Resonant pulse excitation leading to ionization

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Laser excitation of a two-level atom is analytically studied for arbitrary pulse shape, area, and energy. The ionization mechanism is modeled as a homogeneous width γ of the atomic excited state. A transformation of the optical Bloch equations allows the dynamics to be completely described by a single equation for the area of the pulse at time t, $\Phi(t)$. Analytic solutions for the total bound population for a wide variety of pulse shapes can be found in two regions: $\Phi(t) \approx \sin\Phi(t)$ and $\Omega(t) > \gamma$, where $\Omega(t)$ is the resonance Rabi frequency. The generalization of the two-level atom results to the N-level atom is discussed.

Ever since the classic work of Keldysh,[1] theoretical research on multiphoton ionization of atoms consisted primarily of some form of perturbation theory calculation of the ionization rate.[2] However, a dynamic study of two-photon ionization by Beers and Armstrong (BA)[2(b)] shows that rates are valid in two limiting cases: the weak-field limit and the strong-field limit.[3] In intermediate regions, the simulated coherent pumping of population between the ground and excited states makes the ionization dynamics a very complicated function of time. Shore and Ackerhalt (SA)[4] recently made a computer study of multiphoton ionization dynamics of three-level atoms. Their analysis of the population flow shows, in particular, in which parameter regimes bottlenecking is minimized.

In all these studies, the laser has a square pulse shape:

$$\Omega(t) = \begin{cases} 0, & t < t_0 \\ \Omega, & t_0 \leq t \leq t_0 + T \\ 0, & t_0 + T < t, \end{cases}$$

where $\Omega(t)$ is the on-resonance Rabi frequency,[5] t_0 is the initial pulse time, and T is the time duration of the pulse. This special, but artificial, pulse shape has the advantage that exact analytic solutions are easily found for many problems. Crance and Feneuille (CF)[6] extended this multiphoton ionization research by studying the effect of pulse shape on multiphoton ionization using the model of BA. They analytically compute square-pulse solutions, but use the computer to generate solutions for other selected shapes in the weak field-limit of BA.

Since the work of CF is very closely related to the present work, it deserves a more detailed discussion concerning some basic differences. The CF model is identical to the model of BA, in which the atom consists of two bound states and a continuum of states. The laser both excites the atom and ionizes the excited atom. Both the two-step and direct paths to the continuum are taken into account. The shape of the laser pulse affects both the excitation and ionization steps. In our model,[4] the atom consists of two bound states and essentially a third level which represents the continuum. The laser pulse only excites the atom. Ionization is due to some mechanism other than the exciting pulse which is time independent throughout the duration of the excitation. Ionization occurs only from the atomic excited state, giving that state a width proportional to the inverse of the ionization rate.

These models represent different physical situations making our work complementary to the work of CF. Because our equations of motion are simpler, than those of CF, in that our ionization rate is independent of the pulse excitation, we are able to find general analytic solutions in certain parameter regimes. In addition, we find that the optical Bloch equations can be cast into the form of a single equation for the area of the pulse $\Phi(t)$, giving us a physically intuitive picture of the ionization process.[7] The two regimes for which analytic solutions are easily found are $\Phi(t) \simeq \sin\Phi(t)$ and $\Omega(t) > \gamma$, where γ is the homogeneous width of the atomic state due to the ionization mechanism.

For $\Omega(t) > \gamma$ the ionization is described by a single rate constant $\frac{1}{2}\gamma$, which is the strong-field limit of BA. In the region $\Phi(t) \approx \sin\Phi(t)$, the total ionization depends only on the integrated square of the Rabi frequency or equivalently on the integrated pulse intensity. For a square pulse, the ionization is described by a single rate constant, Ω^2/R, which is the weak-field limit of BA. These two-level-atom results apply also to the case of multiphoton excitation and ionization where no intermediate state resonances exist, but where the N-photon transition is resonant. In this instance, the total ionization depends on the integrated product of each laser's intensity.

