Editors' Suggestion

Reconciliation of effective Hamiltonians for intense light-matter interaction

Jakob Nicolai Bruhnke and Jan Marcus Dahlström*
Department of Physics, Lund University, 22100 Lund, Sweden

(Received 6 May 2026; accepted 20 August 2026; published 16 September 2026)

Essential-state models are central for quantum control and technology in broad regimes of light-matter interaction. The canonical effective Hamiltonian is obtained equivalently from adiabatic elimination, the Markov approximation, and the pole approximation. These approximations are known to break down at high intensities, significantly limiting their applicability to moderate light-matter interaction. We show how this limitation can be addressed by applying quasidegenerate Rayleigh-Schrödinger perturbation theory (QDRSPT). We reconcile QDRSPT with adiabatic elimination and propose a quasidegenerate extension of adiabatic elimination that is robust when the detuning of the essential states is non-negligible. The accuracy of QDRSPT is demonstrated in both the low- and high-frequency regimes, showing excellent agreement with Floquet calculations at high intensities. The crucial corrections to adiabatic elimination make the eigenvectors of the effective Hamiltonian nonorthogonal. Physically, this allows us to account for the asymmetric strength with which different essential states couple to the nonessential states. We expect that our systematic approach to effective Hamiltonians from QDRSPT will constitute a new state of the art in intense light-matter interaction and quantum optics with novel forms of strong coupling and quantum control phenomena being conceivable.

DOI: 10.1103/jkmy-8zwr

I. INTRODUCTION

Coherent control is centered around the goal of precisely controlling electrons in quantum systems such as atoms, molecules, and condensed matter. In the low-frequency regime, where the photon energy is small compared to the binding energy, this control is routinely exerted through coherent driving of bound-bound and bound-continuum transitions by intense fields, giving rise to a rich landscape of nonlinear quantum phenomena. These include the Autler-Townes-like splittings in resonant multiphoton ionization of alkali-metal atoms [1] and the associated spin-orbit dynamics [2,3], with recent work exploring spin polarization [4], signatures beyond the rotating-wave approximation [5], coherent control of Stark shifts through dynamic interference [6–8], and two-photon Rabi oscillations in alkali-metal atoms [9]. As the laser field strength is increased further, we enter the strong-field regime of light-matter interaction, in which we stop thinking about electronic populations in field-free states and start thinking in terms of electronic trajectories, leading to high-harmonic generation [10] and attosecond physics [11].

Recent advancements in free-electron laser science [12–14] have made it possible to drive coherent processes with extreme ultraviolet (XUV) light [15–17]. By now, intensities around 10^{14} W cm^{-2} are routinely reached in experiments. At these intensities, nonlinear processes can become not only relevant, but even dominant. For example, for the resonant $1s^2 \leftrightarrow 1s4p$ transition, two-photon nonresonant ionization from $1s^2$ is more likely than single-photon ionization from $1s4p$ beyond 2×10^{13} W cm^{-2} [15]. Closely related is the dressed-atom stabilization in helium, in which, through interference of the two possible ionization pathways (resonant and nonresonant), one of the dressed states becomes stabilized against ionization [18,19]. The most prototypical nonlinear effect is surely a two-photon transition. This too can be driven with intense high-frequency fields [20–23]. Notably, at the intensities required to drive these two-photon transitions at ultrafast timescales (less than or equal to 100 fs), the excited electron begins to follow the instantaneous electric field in what is known as counterrotating oscillations [22], signifying the breakdown of the rotating-wave approximation and the onset of strong-field effects. We also want to mention the work from the Stanford Linear Accelerator Center on x-ray Autler-Townes doublets and Mollow triplets [24].

This wealth of nontrivial effects demands sophisticated theoretical machinery. For some problems, one must resort to numerical solutions of the semiclassical time-dependent Schrödinger equation (TDSE), while for others, the well-established essential-state approach has the advantage that the atom is reduced to a few-level system. The latter enables us to gain a quantitative understanding of the dynamics driven by the semiclassical field and connect better to Floquet or quantum optics descriptions [25–27]. The effects of other states can sometimes be completely ignored; other times it can be parametrized. The effective Hamiltonian H_{eff} for this few-level system should then contain all the important physics, and

*Contact author: marcus.dahlstrom@fysik.lu.se

its diagonalization gives rise to combined eigenstates of light and matter, referred to as the dressed states. Due to their flexibility, their predictive power, and the ease at which physical interpretations may be assigned to calculations, essential-state approaches have enjoyed popularity for several decades across various research fields. These include the area of quantum control [28], where Raman transitions are exploited for precise population transfer [29,30] and two-photon transitions are leveraged to populate high Rydberg states [31].

Parametrizing the influence of nonessential states routinely proceeds via an approximation known as adiabatic elimination [26], which has been shown to be equivalent to the Markov approximation [32] and the pole approximation [33]. We will thus refer to them as the AMP approximation and denote this canonical effective Hamiltonian by H_{AMP}. While in the 1980s, experimental constraints informed the opinion that higher-order corrections to adiabatic elimination were too small to be of interest [34], this position must be reconsidered, with several proposals on higher-order effective Hamiltonians having been put forward in the past decade [32,35–37].

However, several questions remain concerning the AMP approximation. One such question is how to choose the "correct" interaction picture for the adiabatic elimination or Markov approximation or, equivalently, how to choose the correct energy in the pole approximation. In previous studies, guidelines were suggested [32,33], but it is unsatisfactory that such a fundamental question should not have a response based on the theory of effective Hamiltonians. A consequence of projecting the full light-matter Hamiltonian onto a Hilbert subspace is that the resulting effective Hamiltonian has nonorthogonal eigenstates [32]. This is commonly ignored, presumably because the terms that give rise to the nonorthogonality are neglected in adiabatic elimination and appear only in higher-order effective Hamiltonians [32]. Here we will detail their physical significance and interpretation.

In this paper we show that the open questions of the AMP approximation can be resolved by applying quasidegenerate Rayleigh-Schrödinger perturbation theory (QDRSPT) [38]. This approach provides a clear pathway to obtain effective Hamiltonians for nearly degenerate systems. The quasidegenerate nature of QDRSPT enables us to consistently describe detunings; this we show to resolve the debate on the choice of the interaction picture. The corrections to the AMP approximation lead to the nonorthogonality of the eigenstates of the effective Hamiltonian. We will show that the nonorthogonality expresses itself through the transient population of nonresonant states. In this way, ambiguities of the AMP approximation are clarified.

Our perspective is naturally limited. Our focus on QDRSPT implies that we ignore the significant body of work on time-averaged effective Hamiltonians. These averaging procedures are based on either the Dyson expansion [39,40] or the Magnus expansion [25,41] and have recently been utilized for the description of counterrotating oscillations in quantum control theory [42,43]. Furthermore, the Schrieffer-Wolff transformation [44] yields an effective Hamiltonian in terms of operator corrections instead of energy corrections, which is particularly useful in quantum optics. Other names for the Schrieffer-Wolff transformation are van Vleck perturbation theory [45] and block diagonalization [46]; related methods are Klimov's method of small rotations [47,48] and the Morris-Shore transformation [35,37,49] (see also Refs. [28,36]). A further limitation is that QDRSPT is a time-independent theory and thus only applies to time-independent light-matter Hamiltonians. As soon as the laser field is shaped temporally by an envelope or the laser pulse is chirped, we require a time-dependent approach. For effective Hamiltonians obtained via adiabatic elimination, envelopes are convenient to incorporate [9,29,50–53]. We are only aware of one proposal that extends this approach to perturbative effective Hamiltonians beyond adiabatic elimination [25], but it was never applied. Incorporating chirp is even more elusive and will be addressed elsewhere.

Our work is structured as follows. We start with an elaborate account of QDRSPT, in which we introduce the reduced wave operator as the central object in perturbation theory. We further discuss the straightforward extension for complex-scaled Hamiltonians, which allow us to treat ionization through a monotonic decrease in norm. In the results, we first compare the usual effective Hamiltonian from adiabatic elimination to that obtained through QDRSPT. This reveals both the physical significance of the nonorthogonality of eigenstates and a nondegenerate model space. We provide three examples where the AMP approximation fails to capture physical properties: a toy model system, rubidium in an IR field, and helium in an XUV field. Our effective Hamiltonian calculations are supported by Floquet calculations [54], where the atomic parameters of rubidium are obtained through a single-active-electron potential [55], and those of helium obtained through many-body configuration-interaction singles calculations [56,57]. Atomic units are used unless otherwise stated: $e = \hbar = m = 1/4\pi\epsilon_0 = 1$.

II. THEORY

Our starting point is the time-independent light-matter Hamiltonian

$$H = H_{\mathrm{atom}} + H_{\mathrm{field}} + V \equiv H_0 + V. \tag{1}$$

Here V is the light-matter interaction, which in the length gauge and for monochromatic fields is proportional to the electric-field amplitude $\mathcal{E}_0$. For this section, it is inconsequential at what level of theory H is obtained, be it quantum optics, Floquet theory, or simply within the rotating-wave approximation in the rotating frame. We assume the uncoupled eigenvalue problem $H_0|n\rangle = E_n|n\rangle$ to be solved and write H in the eigenbasis of H_0. We refer to eigenvalues of H as the dressed energies, and the eigenstates as the dressed states. Let us for now assume that H is Hermitian and has a discrete spectrum.

In the upcoming sections, we first derive the effective Hamiltonian as a formal object. Then we derive the perturbative expansion for the effective Hamiltonian within QDRSPT. We then elaborate on Floquet theory, which offers a pathway to obtain a time-independent light-matter Hamiltonian. Finally, we show the required adjustments that allow us to describe ionization through complex-scaling techniques. In our account, we stay close to the approach detailed by Lindgren and Morrison for the atomic many-body problem [38,58],

but with electron-electron correlation being replaced by the light-matter interaction.

A. What is an effective Hamiltonian?

An effective Hamiltonian is an operator that reproduces a subset of the true eigenvalues of the full Hamiltonian H. It acts not in the full Hilbert space, but rather in a Hilbert subspace $\mathcal{P}$, referred to as the model space, where $p := \dim(\mathcal{P})$. The model space is spanned by the essential states. We choose the essential states as those that the electric field couples (near-)resonantly from the ground state (GS). All other states are referred to as nonessential. The orthogonal space, spanned by nonessential states, is denoted by $\mathcal{Q} = \mathcal{P}^{\perp}$ so that $\mathcal{P} \oplus \mathcal{Q}$ is the full Hilbert space. We define the projectors onto $\mathcal{P}$ and $\mathcal{Q}$ via

$$P = \sum_{n\in\mathcal{P}} |n\rangle\langle n|, \quad Q = 1 - P, \tag{2}$$

respectively. From this construction, it follows that $[H_0, P] = [H_0, Q] = 0$, as well as $PQ = QP = 0$, $P^2 = P$, $Q^2 = Q$, $PHQ = PVQ$, and $QHP = QVP$.

B. Formal definition of the effective Hamiltonian

We assume that p eigenstates $|\Psi_k\rangle$ $(k = 1, \ldots, p)$ of the full Hamiltonian $H|\Psi_k\rangle = \lambda_k|\Psi_k\rangle$ have their major part within the model space. Their projections onto $\mathcal{P}$ are

$$|\psi_k\rangle = P|\Psi_k\rangle, \quad k = 1, \ldots, p. \tag{3}$$

We now define the wave operator $\hat{\Omega}$, denoted by a hat in order to distinguish it from the Rabi frequency Ω, which reverses the above projection:

$$|\Psi_k\rangle = \hat{\Omega}|\psi_k\rangle, \quad k = 1, \ldots, p. \tag{4}$$

The wave operator $\hat{\Omega}$, when acting on any eigenstate projection $|\psi_k\rangle$, $k = 1, \ldots, p$, yields the respective full eigenstate $|\Psi_k\rangle$ of H. We can rewrite $\hat{\Omega} = P + \chi$, where $\chi = Q\chi P$ is the reduced wave operator, also called the correlation operator, which upon application to a $\mathcal{P}$-space state yields the corresponding components in the complementary space $\mathcal{Q}$, i.e.,

$$Q|\Psi_k\rangle = \chi|\psi_k\rangle, \quad k = 1, \ldots, p. \tag{5}$$

Notably, χ is nilpotent, $\chi^2 = 0$. By applying P to the time-independent Schrödinger equation (TISE) from the left,

$$PH\hat{\Omega}|\psi_k\rangle = E_k|\psi_k\rangle, \quad k = 1, \ldots, p, \tag{6}$$

we can identify

$$H_{\text{eff}} = PH\hat{\Omega} = PH(P + \chi) = PH_0P + PVP + PV\chi \tag{7}$$

as the energy-independent effective Hamiltonian with eigenfunctions $|\psi_k\rangle$ and eigenvalues E_k $(k = 1, \ldots, p)$. Hence, the task of finding H_{eff} is formally reduced to finding the reduced wave operator. In this work the mathematical properties of χ (i.e., existence and uniqueness) are assumed; we refer the reader to Refs. [36,59,60] for more information.

