

Intensity-dependent interferences in strong-field Rydberg-state excitation

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We conduct a systematic investigation of the oscillations of the Rydberg-state populations as a function of intensity arising from strong-field multiphoton excitation of Rydberg atoms. By numerically solving the time-dependent Schrödinger equation and by constructing a quantum model based on Freeman resonances, we elucidate the underlying physical mechanisms governing the intensity-dependent oscillatory behavior of the Rydberg-state population dynamics. Our analysis shows that these oscillations originate from the synergistic interplay of three distinct quantum-interference mechanisms: (1) interference between excitation pathways at the rising and falling edges of the laser pulse (long-time interference), (2) interference between different optical cycles within the same side of the laser pulse (short-time interference), and (3) interference within individual optical cycles (intracycle interference). Additionally, we find that ground-state depletion has varying degrees of influence on the oscillation patterns formed by the three interference mechanisms under different laser-intensity conditions.

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I. INTRODUCTION

Rydberg states, which represent highly excited electronic states of atoms or molecules characterized by large principal quantum numbers, have attracted considerable interest, especially since the discovery of their unexpected survival in strong laser fields [1–5]. It is generally understood that two distinct physical mechanisms contribute to Rydberg-state excitation, with the Keldysh parameter [6] γ serving as the distinguishing criterion. When $\gamma \ll 1$, i.e., in the tunneling regime, electrons first undergo ionization and subsequently are captured into Rydberg states under the combined influence of the laser field and the Coulomb potential. This is recognized as frustrated tunneling ionization [7–15]. Conversely, when $\gamma \gg 1$, i.e., in the multiphoton regime, Rydberg states can be populated through the absorption of multiple photons in a resonant transition, a process termed multiphoton resonance (MPR) [16–19]. These two distinct mechanisms compete with each other depending on the laser parameters [20,21]. One especially intriguing phenomenon in the MPR

process is the appearance of Stückelberg oscillations in the intensity dependence of the Rydberg-state population [22]. Stückelberg oscillations were first predicted in the context of atomic collisions nearly a century ago [23]. They arise whenever a transient resonance condition reappears after a well-defined time interval. This phenomenon is closely related to the Landau-Zener model [24,25], which describes nonadiabatic transitions between quantum states. While multiphoton ionization has been extensively studied [3,26], investigations on the MPR excitation dynamics remain limited owing to the experimental difficulties associated with bound-state detection.

Recently, Stückelberg oscillations in multiphoton Rydberg excitation have been experimentally studied through all-optical measurements of free-induction decay emission [22]. These measurements revealed a distinctive pattern of intensity-dependent oscillations in the population of excited states. However, a complete understanding of the mechanism behind these oscillations has remained elusive, particularly an abrupt change of the oscillation period observed at higher laser intensities. Bengs *et al.* [22] and Zhao *et al.* [27] demonstrated that in multiphoton excitation dynamic interference originates from quantum superposition of contributions of excitation pathways at the rising and falling edges of the laser pulse, which produces fast oscillations in the intensity-dependent yield of excited states. This dynamic interference, a concept introduced to describe interference effects arising from the time-dependent nature of the interaction, can manifest itself through excitation pathways that are realized at different times within a single optical cycle or pathways

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spanning multiple optical cycles [28–30]. Their analysis revealed that when the laser intensity exceeds the eight-photon ionization threshold, ground-state depletion that builds up during the rising edge of the pulse effectively closes the excitation pathway at the falling edge, fundamentally altering the interference mechanism and causing an abrupt increase in the oscillation period. Depletion of the ground state at high intensities affects the interference pattern by eliminating one of the two “arms” of the interferometer. Nevertheless, this explanation alone does not fully account for the observed changes in the oscillation period. Moreover, the role of different multiphoton pathways and their interference has not been thoroughly investigated.

In this work, we present a comprehensive theoretical analysis of the various interference mechanisms in multiphoton Rydberg excitation. By combining accurate numerical simulations using the time-dependent Schrödinger equation (TDSE) and an analytical model, we elucidate the mechanisms responsible for the oscillation patterns across different intensity regimes. Our results reveal a complex interplay of three distinct interference mechanisms: (1) long-time interference between excitation pathways in the first half (the rising or leading edge) and the second half (the falling or trailing edge) of the laser pulse, (2) short-time interference between different optical cycles within the same half of the laser pulse, and (3) intracycle interference within individual optical cycles on one or the other side of the pulse. Furthermore, we demonstrate that ground-state depletion inevitably disrupts these interference mechanisms as the laser intensity increases, thereby altering the resultant oscillation patterns.

This paper is organized as follows: In Sec. II, we describe the theoretical methods used, including the TDSE approach and our analytical model. Section III presents our results and analysis, focusing on the intensity-dependent oscillation patterns and the interplay between the different interference mechanisms. Finally, in Sec. IV, we summarize our findings and discuss their implications for understanding strong-field multiphoton excitation processes. Appendix A presents the explicit form of the transition matrix element for the case of an infinitely extended monochromatic driving field, Appendix B provides a comparison with the TDSE and the two-level model calculations in Ref. [22], Appendix C derives explicit expressions for the period of the intensity oscillations for the case of a triangular pulse shape, and Appendix D presents the oscillation structures for shorter pulse durations. Atomic units $\hbar = 1$, $m = 1$, and $e = 1$ are used unless otherwise stated.

II. THEORETICAL METHODS

A. Time-dependent Schrödinger equation

The three-dimensional TDSE describing the interaction of the hydrogen atom with a laser pulse can be written as

$$i \frac{\partial \psi(\mathbf{r}, t)}{\partial t} = \left[-\frac{1}{2} \nabla^2 - \frac{1}{r} + \mathbf{r} \cdot \mathbf{E}(t) \right] \psi(\mathbf{r}, t). \quad (1)$$

The laser field is characterized by the vector potential $\mathbf{A}(t) = A_0 f(t) \cos(\omega t) \hat{\mathbf{z}}$ along the z axis, where A_0 is the vector-potential amplitude, ω the carrier frequency, and $f(t)$ the pulse-envelope function. The corresponding electric field is

given by $\mathbf{E}(t) = -\frac{\partial \mathbf{A}(t)}{\partial t}$. We use a sine-square laser envelope $f(t) = \sin^2(\pi t/40T)$ with a duration of 40 optical cycles, where $T = 2\pi/\omega$ represents the optical period of the carrier wave.

The time-dependent wave function is expressed in terms of a B -spline basis and spherical harmonics as [31]

$$\Psi(\mathbf{r}, t) = \sum_{\mu l m} C_{\mu l m}(t) \frac{B_{\mu}^k(r)}{r} Y_{lm}(\theta, \phi), \quad (2)$$

where $C_{\mu l m}(t)$ are the time-dependent expansion coefficients, $B_{\mu}^k(r)$ represents the μ th B -spline basis function of order k in the radial direction [31,32], and $Y_{lm}(\theta, \phi)$ are the spherical harmonics describing the angular dependence with orbital and magnetic quantum numbers l and m .