In Sec. I we derive the equations of motion describing the ionization dynamics. Analytic solutions are computed in Sec. II. In Sec. III we discuss the application of these results to multiphoton excitation and ionization. In particular, two-photon Doppler-free excitation and ionization are described. In Appendix A analytic solutions for several pulse shapes are computed. In particular, we present solutions for the hyperbolic secant pulse shape of Ref. 7. In Appendix B the Bloch equations are replaced by rate equations. The solutions are identical to those found in the limit $\Phi(t) \simeq \sin\Phi(t)$.

I. EQUATIONS OF MOTION

The dynamics of excitation and ionization of a two-level atom can be described by a generalized form of the optical Bloch equations[8]:

$$\dot{u} = -\Delta v - \tfrac{1}{2}\gamma u , \tag{1.1}$$

$$\dot{v} = \Delta u + \Omega(t)(\rho_{22} - \rho_{11}) - \tfrac{1}{2}\gamma v , \tag{1.2}$$

$$\dot{\rho}_{11} = \tfrac{1}{2}\Omega(t) v , \tag{1.3}$$

$$\dot{\rho}_{22} = -\tfrac{1}{2}\Omega(t) v - \gamma\rho_{22} . \tag{1.4}$$

The off-diagonal density matrix elements $(\rho_{12}+\rho_{21})$ and $i(\rho_{12}-\rho_{21})$ are represented by u and v, respectively. The diagonal density matrix elements are ρ_{22} and ρ_{11} where ρ_{22}, ρ_{11} is the population in the upper, lower atomic state. The laser-atom detuning, ionization rate, and resonance Rabi frequency are Δ, γ, and $\Omega(t)$. The shape of the pulse is given by specifying the time dependence of $\Omega(t)$. The homogeneous width of the upper state, γ, is due to an ionization mechanism which is totally independent of the excitation pulse and due to another laser, collisions, or dc field. The rate γ is assumed constant over the entire duration of the pulse excitation. From this point on, we will only consider resonant excitation, $\Delta = 0$. Since $\rho_{11}(t) = 1$ for $t \leq t_0, u(t) = 0$ for all t.

By defining the new quantities

$$n = \rho_{22} + \rho_{11} , \quad w = \rho_{22} - \rho_{11} , \tag{1.5}$$

where n is the total bound population and w is the population inversion, we obtain

$$\dot{v} = \Omega(t) w - \tfrac{1}{2}\gamma v , \tag{1.6}$$

$$\dot{w} = -\Omega(t) v - \tfrac{1}{2}\gamma(n + w) , \tag{1.7}$$

$$\dot{n} = -\tfrac{1}{2}\gamma(n + w) . \tag{1.8}$$

If $\gamma = 0$, no ionization, then all the population remains in the two-level atom, $n(t) = 1$. The solutions to (1.6) and (1.7) in this case are well known:

$$v(t) = -\sin\Phi(t) , \tag{1.9}$$

$$w(t) = -\cos\Phi(t) , \tag{1.10}$$

for $t \geq t_0$ where

$$\dot{\Phi}(t) = \Omega(t) \tag{1.11a}$$

or

$$\Phi(t) = \int_{t_0}^{t} \Omega(t')\, dt' \tag{1.11b}$$

and $\Phi(t_0) = 0$.[7] $\Phi(t)$ is the area of the pulse at time t. It represents the angle through which the Bloch vector is rotated (see Ref. 5). When $\Phi = n2\pi, \Phi = (2n+1)\pi, (n = 0, 1, 2, \ldots)$, the atom is in the ground, excited state. The length of the Bloch vector is unity: $v^2 + w^2 = 1$. As we will show, even in the presence of ionization population loss, the Bloch vector picture of atomic excitation can be used giving an intuitive physical description of the ionization dynamics.

Making the transformation

$$\alpha = \bar{\alpha} e^{-\gamma/2(t-t_0)} , \tag{1.12}$$

where α represents v, w, n, the common decay proportion to $\frac{1}{2}\gamma$ is removed from Eqs. (1.6)–(1.8). Since ionization removes population from the two-level atom, we recognize that after a sufficiently long pulse, all the population will be ionized making $v(\infty) = w(\infty) = n(\infty) = 0$. Since n is the total bound population in the two-level atom, it reflects the population lost to ionization, $\rho_I = 1 - n$. The excitation dynamics are described by w and v which are also affected by the ionization population loss. Therefore, making a transformation of the form

$$\alpha' = \bar{\alpha}/\bar{n} , \tag{1.13}$$

where α represents v, w the ionization population loss dynamics are now taken into account allowing the magnitude of a new Bloch vector to be preserved: $(v')^2 + (w')^2 = 1$.