While H is Hermitian, H_{eff} is not. This is quite intuitive: H has eigenvectors $|\Psi_k\rangle$, which are orthogonal. Meanwhile, H_{eff} has the eigenstates $|\psi_k\rangle = P|\Psi_k\rangle$, which by construction are not orthogonal. Hence, H_{eff} must generally be non-Hermitian, although it still has real eigenvalues λ_k. Another way to see this is to realize that due to $\chi^2 = 0$, H_{eff} can be expressed as a similarity transform of H,

$$H_{\text{eff}} = P\mathrm{e}^{-\chi}H\mathrm{e}^{\chi}P. \tag{8}$$

Since $\mathrm{e}^{\chi} = 1 + \chi$ is not unitary, H_{eff} is generally non-Hermitian [61].

The choice of basis in quantum mechanics is of course inconsequential to the observables. Therefore, it is possible to find a Hermitian effective Hamiltonian with the same eigenvalues λ_k as H_{eff} from Eq. (7). To construct this Hermitian effective Hamiltonian, which we denote via $\mathcal{H}_{\text{eff}}$, we notice that the properties of χ and the orthogonality of the full eigenstates of H yield

$$\begin{aligned}\delta_{nm} = \langle\Psi_n|\Psi_m\rangle &= \langle\Psi_n|P|\Psi_m\rangle + \langle\Psi_n|Q|\Psi_m\rangle \\ &= \langle\psi_n|P + \chi^{\dagger}\chi|\psi_m\rangle.\end{aligned} \tag{9}$$

Note that $P + \chi^{\dagger}\chi$ is Hermitian. This suggests that the $\mathcal{P}$-space eigenfunctions defined by

$$|\phi_k\rangle = (P + \chi^{\dagger}\chi)^{1/2}|\psi_k\rangle =: \eta|\psi_k\rangle, \quad \langle\phi_k| = \langle\psi_k|\eta \tag{10}$$

are orthogonal. Hence,

$$\mathcal{H}_{\text{eff}} = \eta H_{\text{eff}}\eta^{-1} \tag{11}$$

must be Hermitian, with orthogonal eigenstates $|\phi_k\rangle$. Note that $\mathcal{H}_{\text{eff}}$ is not "manifestly Hermitian," which is a common phrase used to express that its Hermiticity cannot be seen by eye [61–63], but that it must instead be proven explicitly (see, for instance, Ref. [36]). The implication of this is profound: $\mathcal{H}_{\text{eff}}$ is only exactly Hermitian if χ is the exact reduced wave operator and H_{eff} is the exact effective Hamiltonian. If either H_{eff} or χ is obtained approximately, $\mathcal{H}_{\text{eff}}$ will only be approximately Hermitian.

For context, we finally note that $\mathcal{H}_{\text{eff}}$ is the object obtained via the Schrieffer-Wolff transformation and its related variants [37,45–47,49]. In the context of light-matter interaction, we want to highlight in particular the recent work of Sanz *et al.* [36], who treat adiabatic elimination as a singular perturbation problem, with the reduced wave operator χ (in their work denoted by B) as the central object, connecting both H_{eff} and $\mathcal{H}_{\text{eff}}$. Even more recently, Huang *et al.* employed the effective Hamiltonian $\mathcal{H}_{\text{eff}}$ in quantum control problems, focusing on (among other factors) the fast dynamics due to counterrotating oscillations in few-level systems [28].

C. Quasidegenerate Rayleigh-Schrödinger perturbation theory

Quasidegenerate Rayleigh-Schrödinger perturbation theory offers the possibility to calculate effective Hamiltonians beyond the pole approximation in a perturbative manner. The quasidegeneracy refers to the model space: $PH_0P \neq E_0P$. This is the case in light-matter interaction when the light is not exactly resonant, but slightly detuned. We will show that it is a strength of QDRSPT that the description of detuning is natural in the formalism, while the AMP effective Hamiltonian is not suited for non-negligible detunings.

In the preceding section we showed that the problem of finding the effective Hamiltonian can be reformulated to the problem of finding the reduced wave operator. With the

definitions $Q|\Psi\rangle = \chi P|\Psi\rangle$ and $\chi = Q\chi P$, we find immediately that the reduced wave operator fulfills the decoupling equation [61]

$$\begin{aligned} 0 &= Q(\mathrm{e}^{-\chi} H \mathrm{e}^{\chi})P = Q(1-\chi)H(1+\chi)P \\ &= QVP + QH\chi - \chi HP - \chi V\chi. \end{aligned} \tag{12}$$

For a proof, let $Q(1-\chi)H(1+\chi)P$ act on an arbitrary eigenstate $|\Psi_k\rangle$ of the full Hamiltonian.

Simple rearrangements yield the generalized Bloch equation

$$[\chi, H_0] = QV\hat{\Omega} - \chi V\hat{\Omega}. \tag{13}$$

We now construct a perturbative series in $\hat{\Omega}$, and thus χ,

$$\hat{\Omega} = P + \chi = P + \chi^{[1]} + \chi^{[2]} + \cdots, \tag{14}$$

where we identify $\chi^{[0]} = P$. In the context of light-matter interaction, $\chi^{[n]}$ contains all processes in which n photons are exchanged in order to get from a state $|p\rangle \in \mathcal{P}$ to a state $|q\rangle \in \mathcal{Q}$. We insert Eq. (14) into Eq. (13) and equate terms of the same order in V:

$$[\chi^{[1]}, H_0] = QVP, \tag{15}$$

$$[\chi^{[2]}, H_0] = QV\chi^{[1]} - \chi^{[1]}VP, \tag{16}$$

$$\vdots$$

$$[\chi^{[n]}, H_0] = QV\chi^{[n-1]} - \sum_{m=1}^{n-1} \chi^{[n-m]} V \chi^{[m-1]}. \tag{17}$$

We obtain an explicit formula for $\chi^{[n]}$ by projecting from the left with a $\mathcal{Q}$-space state $\langle q|$ and from the right with a $\mathcal{P}$-space state $|p\rangle$. Defining $\chi^{[n]}_{qp} := \langle q|\chi^{[n]}|p\rangle$, we obtain

$$\chi^{[n]}_{qp} = \frac{1}{E_p - E_q} \langle q| \left(QV\chi^{[n-1]} - \sum_{m=1}^{n-1} \chi^{[n-m]} V \chi^{[m-1]} \right) |p\rangle. \tag{18}$$

For notational convenience, we define elementwise multiplication in the atom-photon state basis via $\odot$ and write the resolvent matrix G via its elements

$$G_{qp} = \frac{1}{E_p - E_q}, \tag{19}$$

after which we can write

$$\chi^{[n]} = G \odot \left(QV\chi^{[n-1]} - \sum_{m=1}^{n-1} \chi^{[n-m]} V \chi^{[m-1]} \right). \tag{20}$$

The effective Hamiltonian of RSPT of nth order therefore reads

$$H^{(n)}_{\mathrm{RS}} = PH_0P + PVP + \sum_{k=1}^{n-1} PV\chi^{[k]}, \tag{21}$$

where the term $PV\chi^{[k-1]} \propto {\mathcal{E}_0}^k$. Note that the convergence of the Rayleigh-Schrödinger perturbative series for $n \to \infty$ cannot be taken for granted, but robust results will often be achieved with truncation at a low orders [64–66].

If the model space is degenerate, i.e., $PH_0P = E_0P$, the expressions simplify to

$$\chi^{[n]} = G_{\mathcal{Q}} \left(QV\chi^{[n-1]} - \sum_{m=1}^{n-1} \chi^{[n-m]} V \chi^{[m-1]} \right), \tag{22}$$

where we defined the $\mathcal{Q}$-space resolvent

$$G_{\mathcal{Q}} := \frac{Q}{E_0 - QH_0Q}. \tag{23}$$

It should be noted here that while the matrix elements of $H^{(n)}_{\mathrm{RS}}$ can of course be written down with pen and paper, the number of terms quickly become prohibitive for $n \geqslant 4$. The utility of QDRSPT in light-matter interaction comes from the ease and speed at which it can be calculated in high-level programming languages such as python. Furthermore, since the kth term of H_{RS} is proportional to $\mathcal{E}_0^k$, we can obtain H_{RS} at any field strength $\mathcal{E}_0$ by calculating the perturbative expansion of H_{RS} at a single $\mathcal{E}_0$ once (the limit being of course the convergence of H_{RS} at high $\mathcal{E}_0$). In this way, QDRSPT provides a generalization to the perturbative AMP effective Hamiltonian, where scaling parameters have previously been provided [18].

D. Floquet theory

So far, we have left the light-matter Hamiltonian H unspecified. In this section we offer a brief overview of Floquet theory, which lets us obtain a time-independent, semiclassical light-matter Hamiltonian H_F. This Hamiltonian can be obtained from the semiclassical, periodic $H(t) = H_0^{(\mathrm{atom})} + z\mathcal{E}_0 \cos(\omega t)$ via Fourier techniques. Here $\mathcal{E}_0$ is the electric-field amplitude and z the dipole operator with matrix elements z_{ij}. Alternatively, the quantum-optics Hamiltonian can be transformed into H_F for a coherent photon field in the limit of infinite cavity volume and infinite photon numbers [67,68]. These assumptions tend to be valid in intense fields. Recently, strong-field physics and quantum optics have begun to merge [69–74], an exciting development which lies outside the scope of this work. For reviews and book chapters on Floquet theory, we refer the reader to Refs. [75,76]. While we will write down the equations for linearly polarized light, Floquet theory can be elegantly formulated just as well for circularly polarized light [77]. Further, many-mode Floquet theory allows for the convenient description of bichromatic pulses [78].

We choose the semiclassical Hamiltonian $H(t)$ as our starting point. The Floquet theorem permits us to expand the time-dependent wave function in the form

$$|\Psi^{(\lambda)}(t)\rangle = \mathrm{e}^{-\mathrm{i}\lambda t} \sum_{k=-\infty}^{\infty} \mathrm{e}^{\mathrm{i}k\omega t} \left|\phi_k^{(\lambda)}\right\rangle, \tag{24}$$

labeled by the quasienergy λ. Upon substitution into the TDSE,

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}|\Psi(t)\rangle = H(t)|\Psi(t)\rangle, \tag{25}$$

we can group the resulting equations by the harmonic component to obtain the set of equations

$$\left(H_0^{(\mathrm{atom})} + k\omega\right)\left|\phi_k^{(\lambda)}\right\rangle + \frac{\mathcal{E}_0 z}{2}\left(\left|\phi_{k+1}^{(\lambda)}\right\rangle + \left|\phi_{k-1}^{(\lambda)}\right\rangle\right) = \lambda\left|\phi_k^{(\lambda)}\right\rangle, \tag{26}$$

where k runs over all negative and positive indices. The harmonic vectors $|\phi_k^{(\lambda)}\rangle$ are then collected as a column vector $|\phi^{(\lambda)}\rangle$, known as the quasienergy state, so that we obtain the simple eigenvalue equation

$$H_F|\phi^{(\lambda)}\rangle = \lambda|\phi^{(\lambda)}\rangle, \tag{27}$$

where λ is an eigenvalue of H_F. The eigenvectors and eigenvalues are often referred to as dressed states and dressed energies due to their close connection with quantum optics. The Floquet Hamiltonian can be written in an extended Hilbert space (known as the Sambe space) via

$$H_F = H_0^{(\mathrm{atom})} \otimes 11 + \omega 11 \otimes D + \frac{\mathcal{E}_0}{2} z \otimes F, \tag{28}$$

where $D = \mathrm{diag}(\ldots, -k, \ldots, 0, \ldots, k', \ldots)$ and F has ones on the sub- and superdiagonals ($F_{ij} = \delta_{i+1,j} + \delta_{i,j+1}$). In perturbation theory, we will then take $H_0^{(\mathrm{atom})} \otimes 11 + \omega 11 \otimes D$ as the unperturbed Hamiltonian H_0 whose solution is known, and $\frac{\mathcal{E}_0}{2} z \otimes F$ as the perturbation V. The unperturbed states are defined as $|n, k\rangle = |n\rangle \otimes |k\rangle$, with $H_0^{(\mathrm{atom})}|n\rangle = E_n|n\rangle$, and $D|k\rangle = k|k\rangle$. In analogy to quantum optics, we call them the (uncoupled) atom-photon states, which should however not cover up the fact that Floquet theory is a semiclassical theory.

