Dressed bound states for attosecond dynamics in strong laser fields

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Abstract

We propose a theoretical approach for the interpretation of pump–probe measurements where an attosecond pulse is absorbed in the presence of an intense laser pulse. This approach is based on abstractly defined dressed bound states, which capture the essential aspects of the interaction with the laser pulse and facilitate a perturbative description of transitions induced by the attosecond pulse. Necessary properties of dressed bound states are defined and various choices are discussed and compared to accurate numerical solutions of the time-dependent Schrödinger equation. © 2012 Elsevier B.V. All rights reserved.

1. Introduction

The ionization of atoms and molecules in the field of an intense laser pulse is one of the most fundamental phenomena in the interaction of light with matter. Despite decades of intensive experimental and theoretical research, there are still open conceptual questions, highlighted by recent experiments [1,2]. A key question is whether one can define the instantaneous ionization probability in the presence of an ionizing field. A closely related question is to which extent the detachment of an electron by multiphoton absorption or quantum tunneling can be described by an ionization rate (see, for example, [3,4]). Such a description is common, but it can be made rigorous only in stationary or quasi-stationary situations. Two such cases are the static and time-periodic external electric fields. In static fields, the system decays exponentially at a well-defined rate. In time-periodic fields, Floquet theory allows defining cycle-averaged decay rates (see e.g. [5] or [6] for a recent application). In both situations, real and imaginary parts of complex energies give energy positions and decay widths. Non-Hilbert space resonance states can be associated with the complex energies, either by using complex scaling [7,8] or by iteratively adapting the boundary conditions [Siegert boundary conditions] [6]. None of these approaches, however, provides a fully satisfactory framework for investigating the interaction of few- to single-cycle ionizing pulses with matter. At the same time, the establishment of attosecond absorption spectroscopy [9,10] and the recent advances in the generation of optical waveforms [1,11] call for a deeper understanding of dynamics unfolding within a laser cycle. Such understanding implies the development of approximate analytical theories, and the aim of this article is to make a step towards creating a useful theoretical framework.

Our work is closely related to the development of adiabatic or quasistatic approximations to strong-field ionization, such as described in [12] and the papers cited therein. In contrast to that research, we make no attempt to analytically describe strong-field ionization. Instead, we search for an analytical description of resonant transitions that an attosecond pulse of ultraviolet (UV) or extreme-ultraviolet (XUV) radiation drives in the presence of a strong near-infrared field.

2. Dressed bound states

Let us consider a quantum system (atom, ion, molecule etc.) described by a Hamiltonian $\hat{H}_0$. In the absence of any external field, this system has a set of orthonormal bound stationary states that satisfy the stationary Schrödinger equation:

$$\hat{H}_0|n\rangle = \epsilon_n|n\rangle.$$  \hspace{1cm} (1)

A typical attosecond measurement consists in letting the system interact with the electric fields of an intense laser pulse $E(t)$, which may be regarded as a pump pulse, and a relatively weak probe pulse (or a train of pulses) $E_{\text{probe}}(t)$. An XUV probe pulse may be as short as a few tens of attoseconds [13,14]. Varying the delay between the two pulses with an attosecond accuracy and observing the outcomes of their interaction with a quantum system provides valuable insights into phenomena triggered and steered by these two pulses [15]. If a significant fraction of atoms or molecules is ionized within a single half-cycle of the laser pulse, the interaction with the pulse is highly non-perturbative, so that an accurate description of this interaction calls for a numerical solution of the time-dependent Schrödinger equation (TDSE). We assume that such a solution

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|ψ_u(t)⟩ is available, and one of our goals is to interpret strong-field dynamics in terms of some dressed states. These states, if introduced properly, may serve as a convenient starting point for building approximate models describing the interaction with the probe pulse. Before we discuss different approaches to defining such dressed states, let us list a few requirements that they should fulfill.

Let |ψ_{0,n}(t)⟩ be a bound dressed state associated with a stationary state |n⟩. This means that ⟨ψ_{0,n}(t_0)| = |n⟩ at some moment t_0 before the interaction with external fields. As a first requirement, we demand that dressed states should form an orthonormal set of wave vectors at all times:

\[ \langle ψ_{0,n}(t)|ψ_{0,m}(t)⟩ = δ_{mn}. \] (2)

Orthonormality is not only convenient for computations, but it is also needed for the physical interpretation of the states: only for orthonormal states does finding a system in state |ψ_{0,n}(t)⟩ with probability \( |⟨ψ_{0,n}(t)|ψ_{0,n}(t)⟩|^2 = 1 \) imply that all other probabilities are 0.