The equations of motion for these new variables v', w', and for $\bar{n}$ are

$$\dot{v}' = [\tfrac{1}{2}\gamma v' + \Omega(t)] w' , \tag{1.14}$$

$$\dot{w}' = -[\tfrac{1}{2}\gamma v' + \Omega(t)] v' , \tag{1.15}$$

$$\dot{\bar{n}} = -\tfrac{1}{2}\gamma w' \bar{n} \tag{1.16}$$

where (1.14) and (1.15) are of the same form as (1.6) and (1.7) with $\gamma = 0$. Consequently, the solutions to (1.14) and (1.15) are identical in form to (1.9) and (1.10):

$$v'(t) = -\sin\Phi(t) , \tag{1.17}$$

$$w'(t) = -\cos\Phi(t) , \tag{1.18}$$

where $\Phi(t_0) = 0$. The equation for Φ is

$$\dot{\Phi}(t) = \tfrac{1}{2}\gamma v' + \Omega(t) = -\tfrac{1}{2}\gamma \sin\Phi(t) + \Omega(t) . \tag{1.19}$$

The solution for $\bar{n}(t)$ is

$$\bar{n}(t) = \exp\left(-\frac{\gamma}{2} \int_{t_0}^{t} dt'\, w'(t')\right) . \tag{1.20}$$

Substituting (1.18) and the transformation (1.12) into (1.20), we find

$$n(t)=\exp\left(-\frac{\gamma}{2}\int_{t_0}^{t}dt'[1+w'(t')]\right)$$
$$=\exp\left(-\frac{\gamma}{2}\int_{t_0}^{t}dt'[1-\cos\Phi(t')]\right), \qquad (1.21)$$

where $[1+w'(t')]/2$ represents the population in the excited state at time t' normalized to $\bar{n}$. The solutions for v and w are

$$v(t)=-\sin\Phi(t)n(t), \qquad (1.22)$$

$$w(t)=-\cos\Phi(t)n(t), \qquad (1.23)$$

where the solutions to (1.21)–(1.23) are obtained after solving the single equation for $\Phi(t)$, Eq. (1.19).

The essential differences between the solutions without ionization, (1.9)–(1.11), and the solutions with ionization, (1.21)–(1.23) and (1.19), are the population normalization to n and the modified equation for the area $\Phi(t)$.

The physics of solutions (1.21)–(1.23) is straightforward: $w(t)$ and $v(t)$ are the usual components of the Bloch vector normalized to the total bound population. $n(t)$ decays at a time dependent rate which depends at any instant in time on the normalized population which is in the excited state. $\Phi(t)$ determines both the usual dynamics of w, v, and also the population loss from the upper state.

The dynamics of $\Phi(t)$ can be easily understood from (1.19): $\Phi(t)$ grows as the electric field envelope of the pulse grows, but always experiences a decay toward stable pulse areas, $\Phi=n2\pi(n=0,1,2,\ldots)$, due to the population ionization loss. The stable areas obviously correspond to all the population in the ground state. Unstable areas occur for $\Phi=(2n+1)\pi$ $(n=0,1,2)$ which correspond to all the population in the excited state.

For pulses whose duration is short with respect to $1/\gamma$ ionization takes place only after the pulse has passed. For $t\geq t_0+T$

$$n(t)=e^{-\bar{\gamma}(t-t_0-T)}, \qquad (1.24)$$

where the ionization rate $\bar{\gamma}$ is a function of the population which remains in the excited state after the pulse has passed:

$$\bar{\gamma}=\frac{1+w'(t_0+T)}{2}=\frac{1-\cos\Phi(t_0+T)}{2}. \qquad (1.25)$$

In Sec. II we will discuss pulses whose durations are on the same order or greater than $1/\gamma$.