E. Complex scaling

In the form proposed by Shirley, Floquet theory was only applicable to bound states [67]. In the following decades, Floquet theory was extended to also allow for the treatment of the continuum via complex-scaling techniques [79,80]. These techniques rotate the spectrum of H_F into the complex plane by an angle θ so that the continuum may be discretized by expanding into an L^2 basis. Often, the transform is only applied beyond a certain radius, which is known as exterior complex scaling [81]. The Floquet Hamiltonian of Eq. (28) then becomes complex symmetric with eigenvalues $E - \frac{\mathrm{i}}{2}\Gamma$, where $E \in \mathbb{R}$ is the dressed energy and $\Gamma \in \mathbb{R}$ is the decay rate (equivalently, the width) of the state.

Traditionally, the complex-scaled Floquet Hamiltonian H_F^θ was constructed via expansion into Sturmian functions [82] or Laguerre functions [79]. Another approach is to solve the TISE with complex scaling in another suitable basis, such as B splines, and subsequently construct H_F^θ from the complex-scaled atomic parameters [22].

Dealing with a complex-symmetric Hamiltonian H necessitates dealing with non-Hermitian quantum theory [83]. Under the usual scalar product $\langle\cdot|\cdot\rangle$, the adjoint $\langle\cdot|$ of the ket vector $|\cdot\rangle$ is given by $\langle\cdot| = (|\cdot\rangle)^\dagger$. Clearly, the eigenvectors of a complex-symmetric H are not orthogonal under this scalar product. To recover the concept of orthogonality for complex-symmetric Hamiltonians, we define the bra vector via the transpose $\langle\cdot| := (|\cdot\rangle)^T$. Then, if $|n\rangle$ and $|m\rangle$ are eigenfunctions of a complex-symmetric H with $H|n\rangle = \lambda_n|n\rangle$, $\lambda_n \in \mathbb{C}$, we have $\langle n|m\rangle = \langle m|n\rangle$ and $\langle n|H|m\rangle = \langle m|H|n\rangle$, leading to the orthogonality relation [84]

$$(\lambda_n - \lambda_m)\langle n|m\rangle = 0, \tag{29}$$

which guarantees the orthogonality of the eigenvectors of a symmetric Hamiltonian. Further, we obtain the usual closure identity $1 = \sum_n |n\rangle\langle n|$. We stress that these considerations only apply for a complex-symmetric H. For more general non-Hermitian H, the c-product with parentheses $(\cdot|\cdot)$ needs to be introduced, which is based on the concept of left and right eigenvectors and implies that the complex conjugation of the bra vector is performed only on those parts of the wave function that would be complex without complex scaling [83]. Complex-symmetric H are obtained if the light is linearly polarized along z and also in circular polarization if the Hamiltonian is transformed into a suitable rotating frame [77]. We can conveniently adapt QDRSPT to obtain a complex-scaled H_{eff} from H_F^θ [76,85].

Clearly, we have to justify this introduction of ambiguous notation, in which the Dirac brackets are reused for the modified scalar product. The advantage lies in the ability to generalize the following theoretical explorations. All upcoming formulas may be interpreted within either Hermitian or non-Hermitian quantum theory, simply by exchanging Hermitian conjugates and transposes, both for operators and for bra vectors. For example, for Hermitian H, the norm of the wave function $|\Psi\rangle$ can be expressed through the $\mathcal{P}$-space wave function $|\psi\rangle := P|\Psi\rangle$ as

$$\langle\Psi|\Psi\rangle = \langle\psi|P + \chi^\dagger\chi|\psi\rangle. \tag{30}$$

For complex-symmetric H, it is given by

$$\langle\Psi|\Psi\rangle = \langle\psi|P + \chi^T\chi|\psi\rangle, \tag{31}$$

where the use of the c-product was implied.

Recall that for Hermitian H, the effective Hamiltonian H_{eff} is generally non-Hermitian, while $\mathcal{H}_{\mathrm{eff}}$ is Hermitian. Analogously, for complex-symmetric H, the effective Hamiltonian H_{eff} is generally complex nonsymmetric, while $\mathcal{H}_{\mathrm{eff}}$ is complex symmetric.

III. RESULTS

A. Relation to the AMP approximation and the role of nonorthogonal eigenstates

In this section we compare the effective Hamiltonian from the AMP approximation to that of QDRSPT and propose a resolution to a longstanding debate on the proper choice of the interaction picture for the AMP approximation. Note that our expressions in this section are for Hermitian Hamiltonians $H = H_0 + V$. They can be easily reformulated for complex-symmetric Hamiltonians.

The AMP effective Hamiltonian can be formulated as

$$H_{\mathrm{AMP}} = PHP + PV\frac{Q}{E_0 - QHQ}VP; \tag{32}$$

we refer to Appendix for derivations using adiabatic elimination, the Markov approximation, and the pole approximation. The energy E_0 is in principle a free parameter. However, it is clear from the AMP approximation that E_0 should be chosen to lie in the vicinity of the $\mathcal{P}$-space spectrum. For sufficiently

weak interactions, we can expand the inverse $[E_0 - QHQ]^{-1}$ in the interaction,

$$\frac{Q}{E_0 - QHQ} = G_\mathcal{Q} + G_\mathcal{Q} V G_\mathcal{Q} + G_\mathcal{Q} V G_\mathcal{Q} V G_\mathcal{Q} + \cdots, \tag{33}$$

so that the AMP effective Hamiltonian in nth-order perturbation theory reads

$$H^{(n)}_{\mathrm{AMP}} = PHP + \sum_{k=1}^{n-1} PV(G_\mathcal{Q} V)^k P, \tag{34}$$

from which we can see by eye that the corresponding reduced wave operator is defined via the recursion

$$\chi^{[n]} = G_\mathcal{Q} V \chi^{[n-1]}, \tag{35}$$

with $\chi^{[1]} = G_\mathcal{Q} V P$. Equation (35) should be compared with the expression for $\chi^{[n]}$ from degenerate RSPT, Eq. (22). The approximations of H_{AMP} enter through the neglect of $G_\mathcal{Q} \sum_{m=1}^{n-1} \chi^{[n-m]} V \chi^{[m-1]}$.

Note how the effective interaction always follows the pattern

$$\mathcal{P} \to \mathcal{Q} \to \underbrace{\cdots}_{\text{only via } \mathcal{Q}} \to \mathcal{Q} \to \mathcal{P}, \tag{36}$$

in other words, the only allowed $\mathcal{P}$-space states are the initial and final states. These paths are necessarily always symmetric, which provides the physical explanation as to why H_{AMP} is Hermitian. We deduce that the attribute of all intermediate states lying in $\mathcal{Q}$ is the defining attribute for the AMP effective Hamiltonian.

It is instructive to compare this perturbation series with the one that we showed for the degenerate Rayleigh-Schrödinger expansion in Eq. (22). First, we notice that in degenerate RSPT, E_0 is unambiguously defined as the unperturbed $\mathcal{P}$-space eigenenergy $PH_0P = E_0P$. The lowest-order correction to H_{AMP} comes in the third order of the effective Hamiltonian, i.e., in second order of the reduced wave operator, $\chi^{[2]}$, where we subtract $G_\mathcal{Q}\chi^{[1]}VP$ from $G_\mathcal{Q}V\chi^{[1]}$. In the effective Hamiltonian, this term appears in third order as $-PVG_\mathcal{Q}\chi^{[1]}VP$. This is is the first term that causes the eigenstates of H_{eff} to be nonorthogonal. It represents a three-photon pathway of the kind $\mathcal{P} \to \mathcal{P} \to \mathcal{Q} \to \mathcal{P}$, where a first-order coupling in the $\mathcal{P}$ space is composed together with a second-order coupling via the $\mathcal{Q}$ space (we read $-PVG_\mathcal{Q}\chi^{[1]}VP$ from right to left). For a two-level $\mathcal{P}$ space with degenerate states $|a\rangle$ and $|b\rangle$ of opposite parity (unperturbed energy E_0), the off-diagonals of the three-photon pathway read

$$(-PVG_\mathcal{Q}\chi^{[1]}VP)_{ba} = -\sum_q \frac{V_{bq}V_{qb}}{(E_0 - E_q)^2} V_{ba}, \tag{37}$$

$$(-PVG_\mathcal{Q}\chi^{[1]}VP)_{ab} = -\sum_q \frac{V_{aq}V_{qa}}{(E_0 - E_q)^2} V_{ab}. \tag{38}$$

While $V_{ba} = V^\dagger_{ab}$, in general the two-photon couplings $V_{bq}V_{qb} \neq V_{aq}V_{qa}$ are not equal. This elucidates the origin of the non-Hermiticity. Only if $|a\rangle$ and $|b\rangle$ couple equally strongly to the $\mathcal{Q}$ space will H_{RS} be Hermitian. In general systems, the coupling from $\mathcal{P}$ to $\mathcal{Q}$ is asymmetric [22].

B. Quasidegenerate AMP approximation

In the literature on the AMP approximation, it has been discussed at length how to choose E_0 or, equivalently, how to choose the correct interaction picture in terms of the energy offset on the Hamiltonian's diagonal [32,33,41]. The most common proposal is to choose it as the center of the $\mathcal{P}$-space spectrum [33]. Here we propose to just take the nondegeneracy into account properly, by calculating the AMP effective Hamiltonian in accordance to QDRSPT as

$$H^{(n)}_{\mathrm{QDAMP}} = PHP + \sum_{k=1}^{n-1} PV[G \odot (QV)]^k P, \tag{39}$$

where G is defined as in Eq. (19) by the unperturbed $\mathcal{P}$-space eigenvalues. The defining equation for the reduced wave operator in this quasidegenerate AMP approximation then becomes

$$\chi^{[n]} = G \odot (QV\chi^{[n-1]}), \tag{40}$$

with $\chi^{[1]} = G \odot (QVP)$, which may be compared to Eq. (35).

The effective Hamiltonian of Eq. (39) treats all essential states on an equal footing and therefore does not have an ambiguous parameter E_0. Crucially, it retains the defining property of the AMP approximation: All intermediate states are in the $\mathcal{Q}$ space. We provide in Sec. IV B a realistic example for a strongly coupled rubidium atom, where the detuning, i.e., nondegeneracy, needs to be taken into account in order to accurately capture the physics.

We should note that H_{QDAMP} from Eq. (39) is non-Hermitian despite only allowing paths via the $\mathcal{Q}$ space, i.e., those of Eq. (36). For example, in second order, we have

$$\left(H^{(2)}_{\mathrm{QDAMP}}\right)_{pp'} = H_{pp'} + \sum_q \frac{V_{pq}V_{qp'}}{E_{p'} - E_q}, \tag{41}$$

$$\left(H^{(2)}_{\mathrm{QDAMP}}\right)_{p'p} = H_{p'p} + \sum_q \frac{V_{p'q}V_{qp}}{E_p - E_q}, \tag{42}$$

which means that $(H_{\mathrm{QDAMP}})_{pp'} \neq (H_{\mathrm{QDAMP}})^\dagger_{p'p}$ because $E_p \neq E_{p'}$. This non-Hermiticity expresses the most simple source of asymmetry in the coupling to the $\mathcal{Q}$ space: Since $|p\rangle$ and $|p'\rangle$ are nondegenerate, the detuning of $|p\rangle$ with any $\mathcal{Q}$-space state will be different than the detuning of $|p'\rangle$ with that same $\mathcal{Q}$-space state. Hence, the effective coupling must differ in general.