The calculations are performed in the dipole approximation and length gauge, as indicated by the interaction term $\mathbf{r} \cdot \mathbf{E}(t)$ in Eq. (1). We use a maximum angular momentum $l_{\max} = 50$ and a radial grid extending to $r_{\max} = 1200$ a.u. with 2400 B -spline basis functions of order $k = 9$. A complex absorbing potential is applied in the region $r > 1000$ a.u. to prevent unphysical reflections from the boundary. The time evolution of the wave function is evaluated on the basis of the Crank-Nicolson method [32,33]. The time step is $\Delta t = 0.1$ a.u., chosen to ensure numerical convergence based on systematic tests verifying that this value is sufficiently small compared to the optical period ($T \approx 55$ a.u.). After the pulse, the population of a specific bound state ϕ_{nlm} is calculated by projecting the final wave function onto the field-free eigenstate,

$$P_{nlm} = |\langle \phi_{nlm} | \psi(t_f) \rangle|^2, \quad (3)$$

where t_f is a time after the end of the pulse. The total population of a Rydberg state with principal quantum number n is obtained by summing over all allowed angular-momentum quantum numbers

$$P_n = \sum_{l, m} P_{nlm}. \quad (4)$$

Here, P_{nlm} and P_n represent the population probability of the corresponding quantum states, i.e., the fractional occupation of these states for a single atom. Throughout this work, we use the terms “population” and “probability” interchangeably.

B. Analytical model

To gain deeper insight into the interference mechanisms, we develop an analytical model based on the transition amplitude from the ground state to a Rydberg state. The exact transition amplitude can be expressed as

$$M_{nlm} = \lim_{t \rightarrow \infty, t' \rightarrow -\infty} \langle \Psi_{nlm}(t) | U(t, t') | \Psi_g(t') \rangle, \quad (5)$$

where Ψ_g and Ψ_{nlm} are the ground-state and the Rydberg-state wave functions, respectively, in the absence of the field and $U(t, t')$ is the complete time-evolution operator. With the help of the Dyson equation [34,35] for the time-evolution operator,

$$U(t, t') = U_0(t, t') - i \int_{t'}^t d\tau U(t, \tau) H_I(\tau) U_0(\tau, t'), \quad (6)$$

where U_0 is the field-free evolution operator and H_I the interaction Hamiltonian, the transition amplitude can be expanded

as

$$M_{nlm} = \lim_{t \rightarrow \infty} \langle \Psi_{nlm}(t) | \Psi_g(t) \rangle - i \int_{-\infty}^{\infty} d\tau \langle \Psi_{nlm}(t) | U(t, \tau) H_I(\tau) | \Psi_g(\tau) \rangle. \quad (7)$$

Due to the orthogonality of the atomic states, the first term vanishes. Using the dressed-state approximation [14], the transition amplitude to first order can be written as

$$M_{nlm}^{(1)} = -i \int_{-\infty}^{\infty} d\tau \langle \Psi_{nlm}^d(\tau) | H_I(\tau) | \Psi_g(\tau) \rangle, \quad (8)$$

where Ψ_{nlm}^d represents the dressed Rydberg state, which we approximate by

$$\langle \mathbf{r} | \Psi_R^d(\tau) \rangle = \phi_{nlm}(\mathbf{r}) e^{-iE_n \tau} e^{i\mathbf{r} \cdot \mathbf{A}(\tau)} e^{-i \int_{-\infty}^{\tau} d\tau' A^2(\tau')/2}, \quad (9)$$

where $\phi_{nlm}(\mathbf{r})$ is the field-free Rydberg-state wave function and E_n is the Rydberg-state energy level. To incorporate the effects of ground-state depletion at high intensities, we modify this expression to include a time-dependent prefactor:

$$M_{nlm}^{(1)'}(t) = -i \int_{-\infty}^{\infty} d\tau \sqrt{P_g(\tau)} \langle \Psi_{nlm}^d(\tau) | H_I(\tau) | \Psi_g(\tau) \rangle, \quad (10)$$

where $P_g(\tau)$ is the time-dependent ground-state population, which can be either obtained from TDSE calculations or artificially prescribed to test specific hypotheses. This accounts for the decreasing ground-state population as ionization and excitation proceed. For the results presented in Figs. 2(a) and 3, we use the unmodified form of Eq. (8) to analyze the intrinsic interference mechanisms, while for Figs. 2(b) and 5 we include the time-dependent depletion factor. For a monochromatic field, by utilizing the Jacobi-Anger expansion, $e^{-i\mathbf{r} \cdot \mathbf{A}(\tau)}$ can be expressed as

$$e^{-i\mathbf{r} \cdot \mathbf{A}(\tau)} = \sum_{N=-\infty}^{\infty} J_N(rA_0 \cos \theta) \exp(-iN\omega\tau), \quad (11)$$

where θ is the angle between $\mathbf{r}$ and the polarization direction, and J_N are Bessel functions of the first kind.

For a linearly polarized laser and considering a hydrogen atom initially in the $1s$ state, the first-order transition matrix element can be expressed as

$$\begin{aligned} M_{nlm}^{(1)} &= -i \int_{-\infty}^{\infty} d\tau \langle \Psi_{nlm}^d(\tau) | H_I(\tau) | \Psi_g(\tau) \rangle \\ &= \sum_{N=-\infty}^{\infty} \int_{-\infty}^{\infty} d\tau E(\tau) e^{-iN\omega\tau} e^{iS_n(\tau)} \int_0^{\infty} \int_0^{\pi} \int_0^{2\pi} \\ &\quad \times R_{nl}(r) Y_{lm}^*(\theta, \varphi) J_N(rA_0 \cos \theta) r \cos \theta R_{10}(r) \\ &\quad \times Y_{00}(\theta, \varphi) r^2 dr \sin \theta d\theta d\varphi, \end{aligned} \quad (12)$$

where $R_{nl}(r)$ and $R_{10}(r)$ are the respective radial wave functions of the final and the initial state. Here,

$$S_n(\tau) = \frac{1}{2} \int_{-\infty}^{\tau} A^2(t) dt + (E_n + I_p)\tau \quad (13)$$

and I_p is the ionization potential. The angular integral, which involves both the $\cos \theta$ from the dipole interaction and the

$\cos \theta$ argument in the Bessel function, can be expressed as

$$\begin{aligned} I_{\Omega} &= \int_0^{\pi} \int_0^{2\pi} Y_{lm}^*(\theta, \varphi) Y_{00}(\theta, \varphi) J_N(rA_0 \cos \theta) \cos \theta \sin \theta d\theta d\varphi \\ &= \frac{1}{\sqrt{4\pi}} \delta_{m,0} \int_{-1}^1 P_l(\cos \theta) J_N(rA_0 \cos \theta) \cos \theta d(\cos \theta). \end{aligned} \quad (14)$$

To determine the selection rules, we use the series expansion of the Bessel function expressed as

$$J_N(x) = \left(\frac{x}{2}\right)^N \frac{1}{\Gamma(N+1)} {}_0F_1\left(N+1; -\frac{x^2}{4}\right). \quad (15)$$

This expansion reveals that $J_N(rA_0 \cos \theta)$ contains only powers of $\cos \theta$ with the same parity as N . For the integral (14) to be nonzero, we need the integrand to have even parity, which requires that N and l have opposite parity.

