PHYSICAL REVIEW A, VOLUME 61, 031402(R)

Multistate non-Hermitian Floquet dynamics in short laser pulses

H. C. Day,[1,*] Bernard Piraux,[2] and R. M. Potvliege[1]
[1]Physics Department, University of Durham, Science Laboratories, Durham DH1 3LE, United Kingdom
[2]Laboratoire de Physique Atomique et Moléculaire, Université Catholique de Louvain, 2 Chemin du Cyclotron, B-1348 Louvain-la-Neuve, Belgium

(Received 3 August 1999; revised manuscript received 19 November 1999; published 3 February 2000)

A time-dependent non-Hermitian close-coupling method is developed for describing the interplay of decaying atomic dressed states in short laser pulses. This interplay accounts for a variety of phenomena. It is shown, in particular, that the probability that the atom is excited off-resonance is modulated by Stueckelberg oscillations, that the ionization yield may be reduced by dynamical coupling with a resonant and rapidly decaying dressed state, and that adiabatic stabilization of circular states subsists down to very short pulse durations.

PACS number(s): 32.80.Rm, 42.50.Hz

In many cases, the resonance poles of the S matrix can be located by complex scaling or by solving the time-independent Schrödinger equation subject to Siegert-Gamow boundary conditions [1]. Both methods have been used in a number of studies of multiphoton processes occurring in atoms (or molecules) placed in an intense laser field, since the demonstration by Chu and Reinhardt [2] of the feasibility of such calculations in that context. In their non-Hermitian Floquet theory, obtaining the wave function is reduced to a time-independent problem by making the assumption that the atom is illuminated by a stationary laser field, described, for example, by a vector potential $\mathbf{A}(t)=\mathbf{A}_0\sin(\omega t)$ of constant amplitude. The perfect periodicity of the field makes it possible to obtain solutions of the Schrödinger equation in the Floquet form

$$\Psi(\boldsymbol{r},t)=\exp(-iEt/\hbar)F(\boldsymbol{r},t). \tag{1}$$

$F(\boldsymbol{r},t)$ is periodic in time and has the same period as the laser field. The quasienergy E and the Fourier components of $F(\boldsymbol{r},t)$ satisfy a system of partial differential equations that, in conjunction with either one of the two techniques mentioned above, can be tackled by computational methods similar to those used in atomic-structure calculations or in calculations of cross sections for electron-atom scattering [3]. In zero field, $F(\boldsymbol{r},t)$ simply is the wave function of an eigenstate of the atomic Hamiltonian and E the corresponding eigenenergy. A nonzero field couples the bound states to the continuum and turns the bound-state poles of the S matrix into resonance poles coinciding with complex quasienergies. It is tempting to interpret the associated wave functions as describing dressed atomic bound states decaying by multiphoton ionization. There are, however, two difficulties to this: the incident field is never stationary in an actual experiment and the Floquet wave function of any dressed bound state, if not complex-scaled, diverges exponentially at large distances. Nonetheless, many authors have postulated that the time-dependent electronic wave packet can be represented by a Floquet wave function (or approximation thereof) or by a linear superposition of Floquet wave functions, evolving (adiabatically or not) under the temporal variation of the intensity of a laser pulse [4]. The atomic dynamics can then be reduced to transfers of population between—and relaxation into—decaying dressed states. Within various approximations, this approach has been used, in particular, to study the formation of resonance enhancements observed in photoelectron energy spectra.

In this Rapid Communication, we give a consistent formulation of the dynamical coupling of Floquet resonance wave functions, based on a multi-dressed-state close-coupling method proposed some time ago [5] and here generalized to the non-Hermitian case. The results obtained in this way are in excellent agreement with those of fully time-dependent computations. Within this framework, we show that the interplay of just two decaying dressed bound states accounts for a variety of short-pulse phenomena, besides the appearance of resonance structures in the above-threshold ionization spectra. In particular, we show that the probability that the atom is excited off-resonance is modulated by Stueckelberg oscillations, that resonance enhancements in the ionization yield may disappear by dynamical coupling with rapidly decaying dressed states, and that adiabatic stabilization can subsist down to very short pulse durations.