C. Validity of the effective Hamiltonian approach

In the well-explored V-type three-level system, the validity condition for adiabatic elimination is that the coupling Ω to the intermediate state should be much smaller than the detuning Δ of the intermediate state, $|\Omega/\Delta| \ll 1$ [33]. When we have many intermediate states, the natural generalization is to search for the bound $\mathcal{Q}$-space state $|q\rangle$ that maximizes the ratio $|\Omega_{pq}/\Delta_{pq}|$ for any state $|p\rangle \in \mathcal{P}$ [25].

We can formalize this condition through the reduced wave operator χ. For this, we notice that in QDRSPT, the first order $\chi^{[1]}$ is defined via its matrix elements

$$\chi^{[1]}_{qp} = \frac{V_{qp}}{E_p - E_q}, \tag{43}$$

in which $\Delta_{pq} \equiv E_p - E_q$ is the detuning of $|q\rangle$ with respect to $|p\rangle$. Hence, the formal version of the condition will be $\|\chi^{[1]}\|_{\max} \ll 1$. It should be noted that this condition applies only to the bound states. The continuum is of course always resonant, but does not cause singularities due to the imaginary part. Hence, only the projection of χ on the bound states should be considered. The condition then answers the following question: How strongly at most can a $\mathcal{P}$-space state couple to a bound $\mathcal{Q}$-space state via one photon?

Clearly, the condition can trivially be generalized to answer the question of how strongly at most a $\mathcal{P}$-space state can couple to a bound $\mathcal{Q}$-space state via up to N photons, $\|\sum_{n=1}^{N}\chi^{[n]}\|_{\max} \ll 1$, or, without specifying the order,

$$\|\chi\|_{\max} \ll 1. \tag{44}$$

Thus, the reduced wave operator, which couples the $\mathcal{P}$ space to the $\mathcal{Q}$ space, provides information on the validity and eventual breakdown of the effective Hamiltonian approach.

IV. EXAMPLES

With the following examples, we aim to illustrate three main points: (i) the physical implication of the nonorthogonal eigenstates of H_{RS}, (ii) a system in which the nondegeneracy of the $\mathcal{P}$ space makes the AMP approximation fail at already low intensities, while the quasidegenerate AMP approximation is reliable at those intensities, and (iii) a system in which the eigenstates of H_{RS} are significantly nonorthogonal. To this end, we analyze Rabi oscillations in a simple model system in Sec. IV A, investigate rubidium in an IR field in Sec. IV B, and finally present results for helium strongly coupled by the XUV field in Sec. IV C. In these examples, we focus on the quasienergy structure of the systems, while the time-dependent implications are only discussed briefly in Sec. IV A to illustrate consequences of the nonorthogonal eigenstates of H_{RS}. Time-dependent effective Hamiltonian calculations will be presented elsewhere, where we will further include envelopes and chirps in a general formalism.

A. Rabi oscillations in a model system

It is now clear that H_{eff} should be non-Hermitian in general. In this example, we will demonstrate the implications, for both the dressed energies and the time evolution, using a minimal model system. We consider three atomic levels: a GS $|a\rangle$, an excited state (ES) $|b\rangle$, and a third state $|q\rangle$. The GS with energy $E_a = 0$ is resonantly coupled to the ES with Rabi frequency $\Omega_{ab} = \Omega_{ba} = \mathcal{E}_0 z_{ab}$. The frequency of the field is hence $\omega = E_b - E_a$. The third state $|q\rangle$ couples to the ES with $\Omega_{bq} = \Omega_{qb} = \mathcal{E}_0 z_{qb}$; the latter two atomic states are furthermore degenerate, $E_q = E_b$. Thus, $|b\rangle$ couples to $|q\rangle$ nonresonantly, via both absorption and emission with equal strength. As such, the minimal relevant block of the Floquet Hamiltonian H has four levels, where $|a, N\rangle$ and $|b, N-1\rangle$ make up the $\mathcal{P}$ space and $|q, N-1\pm1\rangle$ make up the $\mathcal{Q}$ space:

$$H = \begin{pmatrix} 0 & \Omega_{ab}/2 & 0 & 0 \\ \Omega_{ab}/2 & 0 & \Omega_{qb}/2 & \Omega_{qb}/2 \\ 0 & \Omega_{qb}/2 & -\omega & 0 \\ 0 & \Omega_{qb}/2 & 0 & \omega \end{pmatrix}. \tag{45}$$

First, we note that due to the degeneracy of the $\mathcal{P}$ space, QDRSPT yields the same results as degenerate RSPT. Interestingly, since the two $\mathcal{Q}$-space states have opposite detunings $\pm\omega$, their contributions to the Stark shift of the ES cancels out. Further, the GS does not couple to the $\mathcal{Q}$ space, and the $\mathcal{Q}$-space states do not couple among themselves. This means that the effective Hamiltonian from adiabatic elimination reads, in infinite order,

$$H_{\mathrm{AMP}}^{(\infty)} = \begin{pmatrix} 0 & \Omega_{ab}/2 \\ \Omega_{ab}/2 & 0 \end{pmatrix}. \tag{46}$$

In other words, all terms beyond first order vanish. In RSPT, we further consider terms beyond the AMP approximation, which are nonzero in this model system. In third order, we have

$$H_{\mathrm{RS}}^{(3)} = \begin{pmatrix} 0 & \frac{\Omega_{ab}}{2} \\ \frac{\Omega_{ab}}{2}\left(1 - \frac{\Omega_{qb}^2}{2\omega^2}\right) & 0 \end{pmatrix}, \tag{47}$$

and in fifth order

$$H_{\mathrm{RS}}^{(5)} = \begin{pmatrix} 0 & \frac{\Omega_{ab}}{2} \\ \frac{\Omega_{ab}}{2}\left(1 - \frac{\Omega_{qb}^2}{2\omega^2} - \frac{\Omega_{qb}^2\Omega_{ab}^2}{8\omega^4} + \frac{\Omega_{qb}^4}{4\omega^4}\right) & 0 \end{pmatrix}. \tag{48}$$

As parameters, we choose $\omega = 1$ a.u., $z_{ab} = 1$ a.u., and $z_{qb} = 6$ a.u., which are typical for XUV-driven Rabi oscillations, where the ES couples to a degenerate Rydberg state $|q\rangle$ much more strongly than to the GS. Connecting our model with realistic atoms, $|q\rangle$ represents a manifold of Rydberg states. In real atoms, these are not perfectly degenerate with $|b\rangle$. This complicates the expressions significantly but does not fundamentally alter the physics.

In Fig. 1 we compare the properties and time evolution due to the H (solid lines) with those from $H_{\mathrm{AMP}}^{(\infty)}$ (dash-dotted lines), $H_{\mathrm{RS}}^{(3)}$ (dashed lines), and $H_{\mathrm{RS}}^{(5)}$ (dotted lines). In Fig. 1(a) the dressed energy E_+ of the dressed state $|+\rangle$ is compared within the models. Expectedly, $H_{\mathrm{AMP}}^{(\infty)}$ struggles at high intensities, while the $H_{\mathrm{RS}}^{(n)}$ in both third and fifth order capture the dressing accurately. Note that $E_- = -E_+$ for both H and the effective Hamiltonians.

To gauge the non-Hermiticity of $H_{\mathrm{RS}}^{(n)}$, we define an effective asymmetry of the absolute value of the off-diagonal elements,

$$\mathcal{A} := \left| \frac{|H_{\mathrm{eff}}|_{ba} - |H_{\mathrm{eff}}|_{ab}}{|H_{\mathrm{eff}}|_{ba} + |H_{\mathrm{eff}}|_{ab}} \right|, \tag{49}$$

and show the results in Fig. 1(b) for $H_{\mathrm{RS}}^{(3)}$ (dashed line) and $H_{\mathrm{RS}}^{(5)}$ (dotted line). Of course, $H_{\mathrm{AMP}}^{(\infty)}$ (dash-dotted line) is symmetric, so $\mathcal{A} = 0$. The exact effective asymmetry from H (solid line) can be obtained from the exact effective Hamiltonian H_{eff}, which we obtain using the exact dressed energies $E_\pm$ and the projected dressed states $|\psi_\pm\rangle = P|\Psi_\pm\rangle$ via the similarity transform

$$H_{\mathrm{eff}} = (|\psi_-\rangle, |\psi_+\rangle)\begin{pmatrix} E_- & 0 \\ 0 & E_+ \end{pmatrix}(|\psi_-\rangle, |\psi_+\rangle)^{-1}. \tag{50}$$

The exact $E_\pm$ and $|\Psi_\pm\rangle$ were obtained through diagonalization of H.

The time evolution due to H at $\mathcal{E}_0 = 0.1$ a.u. is shown in Fig. 1(c). It is here that the role of the $\mathcal{Q}$ space becomes

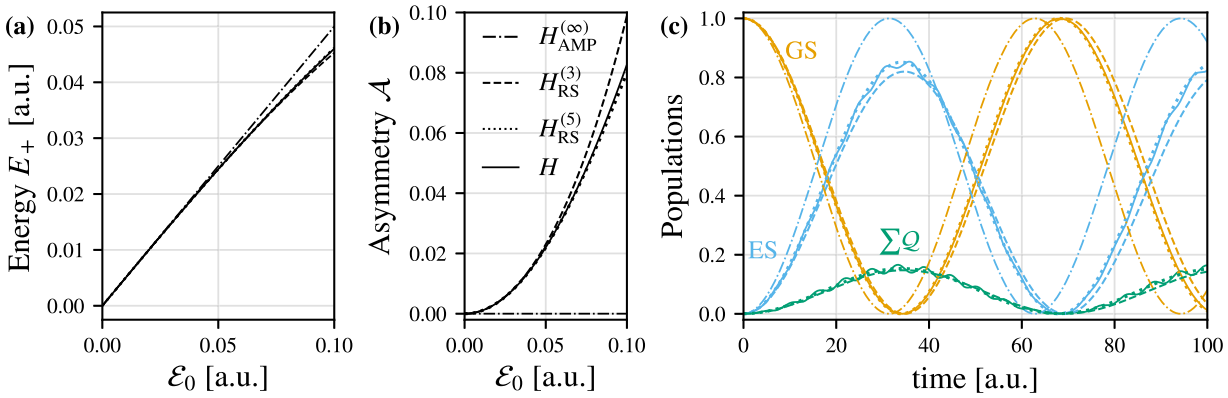

FIG. 1. Properties of the minimal model Hamiltonian H (solid lines) of Eq. (45), compared with the effective Hamiltonians $H_{\mathrm{AMP}}^{(\infty)}$ (dash-dotted lines), $H_{\mathrm{RS}}^{(3)}$ (dashed lines), and $H_{\mathrm{RS}}^{(5)}$ (dotted lines). (a) Plot of the dressed energy E_+ against field strength $\mathcal{E}_0$. (b) Plot of the effective asymmetry defined in Eq. (49) against $\mathcal{E}_0$. (c) Plot of the populations of the GS (yellow lines) and the ES (blue lines) and the sum of the $\mathcal{Q}$-space populations (green lines).

obvious. It is clearly incorrect to assume no population in the $\mathcal{Q}$ space, which is done in the AMP approximation. The norm in the entire Hilbert space must be conserved, i.e., the full wave function $|\Psi(t)\rangle \in \mathcal{P} \oplus \mathcal{Q}$ fulfills $\langle\Psi(t)|\Psi(t)\rangle = 1$. The full wave function is made up of its two projections $|\Psi(t)\rangle = P|\Psi(t)\rangle + Q|\Psi(t)\rangle$ and using the reduced wave operator, we may write $|\Psi(t)\rangle = (P + \chi)P|\Psi(t)\rangle$.

Inspecting Fig. 1(c), we observe the GS (yellow) undergoing usual resonant Rabi oscillations. Meanwhile, the coupling of the ES (blue) to $|q\rangle$ (sum of populations of $|q, N-1\pm1\rangle$ in green) leads to a transient population in $\mathcal{Q}$. In fact, for this simple model system, with the initial population in the GS, we can identify that the maximum excited-state population $\max[|b(t)|^2]$ is determined by $\max[|b(t)|^2] = |(H_{\mathrm{eff}})_{ba}/(H_{\mathrm{eff}})_{ab}|$. Clearly, $H_{\mathrm{AMP}}^{(\infty)}$ does not adequately capture the population dynamics of this system. In comparison, already $H_{\mathrm{RS}}^{(3)}$ provides decent agreement, while $H_{\mathrm{RS}}^{(5)}$ is excellent in predicting both the Rabi frequency and the magnitude of the excited state.