II. ANALYTIC SOLUTIONS

The general solutions, (1.21)–(1.23), and the equation for the area, (1.19), are so physically transparent that further, more detailed solutions might seem unnecessary. However, in two regimes, general analytic solutions can be found. In this section, for these two regimes, we will solve for $\Phi(t)$ and for the total number of ions produced by the pulse, $\rho_I(\infty)$. Since $\rho_I(\infty)=1-n(\infty)$, we will only need to solve for $n(t)$.

Consider the case where the pulse area never becomes greater than roughly $\frac{1}{5}\pi$, then $\sin\Phi(t)\approx\Phi(t)$. Equation (1.19) becomes

$$\dot{\Phi}=-\tfrac{1}{2}\gamma\Phi+\Omega(t). \qquad (2.1)$$

The solution to (2.1) is

$$\Phi(t)=\int_{t_0}^{t}dt'\,e^{-\gamma(t-t')/2}\Omega(t'), \qquad (2.2)$$

and the solution for $n(t)$ is

$$n(t)=\exp\left[-\frac{\gamma}{2}\int_{t_0}^{t}dt'\,(\Phi^2(t')/2)\right]. \qquad (2.3)$$

For pulses which can be modeled for some time period as a polynomial or an exponential (2.2) and (2.3) can be integrated exactly. Some examples will be given in Appendix A.

If the time rate of change of $\Omega(t)$ is sufficiently slow with respect to γ, then the exponential in (2.3) decays very fast. Equation (2.2) can be approximately integrated for $t>1/\gamma$:

$$\Phi(t)\simeq 2\Omega(t)/\gamma. \qquad (2.4)$$

Substituting (2.4) into (2.3), we find

$$n(t)\cong\exp\left(-\int_{t_0}^{t}dt'\,\frac{\Omega^2(t')}{\gamma}\right) \qquad (2.5)$$

which for a square pulse becomes

$$n(t)\simeq e^{-\Omega^2(t-t_0)/\gamma}. \qquad (2.6)$$

The rate Ω^2/γ is the weak-field-limit perturbation theory rate of BA. Assuming the pulse area is never greater than $\frac{1}{5}\pi$ requires from (2.5), that

$$|\Omega(t)|_{\max}\leq\tfrac{1}{10}\pi\gamma. \qquad (2.7)$$

Since the integral in (2.5) is an integral of the laser's intensity, we see that the ionization depends only on the total energy in the pulse. The relation of these results to multiphoton excitation and ionization will be discussed in Sec. III. In Appendix B this result, (2.5), will be shown equivalent to the result obtained using rate equations.

In the limit $|\Omega(t)|>\frac{1}{2}\gamma$, the ionization loss in (1.19) is very small such that the pulse area satisfies (1.11b). Since $\Phi(t)$ in (1.21) oscillates very fast with respect to γ, the integral of $\cos\Phi(t)$ is negligible making the bound-state population decay at the rate $\frac{1}{2}\gamma$:

$$n(t)\simeq e^{-\gamma t/2} \qquad (2.8)$$

which is the strong field limit of BA.

In intermediate regions where we are not in either the weak-field or strong-field limits, solutions can be found by numerical integration, modeling the pulse using a square-pulse shape which has an exact solution, or by breaking the pulse into pieces where the area growth of each piece is smaller than $\frac{1}{5}\pi$ such that the ideas presented in this section can be utilized.

III. MULTIPHOTON EXCITATION AND IONIZATION

We studied in Secs. I and II a two-level atom excited by a laser pulse of arbitrary shape and ionized by some mechanism other than the excitation pulse. The pulse shape appears in the equations as a time-dependent Rabi frequency. The detuning of the laser from the atomic transition frequency is zero. In the case where the pulse area never becomes greater than roughly $\frac{1}{5}\pi$, the ionization depends only on the integrated square of the Rabi frequency. This case corresponds physically to the region where ionization occurs more rapidly than the pulse can excite the atom.

CF point out that the two-level model can describe an N-level model if the intermediate atomic levels are all very far off-resonance.[9] The population in this case simply cycles back and forth between the ground level and level N at an effective Rabi frequency: for the three-level model

$$|\Omega(t)| = |\Omega_1(t)\Omega_2(t)/\Delta_1| \ . \tag{3.1}$$

The index 1, 2 refers to the first, second transition. Δ_1 is the laser-atom detuning for transition 1. For the two-photon resonant case of interest here, $\Delta_1 = -\Delta_2$.