The Floquet multistate method we are concerned with is a particular case of a general non-Hermitian close-coupling formalism applicable to any quantum system in which a few unstable states strongly interact with one another upon the temporal variation of a parameter of the Hamiltonian [6]. Here, the varying parameter is the vector potential amplitude A_0. We assume that the atom can be described in sufficiently good approximation by a finite superposition of Floquet wave functions of decaying dressed bound states of the form

$$\Psi(\boldsymbol{r},t)=\sum_j a_j(t)\,F_j(\boldsymbol{r},t). \tag{2}$$

The method aims at calculating the coefficients $a_j(t)$. It can be sketched as follows. First, recall that two Floquet wave functions can be associated with any bound state quasienergy E_j, i.e.,

$$\Psi_j(\boldsymbol{r},t)=\exp(-iE_jt/\hbar)F_j(\boldsymbol{r},t) \tag{3}$$

---

[*] Present address: Flint Research Institute, 132 Heathfield Road, Keston, Kent BR2 6BA, United Kingdom.

and its adjoint,

$$\Psi_j^\dagger(\mathbf{r},t)=\exp(-iE_j^* t/\hbar)F_j^\dagger(\mathbf{r},t). \qquad (4)$$

They satisfy, respectively, *out*going and *in*going Siegert-Gamow boundary conditions in the open channels. Both $F_j(\mathbf{r},t)$ and $F_j^\dagger(\mathbf{r},t)$ are periodic in time with period $T=2\pi/\omega$, and are defined so as to reduce to a same field-free wave function in the limit $A_0\rightarrow 0$. Different pairs of wave functions are biorthogonal, in the sense that

$$\langle\langle F_j^\dagger|F_n\rangle\rangle\equiv\frac{1}{T}\int_0^T dt\int d\mathbf{r}\,F_j^{\dagger *}(\mathbf{r},t)F_n(\mathbf{r},t)=0 \qquad (5)$$

if $E_j\neq E_n$ [7]. It is useful to impose the normalization condition $\langle\langle F_j^\dagger|F_j\rangle\rangle=1$. Using this and the Schrödinger equation, the coefficients $a_j(t)$ are found to obey a simple system of coupled differential equations under adiabatic temporal variations of A_0, namely,

$$i\hbar\frac{da_j}{dt}=E_j a_j(t)-i\hbar\frac{dA_0}{dt}\sum_n \langle\langle F_j^\dagger|F_n'\rangle\rangle\, a_n(t). \qquad (6)$$

In these equations, F_n' denotes the derivative of $F_n(\mathbf{r},t)$ with respect to A_0, and the field is treated as stationary when performing the cycle-averaging involved in the calculation of the (complex) matrix elements $\langle\langle F_j^\dagger|F_n'\rangle\rangle$. Provided the initial state is properly normalized, the probability that the atom is in the field-free bound state j at the end of the pulse is $|a_j(t\rightarrow\infty)|^2$ and the probability that it is ionized is $1-\Sigma_j|a_j(t\rightarrow\infty)|^2$.

It is only in zero field that the coefficients $a_j(t)$ can be interpreted unambiguously as defining occupation amplitudes, since, in the case we are concerned with, their modulus may temporarily exceed unity when the field is acting. By contrast, the sum $\Sigma_j|a_j(t)|^2$ would remain equal to 1 at all times if the atom was prevented to decay, for instance, by the imposition of bound-state boundary conditions to the wave functions [8]. The corresponding relation reads $\Sigma_j a_j^{\dagger *}(t)a_j(t)\equiv 1$ when photoionization is allowed; it is fulfilled at all times by the solutions of Eq. (6) and those of the system

$$i\hbar\frac{da_j^\dagger}{dt}=E_j^* a_j^\dagger(t)-i\hbar\frac{dA_0}{dt}\sum_n \langle\langle F_j|F_n^{\dagger\prime}\rangle\rangle\, a_n^\dagger(t), \qquad (7)$$

if both systems are solved subject to the same initial conditions. However, the products $a_j^{\dagger *}(t)a_j(t)$ are, in general, complex.