We note that the full system was evolved with $U(t,0) = \exp(-\mathrm{i}Ht)$, which gives rise to a cycle-averaged time evolution, in which the interference due to $|q,N\rangle$ and $|q,N-2\rangle$ is neglected [54,67]. These interferences lead to the appearance of counterrotating oscillations, where the ES exchanges population with the $\mathcal{Q}$ space on subcycle timescales. Note that this also necessitates accounting for the transient population in the counterrotating ES $|b,N-1\pm2\rangle$ [22].

Due to the large detuning of $|q,N-1\pm1\rangle$, their dressing is negligible and all of their population is transient. Transient is however not the same as negligible, which the AMP approximation implies. This population can be predicted by acting on the two-level essential-state wave function $|\psi(t)\rangle = [a(t), b(t)]^T$ with the reduced wave operator, $Q|\Psi(t)\rangle = \chi|\psi(t)\rangle$. Up to second order in χ, we obtain

$$\langle q, N-1\pm1|\chi|\psi(t)\rangle = -\frac{\Omega_{bv}\Omega_{ba}}{4\omega^2}a(t) \mp \frac{\Omega_{qb}}{2\omega}b(t). \quad (51)$$

We use this expression to obtain the $\mathcal{Q}$-space population $\sum_\pm |\langle q, N-1\pm1|\chi|\psi(t)\rangle|^2$ from the $\mathcal{P}$-space population predicted by $H_{\mathrm{RS}}^{(3)}$ and $H_{\mathrm{RS}}^{(5)}$. We find excellent agreement between the exact $\mathcal{Q}$-space population from H and the predictions from RSPT.

B. Quasienergies of rubidium in an IR field

As soon as the essential states are significantly detuned with respect to the optical transition, we should treat the nondegeneracy of the Hamiltonian properly. One system where we can demonstrate the effectiveness of the quasidegenerate approach is the rubidium atom, driven by 800 nm linearly polarized light (corresponding to 1.55 eV). This showcases the power of the effective Hamiltonian approaches in the more traditional long-wavelength regime of strong-field physics.

The field couples the $5s$ GS to the $5p$ ES via one photon, creating two dressed states $|\pm\rangle$. The $5p$ state ionizes then via two photons. Interestingly, the unperturbed $5d$ and $7s$ states are nearly resonant to a one-photon transition from $5p$, with detunings between 0.1 and 0.2 eV. As the intensity increases, the $|+\rangle$ state mixes strongly first with the dressed $5d$ state and subsequently with the dressed $7s$ state. This constitutes two avoided crossings as the intensity is increased. For this reason, an accurate description of this system requires us to calculate a four-level effective Hamiltonian, with the unperturbed basis $|5s,N\rangle$, $|5p,N-1\rangle$, $|5d,N-2\rangle$, and $|7s,N-2\rangle$.

We construct the full light-matter Hamiltonian H within Floquet theory, where we include all atom-photon states in the basis that are accessible from the essential states via an up to four-photon transition. The required complex-scaled atomic parameters are obtained by solving the TISE for a rubidium model potential [55] with exterior complex scaling [81]. Thus, we are invoking a single-active-electron approximation, in which the $5s$ valence electron is propagated in an effective potential generated by the inner-shell electrons [86–88]. The quasienergies of H are then obtained through sparse matrix diagonalization techniques [89].

We compare in Fig. 2 the exact quasienergies $E - \mathrm{i}\Gamma/2$ to those predicted by three different four-level effective Hamiltonians, obtained perturbatively. Figures 2(a)–2(c) show the real part E, while Figs. 2(d)–2(f) show the decay rate Γ on a logarithmic scale. In Figs. 2(a) and 2(d) we present seventh-order calculations in QDRSPT using Eq. (21). In Figs. 2(b) and

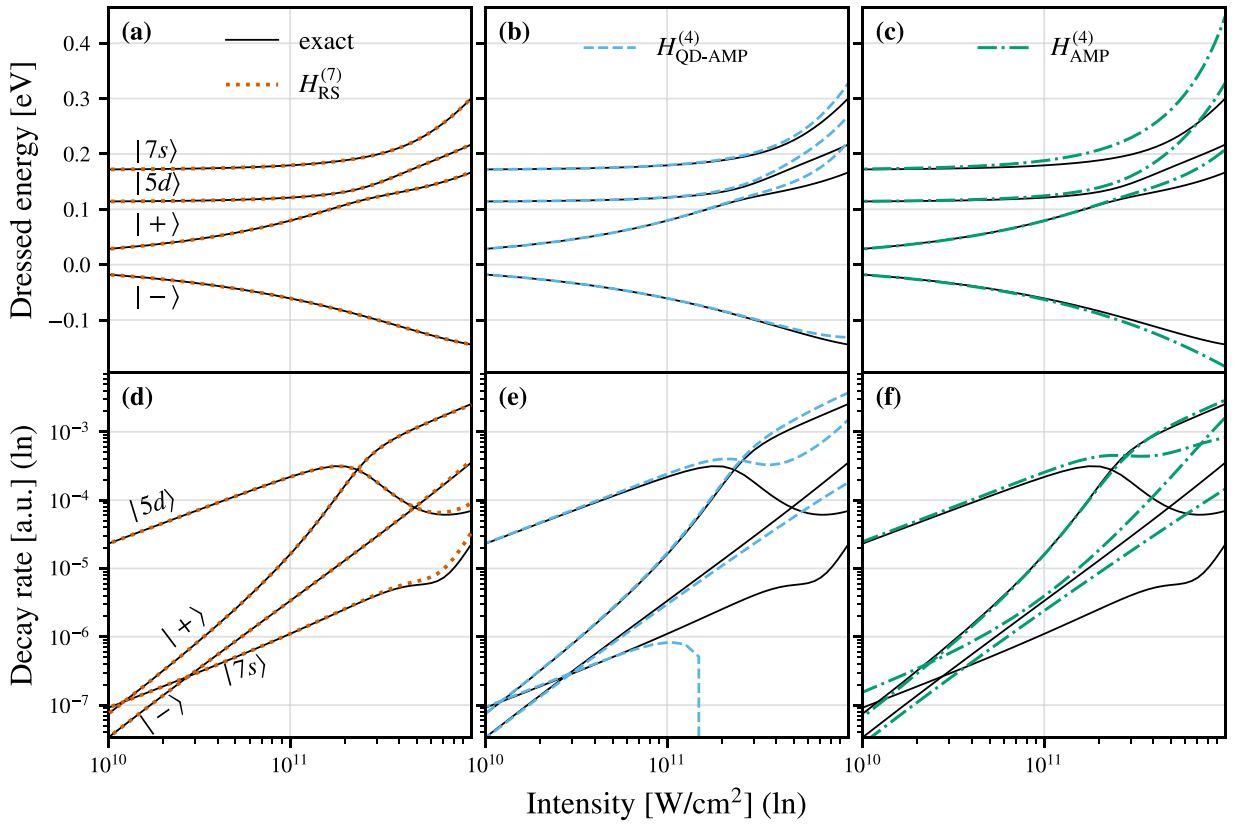

FIG. 2. Quasienergies $E - i\Gamma/2$ of rubidium in a monochromatic, linearly polarized IR field. (a)–(c) Real part of the quasienergies E and (d)–(f) decay rate Γ. The black solid lines are exact calculations obtained from Floquet theory. In (a) and (d) the exact quasienergies are compared to the effective Hamiltonian from QDRSPT in seventh order (red dotted lines) [see Eq. (21)]. In (b) and (e) we compare the exact solution to the effective Hamiltonian from QDRSPT within the pole approximation in fourth order (blue dashed lines) [see Eq. (39)]. In (c) and (f) we compare the exact solution to the effective Hamiltonian from adiabatic elimination in fourth order (green dash-dotted lines) [see Eq. (34)]. We label the dressed states as $|+\rangle$, $|-\rangle$, $|5d\rangle$, and $|7s\rangle$. The labeling of $5d$ and $7s$ refers to their field-free limit; once the $5d$ and $7s$ states are dressed, they mix strongly with other states.

2(e) the effective Hamiltonian of the quasidegenerate AMP approximation, Eq. (39), is shown in fourth order, while Figs. 2(c) and 2(f) contain results from the traditional AMP effective Hamiltonian, Eq. (34), also in fourth order. For the latter, we choose the free parameter E_0 to be the center of the unperturbed $\mathcal{P}$-space spectrum according to common practice. We choose fourth order for H_{QDAMP} and H_{AMP} since higher orders are not common in the literature [18,20,52,84,90]. Further, higher orders do not substantially improve the agreement. In contrast, seventh order in QDRSPT is required to get good agreement of the decay rates with the exact solution.

Quasidegenerate RSPT yields accurate results for all calculated intensities. Notably, even the complex ionization dynamics at high intensities is reproduced rather well. Applying our quasidegenerate AMP approximation yields very good results for both the quasienergies and decay rates at low intensities. Beyond the first avoided crossing of $|+\rangle$ with the dressed $|5d\rangle$, the accuracy quickly worsens, even predicting unphysical negative decay rates for one of the dressed states (this can be improved by including higher-order terms). The AMP approximation is unsuited for this problem, most notably for predicting the ionization dynamics; the dressed energies are however predicted reasonably well up to the first avoided crossing.

At $I_0 < 10^{11}\,\mathrm{W\,cm^{-2}}$ we can accurately model the two-level dynamics of the two dressed states $|\pm\rangle$. For $10^{11}\,\mathrm{W\,cm^{-2}} < I_0 < 10^{12}\,\mathrm{W\,cm^{-2}}$, the two avoided crossings lead to an effective four-level system. Beyond $I_0 > 10^{12}\,\mathrm{W\,cm^{-2}}$, the coupling between $\mathcal{P}$ and $\mathcal{Q}$ becomes so substantial that the effective Hamiltonian approach breaks down, and many-level dynamics will dominate, marking the onset of strong-field effects.

As argued previously, it is not feasible to write out the expansion for seventh-order QDRSPT by hand. Instead, the expansion is numerically evaluated. A natural question is if the cost at higher orders becomes prohibitive, so that a simple diagonalization of the sparse Floquet Hamiltonian may be competitive. To answer this, we have performed a simple benchmark of the calculations that produced Fig. 2. In that example, our Hilbert space was spanned by 5088 atom-photon states. For the numerical implementation of QDRSPT, the limiting factor is repeated sparse matrix-vector multiplication. We are able to compute the expansion of $H^{(7)}_{\mathrm{RS}}$ within 53 ms. Note that since we obtain $H^{(7)}_{\mathrm{RS}}$ as an expansion of the field strength, one calculation yields the effective Hamiltonian at all field strengths. Converging on the two eigenvalues $E_\pm$ at $I_0 = 10^{12}\,\mathrm{W\,cm^{-2}}$ in Fig. 2 using the implicitly restarted Arnoldi method, implemented in

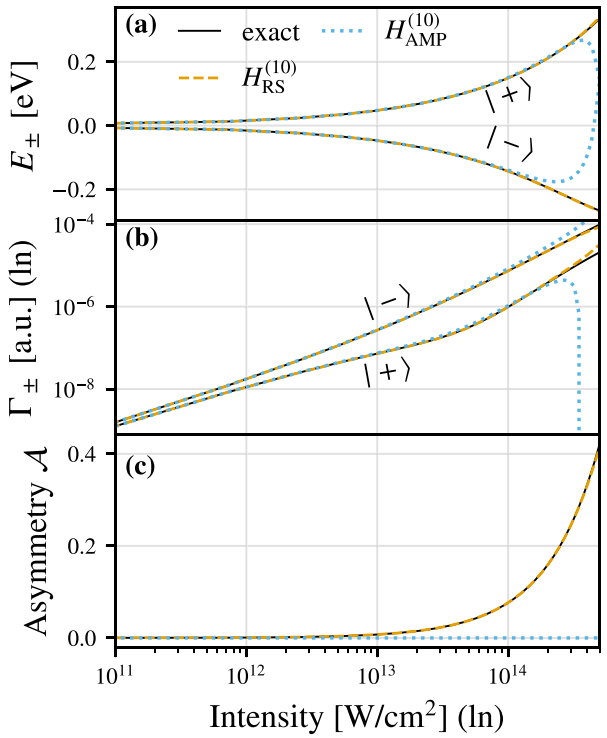

FIG. 3. Quasienergies $E_\pm - \mathrm{i}\Gamma_\pm/2$ of helium in a monochromatic, linearly polarized XUV field, resonant with the $1s^2 \leftrightarrow 1s3p$ transition. (a) Real part of the quasienergies $E_\pm$ (b) decay rate $\Gamma_\pm$. The black solid lines are exact calculations obtained from Floquet theory. We compare the exact quasienergies to the effective Hamiltonian from RSPT in tenth order (yellow dashed lines) [see Eq. (21)] and to the AMP effective Hamiltonian in tenth order (blue dotted lines) [see Eq. (34)] (since the field is exactly degenerate, $H_{\mathrm{AMP}} = H_{\mathrm{QDAMP}}$). We label the dressed states as $|+\rangle$ and $|-\rangle$. (c) Asymmetry of the Hamiltonian, where the asymmetry is defined in Eq. (49). The exact asymmetry was obtained by calculating the effective Hamiltonian from diagonalization of the Floquet Hamiltonian [see Eq. (50)].

scipy via the method `scipy.sparse.linalg.eigs` [89], takes 4 s in comparison. These numbers were obtained on a laptop processor (11th Gen Intel® Core™ i7-1185G7 @ 3.00 8 × 8), evaluated with `%timeit` in a jupyter notebook running python, and averaged over seven runs each (see Ref. [91] for details).