The requirement that the N-level model dynamics reduce to two-level model dynamics means that

$$|\Delta_1| > \Omega_1, \Omega_2 \tag{3.2}$$

which from (3.1) implies

$$\Omega < \Omega_1, \Omega_2 \, . \tag{3.3}$$

In general, the effective Rabi frequency is always smaller than the individual transition Rabi frequencies because the laser-atom intermediate state detunings are large with respect to the transition Rabi frequencies.

In most cases the ionization rates will compare in magnitude with the transition Rabi frequencies. Therefore, for N-photon resonant excitation and ionization, the effective Rabi frequencies will be much smaller than the rate of ionization making the results (2.2)–(2.5) applicable in this general case. Using (2.5) and (3.1), we find the ionization for a three-level atom:

$$n(t) \simeq \exp\left(-\int_{t_0}^{t} dt' \ \frac{\Omega_1^2(t')\Omega_2^2(t')}{\Delta_1^2 \gamma}\right), \tag{3.4}$$

where the total ionization depends on the overlap of the pulse intensities or for a single pulse excitation on the square of the pulse intensity. Equation (3.4) can be easily generalized for the N-level model.

Our model requires that level N be resonant with the sum of the exciting lasers' transition frequencies giving an $(N-1)$ photon resonance. For laser isotope separation and other multiphoton excitation and ionization processes, the atomic system is in the vapor phase resulting in a large Doppler broadening of the atomic transitions. Our model would not apply to this case unless the ionization rate is sufficiently large. However, for $3, 5, 7 \cdots$ state models it is possible to use nearly equal frequency counter-propagating pulses to excite the $(N-1)$ photon resonance for all the atoms in the Doppler profile. Doppler-free excitation is presently a very valuable spectroscopic tool.[10] The three-level model is, therefore, the simplest model for which Doppler broadening can be overcome, making our result (3.4) valid.

CONCLUSIONS

Analytic solutions can be found for the pulse-excitation and ionization problem using the model of SA for a two-level model independent of pulse shape in two limiting cases: the area of the pulse $\Phi(t)$ never becomes greater than $\frac{1}{5}\pi$ and the magnitude of the Rabi frequency is always greater than the ionization rate.

The solution for the bound population in the case where $|\Phi(t)|_{\max} \leq \frac{1}{5}\pi$ depends on the time integrated intensity of the pulse and is independent of the pulse shape.

The two-level-model solutions are valid for the N-level model if all the intermediate level laser-atom detunings are far from resonance and if the $(N-1)$ photon detuning from level N is exactly on resonance. This condition on the laser-atom detunings can be easily satisfied for all atoms in the Doppler velocity distribution if level $N = 3, 5, 7 \ldots$ and if the excitation pulses are counter propagating such that each two-photon excitation is Doppler free.

The effective Rabi frequency for the N-level model will always be much smaller than any single-step Rabi frequency due to the large intermediate level laser-atom detunings. The typical order of magnitude for the ionization rate γ is similar to the order of magnitude of the single-step Rabi frequency making it much greater than the effective N-level Rabi frequency. This region corresponds to the case where $|\Phi(t)| \leq \frac{1}{5}\pi$. Therefore, for the

N-level model the total bound population depends on the integrated overlap of the pulses' intensities and is independent of their specific shape.

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APPENDIX A

In this Appendix we will apply the analytic formulas derived in Sec. II to two selected pulse shapes. We will consider only the case where the ionization is sufficiently fast such that the pulse area is never greater than $\frac{1}{5}\pi$. We will use these example pulse shapes to compare the results (2.1)–(2.3) and (2.4) and (2.5). Since all the integrals can be found in standard tables, we will only give the final results in all cases.