For analyzing the time-dependent field interaction, the electric field of a monochromatic field can be expressed as

$$\mathbf{E}(t) = \mathbf{E}_0 \sin \omega t = \mathbf{E}_0 (e^{i\omega t} - e^{-i\omega t}) / (2i). \quad (16)$$

The phase of the time-dependent part of the integral in Eq. (12) then is

$$S'_n(\tau) = S_n(\tau) - N'\omega\tau = \int_{-\infty}^{\tau} \frac{A^2(t)}{2} dt + (E_n + I_p - N'\omega)\tau, \quad (17)$$

where $N' = N \pm 1$ represents the number of absorbed photons, which indicates that the numbers N' and l must have the same parity. This is consistent with the selection rules for multiphoton transitions. In Appendix A, we display an alternative form of the matrix element $M_{nlm}^{(1)}$, which also clearly exhibits the selection rules.

III. RESULTS AND DISCUSSION

A. Population of the Rydberg state from the TDSE

Figure 1(a) shows the distribution of Rydberg states with principal quantum number n as a function of laser peak intensity, calculated by the TDSE approach. The laser wavelength is 400 nm and the pulse duration corresponds to 40 optical cycles. According to energy conservation, at least five photons are required for ionization from the ground state of hydrogen. We also mark the laser intensities corresponding to the opening of higher-order resonances by the white dashed lines in Fig. 1(a).

A striking feature in Fig. 1(a) is the appearance of clear fringes in the n distribution as the laser intensity increases. These intensity-dependent oscillations result from the interference of different excitation pathways. Notably, there are approximately eight interference fringes in the five-photon resonance regime and slightly fewer in the six-photon resonance regime. However, when the laser intensity increases into the seven-photon resonance regime, the number of fringes abruptly decreases to four and less.

This sudden change in the oscillation pattern suggests a fundamental shift in the underlying interference mechanism. Previous studies attributed this transition to the complete depletion of the ground-state population before the pulse

reaches its peak intensity under high-intensity conditions, accordingly eliminating excitation pathways with contributions from the second half of the pulse and fundamentally altering the interference mechanism. Zhao *et al.* [27] proposed that in the seven-photon resonance regime, due to ground-state depletion, the long-time interference originating from both sides of the pulse disappears, leaving only interference between electron wave packets excited during the rising edge of the pulse through absorption of five, six, and seven photons. According to the transition selection rule mentioned above, for an excited state with given l only excitation pathways with the same parity of N can interfere with each other.

To further analyze the fine structure and the mechanism of these oscillations, we perform further investigations by directly manipulating the ground-state population and the excitation pathways within our theoretical model.

B. Population of the Rydberg state from the model

To systematically investigate the interference mechanisms, we employ our analytical model to calculate the excitation probability as a function of laser peak intensity. Figure 2(a) shows results for the $15s$ (black line) and $15p$ (red line) states using the same parameters as in Fig. 1. Importantly, these calculations do not include ground-state depletion effects, allowing us to first identify the intrinsic interference mechanisms before examining how depletion modifies them.

Intrinsic interference patterns without depletion. For the low-intensity five-photon-dominated resonance regime with laser peak intensity $\sim 110 \text{ TW/cm}^2 < I < \sim 320 \text{ TW/cm}^2$ and the six-photon-dominated resonance regime with $< \sim 320 \text{ TW/cm}^2 < I < \sim 530 \text{ TW/cm}^2$, our model (without depletion) reproduces the TDSE results remarkably well, as shown in Fig. 2(a), capturing approximately eight and seven oscillation peaks, respectively. This agreement demonstrates that our analytical framework correctly describes the fundamental interference mechanisms in these regimes. Moreover, within each photon-number plateau, we observe that the oscillation period gradually decreases with increasing intensity—a feature that will be explained in Sec. III C through the phase-slope mechanism.

Discrepancy at high intensities and role of ground-state depletion. In the high-intensity seven-photon and eight-photon resonance regimes with $I > \sim 530 \text{ TW/cm}^2$, our theoretical model predicts more oscillation peaks than observed in the TDSE calculations. This systematic discrepancy indicates the emergence of physics beyond our initial model formulation. To test the hypothesis that ground-state depletion is responsible, we artificially suppress contributions from the falling edge of the pulse by setting the ground-state population to zero after the pulse center. Figure 2(b) shows that this modification reduces the number of oscillation peaks by approximately half [black dashed line compared to the red solid line from panel (a)], bringing the model into qualitative agreement with TDSE results at high intensities.

Physical interpretation. This comparison reveals that as the laser peak intensity increases into the seven-photon resonance regime, the ground state becomes significantly depleted during the rising edge of the pulse. This depletion eliminates the excitation pathway during the falling edge, thereby

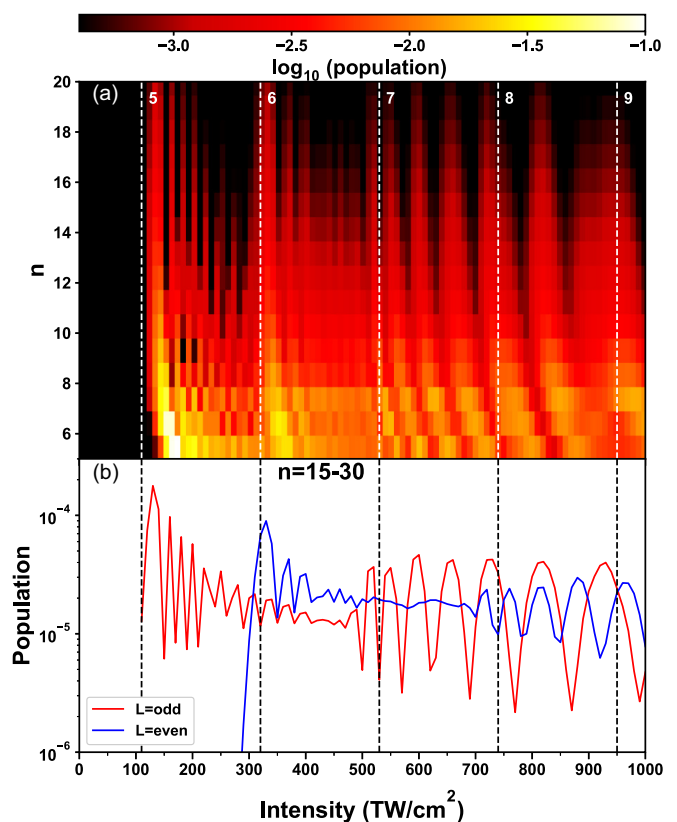


FIG. 1. Results from TDSE simulations for the H atom. (a) Population distribution of Rydberg states with $5 \leq n \leq 20$ as a function of the laser peak intensity. The white dashed lines indicate the peak intensities corresponding to the opening of the multiphoton resonances with five, six, and more photons, as marked at the upper ends of the lines. (b) Population of Rydberg states with odd l (red line) and even l (blue line) as a function of laser intensity, obtained by summing over $15 < n < 30$.

suppressing the long-time interference between contributions from both sides of the pulse [mechanism (1) as defined in the Introduction]. The resulting reduction in oscillation number provides direct computational evidence—rather than merely correlation-based inference—for the role of ground-state depletion in modifying the interference structure. However, even the complete suppression of the falling-edge contribution in Fig. 2(b) does not fully explain all features observed in the TDSE results, motivating a more detailed analysis of how depletion affects the remaining interference mechanisms, which we present in Sec. III C.

