Analytic Solution for Strong-Field Quantum Control of Atomic Wave Packets

Luís E. E. de Araujo, Ian A. Walmsley, and C. R. Stroud, Jr.
The Institute of Optics, University of Rochester, Rochester, New York 14627
(Received 30 June 1997)

A simple analytic formula is derived for the impulse that will produce approximately, for arbitrarily large population transfer, a specified wave packet in a Rydberg atom. This solution to the nonlinear control problem is obtained in the limit that the duration of the pulsed control field is less than the Kepler period of the system. The validity of our solution is tested by comparison with a direct numerical integration of Schrödinger's equation, and it is found to yield the target state and population transfer with very high fidelity. [S0031-9007(98)06764-7]

PACS numbers: 32.80.Qk, 42.50.Hz, 42.50.Vk

Perhaps the ultimate goal of engineering is to develop the ability to control matter at its most fundamental level—that is, to have control over the quantum state of one or more degrees of freedom of an atom or molecule. The preparation of particular quantum states of atoms and molecules is important in a number of emerging areas, including quantum computation [1] and nanoscale technology [2]. For this reason, a number of quantum control theories have been developed in the past few years by extending the control theory of classical systems into the quantum domain [3–5]. These theories, which seek a prescription for the control field that prepares a specific state of an internal degree of freedom of an atom or molecule, have led to new insights into the way in which matter can be manipulated that may have important consequences for chemistry, biology, material science, and quantum communications and computing. Moreover, they allow one to determine, for the regime of small excitation, a globally optimal classical driving force that produces the specific target state in a canonical bound quantum system.

If, however, quantum state engineering in matter is to have a significant impact, it is imperative to find a solution to the quantum control problem for large population transfer from the initial state to the target state. In this regime the problem of quantum control is highly nonlinear. Because of this, the regime of strong excitation has been explored only by iterative numerical techniques [6–8], sometimes beginning from an optimized weak-field solution.

In this paper we propose a general approach to the quantum control of matter that allows a driving force to be found simply, even for highly nonlinear excitation, for a target state that is a bound state of the system. The key to obtaining this solution is to simplify the problem by limiting the duration of the driving force to less than one characteristic period of the system (for an atomic electron Rydberg wave packet this would be the Kepler period, for example). The reason is that for times less than the characteristic period, the particle does not have the opportunity to reach the system's boundary, and acts essentially as a classical free particle. We will discuss the technique with respect to the hydrogen atom but it can be readily extended to a variety of systems consisting of a single ground state and a manifold of excited states. Many quantum systems have at least part of their structure of this type. Each energy level within the upper manifold is connected to the ground state by time-dependent interaction, but no direct interaction is allowed between different levels of the excited state manifold. In the case of a hydrogen atom driven by an external optical field, the ground and excited states are electronic states connected by a dipole transition.

We want to find the electric field that will create a target wave packet centered at some principal quantum number $\bar{n}$. The Hamiltonian describing the interaction of the atom with an external classical driving field $E(t)$ can be written as

$$\hat{H} = \hbar \sum_{m=2}^{\infty} \omega_m |m\rangle\langle m| + \sum_{m=2}^{\infty} d_m E(t) \left(|1\rangle\langle m| + |m\rangle\langle 1|\right), \tag{1}$$

where $|1\rangle$ and $|m\rangle$ are the ground and excited electronic states, respectively. The eigenfrequency of state $|1\rangle$ is defined to be zero while that of state $|m\rangle$ is given by $\omega_m = [1 - m^{-2}]\bar{n}^3\omega/2$. When $\bar{n} \gg 1$, $\omega \equiv e^2/\hbar a_0 \bar{n}^3$ corresponds to the Kepler frequency of the wave packet. For hydrogen, the electric dipole moments are given by $d_m \approx e a_0 / m^{3/2}$. The electric field is written as $E(t) = E_0[f(t)e^{i\omega_L t} + \text{c.c.}]$; here $|f(t)|$ is the dimensionless slowly varying pulse envelope and ω_L is the carrier frequency.

The state of the system at time t can be expanded in terms of the unperturbed eigenstates of the atom: $|\Psi\rangle = a(t)|1\rangle + \sum_{m=2}^{\infty} b_m(t) e^{i\omega_m t}|m\rangle$. Substituting this into Schrödinger's equation yields, in the rotating wave approximation, a set of coupled differential equations for the probability amplitudes:

$$\dot{b}_n = -i\Omega_n f^*(t) a(t) e^{i\delta_n t}, \tag{2}$$

$$\dot{a} = -i \sum_{m=2}^{\infty} \Omega_m f(t) b_m(t) e^{-i\delta_m t}, \tag{3}$$

where $\Omega_m = d_m E_0/\hbar$ is the Rabi frequency and $\delta_m = \omega_m - \omega_L$ the detuning for the transition. The carrier frequency is taken to be equal to $\omega_{\bar{n}}$.