Let us consider the hyperbolic secant pulse shape[7]:

$$\Omega(t) = A\gamma/4\cosh(\gamma t/2)\,, \tag{A1}$$

where the pulse area θ (defined in the absence of ionization) and energy E are

$$\theta = \int_{-\infty}^{\infty} dt'\,\Omega(t') = A\pi\,, \tag{A2}$$

$$E \sim \int_{-\infty}^{\infty} dt'\,\Omega^2(t') = A^2\gamma/4\,. \tag{A3}$$

The pulse width has been chosen proportional to γ so that the integral in (2.2) can be evaluated for the time-dependent area in the presence of ionization:

$$\Phi(t) = \int_{-\infty}^{t} dt'\,\frac{e^{-\gamma(t-t')/2}A}{\cosh(\gamma t'/2)}\frac{\gamma}{4} = \tfrac{1}{2}A\,e^{-\gamma t/2}\ln(1+e^{\gamma t})\,. \tag{A4}$$

We must require that $\Phi(t)$ is never greater than $\frac{1}{5}\pi$:

$$\Phi(t)_{\max} \simeq \tfrac{1}{4}A\ln 5 = 0.4A \leq \tfrac{1}{5}\pi \tag{A5}$$

or using (A2)

$$\theta \leq 1.57\pi\,. \tag{A6}$$

An atom not undergoing ionization could have its Bloch vector turned through an angle as large as 1.57π, but an atom with ionization rate γ will only have its Bloch vector turned through an angle as large as $\frac{1}{5}\pi$.

The total bound population after the pulse has passed is found using (2.3):

$$n(\infty) = \exp\left(-\frac{\gamma}{4}\left(\frac{A}{2}\right)^2\int_{-\infty}^{\infty} dt'\,e^{-\gamma t'}[\ln(1+e^{\gamma t'})]^2\right) = e^{-(1/3)(A\pi/4)^2}\,. \tag{A7}$$

For a π pulse ($A = 1$) we find $n(\infty) = 0.81$, or 19% of the atoms are ionized.

Let us compare this result with the general result (2.5):

$$n(\infty) = \exp\left\{-\int_{-\infty}^{\infty} dt'\gamma\left(\frac{A}{4}\right)^2\left[1/\cosh\left(\frac{\gamma+1}{2}\right)\right]^2\right\} = e^{-(A/2)^2}\,. \tag{A8}$$

From (2.7) we require $A \leq \frac{2}{5}\pi = 1.26$ or $\theta \leq 1.26\pi$ which is somewhat more restrictive than (A6). For a π pulse ($A = 1$) (A8) gives $n(\infty) = 0.78$, or 22% of the atoms are ionized. A comparison of the results (A7) and (A8) shows them to be in relatively good agreement.

Let us consider a square pulse of finite duration:

$$\Omega(t) = 0,\quad t<0,\ t>\tau\,,$$
$$\Omega(t) = \Omega,\quad 0 \leq t \leq \tau\,. \tag{A9}$$

The area θ and energy E of the pulse are

$$\theta = \Omega\tau\,, \tag{A10}$$

$$E = \Omega^2\tau\,. \tag{A11}$$

If the area and energy of the pulse are set equal to (A2) and (A3), then

$$\Omega = A\gamma/4\pi\,, \tag{A12}$$

$$\tau = 4\pi^2/\gamma\,. \tag{A13}$$

Substituting (A9) into (2.2) gives

$$\begin{aligned}\Phi(t) &= \int_0^t dt'\,e^{-\gamma(t-t')/2}\,\Omega u(t-\tau) \\ &= (2\Omega/\gamma)(1-e^{-(\gamma/2)t}),\quad t \leq \tau \\ &= (2\Omega/\gamma)e^{-\gamma t/2}(e^{(\gamma/2)\tau}-1),\quad t \geq \tau\end{aligned} \tag{A14}$$

where $u(t-\tau)$ is a step function.