C. Oscillation pattern and interference mechanism

To relate the oscillation patterns and the interference mechanisms, we define the excitation amplitude $M'(t_1, t_2)$ as

$$M'(t_1, t_2) = -i \int_{t_1}^{t_2} d\tau \langle \psi_R^d(\tau) | H_I(\tau) | \Psi_g(\tau) \rangle, \quad (18)$$

which takes into account excitation events in the time interval $t_1 < \tau < t_2$. We define the magnitude and the phase by writing $M'(t_1, t_2) = |M'(t_1, t_2)| \exp[i\Phi(t_1, t_2)]$. Figure 3(a) shows the cosine of the phase difference between the excitation

amplitudes $M'(T_0, T_0 + \Delta t)$ and $M'(T_0 - \Delta t, T_0)$ within the falling and the rising edges of the laser pulse, namely, $\cos[\Phi(T_0 + \Delta t, T_0) - \Phi(T_0 - \Delta t, T_0)]$, as a function of $\Delta t/T$ and the peak laser intensity. Here, $\Delta t/T$ specifies a time interval on either side of the pulse center located at the time T_0 . In Fig. 3(a), for given intensity the phase difference becomes largely independent of $\Delta t/T$ if it is sufficiently large. Especially, the positions of the bright fringes in Fig. 3(a), where the cosine of the phase difference is close to unity, are in accordance with the positions of the peaks plotted in Fig. 2(a). Furthermore, in Fig. 3(b) we plot the magnitude of the intensity-dependent excitation amplitude $M'(T_0 - \Delta t, T_0)$ from the first half of the pulse. It shows the structural features similar to those in Fig. 3(a). By careful comparison between Figs. 3(a) and 3(b), we find that the former actually exhibits a splitting of each bright fringe, which is absent from the latter. This indicates that the long-time interference causes the splitting of the oscillation peaks, while their positions are already determined by short-time interference during the first half of the pulse.

For a given multiphoton resonance channel, there is a time at which the time-dependent laser intensity starts to exceed the corresponding multiphoton resonance intensity. These times (specified by $\Delta t/T$ relative to the peak T_0) for the five-photon and seven-photon resonance channels are represented by the black lines in Fig. 3(b). As expected, the dominant contributions to the excitation rate come from those times when the intensity exceeds the resonant intensity.

Next, we will give an analysis of the plateaus in Fig. 2 similar to the discussion in Ref. [14]. If the phase of the transition matrix element at a given time is $S_n(t)$, then one optical cycle later it is

$$S_n(t + T) = S_n(t) + (U_p + E_n + I_p)T. \quad (19)$$

The contributions of these trajectories differ by the phase $(U_p + E_n + I_p)T = (U_p + E_n + I_p)2\pi/\omega$ [36]. They add constructively if the phase difference equals an integer multiple of 2π . Since the action only depends on $A^2(t)$, we also have

$$S_n(t + T/2) = S_n(t) + (U_p + E_n + I_p)T/2. \quad (20)$$

The contributions of these trajectories differ by the phase $(U_p + E_n + I_p)T/2 = (U_p + E_n + I_p)\pi/\omega$. For constructive interference to occur again, this phase difference must increase by 2π , which requires a change in the ponderomotive potential $\Delta U_p = 2\omega$. Therefore, the interference of quantum trajectories separated by half an optical cycle reveals a platform modulation with periodicity $\Delta I = 416 \text{ TW/cm}^2$ corresponding to $\Delta U_p = 2\omega$ for our parameters, as shown in Fig. 3(b), while the interference of trajectories separated by a full optical cycle produces a platform modulation with periodicity $\Delta I = 208 \text{ TW/cm}^2$ corresponding to $\Delta U_p = \omega$, as shown in Fig. 3(c).

Another factor to consider is the phase induced by the parity of the initial ground state and the final Rydberg state. As shown in Fig. 2(a), the red line represents transitions between initial and final states with opposite parities, while the black line represents transitions with the same parity. For opposite-parity transitions, an additional phase of π occurs, causing a platform structure displacement of $\Delta U_p = \omega$ compared to same-parity transitions. If we neglect the parity of the wave

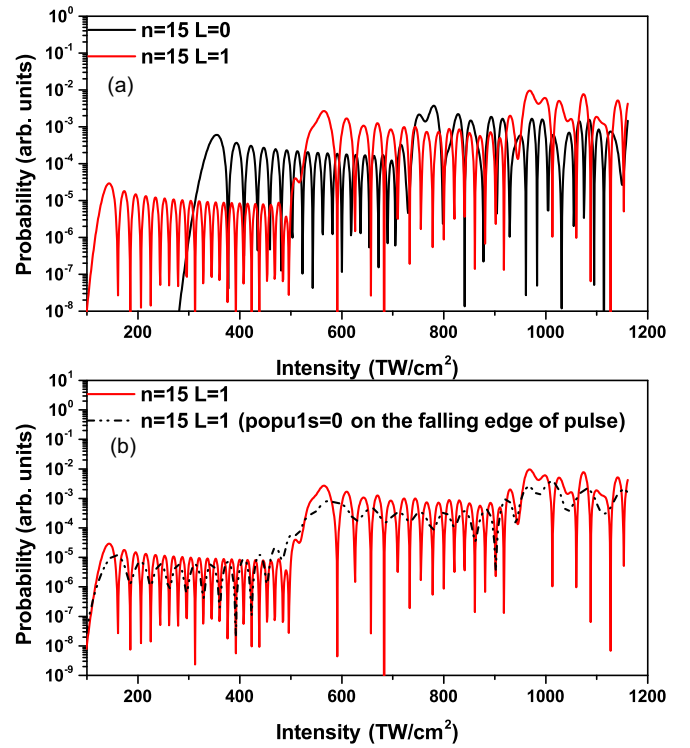


FIG. 2. (a) Excitation probability as a function of laser peak intensity from our theoretical model for the $15s$ excited state (black line) and $15p$ excited state (red line). (b) Excitation probability after considering complete depletion at the falling edge of the pulse (black dashed line). The red line is the same as in panel (a) for comparison.

function by performing spatial integration over half space, an additional platform shifted by $\Delta U_p = \omega$ will be superimposed on the original platform with spacing $\Delta U_p = 2\omega$, ultimately forming a platform structure with spacing $\Delta U_p = \omega$, as illustrated in Fig. 3(d).