C. Quasienergies of helium in an XUV field

In the XUV and x-ray regime, the detuning of intermediate states is usually very large, thus allowing for large intensities upwards of 10^{14} W cm^{-2}. At these large intensities, nonlinear effects, such as nonresonant two-photon ionization from the ground state or counterrotating transitions, strongly shape the dynamics. Here QDRSPT provides clear instructions on how to treat these effects systematically, order by order.

As an example system, we study the $(1+1)$-resonance-enhanced multiphoton ionization process in helium for the resonant $1s^2 \leftrightarrow 1s3p$ transition in Fig. 3. Therefore, QDRSPT is equivalent to degenerate RSPT. We benchmark the RSPT calculations against exact solutions, obtained via Floquet theory. The complex-scaled atomic parameters (energies and dipole moments) are obtained by diagonalizing the Hamiltonian from the configuration-interaction singles method [56,57]. We construct the full light-matter Floquet Hamiltonian H within Floquet theory, where we include all atom-photon states in the basis that are accessible from the essential states via an up to five-photon transition. Note that we do not account for doubly excited states. Although we expect this to be a good approximation for these parameters, it may be a worthwhile future endeavor to study transitions to doubly excited states. The effective Hamiltonian of RSPT is then obtained in tenth order. We obtain a 2×2 complex nonsymmetric effective Hamiltonian of the form

$$H_{\mathrm{RS}} = \begin{pmatrix} S_a - \frac{\mathrm{i}}{2}\gamma_a & (\Omega_{ab} + \mathrm{i}\beta_{ab})/2 \\ (\Omega_{ba} + \mathrm{i}\beta_{ba})/2 & \delta + S_b - \frac{\mathrm{i}}{2}\gamma_b \end{pmatrix}, \tag{52}$$

where $S_{a,b}$ are the Stark shifts, δ is the detuning, $\gamma_{a,b}$ are the ionization rates, and $\Omega_{ab,ba}$ and $\beta_{ab,ba}$ are the real and imaginary parts of the effective Rabi frequency, respectively. Diagonalization of H_{eff} yields the quasienergies $E_\pm - \mathrm{i}\Gamma_\pm/2$ of the two dressed states $|\pm\rangle$.

In Figs. 3(a) and 3(b) we show $E_\pm$ and $\Gamma_\pm$, respectively. We see that at intensities beyond 2×10^{14} W cm^{-2}, the tenth-order AMP effective Hamiltonian, H_{AMP}, ceases to match the exact results, while H_{RS} agrees excellently with the Floquet calculations. In Figs. 3(c) we plot the effective asymmetry of the effective Hamiltonian, defined by Eq. (49). We further obtain the exact effective Hamiltonian nonperturbatively from Floquet theory through sparse matrix diagonalization techniques. By applying Eq. (50), we obtain the exact effective Hamiltonian via similarity transform.

It is clear from Fig. 3(c) that already beyond 10^{13} W cm^{-2}, the nonorthogonality of the $\mathcal{P}$ space becomes non-negligible, and H_{RS} needs to be employed to account for it, even if the eigenenergies of H_{AMP} stay accurate for another order of magnitude of the intensity. The source and physical implication of the effective asymmetry were discussed earlier this section; see especially Fig. 1(c). In our helium $1s^2 \leftrightarrow 1s3p$ system at high intensities, the $1s3p$ state couples strongly to nearby s and d Rydberg states in $\mathcal{Q}$, while the ground state couples only negligibly to $\mathcal{Q}$. This asymmetric coupling is encoded in the effective asymmetry (49) and is the origin for nonreciprocal counterrotating oscillations (see Ref. [22]). Meanwhile, H_{AMP} is complex symmetric by design; therefore its asymmetry is zero for all intensities.

We have additionally performed the same execution speed analysis as for the rubidium system discussed in the previous section. For the helium system, the Hilbert space is spanned by 9786 states, which is approximately double the size of the Floquet Hamiltonian we used to describe rubidium. We are able to compute the expansion of $H_{\mathrm{RS}}^{(10)}$ within 148 ms, while direct diagonalization of the sparse Floquet Hamiltonian takes 17 s in comparison [91] Our analysis implies that as the complexity of the system increases and more states have to be included in $\mathcal{Q}$, it becomes more advantageous to apply QDRSPT instead of diagonalizing the sparse Floquet Hamiltonian.

V. CONCLUSION

Coherent processes in light-matter interaction are routinely described via essential-state approaches, where adiabatic elimination, the Markov approximation, and the pole approximation are the standard approaches used to obtain an effective Hamiltonian H_{AMP} acting in a Hilbert subspace $\mathcal{P}$. However, at high intensities, H_{AMP} fails to describe the physics, both quantitatively and qualitatively. One weak point is that traditional methods such as adiabatic elimination, the pole approximation, and the Markov approximation assume implicitly that the incoming field is resonant, i.e., that the unperturbed states in $\mathcal{P}$ are degenerate. This makes the methods sensitive to an arbitrary energy shift. Most importantly, the adiabatic elimination of nonessential states forces all states in $\mathcal{P}$ to couple with the same strength to the nonessential states in $\mathcal{Q}$. We have demonstrated that this assumption is untenable. Against this backdrop, QDRSPT emerges as a versatile and accurate tool that properly accounts for the detunings of essential states and incorporates the asymmetric coupling from $\mathcal{P}$ to $\mathcal{Q}$ by design. Mathematically, this is encapsulated by the nonorthogonal eigenvectors of the effective Hamiltonian H_{RS}. Through QDRSPT, we have shown that we can describe intricate quasienergy structures in intense fields, both in the traditional low-frequency regime and in the high-frequency domain that was recently made accessible through seeded free-electron lasers.

Even at moderate intensities, where the corrections beyond the AMP approximation are not required, it can still be crucial to incorporate the nondegeneracy in $\mathcal{P}$. To this end, we have introduced the quasidegenerate AMP approximation, in which all essential states are treated on an equal footing. We underpinned this through simulations in rubidium with an IR field, in which the quasidegenerate AMP approximation clearly outperformed the traditional AMP approximation, at no added complexity in the perturbative series.

For higher intensities, the coupling between $\mathcal{P}$ and $\mathcal{Q}$ becomes so strong that QDRSPT becomes indispensable. Such coupling was explored in a model system, where an excited state coupled to a degenerate Rydberg state. This model connects QDRSPT to the recent predictions of giant nonreciprocal counterrotating oscillations [22]. The predictive power of QDRSPT comes at a price: Already at low orders ($n \geqslant 4$), the number of terms in the perturbative expansion becomes so large that purely analytical endeavors with pen and paper are unproductive. Thus, QDRSPT does not make small-scale essential-state approaches obsolete. Instead, QDRSPT is useful especially in an intermediate regime, where the physical processes of interest are still due to only a handful of states, but the intensity is so high that more sophisticated approaches than the (quasidegenerate) AMP approximation are required.

It is important to note that the calculation of H_{RS} from the full Hamiltonian is computationally very cheap, being up to 200 times faster than a brute diagonalization of the Floquet Hamiltonian in our tests. Furthermore, by yielding the effective Hamiltonian for arbitrary field strengths from a single calculation, QDRSPT allows us to scan large parameter spaces of frequency and field strength. To provide an outlook, this allows us to extend the effective Hamiltonian formalism to time-dependent envelopes and chirps. Results in that direction were recently presented by us in Ref. [92] and will be elaborated upon elsewhere. By obtaining the essential-state dynamics, we can efficiently calculate experimentally relevant observables such as photoelectron spectra for systems that would otherwise require the costly propagation of the full TDSE or diagonalization of large matrices in Floquet theory [93]. Other observables that can and should be modeled include the absorption and stimulated emission, for which we expect that the theory of effective operators [94] will prove helpful. More recently, the field of intense quantum optics was established [72]. In this field, new theoretical tools are being developed, in which the single-atom response to quantum light is modeled through appropriate averaging over semiclassical calculations [95–97]. If resonant atomic transitions are targeted with intense quantum light, we expect QDRSPT to be of great use in this area to make quantitative studies numerically feasible.

ACKNOWLEDGMENTS

We acknowledge Edvin Olofsson for insightful discussions. J.M.D. acknowledges support from the Knut and Alice Wallenberg Foundation through Grant No. 2024.0212 and the Swedish Research Council through Grant No. 2024-04247.

DATA AVAILABILITY

The code that generates the data and figures presented in this work is openly available [91].

APPENDIX: EFFECTIVE HAMILTONIANS FROM ADIABATIC ELIMINATION, THE MARKOV APPROXIMATION, AND THE POLE APPROXIMATION

1. Energy domain: The pole approximation

The pole approximation can be derived in several ways. We choose a conceptually simple path, starting with the TISE $H|\Psi\rangle = E|\Psi\rangle$. We obtain a $\mathcal{P}$-space and a $\mathcal{Q}$-space TDSE by inserting $P + Q = 1$ before $|\Psi\rangle$ and then projecting from the left with either P or Q:

$$PHP|\Psi\rangle + PHQ|\Psi\rangle = EP|\Psi\rangle, \tag{A1}$$

$$QHQ|\Psi\rangle + QHP|\Psi\rangle = EQ|\Psi\rangle. \tag{A2}$$

Due to the properties of the projectors P and Q, we have $PHQ = PVQ$ and $QHP = QVP$. The $\mathcal{P}$-space TDSE (A1) still depends on $Q|\Psi\rangle$. By rearranging Eq. (A2), we can express $Q|\Psi\rangle$ as a function of $P|\Psi\rangle$, which we insert into Eq. (A1) in order to obtain a TISE for the $\mathcal{P}$-space dynamics,

$$\left(PHP + PV\frac{Q}{E - QHQ}VP\right)|\Psi\rangle = EP|\Psi\rangle. \tag{A3}$$

Here we identify the Bloch-Horowitz Hamiltonian [98] (also called the Feshbach operator [99])

$$H_{\mathrm{BH}}(E) = PHP + PV\frac{Q}{E - QHQ}VP, \tag{A4}$$

which is an energy-dependent effective Hamiltonian that appears in Brillouin-Wigner perturbation theory [38,100]. Since $H_{\mathrm{BH}}(E)$ is energy dependent, Eq. (A3) is a nonlinear

eigenvalue problem and must be solved self-consistently one eigenenergy at a time [38]. Note that Eq. (A3) is formally equivalent to the full TISE, given that no approximations have been made thus far.

The pole approximation is arguably the most simple method to eliminate the energy dependence of $H_{\mathrm{BH}}(E)$ and has been used to great effect, for example, to explain phenomena in resonant multiphoton ionization [18,90,101] or to predict the bound-state dynamics in a low-energy subspace [102]. It can be regarded as the implementation of adiabatic elimination in the energy domain [33]. The energy dependence of $H_{\mathrm{BH}}(E)$ is weak if the $\mathcal{P}$-space eigenvalues are close to another and well separated from the $\mathcal{Q}$-space spectrum. In this case, the pole approximation consists of evaluating $H_{\mathrm{BH}}(E)$ at a fixed energy E_0 that lies approximately in the center of the $\mathcal{P}$-space spectrum [27]:

$$H_{\mathrm{AMP}} = H_{\mathrm{BH}}(E_0) = PHP + PV\frac{Q}{E_0 - QHQ}VP. \tag{A5}$$

The optimal choice of E_0, where the eigenvalues of H_{AMP} are closest to the true eigenvalues, is impossible to determine *a priori*.