We now let A_0 vary arbitrarily fast, while still calculating the coefficients $a_j(t)$ and the matrix elements $\langle\langle F_j^\dagger|F_n'\rangle\rangle$ as described above. We start by examining the formation of residual populations left by short laser pulses in bound states other than the initial one. This process is usually discussed in terms of population transfers at multiphoton resonances, or, if off-resonance, in terms of Raman transitions made possible by the nonzero bandwidth of the field. However, there is no formal distinction between these two situations in the multistate Floquet approach. Results from the Floquet close-coupling calculations are presented in Fig. 1, for a resonant case, and in Fig. 2, for a nonresonant case. Results obtained by direct solution of the time-dependent Schrödinger equation are also given [9]. The pulses used in these calculations, as in the rest of the paper, have a finite duration, with a temporal envelope $A_0(t)=A_{00}\sin^2\Omega t$, and are linearly polarized. Figure 1 concerns an isolated Stark-shift-induced three-photon resonance between the $1s_0$ dressed state and the $3p_0$ dressed state occurring in atomic hydrogen at about 3×10^{13} W/cm^2 intensity, at 300-nm wavelength [10]. The resonance is marked by an enhancement of the rate of ionization from the $1s_0$ dressed state, but not by an avoided crossing between the real parts of the corresponding quasienergies. (The $3p_0$ quasienergy has a much larger ionization width.) Despite the vanishing energy gap, the dynamical effect of the resonance is not negligible. If the atom is initially in the $1s_0$ state, it manifests by a small (but significant) residual population left in the $3p_0$ state at the end of the pulse. This population is well reproduced by coupling the $1s_0$ and $3p_0$ dressed states, down to pulse durations of but a few optical periods. Its behavior, for varying Ω, is marked by "Stueckelberg oscillations," as has been described for

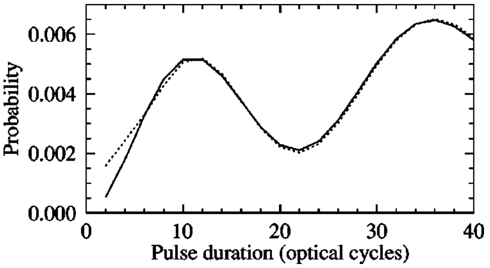

FIG. 1. Probability that a hydrogen atom initially in the $1s_0$ state is in the $3p_0$ state at the end of a pulse of 300-nm wavelength and 8×10^{13} W/cm^2 peak intensity vs the duration of the pulse. Solid line, two-state $(1s,3p)$ Floquet results; dotted line, full time-dependent results.

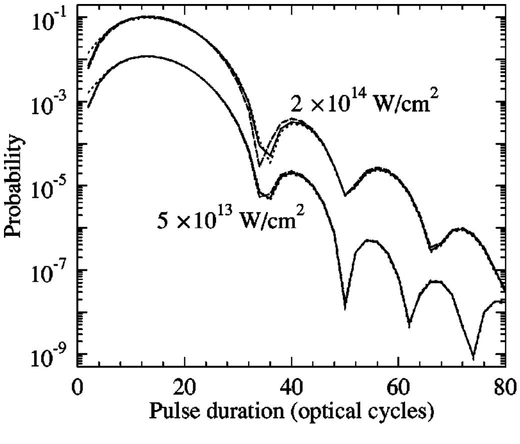

FIG. 2. Probability that a hydrogen atom initially in the $5g_4$ state is in the $6g_4$ state at the end of a pulse of 620-nm wavelength and 5×10^{13} or 2×10^{14} W/cm^2 peak intensity vs the duration of the pulse. Solid lines, three-state $(5g,6g,7g)$ Floquet results; broken lines, two-state $(5g,6g)$ Floquet results; dotted lines, full time-dependent results.

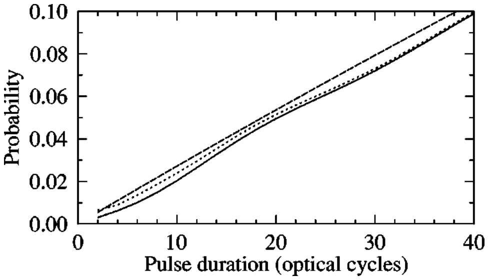

FIG. 3. Probability that a hydrogen atom initially in the $1s_0$ state is ionized by a pulse of 300-nm wavelength and 8×10^{13} W/cm^2 peak intensity vs the duration of the pulse. Solid line, two-state ($1s$,$3p$) Floquet results; broken line, one-state ($1s$) Floquet results; dotted line, full time-dependent results.

