, 0\rangle \approx 0$) to obtain a compact form for our transition probability:

$$|U_{ba}(t)|^2 = |b(t)|^2 [1 + |\Lambda^{(0)}|^2 + |\Lambda^{(-4)}|^2 + 2\Re(\Lambda) \cos(2\omega t)]. \quad (6)$$

Here, $\Re$ refers to the real part. $\Lambda^{(k)} := \langle b, k | \chi | b, -2 \rangle$, and thus $|\Lambda^{(k)}|^2 |b(t)|^2$ represents the optical cycle-averaged population of the CR state $|b, k\rangle$, $k = 0, -4$. Further, $\Lambda := \Lambda^{(0)} + \Lambda^{(-4)}$ is the characteristic quantity determining the magnitude of the CR oscillations. The maximum span of the CR oscillations is given by

$$M_{\text{max}} = 4|\Re(\Lambda)| \max(|b(t)|^2). \quad (7)$$

Resonant parameter space. While previous studies have evidenced weak CR oscillations at short optical wavelengths [10–13], the question of their enhancement in the strong coupling regime has been unexplored and is the main target of our analysis. Hence, we must first find the regime in which Rabi oscillations can be resonantly driven at high intensity. In Fig. 1, we showed results for the resonantly driven two-photon transition in hydrogen ($1s \leftrightarrow 2s$) [panel (a)] and helium: $1s^2 \leftrightarrow 1s2s$ [panel (b)], $1s^2 \leftrightarrow 1s3d$ [panel (c)], for rectangular pulses. We found the Stark-shifted resonance by calculating the maximum population of the ES $\max(|b(t)|^2)$ depending on ω and I_0 . The results are shown in Fig. 2 for the $1s^2$ - $1s3d$ transition. The resonance condition is extremely narrow at low intensities. With higher intensities, the condition becomes more lenient, allowing for detunings of ~ 5 meV or more.

Giant CR oscillations. In Fig. 3(a), we show evidence for giant CR oscillations ($M_{\text{max}} = 62\%$) in the $1s3d$ state at $I_0 = 10^{14} \text{ Wcm}^{-2}$ and $\omega = 11.8017 \text{ eV}$ (blue area). The mean populations (dashed) and the analytically calculated outlines of the CR oscillations (dash-dotted) reproduce the TDCIS calculations accurately. In panel (b), we focus on a single femtosecond to show the subcycle modulation of the ES on the attosecond timescale. Here, we further plot $|U_{ba}(t)|^2$ from Eq. (6) to show the excellent—subcycle—agreement of the theoretical model with numerical TDCIS simulations.

The $1s^2$ - $1s3d$ system displays complete nonreciprocity in the CR oscillations: The GS evolves only with the optical cycle-averaged population $|a(t)|^2$ due to $|a, 0\rangle$, whereas the

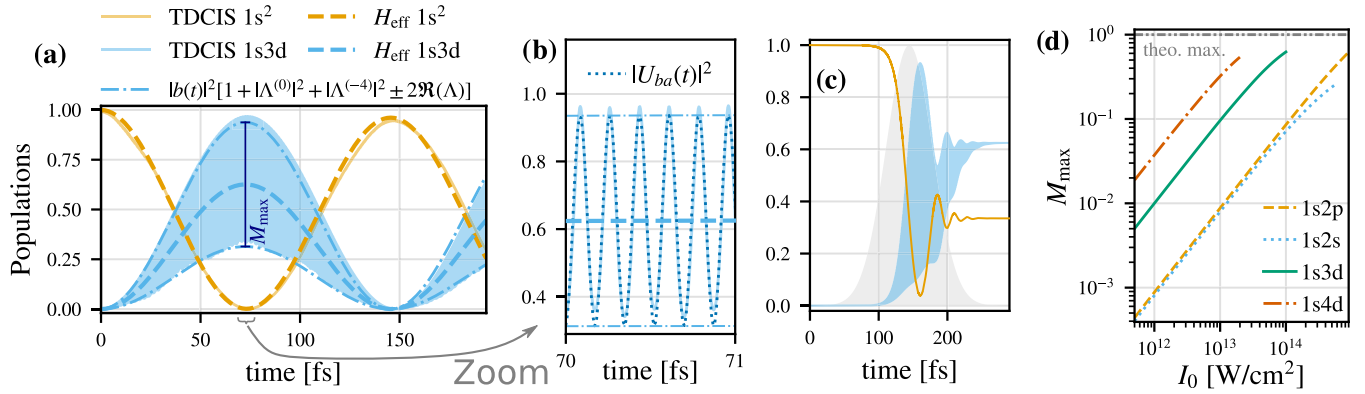


FIG. 3. (a) 2-PROs from $1s^2$ to $1s3d$, induced by a monochromatic field, from *ab initio* (solid) and H_{eff} (dashed) calculations. CR oscillations are the blue area. Outlines of the CR oscillations are in dash-dotted. M_{max} is given by Eq. (7). There is no yellow area due to absence of CR oscillations in the GS. (b) Closed-up on 1 fs, additionally plotting $|U_{ba}(t)|^2$ from Eq. (6) (dotted). (c) Results for a Gaussian envelope (shaded gray). (d) Maximum span of CR oscillations M_{max} against intensity for one-photon transition to $1s2p$, and two-photon transition to $1s2s$, $1s3d$, and $1s4d$. Theoretical maximum is at $M_{\text{max}} = 1$ (representing CR oscillations that lead to a complete population transfer on subcycle scales). See main text for laser parameters in panels (a) and (c).