2. Time domain: Adiabatic elimination and the Markov approximation

In analogy with the time-independent case, we partition the TDSE for a time-independent Hamiltonian, $\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}|\Psi(t)\rangle = H|\Psi(t)\rangle$, into

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}P|\Psi(t)\rangle = PHP|\Psi(t)\rangle + PVQ|\Psi(t)\rangle, \tag{A6}$$

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}Q|\Psi(t)\rangle = QVP|\Psi(t)\rangle + QHQ|\Psi(t)\rangle. \tag{A7}$$

In the Markov approximation, we first formally solve Eq. (A7),

$$Q|\Psi(t)\rangle = -\mathrm{i}\int_0^t \mathrm{d}t' \mathrm{e}^{-\mathrm{i}QHQ(t-t')}QVP|\Psi(t')\rangle, \tag{A8}$$

in order to obtain the $\mathcal{P}$-space TDSE

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}P|\Psi(t)\rangle = PHP|\Psi(t)\rangle - \mathrm{i}PVQ\int_0^t \mathrm{d}t' \mathrm{e}^{-\mathrm{i}QHQ(t-t')}QVP|\Psi(t')\rangle. \tag{A9}$$

We now apply the Markov approximation $P|\Psi(t')\rangle \approx P|\Psi(t)\rangle$. This amounts to neglecting the "history" of $P|\Psi(t')\rangle$ before t. The integral becomes solvable, yielding

$$\int_0^t \mathrm{d}t' \mathrm{e}^{-\mathrm{i}QHQ(t-t')} = \frac{1 - \mathrm{e}^{-\mathrm{i}QHQt}}{\mathrm{i}QHQ} \approx -\frac{\mathrm{i}}{QHQ}, \tag{A10}$$

where we coarse grained $\mathrm{e}^{-\mathrm{i}QHQt} \approx 0$. The physical reasoning here is that the evolution due to the $\mathcal{Q}$-space states is highly nonresonant, therefore only giving rise to rapid oscillations which average out. Coarse graining is of course essential to arrive at a true effective Hamiltonian, which drives the dynamics only due to its own eigenvalues, the dressed energies.

Inserting in Eq. (A6) yields the usual effective Hamiltonian from adiabatic elimination [32],

$$H_{\mathrm{AMP}} = PHP - PV\frac{Q}{QHQ}VP. \tag{A11}$$

Note that H_{AMP} is also obtained swiftly when setting the driving force zero, $\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}Q|\Psi(t)\rangle \approx 0$. This canonical approach to adiabatic elimination amounts to only considering the dc response [103]. Of course, if we shift the original Hamiltonian by $H \to H - E_0$, this will not affect the dynamics of the full system, since it corresponds to a time-dependent unitary transformation. In adiabatic elimination however, it will shift the $\mathcal{Q}$-space resolvent, leading to

$$H_{\mathrm{AMP}} = P(H - E_0)P + PV\frac{Q}{E_0 - QHQ}VP, \tag{A12}$$

which generates a different dynamics than Eq. (A11). Of course, the energy shift $PHP \to P(H - E_0)P$ is still inconsequential, so Eqs. (A5) and (A12) give rise to the same physics.

We note finally that Paulisch *et al.* have proposed a higher-order Markov approximation leading to an effective Hamiltonian beyond the AMP approximation [32]. Their higher-order effective Hamiltonian is notably nonperturbative, making it less suitable for the description of large atomic systems with thousands of basis states. We have previously shown how the higher-order Markov approximation is equivalent to a higher-order pole approximation in first order [22]. Connecting it to QDRSPT, the effective Hamiltonian from the higher-order Markov approximation is a partial resummation of specific terms of the Rayleigh-Schrödinger perturbative expansion. By constructing an inductive proof, we have managed to show that the infinite-order Markov approximation is equivalent to the Krenciglowa-Kuo iterative procedure from nuclear structure theory [104,105]; it is however outside the scope of this work. If the procedure converges, the exact effective Hamiltonian H_{eff} is obtained.

[1] M. Wollenhaupt, A. Assion, O. Bazhan, C. Horn, D. Liese, C. Sarpe-Tudoran, M. Winter, and T. Baumert, Control of interferences in an Autler-Townes doublet: Symmetry of control parameters, Phys. Rev. A **68**, 015401 (2003).

[2] T. Bayer, D. Gräfing, S. Kerbstadt, D. Pengel, K. Eickhoff, L. Englert, and M. Wollenhaupt, Time-resolved 3D imaging of ultrafast spin–orbit wave packet dynamics, New J. Phys. **21**, 033001 (2019).

[3] W. Li, J. Zhang, Y. Jin, D. Zhang, D. Ding, and K. Ueda, Partial-wave resolved spin-orbit dynamics, Phys. Rev. Lett. **135**, 143201 (2025).

[4] L. Zhang, S. Carlström, O. Smirnova, M. Ivanov, and D. Ye, Spin polarization in strong-field ionization as sensor of trapped electron orbits, Phys. Rev. Lett. **135**, 193201 (2025).

[5] M. Anand, S. Pabst, O. Kwon, and D. E. Kim, Attosecond counter-rotating-wave effect in xenon driven by strong fields, Phys. Rev. A **95**, 053420 (2017).

[6] M. Bertolino, S. Carlström, J. Peschel, F. Zapata, E. Lindroth, and J. M. Dahlström, Thomas-Reiche-Kuhn correction for truncated configuration-interaction spaces: Case of laser-assisted dynamical interference, Phys. Rev. A **106**, 043108 (2022).

[7] F. Vismarra *et al.*, Dynamic interference of chirped photoelectrons, Phys. Rev. Lett. **135**, 033202 (2025).

[8] M. Li, M.-F. Xie, H. Wang, L. Jia, J. Li, W. Wang, J. Cai, X. Hong, X. Shi, Y. Lv, X. Zhao, S. Luo, W.-C. Jiang, L.-Y. Peng, and D. Ding, Observation of laser-assisted dynamic interference by attosecond controlled photoelectron spectroscopy, Phys. Rev. Lett. **133**, 253201 (2024).

[9] A. Tóth and A. Csehi, Probing strong-field two-photon transitions through dynamic interference, J. Phys. B **54**, 035005 (2021).

[10] M. Ferray, A. L'Huillier, X. F. Li, L. A. Lompre, G. Mainfray, and C. Manus, Multiple-harmonic conversion of 1064 nm radiation in rare gases, J. Phys. B **21**, L31 (1988).

[11] F. Krausz and M. Ivanov, Attosecond physics, Rev. Mod. Phys. **81**, 163 (2009).

[12] P. Emma *et al.*, First lasing and operation of an ångstrom-wavelength free-electron laser, Nat. Photon. **4**, 641 (2010).

[13] E. Allaria *et al.*, Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet, Nat. Photon. **6**, 699 (2012).

[14] N. S. Mirian *et al.*, Generation and measurement of intense few-femtosecond superradiant extreme-ultraviolet free-electron laser pulses, Nat. Photon. **15**, 523 (2021).

[15] S. Nandi *et al.*, Observation of Rabi dynamics with a short-wavelength free-electron laser, Nature (London) **608**, 488 (2022).

[16] S. Nandi *et al.*, Generation of entanglement using a short-wavelength seeded free-electron laser, Sci. Adv. **10**, eado0668 (2024).

[17] F. Richter *et al.*, Strong-field quantum control in the extreme ultraviolet domain using pulse shaping, Nature (London) **636**, 337 (2024).

[18] E. Olofsson and J. M. Dahlström, Photoelectron signature of dressed-atom stabilization in an intense XUV field, Phys. Rev. Res. **5**, 043017 (2023).

[19] E. Olofsson, E. L. Fulton, R. Tahouri, M. Bertolino, J. M. N. Djiokap, and J. M. Dahlström, Coherent control of ionization via stabilization by resonant pulse pairs, Phys. Rev. Res. **8**, 013199 (2026).

[20] M. Dörr, O. Latinne, and C. J. Joachain, Time evolution of two-photon population transfer between the 1*s* and 2*s* states of a hydrogen atom, Phys. Rev. A **55**, 3697 (1997).

[21] L. P. Yatsenko, B. W. Shore, T. Halfmann, K. Bergmann, and A. Vardi, Source of metastable H(2*s*) atoms using the Stark chirped rapid-adiabatic-passage technique, Phys. Rev. A **60**, R4237 (1999).

[22] J. N. Bruhnke, E. Olofsson, A. Stenquist, and J. M. Dahlström, Giant counter-rotating oscillations on the attosecond timescale, Phys. Rev. Res. **7**, L042019 (2025).

[23] S. Kumar, J. D. Piel, C. H. Greene, and N. Shivaram, Broadband femtosecond lasers enable efficient two-photon excitation of the ultranarrow linewidth singlet 1*s*2*s* state in helium, Phys. Rev. Res. **8**, 013009 (2026).

[24] T. M. Linker *et al.*, Attosecond inner-shell lasing at ångström wavelengths, Nature (London) **642**, 934 (2025).

[25] F. H. M. Faisal, *Theory of Multiphoton Processes* (Springer, New York, 1987).

[26] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley-VCH, New York, 1990).

[27] C. Cohen-Tannoudji, G. Grynberg, and J. Dupont-Roc, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley-VCH, New York, 1998).

[28] L. Huang, J. Luneau, J. Schirk, F. Wallner, C. M. F. Schneider, S. Filipp, K. Liegener, and P. Rabl, Theory of multiphoton processes for applications in quantum control Phys. Rev. A **113**, 032620 (2026).

[29] N. V. Vitanov and S. Stenholm, Population transfer via a decaying state, Phys. Rev. A **56**, 1463 (1997).

[30] A. Kinos, L. Rippe, S. Kröll, and A. Walther, Designing gate operations for single-ion quantum computing in rare-earth-ion-doped crystals, Phys. Rev. A **104**, 052624 (2021).

[31] J. Kumlin, C. Braun, C. Tresp, N. Stiesdal, S. Hofferberth, and A. Paris-Mandoki, Quantum optics with Rydberg superatoms, J. Phys. Commun. **7**, 052001 (2023).

[32] V. Paulisch, H. Rui, H. K. Ng, and B.-G. Englert, Beyond adiabatic elimination: A hierarchy of approximations for multi-photon processes, Eur. Phys. J. Plus **129**, 12 (2014).

[33] E. Brion, L. H. Pedersen, and K. Mølmer, Adiabatic elimination in a lambda system, J. Phys. A: Math. Theor. **40**, 1033 (2007).

[34] H. C. Baker, Non-Hermitian quantum theory of multiphoton ionization, Phys. Rev. A **30**, 773 (1984).

[35] B. T. Torosov and N. V. Vitanov, Adiabatic elimination of a nearly resonant quantum state, J. Phys. B **45**, 135502 (2012).

[36] M. Sanz, E. Solano, and Í. L. Egusquiza, in *Applications + Practical Conceptualization + Mathematics = Fruitful Innovation*, edited by R. S. Anderssen, P. Broadbridge, Y. Fukumoto, K. Kajiwara, T. Takagi, E. Verbitskiy, and M. Wakayama, Mathematics for Industry Vol. 11 (Springer, Tokyo, 2016), pp. 127–142.

[37] K. N. Zlatanov, G. S. Vasilev, and N. V. Vitanov, Morris-Shore transformation for nondegenerate systems, Phys. Rev. A **102**, 063113 (2020).

[38] I. Lindgren and J. Morrison, *Atomic Many-Body Theory*, edited by V. I. Goldanskii, R. Gomer, F. P. Schäfer, and J. P. Toennies, Springer Series in Chemical Physics Vol. 13 (Springer, Berlin, 1982).

[39] D. F. James and J. Jerke, Effective Hamiltonian theory and its applications in quantum information, Can. J. Phys. **85**, 625 (2007).

[40] O. Gamel and D. F. V. James, Time-averaged quantum dynamics and the validity of the effective Hamiltonian model, Phys. Rev. A **82**, 052106 (2010).