To understand the origins of the splitting and oscillation pattern, we will investigate the phase of the transition matrix element in terms of the accumulation over time from cycle to cycle for a given photon-number resonance. The expressions of the phase accumulations for the long-time interference of contributions from both sides of the pulse and the short-time interference from its rising edge are

$$\phi_{\text{long-time}} = \frac{E_0^2}{4\omega^2} \int_{t_r}^{t'_r} f^2(t) dt - (N\omega - E_n - I_p)(t'_r - t_r) \quad (21)$$

and

$$\phi_{\text{short-time}} = \frac{E_0^2}{4\omega^2} \int_{t_r}^{t_0} f^2(t) dt - (N\omega - E_n - I_p)(t_0 - t_r), \quad (22)$$

where t_r and t'_r are the instants of time related to the resonance intensity on the rising and falling edges, respectively, while t_0 is located in the first optical cycle near the pulse center in the rising edge. In order to understand the growth characteristic of the phase with the peak laser intensity, we define a

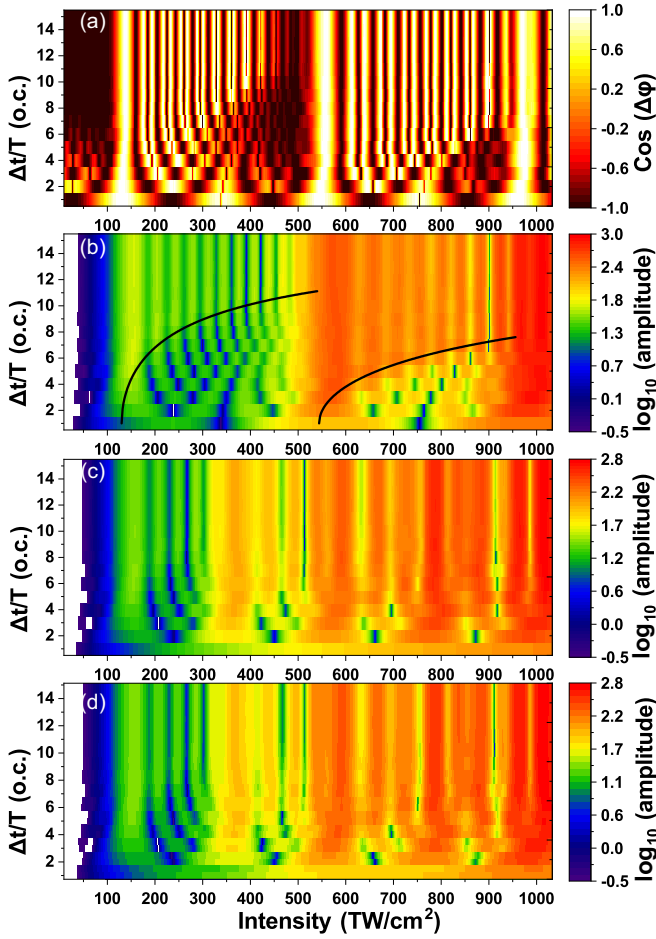


FIG. 3. Results from simulations of the analytical model for the H atom. (a) Cosine of the phase difference between the excitation amplitudes $M'(T_0, T_0 + \Delta t)$ and $M'(T_0 - \Delta t, T_0)$ as a function of $\Delta t/T$ and the laser peak intensity; the color code is given on the right side of panel (a). (b) Excitation amplitude $M'(T_0 - \Delta t, T_0)$ as a function of $\Delta t/T$ and the laser peak intensity. The black lines specify the times (in terms of $\Delta t/T$) when the time-dependent intensity reaches the five-photon and seven-photon resonance intensities, respectively. (c) Same as panel (b), but only considering the leading half of each optical cycle. (d) Same as panel (b), with the spatial integration over half space to neglect wave-function parity.

function $F(t, x) = x f^2(t)$, where $x = I/I_0$ denotes the ratio of the laser peak intensity I to the five-photon resonance intensity $I_0 \approx 110 \text{ TW/cm}^2$. The phase can be geometrically interpreted as the area enclosed by the (rising) edge of the function $E_0^2/(4\omega^2)f^2(t)$ and the line corresponding to the resonance intensity up to the time t , as shown in Fig. 4(a). The second term of the phase corresponds to the rectangular area below the resonance-intensity threshold.

Obviously, for $t_0 \approx T_0$ in Eq. (22), the phase accumulation of the long-time interference (21) is twice that of the short-time interference (22), thereby inducing the splitting of the bright fringes based on short-time interference due to phase evolution, which we observed in the comparison of Figs. 3(a) and 3(b). Furthermore, within each photon-number resonance regime, the slope of the phase with respect to intensity grad-

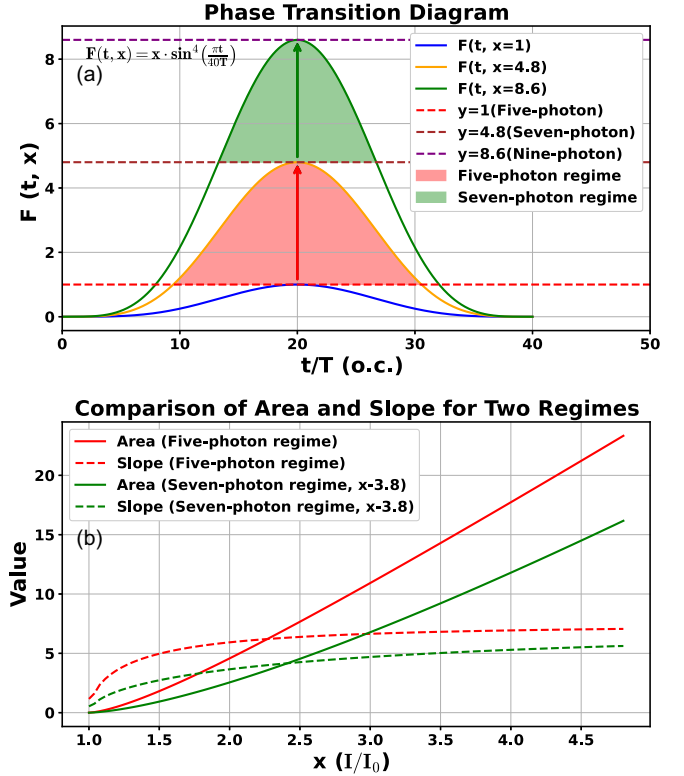


FIG. 4. Schematic diagram of phase variation of the long-time interference on both sides and the short-time interference on the rising edge with laser peak intensity. (a) The function $F(t, x)$ as a function of time. The red region indicates the phase variation in the five-photon regime, and the green region shows the phase variation in the seven-photon regime. The dashed lines represent the resonant laser intensities corresponding to different photon number regimes. (b) The phase variation of the short-time interference on the rising edge with x . The red solid line represents the five-photon regime, while the green solid line represents the seven-photon regime. The dashed lines show the variation in slopes for both regimes. (For ease of comparison between different regimes, the green line has been horizontally shifted along the x axis.)

ually increases [presented by the dashed lines in Fig. 4(b)], meaning that the phase changes more rapidly with increasing intensity. This phenomenon manifests itself in the slow decrease of the oscillation period along each photon-number plateau. It is most clearly seen in Fig. 2.