A strong laser field can significantly ionize a quantum system. As a free electron cannot absorb a photon, the probe pulse predominantly interacts with bound electrons. Therefore, it would be desirable to have dressed states that serve as a basis for describing the bound part of an electron wavefunction even in the presence of a strong external electric field. When this property is satisfied, strong-field ionization should deplete bound dressed states without changing their shape in a fashion similar to the decay of quasi-stationary states.

Except for losses due to ionization, each dressed state should fully account for distortions caused by the laser field. Therefore, we assume that the pump pulse induces no transitions between dressed states. This is the most important requirement that we place on dressed states, and it means that \( |ψ_{0,n}(t_2)|U(t_2, t_1)|ψ_{0,n}(t_1)⟩ \) must be negligibly small if \( m ≠ n \), where \( U(t_2, t_1) \) is the propagator describing the interaction with the laser pulse (the exact definition of the propagator is given below). In particular, we expect that the interaction with the laser pulse brings the quantum system from its initial state |n⟩ to the corresponding dressed state |ψ_{0,n}(t)⟩ multiplied with a probability amplitude \( a_{0,n}(t) \), the modulus of which accounts for the depletion by the laser pulse:

\[ a_{0,n}(t) = \left| \langle ψ_{0,n}(t)|U(t, t_0)|n⟩ \right|. \] (3)

while

\[ \langle ψ_{0,n}(t)|U(t, t_0)|n⟩ = 0 \text{ if } m ≠ n. \] (4)

As long as the above assumptions hold, the effect of the laser field on any dressed bound state can be expressed via the probability amplitudes:

\[ \langle ψ_{0,n}(t_2)|U(t_2, t_1)|ψ_{0,n}(t_1)⟩ = \frac{a_{0,n}(t_2)}{a_{0,n}(t_1)} \] (5)

and the time evolution of the system can be approximated as

\[ U(t_2, t_1) ≈ \sum_n \frac{a_{0,n}(t_2)}{a_{0,n}(t_1)}|ψ_{0,n}(t_1)⟩. \] (6)

Note that the approximate time evolution is no longer unitary as it accounts for ionization by losses from the dressed-state space.

Assuming that time-dependent wave vectors |ψ_{0,n}(t)⟩ satisfying all the above requirements exist, let us use them to develop a general perturbative description of the interaction of a laser-dressed system with a weak high-frequency probe pulse. For simplicity, we assume that the pump (laser) and probe (UV or XUV) pulses are polarized along the z-axis. In the dipole approximation, the length-gauge Hamiltonian takes the form \( H(t) = H_0 + V_L(t) + V_{\text{probe}}(t) \) with

\[ V_L(t) = eE_L(t)\hat{Z}, \] (7)

\[ V_{\text{probe}}(t) = eE_{\text{probe}}(t)\hat{Z}, \] (8)

where \( e > 0 \) is the electron charge and \( \hat{Z} \) is the dipole operator. The interaction with the laser pulse is described by

\[ \frac{ie}{\hbar} \frac{d}{dt} |ψ_{0,n}(t)⟩ = (H_0(t) + V_L(t))|ψ_{0,n}(t)⟩ \] (9)

or, in the operator form,

\[ \frac{ie}{\hbar} \frac{d}{dt} U_{0,n}(t, t_0) = (H_0(t) + V_L(t))U_{0,n}(t, t_0) \] (10)

with \( |ψ_{0,n}(t_0)⟩ = U_{0,n}(t_0, t_0)|n⟩ \) for an initial state |n⟩. A standard approach to developing a perturbation theory with respect to the probe pulse consists in writing the complete TDSE

\[ \frac{d}{dt} |ψ(t)⟩ = \hat{H}(t)|ψ(t)⟩ \] (11)

in the following integral form:

\[ U(t, t_0) = U_{0,n}(t, t_0) - i \frac{e}{\hbar} \int_{t_0}^{t} dt' U(t, t')\hat{V}_{\text{probe}}(t')U_{0,n}(t', t_0). \] (12)