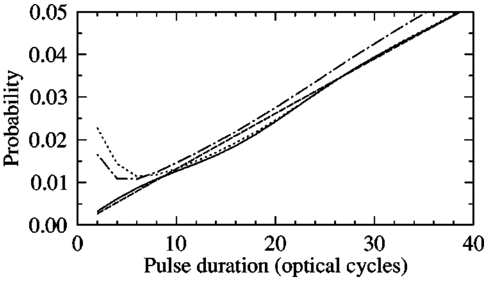

FIG. 5. Probability that a hydrogen atom initially in the $5g_4$ state is ionized by a pulse of 620-nm wavelength and 2×10^{14} W/cm^2 peak intensity vs the duration of the pulse. Solid line, two-state ($5g$,$6g$) Floquet results; broken line, one-state ($5g$) Floquet results; dotted line: full time-dependent results; dash-dotted line: full time-dependent results for a peak intensity of 1×10^{14} W/cm^2.

other resonances of the same nature [11]. These oscillations are due to interferences between transfers made at different times during the pulse, and are a general feature of multistate Floquet dynamics. They can also be observed in Fig. 2, which shows the population left in the $6g_4$ state at the end of a pulse of 620-nm wavelength, for two different peak intensities, the initial state being the $5g_4$. In the dipole approximation, this state is coupled only to bare states with the same magnetic quantum number; the photon energy being larger than the binding energy of any of them, photoionization is never resonant at that wavelength. The residual $6g_4$ population originates from the (weak) coupling between the $5g_4$ and $6g_4$ dressed states. The predictions of a two-state model whereby only these two states are coupled together are indeed in good agreement with the results of the full time-dependent calculation, down to very short pulse durations. It is improved by coupling the $7g_4$ dressed state to the other two. For a given pulse duration, thus a given pulse bandwidth $|dA_0/dt|$ increases with the peak intensity, and with it the probability of population transfer. Because of the Stueckelberg oscillations, however, increasing the pulse bandwidth does not necessarily result in an increase of the probability that the atom is left in the field-free $6g_4$ state.

The ionization yield at 300 nm is presented in Figs. 3 and 4. The figures show results of a one-state Floquet calculation, whereby the atom is described by the single $1s_0$ dressed state, of a two-state Floquet calculation and of a fully time-dependent calculation. The two-state results are in very good agreement with the latter (even for pulses as short as two optical cycles), despite the unphysical asymptotic behavior of the Floquet wave function. An interesting feature of Fig. 3 is that the solid curve is constantly below the dashed curve: despite the larger ionization rate of the $3p_0$ dressed state, coupling it to the $1s_0$ dressed state *reduces* the probability of ionization from the ground state. In other words, the wave packet formed by superposing the two dressed states has a longer lifetime than the isolated $1s_0$ dressed state. The suppression of the resonance enhancement as the pulse duration is shortened is clearly visible in Fig. 4. A study of the state composition of the wave packet reveals that fast variations of the intensity oppose the admixture of the bare $3p_0$ state to the bare $1s_0$ at resonance. In terms of dressed states, the dressed $3p_0$ partly cancels the bare $3p_0$ contribution made by the dressed $1s_0$ to the wave packet. The reduction in the ionization probability can be attributed (heuristically) to this reduction in the weight of the bare $3p_0$ state, since that state ionizes readily by one-photon absorption. The cancellation is less efficient in long pulses owing to the faster decay of the dressed $3p_0$. Conversely, the dressed $3p_0$ component of the wave packet may survive until the end of the pulse if it is short enough, leaving thereby a significant residual population in the field-free $3p_0$ state [12].

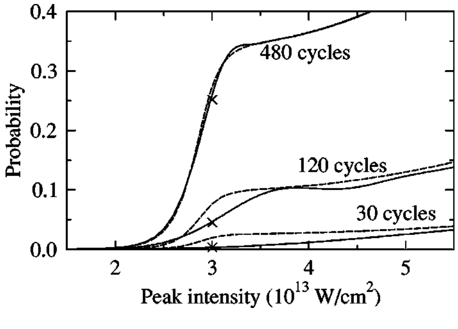

FIG. 4. Probability that a hydrogen atom initially in the $1s_0$ state is ionized by a pulse of 300-nm wavelength and 30, 120, or 480 optical cycles duration vs the pulse peak intensity. Solid lines, twostate ($1s$,$3p$) Floquet results; broken lines, one-state ($1s$) Floquet results; crosses, full time-dependent results.