[41] N. Macrì, L. Giannelli, E. Paladino, and G. Falci, Coarse-grained effective Hamiltonian via the Magnus expansion for a three-level system, Entropy **25**, 234 (2023).

[42] K. D. Barajas and W. C. Campbell, Quantum gate dynamics beyond the rotating wave approximation using multitimescale quantum averaging theory, Phys. Rev. A **113**, L030403 (2026).

[43] K. D. Barajas and W. C. Campbell, Quantum averaging theory for multi-timescale driven quantum systems, Phys. Rev. A **113**, 032210 (2026).

[44] J. R. Schrieffer and P. A. Wolff, Relation between the Anderson and Kondo Hamiltonians, Phys. Rev. **149**, 491 (1966).

[45] I. Shavitt and L. T. Redmon, Quasidegenerate perturbation theories. A canonical van Vleck formalism and its relationship to other approaches, J. Chem. Phys. **73**, 5711 (1980).
[46] L. S. Cederbaum, J. Schirmer, and H.-D. Meyer, Block diagonalisation of Hermitian matrices, J. Phys. A: Math. Gen. **22**, 2427 (1989).
[47] A. B. Klimov and L. L. Sanchez-Soto, Method of small rotations and effective Hamiltonians in nonlinear quantum optics, Phys. Rev. A **61**, 063802 (2000).
[48] A. B. Klimov, L. L. Sánchez-Soto, A. Navarro, and E. C. Yustas, Effective Hamiltonians in quantum optics: A systematic approach, J. Mod. Opt. **49**, 2211 (2002).
[49] J. R. Morris and B. W. Shore, Reduction of degenerate two-level excitation to independent two-state systems, Phys. Rev. A **27**, 906 (1983).
[50] L. G. Hanson, J. Zhang, and P. Lambropoulos, Manifestations of atomic and core resonances in photoelectron energy spectra, Phys. Rev. A **55**, 2232 (1997).
[51] L. P. Yatsenko, V. I. Romanenko, B. W. Shore, T. Halfmann, and K. Bergmann, Two-photon excitation of the metastable 2*s* state of hydrogen assisted by laser-induced chirped Stark shifts and continuum structure, Phys. Rev. A **71**, 033418 (2005).
[52] X. Zhang, Y. Zhou, Y. Liao, Y. Chen, J. Liang, Q. Ke, M. Li, A. Csehi, and P. Lu, Effect of nonresonant states in near-resonant two-photon ionization of hydrogen, Phys. Rev. A **106**, 063114 (2022).
[53] A. Tóth, S. Borbély, Y. Zhou, and A. Csehi, Role of dynamic Stark shifts in strong-field excitation and subsequent ionization, Phys. Rev. A **107**, 053101 (2023).
[54] S.-I. Chu, in *Advances in Atomic and Molecular Physics*, edited by D. R. Bates and B. Bederson (Academic, New York, 1985), Vol. 21, pp. 197–253.
[55] W. Schweizer, P. Faßbinder, and R. González-Férez, Model potentials for alkali metal atoms and Li-like ions, At. Data Nucl. Data Tables **72**, 33 (1999).
[56] J. B. Foresman, M. Head-Gordon, J. A. Pople, and M. J. Frisch, Toward a systematic molecular orbital theory for excited states, J. Phys. Chem. **96**, 135 (1992).
[57] A. Dreuw and M. Head-Gordon, Single-reference *ab initio* methods for the calculation of excited states of large molecules, Chem. Rev. **105**, 4009 (2005).
[58] I. Lindgren, The Rayleigh-Schrodinger perturbation and the linked-diagram theorem for a multi-configurational model space, J. Phys. B **7**, 2441 (1974).
[59] P. Durand, Direct determination of effective Hamiltonians by wave-operator methods. I. General formalism, Phys. Rev. A **28**, 3184 (1983).
[60] C. Brouder, G. H. Duchamp, F. Patras, and G. Z. Tóth, The Rayleigh-Schrödinger perturbation series of quasi-degenerate systems, Int. J. Quantum Chem. **112**, 2256 (2012).
[61] K. Suzuki and R. Okamoto, Degenerate perturbation theory in quantum mechanics, Prog. Theor. Phys. **70**, 439 (1983).
[62] B. H. Brandow, Formal theory of effective π-electron Hamiltonians, Int. J. Quantum Chem. **15**, 207 (1979).
[63] J. P. Killingbeck and G. Jolicard, The Bloch wave operator: Generalizations and applications: Part I. The time-independent case, J. Phys. A: Math. Gen. **36**, R105 (2003).
[64] C. M. Bender and T. T. Wu, Anharmonic oscillator. II. A study of perturbation theory in large order, Phys. Rev. D **7**, 1620 (1973).
[65] E. R. Vrscay and J. Cizek, Continued fractions and Rayleigh–Schrödinger perturbation theory at large order, J. Math. Phys. **27**, 185 (1986).
[66] U. D. Jentschura, Resummation of the divergent perturbation series for a hydrogen atom in an electric field, Phys. Rev. A **64**, 013403 (2001).
[67] J. H. Shirley, Solution of the Schrödinger equation with a Hamiltonian periodic in time, Phys. Rev. **138**, B979 (1965).
[68] S. Guérin, F. Monti, J.-M. Dupont, and H. R. Jauslin, On the relation between cavity-dressed states, Floquet states, RWA and semiclassical models, J. Phys. A: Math. Gen. **30**, 7193 (1997).
[69] M. Lewenstein, M. F. Ciappina, E. Pisanty, J. Rivera-Dean, P. Stammer, T. Lamprou, and P. Tzallas, Generation of optical Schrödinger cat states in intense laser–matter interactions, Nat. Phys. **17**, 1104 (2021).
[70] P. Stammer, J. Rivera-Dean, A. Maxwell, T. Lamprou, A. Ordóñez, M. F. Ciappina, P. Tzallas, and M. Lewenstein, Quantum electrodynamics of intense laser-matter interactions: A tool for quantum state engineering, PRX Quantum **4**, 010201 (2023).
[71] N. Tsatrafyllis, I. K. Kominis, I. A. Gonoskov, and P. Tzallas, High-order harmonics measured by the photon statistics of the infrared driving-field exiting the atomic medium, Nat. Commun. **8**, 15170 (2017).
[72] A. Gorlach, O. Neufeld, N. Rivera, O. Cohen, and I. Kaminer, The quantum-optical nature of high harmonic generation, Nat. Commun. **11**, 4598 (2020).
[73] R. V. Gothelf, C. S. Lange, and L. B. Madsen, High-order harmonic generation in a crystal driven by quantum light, Phys. Rev. A **111**, 063105 (2025).
[74] A. Stenquist, J. N. Bruhnke, F. Zapata, and J. M. Dahlström, Entanglement transfer in a composite electron–ion–photon system, Rep. Prog. Phys. **88**, 080502 (2025).
[75] S.-I. Chu and D. A. Telnov, Beyond the Floquet theorem: Generalized Floquet formalisms and quasienergy methods for atomic and molecular multiphoton processes in intense laser fields, Phys. Rep. **390**, 1 (2004).
[76] C. J. Joachain, N. J. Kylstra, and R. M. Potvliege, *Atoms in Intense Laser Fields* (Cambridge University Press, Cambridge, 2011).
[77] S.-I. Chu, Quasienergy formalism for intense field multiphoton ionization of atoms induced by circularly polarized radiation, Chem. Phys. Lett. **54**, 367 (1978).
[78] T.-S. Ho, S.-I. Chu, and J. V. Tietz, Semiclassical many-mode Floquet theory, Chem. Phys. Lett. **96**, 464 (1983).
[79] S.-I. Chu and W. P. Reinhardt, Intense field multiphoton ionization via complex dressed states: Application to the H atom, Phys. Rev. Lett. **39**, 1195 (1977).
[80] A. Maquet, S.-I. Chu, and W. P. Reinhardt, Stark ionization in dc and ac fields: An L_2 complex-coordinate approach, Phys. Rev. A **27**, 2946 (1983).
[81] B. Simon, The definition of molecular resonance curves by the method of exterior complex scaling, Phys. Lett. **71A**, 211 (1979).
[82] R. M. Potvliege and R. Shakeshaft, Time-independent theory of multiphoton ionization of an atom by an intense field, Phys. Rev. A **38**, 4597 (1988).
[83] N. Moiseyev, *Non-Hermitian Quantum Mechanics* (Cambridge University Press, Cambridge, 2011).

[84] C. R. Holt, M. G. Raymer, and W. P. Reinhardt, Time dependences of two-, three-, and four-photon ionization of atomic hydrogen in the ground 1^2S and metastable 2^2S states, Phys. Rev. A **27**, 2971 (1983).

[85] C. Buth, R. Santra, and L. S. Cederbaum, Non-Hermitian Rayleigh-Schrödinger perturbation theory, Phys. Rev. A **69**, 032505 (2004).

[86] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Above threshold ionization beyond the high harmonic cutoff, Phys. Rev. Lett. **70**, 1599 (1993).

[87] J. B. Watson, A. Sanpera, D. G. Lappas, P. L. Knight, and K. Burnett, Nonsequential double ionization of helium, Phys. Rev. Lett. **78**, 1884 (1997).

[88] H. G. Muller and F. C. Kooiman, Bunching and focusing of tunneling wave packets in enhancement of high-order above-threshold ionization, Phys. Rev. Lett. **81**, 1207 (1998).

[89] P. Virtanen *et al.*, SciPy 1.0: Fundamental algorithms for scientific computing in Python, Nat. Methods **17**, 261 (2020).

[90] B. L. Beers and L. Armstrong, Exact solution of a realistic model for two-photon ionization, Phys. Rev. A **12**, 2447 (1975).

[91] J. N. Bruhnke, Code for the paper "Reconciliation of effective Hamiltonians for intense light-matter interaction", Dataset: Zenodo, Geneva, 2026, doi: https://zenodo.org/records/22009587.

[92] J. N. Bruhnke and J. M. Dahlström, Quantitative analysis of resonant ionization by smooth laser pulses: Connection between effective Hamiltonian theory and strong-field dressed continua, arXiv:2606.21526.

[93] M. Dörr, R. M. Potvliege, and R. Shakeshaft, Multiphoton processes in an intense laser field: III. Resonant ionization of hydrogen by subpicosecond pulses, Phys. Rev. A **41**, 558 (1990).

[94] K. Suzuki and R. Okamoto, Effective operators in time-independent approach, Prog. Theor. Phys. **93**, 905 (1995).

[95] A. Gorlach, M. E. Tzur, M. Birk, M. Krüger, N. Rivera, O. Cohen, and I. Kaminer, High-harmonic generation driven by quantum light, Nat. Phys. **19**, 1689 (2023).

[96] J. M. González-Monge and J. Feist, High-harmonic generation driven by temporal-mode quantum states of light, arXiv:2512.06602.

[97] C. S. Lange and L. B. Madsen, Hierarchy of approximations for describing quantum light from high-harmonic generation: A Fermi-Hubbard-model study, Phys. Rev. A **111**, 013113 (2025).

[98] C. Bloch and J. Horowitz, Sur la détermination des premiers états d'un système de fermions dans le cas dégénéré, Nucl. Phys. **8**, 91 (1958).

[99] H. Feshbach, A unified theory of nuclear reactions. II, Ann. Phys. (NY) **19**, 287 (1962).

[100] P.-O. Löwdin, A note on the quantum-mechanical perturbation theory, J. Chem. Phys. **19**, 1396 (1951).

[101] L. G. Hanson, J. Zhang, and P. Lambropoulos, Theory of core-resonant ionization, Europhys. Lett. **30**, 81 (1995).

[102] G. A. Voth and R. A. Marcus, Adiabatically reduced coupled equations for intramolecular dynamics calculations, J. Chem. Phys. **84**, 2254 (1986).

[103] L. Allen and C. R. Stroud, Broadening and saturation in n-photon absorption, Phys. Rep. **91**, 1 (1982).

[104] E. M. Krenciglowa and T. T. S. Kuo, Convergence of effective Hamiltonian expansion and partial summations of folded diagrams, Nucl. Phys. A **235**, 171 (1974).

[105] K. Takayanagi, Effective Hamiltonian in the extended Krenciglowa–Kuo method, Nucl. Phys. A **864**, 91 (2011).