Here, \( U_{0,n}(t, t_0) \) is the propagator associated with the full Hamiltonian \( H(t) \):

\[ \frac{d}{dt} U_{0,n}(t, t_0) = \hat{H}(t)U_{0,n}(t, t_0) \] (13)

and \( |ψ(t)⟩ = U_{0,n}(t, t_0)|ψ(t_0)⟩ \) for the initial condition \( |ψ(t_0)⟩ = |n⟩ \). Furthermore, if only single-photon processes play a role in the interaction with the probe pulse, then the unknown operator \( U(t, t') \) on the right-hand side of Eq. (12) may be approximated with \( U_{0,n}(t, t') \), which we consider to be known. This yields

\[ |ψ(t)⟩ ≈ |ψ_{0,n}(t)⟩ - i \frac{e}{\hbar} \int_{t_0}^{t} dt' U_{0,n}(t, t')\hat{V}_{\text{probe}}(t')|ψ_{0,n}(t')⟩. \] (14)

The probe pulse may cause bound–bound, as well as bound–continuum transitions. In this paper, we focus on transitions between bound states, leaving the direct photionization by the probe pulse aside. In this case, Eq. (14) can be written as

\[ |ψ(t)⟩ ≈ |ψ_{0,n}(t)⟩ - \frac{i}{\hbar} \sum_{m ≠ n} \int_{t_0}^{t} dt' \left( \hat{V}_{\text{probe}}(t')|ψ_{0,n}(t')⟩\langle ψ_{0,n}(t')|ψ_{0,m}(t')⟩\right). \] (15)

Let us evaluate the probability amplitude that a system, initially prepared in state \( |ψ(t_0)⟩ = |n⟩ \), will be found in state \( |ψ_{0,m}(t)⟩ \) at a later time \( t \). Multiplying both sides of Eq. (15) with \( ⟨ψ_{0,m}(t)| \) from the left, keeping in mind the fact that \( |ψ_{0,n}(t)⟩ ≈ a_{0,n}(t)|ψ_{0,n}(t)⟩ \), and using Eq. (5), we obtain

\[ x_{0,m}(t) = \left⟨ ψ_{0,m}(t) \right| U_{0,n}(t, t_0) \left| 1 \right⟩ \] (16)

\[ \approx - \frac{i}{\hbar} \sum_{n ≠ i} \int_{t_0}^{t} dt' E_{\text{probe}}(t')\frac{a_{0,n}(t')}{a_{0,i}(t')} Z_{0,m}(t'). \]

for \( n ≠ i \), where

\[ Z_{0,m}(t) = \left| ψ_{0,m}(t) \right| Z_0(t). \] (17)

is the transition matrix element between dressed states. Eq. (16) is the desired expression, where the interaction with the probe pulse is accounted for in first-order perturbation theory, while the effect of the strong laser pulse is represented by quantities related to dressed states. Based on Eq. (16), we can evaluate quantities observable in measurements. For example, the probability that, at some final time \( t_f \) after the interaction, the system will be found in a bound stationary state \( |f⟩ \) is given by
As an example that is more relevant to transient absorption spectroscopy, the dipole response due to bound–bound transitions induced by the probe pulse can be evaluated as

$$d(t) = \langle \psi(t) | \mathcal{L} | \psi(t) \rangle \approx \sum_{m} \langle \psi(t) | Z_{mn}(t) | \psi(t) \rangle.$$  

(19)

So far, the dressed bound states were treated as an abstract mathematical concept. It is worth mentioning that without the requirement that dressed states should describe bound electrons, the wavevectors $\langle \psi(t) | \mathcal{L} | \psi(t) \rangle = \hat{U}_{1}(t, t_{0}) | \psi(t) \rangle$ would exactly satisfy the rest of our requirements, except the boundness of the wavefunction. However, such solutions do not satisfy all our requirements including the boundness of the wavefunction, exist, but we can consider several approximations to dressed bound states and use our general theory to investigate their performance in comparison with exact solutions of the TDSE. We will consider four different definitions of dressed bound states.