When transitioning from a lower photon-number resonance regime to a higher one, the phase slope suddenly decreases [as illustrated by the transition from the red dashed line to the green dashed line in Fig. 4(b)], causing the oscillation period to abruptly increase. This explains why the number of oscillation peaks decreases in the transition from the five-photon resonance regime to the seven-photon resonance regime, even in the absence of ground-state depletion, as is evident from Fig. 2. As the pulse duration continues to increase, the oscillation amplitude gradually decreases. In the limit of infinite pulse duration, the time-dependent envelope degenerates into a constant envelope, and the oscillations reduce to the conventional single-peak structure with two-photon spacing. For the special case of a triangular envelope, a quantitative

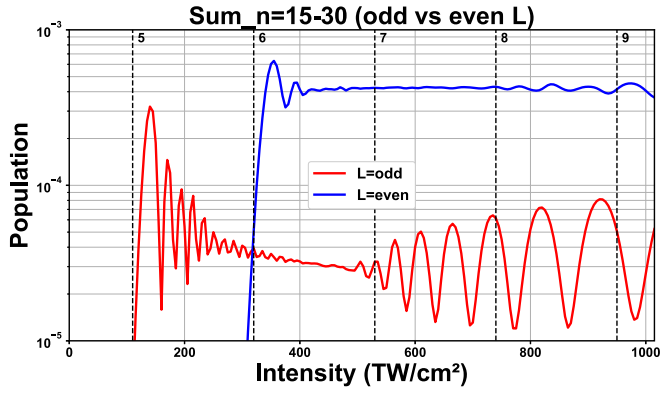


FIG. 5. Population of Rydberg states of the H atom with odd L (red line) and even L (blue line) as a function of laser intensity (obtained by summing over $n = 15$ – 30), calculated using the analytical model corrected for the changing initial-state population as obtained from the TDSE.

analytical discussion is presented in Appendix C. It is worth noting that the choice of a relatively long pulse (40 optical cycles) in our calculations is motivated by the need to clearly resolve the oscillation structures. As derived analytically in Appendix C [Eq. (C5)], the oscillation period increases with decreasing pulse duration. For very short pulses (a few femtoseconds), the oscillation period can become comparable to the plateau spacing, making these two structures difficult to distinguish experimentally. Additional calculations for shorter pulse durations, presented in Appendix D, confirm this effect.

We note that the TDSE results do not consistently reproduce this phenomenon at high intensities. Hence, considering the depletion of the ground state at the falling edge of the pulse in the model is insufficient to explain this discrepancy. Therefore, we further modified the model using the ground-state population distribution results calculated by the TDSE for all intensities and times. The results are shown in Fig. 5, and they are essentially consistent with the TDSE results. They confirm that the depletion of the ground state at high intensities has significantly disrupted the fundamental oscillation pattern originally formed by short-time interference on the rising edge. This leads to an anomalous reversal phenomenon where the oscillation periods increase within the high-intensity plateaus. At high intensities, the ground-state depletion not only eliminates excitation pathways during the falling edge but also progressively erodes the phase accumulation (22) on the rising edge from the pulse center toward earlier times, requiring larger intensity increases to satisfy the constructive interference condition $\phi_{\text{short-time}} = 2\pi n_s$, thereby increasing the oscillation period and even reversing its decreasing trend in the low-intensity region.

Moreover, as shown in Fig. 5, the strong depletion of the ground state under high laser intensity inevitably affects the plateau structure, almost causing it entirely to disappear. This may also be an important reason of why only the first plateau (at low laser intensity) has been observed in experiments, while the subsequent plateaus (at higher laser intensity) have been indistinct [22].

IV. CONCLUSION

We have presented a comprehensive analysis of the intensity-dependent oscillations in multiphoton Rydberg excitation using both TDSE calculations and an analytical model. Our results reveal that the oscillatory structure of the Rydberg-state excitation populations is formed by the interplay of three distinct interference mechanisms: short-time interference, intracycle interference, and long-time interference, each affecting the oscillation structure differently. Short-time interference occurring on the same side of the pulse determines the fundamental oscillation pattern within each plateau, with the oscillation period gradually decreasing as the laser intensity increases. Long-time interference induces a spectral splitting of the oscillations, reducing the oscillation period to half of its original value. Meanwhile, intracycle interference causes the plateau structure to exhibit two-photon spacing, playing a role equivalent to the selection rules determined by the symmetry of wave functions with different parities. When the laser intensity traverses a photon-number resonance threshold, the oscillation period undergoes an abrupt change due to the abrupt change of the accumulated phase difference between the dominant excitation pathways. Furthermore, ground-state depletion substantially modifies all three interference mechanisms and the resulting fine structure of the oscillations of the Rydberg-state population. At high intensity, the ground state becomes completely depleted before the pulse reaches its peak intensity, leading to the suppression of long-time interference between the rising and falling edges and consequently eliminating the oscillation splitting. Simultaneously, ground-state depletion that occurs on the rising edge of the pulse inevitably reverses the fundamental oscillation pattern, transforming the gradual decrease of the oscillation periods within the same plateau with increasing intensity into a gradual increase, and disrupts the short-time and intracycle interference, thereby leading to the suppression of plateau structures in the high-intensity regime. Our analytical framework, based on the strong-field approximation and an analysis of the phase accumulation, is conceptually general and could potentially be extended to other systems exhibiting Rydberg-like characteristics. For instance, Rydberg excitons in semiconductors [37–40] share qualitative features with atomic Rydberg states, including large spatial extent and hydrogenlike energy series. While application to such solid-state systems would require incorporating additional effects such as dielectric screening, effective mass differences, and many-body interactions, the fundamental interference mechanisms identified here should be operational as well and provide insights into the strong-field dynamics. These findings contribute to a deeper understanding of the fundamental quantum dynamics in strong-field interactions and may have implications for the development of spectroscopic techniques based on quantum interference.

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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.