The total bound population remaining at the end of the pulse is found using (2.3):

$$\begin{aligned}n(\tau) &= \exp\left[-\frac{\gamma}{4}\left(\frac{2\Omega}{\gamma}\right)^2\int_0^{\tau} dt'\,(1-e^{-(\gamma/2)t'})^2\right] \\ &= \exp\left\{-\left(\frac{\Omega}{\gamma}\right)^2[\gamma\tau-(3-e^{-(\gamma/2)\tau})(1-e^{-(\gamma/2)\tau})]\right\}\end{aligned} \tag{A15}$$

which using (A13) reduces approximately to

$$n(\tau) \sim \exp\{-(A/4\pi)^2\,[4\pi^2-3]\} \sim \exp[-(A/2)^2]\,. \tag{A16}$$

If the ionization continues after the pulse has passed, then we find

$$\begin{aligned}n(\infty) &= n(\tau)\exp\left[-\frac{\gamma}{4}\int_{\tau}^{\infty} dt'\left(\frac{2\Omega}{\gamma}\right)^2\left(e^{(\gamma/2)\tau}-1\right)^2 e^{-\gamma t'}\right] \\ &= n(\tau)\exp\left[-\left(\frac{\Omega}{\gamma}\right)^2\left(1-e^{-\gamma\tau/2}\right)^2\right]\,.\end{aligned} \tag{A17}$$

Applying (A13) reduces (A17) to

$$n(\infty)=n(\tau)\exp[-(A/4\pi)^2]\sim n(\tau). \qquad \text{(A18)}$$

Essentially no ionization occurs in this special case after the pulse has passed. Since (A16) or (A18) is identical with (A8), we find the ionization to be independent of which shape was chosen for the pulse, the hyperbolic secant, or the square-pulse shape, as long as the energy in the two pulses is the same.

We also considered an exponential pulse:

$$\Omega(t)=0, \quad t\leqslant 0$$

$$\Omega(t)=(A\pi/\tau^2)t\,e^{-t/\tau}, \; t\geqslant 0. \qquad \text{(A19)}$$

The results for this shape were in agreement with the hyperbolic secant and square pulse solutions and will not be given here.

APPENDIX B

In this Appendix we will show that in the case where the area of the pulse in the presence of ionization never is larger than $\frac{1}{5}\pi$, (2.5) is just the solution one would expect using standard rate equations.

We can derive standard rate equations from (1.6)–(1.8) using the Wilcox-Lamb method.[11] The procedure is to set the time derivatives of the off-diagonal density matrix elements to zero. The solutions of these algebraic equations for the off-diagonal density matrix elements are substituted into the equations of motion for the diagonal density matrix elements. Following this procedure (1.6)–(1.8) become

$$\dot{w}=-\frac{2\Omega^2(t)}{\gamma}w-\frac{\gamma}{2}(n+w), \qquad \text{(B1)}$$

$$\dot{n}=-\tfrac{1}{2}\gamma(n+w). \qquad \text{(B2)}$$

We can put Eqs. (B1) and (B2) in a more convenient form by applying transformations (1.12) and (1.13):

$$\dot{w}'=-\frac{2\Omega^2(t)}{\gamma}w'-\frac{\gamma}{2}(1-w'^2) \qquad \text{(B3)}$$

$$\dot{\bar{n}}=-\tfrac{1}{2}\gamma w'\bar{n}. \qquad \text{(B4)}$$

From (2.7) we see that $|\Omega(t)|/\gamma\ll 1$ which implies the dynamics of (B3) is determined from the term proportional to $(1-w'^2)$. If $\Omega(t)$ is set equal to zero in (B3), (B3) has an exact solution, $w'=-1$. We therefore expect w' to always be nearly -1 allowing us to make (B3) into a linear equation:

$$\dot{w}'\simeq-\gamma(1+2\Omega^2(t)/\gamma^2)w'-\gamma \qquad \text{(B5)}$$

which has the approximate steady-state solution

$$w'=-1+2\Omega^2(t)/\gamma^2. \qquad \text{(B6)}$$

We have assumed that γ relaxes w to its steady-state value faster than the rate at which $\Omega(t)$ can change that value.

After inserting (B6) into (B4) and applying the inverse transformation of (1.12), we find

$$n(t)\simeq\exp\left(-\int_{t_0}^{t}dt'\,\frac{\Omega^2(t')}{\gamma}\right) \qquad \text{(B7)}$$

which is the rate equation solution for the total bound population. Equation (B7) is identical with (2.5) showing that we are in the rate equation limit of the Bloch equations when we require $\Phi(t)$ to always be less than $\frac{1}{5}\pi$.

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