As a very crude approximation, one can attempt to use the unperturbed stationary states:

$$| \phi_{\text{stat}}^{(l)}(t) \rangle = | \psi(t) \rangle.$$  

(20)

Another option, frequently encountered in literature, is to use time-dependent eigenstates of the Hamiltonian $\hat{H}(t)$ evaluated in the subspace of unperturbed bound states and normalized to satisfy Eq. (2):

$$\sum_{m} | \langle \psi(t) | \mathcal{L} | \psi(t) \rangle |^{2} = \epsilon_{\text{stat}}^{(l)}.$$  

(21)

In the context of strong-field ionization, these states are known under many different names, such as adiabatic states [12,16], quasi-static states [17,18], phase-adiabatic states [19], field-adapted states [20], and adiabatic-field-following dressed states [21,22]. We will refer to them as “adiabatic states”.

Yet another reasonable approach is to obtain dressed bound states by solving the TDSE in the basis of unperturbed bound states:

$$i \hbar \frac{d}{dt} | \phi_{\text{stat}}^{(l)}(t) \rangle = \sum_{m} | \langle m | H_{0} + \hat{V}_{L}(t) | \psi_{\text{stat}}^{(l)}(t) \rangle |^{2}$$  

(22)

with the initial condition $| \phi_{\text{stat}}^{(l)}(t_{0}) \rangle = | \psi(t) \rangle$. With this choice, we assume that virtual excitations into field-free continuum states do not influence the dynamics. However, contrary to $| \phi_{\text{stat}}^{(l)}(t) \rangle$, these states do not follow the laser field adiabatically, but rather allow for non-instantaneous response to the laser field. We will call them “dynamically dressed states”.

The above three alternatives allow one to evaluate the dressed bound states without solving the TDSE in the full Hilbert space, although such solutions $| \psi(t) \rangle$ are required later to obtain the probability amplitudes $a_{l}(t)$ from Eq. (3). The last option that we are going to consider is different: we will evaluate dressed bound states by projecting solutions of the TDSE obtained in the whole Hilbert space onto the subspace of unperturbed bound stationary states and orthonormalizing them with the aid of the Gram-Schmidt procedure:

$$| \tilde{\psi}_{\text{stat}}^{(l)}(t) \rangle = \sum_{m} \langle m | U_{1}(t, t_{0}) | \psi(t) \rangle | \psi_{\text{stat}}^{(l)}(t) \rangle.$$  

(23)

$$| \tilde{\phi}_{\text{stat}}^{(l)}(t) \rangle = \frac{\langle \psi(t) | \mathcal{L} | \psi(t) \rangle - \langle \psi(t) | \mathcal{L} | \tilde{\psi}_{\text{stat}}^{(l)}(t) \rangle}{\sqrt{\langle \psi(t) | \mathcal{L} | \psi(t) \rangle \langle \psi(t) | \mathcal{L} | \psi(t) \rangle}}.$$  

(24)

where the projector $\tilde{P}_{l}(t)$ is defined as

$$\tilde{P}_{l}(t) = \langle \psi(t) | \mathcal{L} | \psi(t) \rangle - \langle \psi(t) | \mathcal{L} | \tilde{\psi}_{\text{stat}}^{(l)}(t) \rangle.$$  

(25)

We will call $| \tilde{\phi}_{\text{stat}}^{(l)}(t) \rangle$ “projected dynamical states”, as they result from calculations that account for all electron dynamics including strong-field ionization, but the outcomes of these calculations are projected onto the field-free bound-state content. Note that, evaluating the projected dynamical states, the projected dressed ground state is only normalized without any orthogonalization, the first excited dressed state is forced to be orthogonal to the dressed ground state and so on.

3. Numerical results

Unless stated otherwise, we use atomic units (a.u.) in this section: $\hbar = e = m_{e} = 1$, where the units of energy and length are 1 Hartree = 27.21 eV and the Bohr radius $\left(5.29 \times 10^{-11}\text{ m}\right)$, respectively. One atomic unit of the electric field is 5.412 $\times 10^{11}\text{ V/m}$. To compare the different approaches to defining dressed bound states and to illustrate the power of our analytical theory, we solve the TDSE numerically for one electron in one spatial dimension with a soft-core model potential:

$$V(z) = -\frac{1}{\sqrt{z^{2} + a_{0}^{2}}}.$$  

(26)

With the soft-core parameter being equal to $a_{0} = 0.3$, the deepest two energy levels have energies $\epsilon_{1} = -1.75 = 47.49\text{ eV}$ and $\epsilon_{2} = -0.41 = -11.11\text{ eV}$. The TDSE was solved on a grid with a step of $\Delta z = 0.1$ atomic units in a box as large as 819.2 atomic units. We used the first five bound states to investigate various definitions of dressed states. The most excited of these unperturbed states has a binding energy of $\epsilon_{5} = -0.077 = -2.10\text{ eV}$.