Assuming that initially all the population is on the ground state, substitution of the formal solution of Eq. (2) into Eq. (3) yields

$$\dot{a} = -f(t) \int_{-\infty}^{t} ds\, f^*(s) a(s) \xi(t - s). \tag{4}$$

The electronic response function $\xi(t - s)$ consists of two parts: a contribution $\xi_r(t - s) \equiv \sum_{m=N}^{\infty} \Omega_m^2 e^{-i\delta_m(t-s)}$ from states near resonance with the optical field, and another $\xi_{nr}(t - s) \equiv \sum_{m=2}^{N} \Omega_m^2 e^{-i\delta_m(t-s)}$ from states significantly detuned from it. The separation into these contributions occurs at level $N \gg 2$ corresponding to the lowest electronic state occupied by the target wave packet. In any case, $\xi_{nr}(t - s)$ oscillates very rapidly compared to $f^*(s)a(s)$, and the integration over s will be negligible. On the other hand, the resonant contribution consists of a small δ-function-like "spike" centered at $s = t$, with quasiperiodic revivals at $s = t - jT$ (with $j = \pm 1, \pm 2, \ldots$, and $T \equiv 2\pi/\omega$), each of much lower amplitude and broader duration than that at $s = t$. Therefore, by restricting the exciting pulse duration to less than one Kepler period T we can write to a good approximation: $\xi_r(t - s) \approx (2\pi\Omega_{\bar{n}}^2 \eta/\omega)\, \delta(t - s)$. The dimensionless quantity η will be left to be determined later.

Equation (4) can then be solved for $a(t)$, yielding

$$a(t) \approx \exp\left[-(R/2) \int_0^t |f(s)|^2\, ds \right], \tag{5}$$

where $R \equiv (2\pi\Omega_{\bar{n}}^2/\omega)\eta$.

This equation describes the temporal evolution of the probability amplitude of the ground state, and it has been tested against full numerical integration of Eqs. (2) and (3). It is important to point out that in arriving at this result, no approximation was made regarding the strength of the field, except that it should not be so strong that the ground state is depleted significantly during the duration of $\xi_r(t)$, though it may do so over the duration of the control pulse itself. It indicates that no Rabi cycling of the population between the ground and excited states can occur during the first Kepler period. This is because transfer of population back from the bright state excited in the upper manifold to the ground state is suppressed by quantum interference. The short pulse duration implies that there is an uncertainty of the excitation frequency large enough that the discreteness of the manifold cannot be resolved, and the ground state simply depletes as if the manifold were a continuum. As a matter of fact, Eq. (5) is similar to the Fermi golden rule expression for ionization of the atom. In a wave packet picture, the localized state that is excited by the pulse does not complete an entire period of oscillation during the time that the driving field is nonzero, so that there is no possibility that quantum interference of possible transition amplitudes between ground and excited states can occur during the pulse.

In the weak-field regime, where depopulation of the ground state is small, the ground state's amplitude remains fairly constant. Then, any field which produces the correct spectral amplitudes and phases at the particular resonance frequencies ω_m is a solution to the control problem: the form of the field (or more precisely, of its Fourier transform) in between these resonances is irrelevant [9]. In the more general case of significant ground state population depletion, the driving field may be found from the *given* set of target amplitudes $b_n(\tau)$ for the desired wave packet at some target time τ, when the field is again zero.

Substitution of Eq. (5) back into Eq. (2) followed by its integration, allows one to solve for the field once $b_n(\tau)$ is specified. By requiring the field to be zero at the target time, we obtain

$$f(t) = (\Omega_{\bar{n}}/R) B^*(t) e^{G(t)}, \tag{6}$$

where $B(t) = i \sum_{m=N}^{\infty} \lambda_m b_m e^{-i\delta_m t}$, with $b_m = b_m(\tau)$ and $\lambda_m \equiv (\bar{n}/m)^{3/2}$. The function $G(t)$, defined as $G(t) \equiv (R/2) \int_0^t |f(s)|^2\, ds$, is related to the pulse energy up to time t. Taking its derivative with respect to time and using Eq. (6) yields

$$\dot{G}(t) = (\omega/4\pi\eta)\, |B(t)|^2 e^{2G(t)}. \tag{7}$$

Equation (7) can be easily integrated for $G(t)$, and substituting the result back into Eq. (6), we find the field that generates the target state in the excited manifold to be

$$f(t) = -i(\eta R_0)^{-1} \frac{B^*(t)}{[1 - (\omega/2\pi\eta) \int_0^t |B(s)|^2\, ds]^{1/2}}. \tag{8}$$

Here $R_0 \equiv 2\pi\Omega_{\bar{n}}/\omega$. The quantity η can now be evaluated by setting $\exp[-2G(\tau)] = 1 - \Delta$, where Δ is the target depletion of the ground state. It is straightforward to show that $\eta^{-1} = (2\pi\Delta)/[\omega \int_0^{\tau} |B(s)|^2\, ds]$. From Eq. (8), the product $E_0 f(t)$ can be evaluated, thus completely determining the driving field $E(t)$.