APPENDIX A: TRANSITION MATRIX ELEMENT FOR AN INFINITELY EXTENDED FIELD

For an infinitely extended field with period T , we exploit the periodicities $A(t + T/2) = -A(t)$, $A^2(t + T/2) = A^2(t)$, $H_I(t + T/2) = -H_I(t)$, etc. to obtain

$$M_{nlm}^{(1)} = -i\frac{\omega}{2} \sum_{k=-\infty}^{\infty} \int_0^{T/2} d\tau \exp[-i(E_n + U_p + I_p)\tau] \\ \times \int d^3\mathbf{r} \Phi_{nlm}^*(\mathbf{r}) H_I(\mathbf{r}, \tau) \Phi_g(\mathbf{r}) \{-(1 + (-1)^l)i \\ \times \sin(\mathbf{r} \cdot \mathbf{A}(\tau)) \delta(E_n + U_p + I_p - 2k\omega) + (1 - (-1)^l) \\ \times \cos(\mathbf{r} \cdot \mathbf{A}(\tau)) \delta(E_n + U_p + I_p - (2k + 1)\omega)\},$$

where we used that

$$\sum_{k=-\infty}^{\infty} e^{ik\pi x} = 2 \sum_{k=-\infty}^{\infty} \delta(x - 2k). \quad (\text{A1})$$

This shows that for given l Rydberg resonances are separated by intensity intervals of 416 TW/cm² corresponding to $\Delta U_p = 2\omega$ for our parameters. It also illustrates the selection rules: Excitation to a Rydberg state with even (odd) l requires absorption of an even (odd) number of photons.

APPENDIX B: COMPARISON WITH RESULTS OF REF. [22]

For a comparison with previous work, we calculate the populations of the 6s and 7s states in the Ar atom using laser parameters identical to those employed in Ref. [22]: a linearly polarized laser field with wavelength $\lambda = 400$ nm and a Gaussian temporal envelope with full width at half maximum (FWHM) of 40 fs. The results are presented in Fig. 6(a) for the 6s state and Fig. 6(b) for the 7s state. Figure 6 demonstrates that our analytical model is consistent with the calculations of Ref. [22]. Specifically, the intensity-dependent oscillation periods predicted by our model are in good agreement with both their two-level model and their TDSE results.

However, a notable difference appears in the oscillation contrast, which is larger in our model compared with their TDSE calculations. This discrepancy can be attributed to the fact that our model does not account for ionization losses from Rydberg states, which tend to dampen the oscillation amplitudes in the full quantum dynamics [22].

APPENDIX C: INTENSITY-DEPENDENT PERIODS OF THE OSCILLATIONS WITHIN EACH PLATEAU

To derive an analytical expression for the oscillation period, we approximate the squared envelope function $f^2(t)$ as

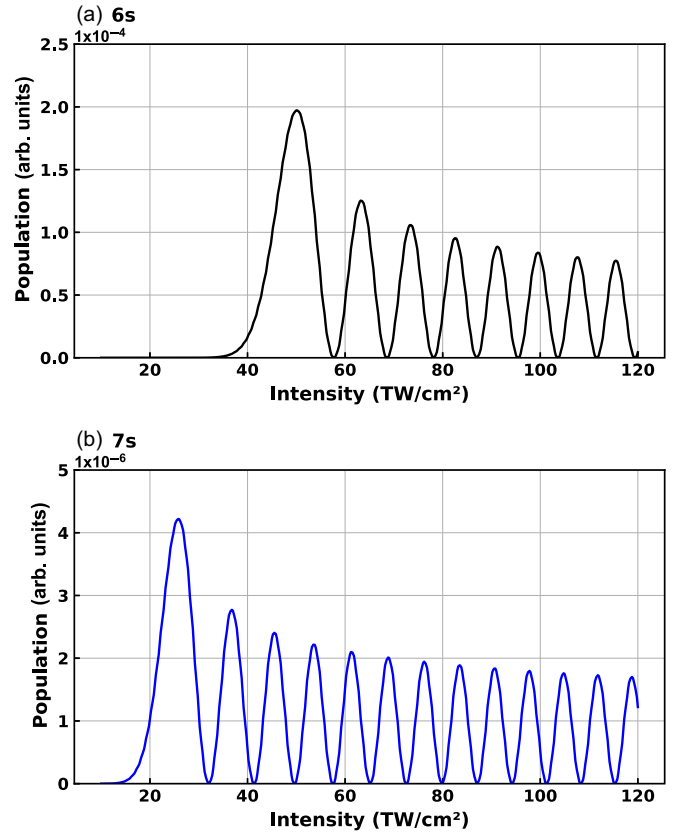


FIG. 6. Results from the analytical model for Ar atoms subjected to a linearly polarized laser field at 400 nm wavelength with a Gaussian envelope (FWHM = 40 fs), using parameters consistent with Ref. [22]. (a) Population of the 6s state as a function of laser peak intensity. (b) Same as panel (a), but for the 7s state.

a piecewise linear function:

$$f^2(t) = \begin{cases} \frac{2U_{p0}}{t_p} t, & t \leq t_p/2, \\ -\frac{2U_{p0}}{t_p} (t - t_p), & t > t_p/2, \end{cases} \quad (\text{C1})$$

where $U_{p0} = E_0^2/(4\omega^2)$ denotes the ponderomotive potential at peak intensity and t_p is the full pulse duration. Under this approximation, the phase accumulation for long-time interference between both sides of the pulse, as given by Eq. (21), can be expressed as

$$\phi_{\text{long-time}} = [U_{p0} - (N\omega - E_n - I_p)] \left(\frac{t_p}{2} - t_r \right), \quad (\text{C2})$$

where $t_r = (N\omega - E_n - I_p)t_p/(2U_{p0})$ represents the time at which the laser intensity on the rising edge reaches the resonance condition.

Applying the constructive interference condition $\phi_{\text{long-time}} = 2\pi n_s$, where n_s is an integer labeling the oscillation order, we obtain

$$U_{p0} = \frac{1}{t_p} [2\pi n_s + N\omega t_p - I_p t_p - E_n t_p \\ + 2\sqrt{\pi} \sqrt{\pi n_s^2 + N\omega n_s t_p - I_p n_s t_p - E_n n_s t_p}]. \quad (\text{C3})$$

The n_s th oscillation period is given by

$$\begin{aligned}\Delta I_{0,n_s} &= \Delta E_{0,n_s}^2 = 4\omega^2 \Delta U_{p0,n_s} \\ &= 4\omega^2 (U_{p0,n_s} - U_{p0,n_s-1}) \\ &= \frac{8\sqrt{\pi}\omega^2}{t_p} [\sqrt{\pi} + \sqrt{n_s(\pi n_s + (N\omega - E_n - I_p)t_p)} \\ &\quad - \sqrt{(n_s - 1)[\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}].\end{aligned}\quad (\text{C4})$$

By taking derivatives with respect to different parameters, we can analyze how the oscillation period varies with these parameters.