We define the light pulses via their vector potentials: $A(t) = -A_{\text{max}} \cos(2 \omega t) \sin(\omega t)$ for $|t| < \pi T/2$ and $A(t) = 0$ for $|t| > \pi T/2$. Here, $A_{\text{max}} = E_{\text{max}} / \omega$ is the peak value reached by the envelope of the vector potential, $E_{\text{max}}$ is the peak value reached by the envelope of the electric field, $\omega$ is the central frequency, and $T$ is related to the full width at half maximum (FWHM) of the pulse intensity as $T = \text{FWHM}/[2 \cos^{-1}(2^{-1/3})]$. Given a vector potential $A(t)$, the electric field is evaluated as $E(t) = -\partial A(t)/(\partial t)$. In our simulations, we used a 3.5-femtosecond laser pulse ($T_{\text{FWHM}} = 100$ fs and a 300-attosecond XUV probe pulse ($T_{\text{probe}} = 10.84 = 0.26 \text{ fs}$). The central wavelength of the laser pulse was set to 760 nanometers ($\omega_{\text{L}} = 0.06 = 2.48 \text{ fs}^{-1}$). The central frequency of the XUV pulse was chosen to be resonant with the transition between the first two stationary states: $\omega_{\text{probe}} = \epsilon_{1}^{(l)} - \epsilon_{0}^{(l)} = 1.34 = 36.38\text{ eV}$. Although we make no attempt to model a realistic atom, the excitation from 0 to 1 may be viewed as analogous to a dipole-allowed transition from an inner-shell orbital of a multi-electron atom to an unoccupied orbital.

In Fig. 1, we compare the amplitudes $|a_{l}^{(l)}(t)|^{2} = |\langle \psi_{\text{stat}}^{(l)}(t) | \hat{\psi}_{\text{stat}}^{(l)}(t) \rangle|^{2}$ evaluated using the four different definitions of dressed bound states introduced in the previous section. In these simulations, we set $n = 1$, that is, each simulation begins with the electron being in first excited state $|1\rangle$. For the peak electric field $E_{\text{max}} = 0.02 = 10^{11}\text{ V/m}$, the ionization probability is as little as 0.3%. If we knew the perfect dressed bound states, then, for such a small ionization probability, our model system would largely remain in $|\psi_{\text{stat}}^{(l)}(t)\rangle$, so that $|a_{l}^{(l)}(t)|^{2}$ would only take values between $|a_{l}^{(l)}(t)|^{2} = 1$ and $|a_{l}^{(l)}(t)|^{2}$, increasing more or less monotonously as the initial state is slightly depleted. Fig. 1 illustrates the well-known fact that the unperturbed bound states are very far from this ideal: $|a_{l}^{(l)}(t)|^{2}$ reaches values below 0.95. This happens mainly because the initial state is distorted by the external field, which reduces the value of $|\langle \psi_{\text{stat}}^{(l)}(t) | \hat{\psi}_{\text{stat}}^{(l)}(t) \rangle|^{2}$. So, even though $|a_{l}^{(l)}(t)|^{2}$ can be interpreted as the probability of being in the unperturbed stationary state, it should not be given the physical meaning of being in some
In this respect, dressed state $|\psi_d^n(t)\rangle$, defined by Eq. (21), performs much better, but it is not perfect either. Even though $|\psi_{u}^{(n)}(t)\rangle$ accounts for the distortion of the initial state in the external field, $|\psi_{d}^{(n)}(t)\rangle$ in Fig. 1a still has pronounced minima at times where $E_{L}(t)$ has its zero crossings, and $|\psi_{d}^{(n)}(t)\rangle^2$ reaches values that are significantly smaller than the final probability of being in state $|1\rangle$. The main reason for this is that $|\psi_{d}^{(n)}(t)\rangle$ is a stationary solution, while the average velocity of a bound electron can become relatively big in an intense laser pulse. As this velocity has maxima at zero-crossings of the electric field, $|\langle \psi_{d}^{(n)}(t)|\psi_{d}^{(n)}(t)\rangle|^{2}$ has minima at these times. This can also be interpreted as a breakdown of the adiabatic (quasistatic) approximation [23]. Also the slow delays between the extrema of $E_{L}(t)$ and the minima of $|\psi_{d}^{(n)}(t)\rangle^2$, visible in Fig. 1a, point out some non-adiabaticity of the electron response.