ES is modulated with CR oscillations that are as large as the Rabi oscillations themselves. Remarkably, despite violent exchange of photons to the Q space via $|b, -2\rangle$, the state coherently returns to the initial $|a, 0\rangle$ in $\mathcal{P}$ after the completed Rabi cycle. The asymmetry stems from the vastly different strengths in which the GS and ES couple to the Q space. In this way, the Rydberg states (e.g., $1s3p$, $1s4p$, $1s5p$, $1s4f$, and $1s5f$) exchange population with the ES on the attosecond timescale. The electron remains bound, yet is highly polarized, switching between d -character and p -/ f -character, following the instantaneous laser field. Unlike how free electrons wiggle in laser fields, the underlying assumption in the strong-field approximation [20], our reported dynamics arise due to simultaneous confinement by the atomic potential and the action of the instantaneous laser field.

Discussion. Following the several dismissals of CR phenomena in the literature [9–11], it is imperative to motivate that the proposed giant CR effects can be generated with realistic pulse forms and observed experimentally through possible detection schemes. We find that the breaking of CR reciprocity is robust against changes in the envelope of the field, as shown in Fig. 3(c) using TDCIS. The simulation was performed with a Gaussian intensity envelope, $I(t) = I_0 e^{-\ln^2[2t/\tau]^2}$, where $I_0 = 2 \times 10^{14} \text{ W cm}^{-2}$ and $\tau = \text{FWHM} = 80 \text{ fs}$ at a frequency $\omega = 11.86 \text{ eV}$, corresponding to $\sim 105 \text{ nm}$. These parameters are reasonable for seeded FEL sources, such as the externally seeded FEL at FERMI [38,68,69].

It is also important to understand the generality of the CR effects. Can they be observed in other UV/XUV-dressed systems? In Fig. 3(d), we compare the maximum span of CR oscillations M_{max} between different experimentally relevant systems. In 1-PROs, $1s^2$ - $1s2p$, the CR oscillations are weaker for a given intensity, so that intensities as large as $9 \times 10^{14} \text{ W cm}^{-2}$ are required to generate giant CR oscillations. We show calculations for the $1s^2$ - $1s2p$ system in the Supplemental Material [49]. Meanwhile, the two-photon transition $1s^2$ - $1s2s$ is too damped, making it unsuitable for our

purposes. A possible way to enhance CR oscillations is to target higher Rydberg states that couple to Q -space states with stronger dipole moments. We demonstrate this in panel (d) by including results for the $1s4d$ state. Generating giant CR oscillations in higher Rydberg states requires less intense, but longer pulses.

Experimental observation. We propose that the giant CR oscillations can be probed experimentally in two complementary ways: in the frequency or time domain. The detection in the frequency domain is possible because the ES amplitude has significant 2ω components [Fig. 3(b)], which appear in the Fourier transform of the ES amplitude (see the Supplemental Material [49]). Higher-order components ($4\omega, 6\omega, \dots$) are also present, as recently investigated by de las Heras *et al.* [13]. In contrast to prior works, the intensity of our 2ω components is $>2\%$, which we surmise is strong enough to significantly affect above-threshold ionization processes, leading to detectable imprints on photoelectron spectra.

The time dependence of the CR oscillations can be probed by a synchronized auxiliary attosecond pulse. Prior work with seeded FELs has shown that it is possible to generate *attosecond waveforms* [70] that open the possibility to temporally resolve the CR oscillations by photoelectron spectroscopy. Studying the influence of giant CR oscillations on photoelectrons in the time and energy domains will be the subject of future work.

Conclusion. CR oscillations are a general, and fundamental, feature beyond the RWA, which can be enhanced to the same magnitude as the coexisting Rabi oscillations. We have demonstrated via *ab initio* simulations that giant CR effects can be induced with intense UV/XUV fields in helium on attosecond timescales. This opens the gates to an unexplored perturbative ultrastrong coupling regime with significant ultrafast effects due to the instantaneous electric field. Contrary to the simple two-level system, where CR oscillations are reciprocal, we find that only the ES is modulated on a subcycle scale, while the GS is effectively shielded from the Q space. This effect is remarkably strong in 2-PROs from $1s^2$ to $1s3d$,

but is found to be a general phenomenon that can be induced for other optical transitions. We have explored these effects quantitatively through a theoretical approach for describing CR oscillations, enabled by effective Hamiltonian theory and Floquet theory. Given the power of Floquet approaches in different areas of physics [44,71,72], and the fundamental importance of the RWA in quantum physics, we expect that our findings will be fruitful for a range of fields.

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Data availability. The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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