Dependence on pulse duration. The derivative with respect to t_p yields

$$\begin{aligned}\frac{\partial \Delta I_{0,n_s}}{\partial t_p} &= -\frac{4\sqrt{\pi}\omega^2}{t_p^2} \left[2\sqrt{\pi} + \frac{n_s[2\pi n_s + (N\omega - E_n - I_p)t_p]}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right. \\ &\quad \left. - \frac{(n_s - 1)[2\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}{\sqrt{(n_s - 1)[\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}} \right].\end{aligned}\quad (\text{C5})$$

Since the derivative

$$\begin{aligned}\frac{\partial}{\partial n_s} \left[\frac{n_s[2\pi n_s + (N\omega - E_n - I_p)t_p]}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right] &= \frac{n_s[4\pi^2 n_s^2 + 6\pi(N\omega - E_n - I_p)n_s t_p + (N\omega - E_n - I_p)^2 t_p^2]}{2[n_s(\pi n_s + (N\omega - E_n - I_p)t_p)]^{3/2}}\end{aligned}\quad (\text{C6})$$

is positive, the expression in brackets in Eq. (C5) is also positive, ensuring that $\partial \Delta I_{0,n_s} / \partial t_p < 0$. Therefore, the oscillation period decreases as the pulse duration increases, as it should.

Dependence on laser frequency. The derivative with respect to ω is

$$\begin{aligned}\frac{\partial \Delta I_{0,n_s}}{\partial \omega} &= \frac{4N\sqrt{\pi}\omega^2}{t_p} \left[\frac{n_s}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right. \\ &\quad \left. - \frac{n_s - 1}{\sqrt{(n_s - 1)[\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}} \right] \\ &\quad + \frac{16\sqrt{\pi}\omega}{t_p} [\sqrt{\pi} + \sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]} \\ &\quad - \sqrt{(n_s - 1)[\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}].\end{aligned}\quad (\text{C7})$$

Since the derivative

$$\begin{aligned}\frac{\partial}{\partial n_s} \left[\frac{n_s}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right] &= \frac{(N\omega - E_n - I_p)n_s t_p}{2[n_s(\pi n_s + (N\omega - E_n - I_p)t_p)]^{3/2}}\end{aligned}\quad (\text{C8})$$

is positive, both terms in Eq. (C7) are positive, yielding $\partial \Delta I_{0,n_s} / \partial \omega > 0$. Consequently, the oscillation period increases with laser frequency.

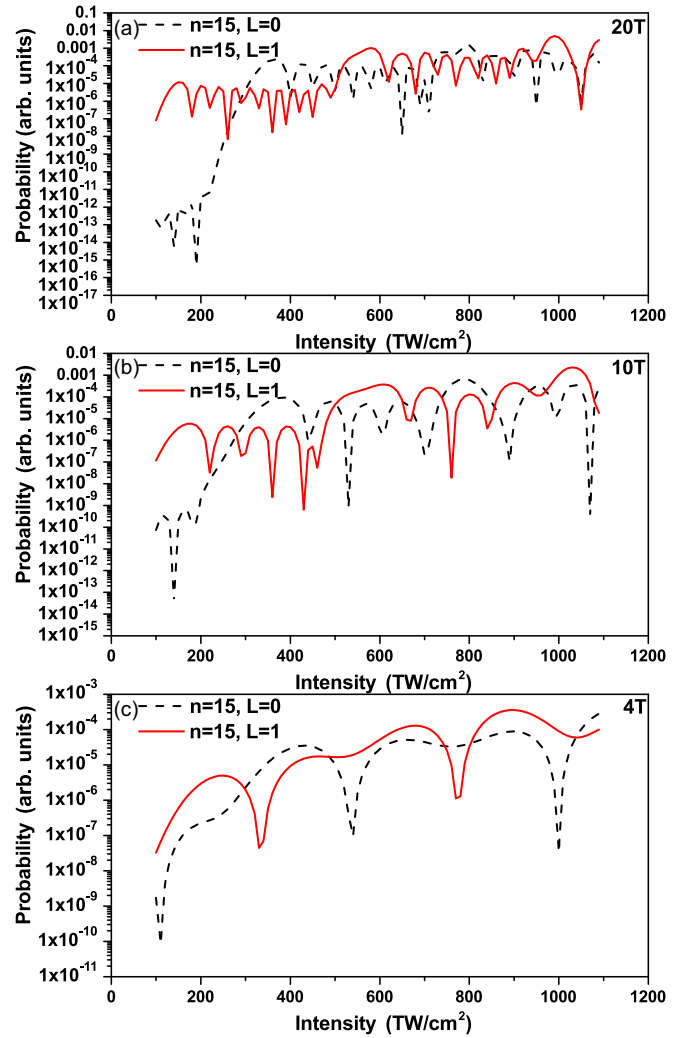


FIG. 7. Excitation probability as a function of laser peak intensity for the $n = 15$, $L = 0$ state (black dashed line) and $n = 15$, $L = 1$ state (red solid line), calculated using the analytical model for three different pulse durations: (a) 20 optical cycles (20T), (b) 10 optical cycles (10T), and (c) 4 optical cycles (4T).

Dependence on oscillation order. The derivative with respect to the oscillation index n_s gives

$$\begin{aligned}\frac{\partial \Delta I_{0,n_s}}{\partial n_s} &= \frac{4\sqrt{\pi}\omega^2}{t_p} \left[\frac{2\pi n_s + (N\omega - E_n - I_p)t_p}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right. \\ &\quad \left. - \frac{2\pi(n_s - 1) + (N\omega - E_n - I_p)t_p}{\sqrt{(n_s - 1)[\pi(n_s - 1) + (N\omega - E_n - I_p)t_p]}} \right].\end{aligned}\quad (\text{C9})$$

The derivative

$$\begin{aligned}\frac{\partial}{\partial n_s} \left[\frac{2\pi n_s + (N\omega - E_n - I_p)t_p}{\sqrt{n_s[\pi n_s + (N\omega - E_n - I_p)t_p]}} \right] &= -\frac{(N\omega - E_n - I_p)^2 t_p^2}{2[n_s(\pi n_s + (N\omega - E_n - I_p)t_p)]^{3/2}}\end{aligned}\quad (\text{C10})$$

is negative, so the expression in brackets in Eq. (C9) is also negative, yielding $\partial \Delta I_{0,n_s} / \partial n_s < 0$. This confirms that the oscillation period gradually decreases as the laser intensity increases (corresponding to a larger oscillation index n_s), in agreement with our numerical observations in Fig. 2.

APPENDIX D: SHORT-PULSE RESULTS

To investigate the effect of pulse duration on the oscillation structures, we perform calculations for three different pulse durations: 20, 10, and 4 optical cycles, in place of the 40 cycles used in the main text, with results shown in Fig. 7. The calculations are performed for $n = 15$ states with both

$L = 0$ (black dashed lines) and $L = 1$ (red solid lines). For the 20-cycle pulse shown in panel (a), for the $L = 1$ state, approximately ten oscillations are visible within the five-photon resonance plateau. When the pulse duration is reduced to ten optical cycles [panel (b)], only about five oscillations remain discernible. For the shortest pulse of four optical cycles [panel (c)], the oscillation structure is greatly suppressed, with only about two broad features remaining within the photon-number plateau. This demonstrates that longer pulses are necessary to clearly resolve the fine oscillation structures arising from the interference mechanisms discussed in Sec. III. These results confirm the analytical predictions from Appendix C, specifically Eq. (C5), which shows that the oscillation period increases with decreasing pulse duration.

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