The intuitively expected step-like decrease of $|a_{d}(t)|^2$, as the laser field depopulates the initial state, is best reproduced by the dynamically dressed states $|\psi_{d}(t)\rangle$ and the projected dynamical states $|\psi_{d}^{(n)}(t)\rangle$, defined by Eqs. (22) and (24), respectively. However, unlike $|\psi_{u}^{(n)}(t)\rangle$ and $|\psi_{d}^{(n)}(t)\rangle$, these dressed states do not necessarily turn into $|n\rangle$ once the laser pulse is gone. In other words, $|\psi_{d}^{(n)}(t)\rangle^2 \neq |\langle n|\psi_{d}^{(n)}(t)\rangle|^2$ at a final time $t_f$. As long as the discrepancy is small, it is legitimate to interpret $|a_{d}(t)|^2$ as the probability of being in the first excited state dressed by the laser field. In these simulations, this discrepancy is much smaller for $|a_{d}^{(n)}(t)|^2$, as compared to $|a_{d}^{(n)}(t)|^2$, which is especially easy to see in Fig. 1b.

The mere fact that, for a certain choice of dressed states, $|a_{d}(t)|^2$ does not show the intuitively expected step-like behavior, does not necessarily mean that these dressed states should not be used in our analytical result (16) for the interaction with a probe pulse. In Fig. 2, we plot the results of simulations where our model atom, initially prepared in state $|1\rangle$, interacts with the near-infrared laser pulse $E_{L}(t)$ and a delayed attosecond XUV pulse $E_{XUV}(t)$ at the end of the interaction with a probe pulse, $a_{d}(t)$ in the denominator of Eq. (16) may become very small. As a result, even small inaccuracies in the approximations that we made deriving Eq. (16) may result in big errors. To avoid this, we restrict the number of states used in Eq. (18):

$$p_{n} = \left| \sum_{n=0}^{\text{max}} z_{n}(t) \langle n|\psi_{d}^{(n)}(t)\rangle \right|^2. \quad (27)$$

The filled circles in Figs. 2a and b represent the results of solving the TDSE on a grid. Comparing these “exact” transition probabilities with the predictions of our analytical model, we clearly see that projected dressed states $|\psi_{d}^{(n)}(t)\rangle$, defined by Eq. (24), give
the most accurate results. At the same time, the unperturbed states $|\phi^{0}_i(t)\rangle = |n\rangle$, which are our most primitive dressed states, also perform surprisingly well. For the calculations with the higher intensity, presented in Fig. 2b, these states even outperform both the adiabatic states $|\phi^0_n(t)\rangle$ and the dynamically dressed states $|\phi^{u}_n(t)\rangle$ in terms of the agreement with the TDSE calculations. For this high intensity, the biggest discrepancy between the exact and analytical results is found for the dynamically dressed states $|\phi^{u}_n(t)\rangle$.

In Fig. 3, we present similar calculations for a case more relevant to possible experiments—in this case, the XUV pulse excites our model atom from its ground state. The figure depicts the probability to find the atom in state $|1\rangle$ after the interaction with the XUV and laser pulses. In contrast to Fig. 2, the unperturbed states $|\phi^{0}_i(t)\rangle$ fail to describe the modulation of the transition probability by the laser field. Both $|\phi^{0}_j(t)\rangle$ and $|\phi^{0}_i(t)\rangle$ yield transition probabilities that agree with the TDSE calculations. The corresponding discrepancies are comparable for the more intense laser pulse (Fig. 3b), but using $|\phi^{0}_i(t)\rangle$ clearly gives more accurate results if the depletion of state $|1\rangle$ is weak (Fig. 3a). Thus, in most cases, the projected dynamical states $|\phi^{u}_j(t)\rangle$, defined by Eq. (24), best satisfy the requirements that we placed on dressed bound states.