Finally, we consider ionization from the $5g_4$ dressed state at 620 nm. This process has been analyzed for relatively long pulse durations (120 cycles) in Ref. [13]. For such pulses, adiabatic stabilization of the $5g_4$ results in a smaller ionization yield at 2×10^{14} W/cm^2 peak intensity than at 1×10^{14} W/cm^2, and the single-state Floquet approximation is very accurate. Figure 5 shows that the same remains true for pulses as short as ten cycles. Population transfer to other dressed bound states does not appear to spoil stabilization. On the contrary, the coupling with the $6g_4$ state reduces the yield below 20 cycles, at 2×10^{14} W/cm^2. This reduction can be simply explained by the greater stability of the $6g_4$ in the field, rather than by a dynamical interference. The yield for ultrashort pulses is underestimated by the multistate Floquet calculation, though, presumably because it neglects Raman transitions to continuum states. These transitions can be expected to be more important here than in the case of Fig. 3, in view of the smaller binding energy of the initial state. Their contribution to the total ionization yield of Fig. 5, estimated from the difference between the time-dependent results and the Floquet results below ten cycles, is roughly proportional to the square root of the peak intensity and inversely proportional to the pulse duration. This dependence is consistent with the dA_0/dt factor in the right-hand side of Eq. (6).

This work has benefited from discussions with M. Dörr and from financial support given to H.C.D. and R.M.P. by EPSRC.

---

[1] N. Moiseyev, Phys. Rep. **302**, 212 (1998).

[2] S.-I Chu and W. P. Reinhardt, Phys. Rev. Lett. **39**, 1195 (1977).

[3] For a recent review, see, e.g., C. J. Joachain, M. Dörr, and N. J. Kylstra, Adv. At. Mol. Opt. Phys. (to be published).

[4] Floquet calculations are usually performed in the approximation whereby the atom is represented by a single dressed state. Examples of multistate Floquet analyses applied to multiphoton ionization can be found in M. S. Pindzola and M. Dörr, Phys. Rev. A **43**, 439 (1991); V. C. Reed, P. L. Knight, and K. Burnett, Phys. Rev. Lett. **67**, 1415 (1991); D. I. Duncan, J. G. Story, and T. F. Gallagher, Phys. Rev. A **52**, 2209 (1994); M. Edwards and C. W. Clark, J. Opt. Soc. Am. B **13**, 101 (1996); P. G. Burke and V. M. Burke, J. Phys. B **30**, L383 (1997); M. Dörr, O. Latinne, and C. J. Joachain, Phys. Rev. A **55**, 3697 (1997); and J. C. Wells, I. Simbotin, and M. Gavrila, *ibid.* **56**, 3961 (1997).

[5] T.-S. Ho and S.-I Chu, Chem. Phys. Lett. **141**, 315 (1987); R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 4597 (1988). See also K. Drese and M. Holthaus, Eur. Phys. J. D **5**, 119 (1999).

[6] See, e.g., H. C. Baker, Phys. Rev. A **30**, 773 (1984); V. V. Kocharovsky, Vl. V. Kocharovsky, and S. Tasaki, Adv. Chem. Phys. **99**, 333 (1997) and references therein.

[7] The wave functions diverging exponentially, the spatial integral must be regularized or be defined by analytic continuation; see, e.g., R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989). As will be discussed elsewhere, the inner product defined by Eq. (5) is equivalent when the Hamiltonian matrix is symmetric (which is the case for monochromatic fields) to the generalized inner product defined by N. Moiseyev, P. R. Certain, and F. Weinhold, Mol. Phys. **36**, 1613 (1978).

[8] The quasienergies would also be real in this case, and there would be no difference between the Floquet wave functions and their adjoints.

[9] Details about the numerical methods can be found in E. Huens, B. Piraux, A. Bugacov, and M. Gajda, Phys. Rev. A **55**, 2132 (1997), and in R. M. Potvliege, Comput. Phys. Commun. **114**, 42 (1998).

[10] By ‘‘$n\ell_m$ dressed state’’ we mean the dressed state originating from the field-free state with quantum numbers n, ℓ, and m. The axis of quantization of the angular momenta is parallel to the direction of polarization.

[11] K. J. Schafer and K. C. Kulander, Laser Phys. **7**, 740 (1997).

[12] This reduction in the ionization probability thus results from the transient formation of a nonstationary wave packet, and is not an instance of population trapping into a quasistationary state similar to that analyzed, e.g., by H. R. Gray, R. M. Whitley, and C. R. Stroud, Opt. Lett. **3**, 218 (1978).

[13] B. Piraux and R. M. Potvliege, Phys. Rev. A **57**, 5009 (1998).