Having identified the most promising definition of dressed bound states, we can interpret the effect of the laser pulse on the probabilities of XUV-induced transitions. The overall decrease of $p_{01}$ with $\tau_{\text{probe}}$ in Fig. 2b, as well as the increase of $p_{10}$ in Fig. 3b are clearly due to the depletion of the first excited state by the laser field. The periodic modulations present in all the figures deserve more consideration. In principle, they may be a consequence of two effects: the Stark shift, which detunes the transition frequency away from the central frequency of the XUV pulse, and the modulation of the dressed transition matrix element (17). In the present example, we find that the dominant contribution is due to the effect of the laser field on the dressed matrix elements. To see this, we plot $|Z_{01}(t)|^2$, evaluated with the aid of $|\phi^{0}_j(t)\rangle$, for different intensities of the laser field. Fig. 4. For the central half-cycles of the laser pulse, the modulation depths of $|Z_{01}(t)|^2$ are very close to those of transition probabilities in Figs. 2 and 3. A strong field tends to reduce $|Z_{01}(t)|^2$ because it displaces the electron wavefunction in the first excited state, while the ground state in the present example has a very small polarizability. As a consequence, the overlap between the two dressed states decreases, which results in a smaller value of the dipole transition matrix element between them.

The relatively fast oscillations of $|Z_{01}(t)|^2$ at the tail of the laser pulse are mainly due to the multiphoton excitation of state $|3\rangle$ from state $|1\rangle$. The energy difference between these two states is $\epsilon_{1}^0 - \epsilon_{3}^0 = -0.113 + 0.408 \approx 0.29$ atomic units; since the transitions from both these states to $|0\rangle$ are dipole-allowed, this leads to quantum beats with a period of $2\pi / 0.29 = 21 \text{ a.u.} \approx 0.5 \text{ fs}$. These quantum beats do not, however, appear in the probabilities of the XUV-induced transitions presented in Figs. 2 and 3 because the XUV pulse, which is resonant with the transition between states $|0\rangle$ and $|1\rangle$, does not have enough bandwidth to drive transitions between states $|0\rangle$ and $|3\rangle$. This fact is successfully accounted for by our analytical theory.

4. Conclusions and outlook

We have theoretically investigated how an ionizing few-cycle near-infrared laser pulse affects single-photon bound–bound transitions driven by an attosecond pulse of extreme ultraviolet radiation. Our approach is based on the assumption that even when the laser field is strong enough to significantly ionize a quantum system, one can describe bound electrons with some dressed bound states, which we introduce in an abstract way by a set of simple requirements listed at the beginning of Section 2. With such dressed states, we have obtained relatively simple analytical results for the perturbative interaction with a probe pulse, such as Eq. (16). For a particular model problem, we have systematically compared four different kinds of dressed bound states and found that the most accurate results are generally obtained with the projected dynamical states (24). These states are easily evaluated from numerical simulations of the interaction with the laser pulse, and they make no assumptions about the adiabaticity of the response to the laser pulse.

As the main observable in our numerical examples, we have chosen the probability to find a single-electron atom in a certain state after the interaction with light pulses. This was done to test our analytical theory in a possibly simple and convincing way.
Exponentially, it is easier to measure the transient absorption of an attosecond pulse in the presence of an ionizing laser field \[10\]. It should be straightforward to employ dressed bound states to theoretically investigate attosecond transient absorption measurements. In this paper, we have only made a first step in this direction by writing an explicit expression for the dipole response associated with an XUV-driven laser-dressed bound–bound transition: Eq. (19).

The main purpose of this work has been to define general properties for dressed states in strong laser-atom interactions and to explore their existence. We have shown that quantum states with properties close to those of idealized dressed bound states can be found. From the fact that, overall, the projected dynamical states \[20\] perform best, while all other states show severe shortcomings in various situations, we can draw the following preliminary conclusions: Not surprisingly, beyond the lowest intensities, the unperturbed states \[21\] are not suitable for the analysis of laser induced dynamics. Adiabatic states \[22\] provide a good qualitative picture, but they fail when the velocity acquired by a bound electron in a strong field becomes significant, which is the case for systems with a reasonable polarizability. The dynamically dressed states \[23\] showed us that admitting dynamics, but restricting it to the field-free bound states, may worsen rather than improve the agreement with the full TDSE: It appears that the restriction of the dynamics actually introduces serious artifacts, even when comparing to the projected dynamical states. In this paper, we have only made a first step in this direction, and possibly find a compromise between accuracy and computational simplicity are left to future work.

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