Equation (8) is the main result of this paper, and it represents a prescriptive solution for creating a particular Rydberg wave packet in hydrogenlike atoms. If the target wave packet is well localized, the function $B(t)$ consists of a series of impulses of decreasing amplitude that gradually broaden into one another. It is clear from the weak-field solution [obtained from Eq. (8) by setting the denominator equal to unity] that each of the isolated impulses contains a complete specification of the target wave packet. In the strong-field regime, only those impulses within a single Kepler period play a role since, once the target depletion of the ground state is reached, the control field is set to zero. Of course, this restricts the set of possible target wave packets to those for which $B(t)$ has this quasiperiodic structure. For example, a wave packet

with population uniformly distributed across states $|75\rangle$ to $|100\rangle$ with constant phase yields a time series that satisfies this criterion, but a state with half its population in state $|75\rangle$ and half in state $|100\rangle$ is not amenable to control using this method. This is because, in the latter case, the field contains structure of duration shorter than the "response" function.

The ability of this approximate driving field to generate the target quantum state in the upper manifold was tested on two prototypical localized states that exhibit both classical and quantum features. The first of these was a Rydberg quasi-coherent-state wave packet centered at $\bar{n} = 85$. Here, the target amplitudes were taken to be $b_{N+j}(\tau) = \{\Delta[(\bar{n} - N)^j \exp(N - \bar{n})/j!]\}^{1/2}$ with $j = 0, 1, \ldots, 25$, and $N = 75$; the other levels were assigned zero amplitudes. This corresponds to a Gaussian radial wave packet with spatial extent of approximately 10% of the Kepler period, at the outer turning point. The driving fields, determined using Eq. (8), are shown in Fig. 1a for both small (3%) and large (99%) ground state depletion. Note that the shapes of the field (although not the pulse energy) in the strong-field case is not radically different from that in the weak-field regime, and the differences make good physical sense. In the strong-field case, the dynamics are easily understood from the following argument. By the time the trailing edge of the pulse arrives at the system, the ground state population is smaller than initially, and there is consequently less absorption than at the leading edge of the pulse. Therefore, the pulse must be more intense at the trailing edge in order to be able to pump whatever population is left in the ground state to the upper manifold. Thus, aside from the practically important problem of developing analytic solutions to nonlinear control problems, these results provide some physical insight into a previous work by Krause *et al.* [6], where optimal control theory was used to generate a minimum uncertainty wave packet in the B state of an I_2 molecule starting in the ground X state. The field was designed in the weak-field regime and then scaled to higher intensities, depleting 100% of the population from the ground electronic state. They too found that only small changes in the pulse shape were required to obtain reasonable target achievements—there was no radical restructuring.

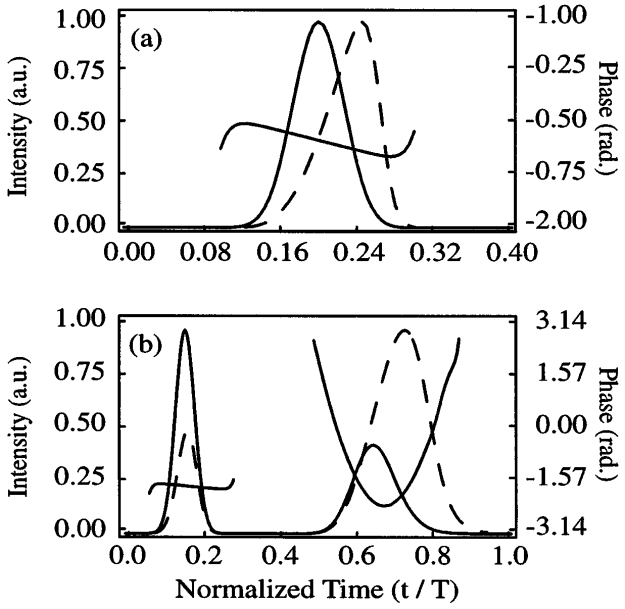

FIG. 1. The driving field that generates (a) a coherent state and (b) a "cat" state with 10% depletion (solid line) and 97% depletion (dashed line). The phase of the field is the same in both strong and weak cases. In (a), the intensity of the weak field corresponds to approximately 3×10^9 W/cm^2 and to 4×10^{11} W/cm^2 for the strong field.

The validity of these approximate solutions was tested by substituting the designed fields back into Schrödinger's equation [Eqs. (2) and (3)] and numerically integrating without any further approximations to find the final state. The continuum was not included in the simulations because the control field intensities were always less than 5×10^{11} W/cm^2 even in the regime of nearly total population transfer to the wave packet state. The actual and target populations and phases, for the strong-field case, are shown in Fig. 2a. In both weak- and strong-response regimes the analytic prediction compares favorably with the full numerical results, with less than 1% difference from the targeted population transfer from the ground state. To quantify the fidelity, we used a generalization of the achievement factor A, defined by [6] $A^2 = Tr(\hat{\rho}\hat{\rho}_T)/(Tr\hat{\rho}^2\ Tr\hat{\rho}_T^2)^{1/2}$, where $\hat{\rho}$ is the density operator associated with the final state in the upper manifold and $\hat{\rho}_T$ is that of the target state. Here, $A = 1$ when $\hat{\rho} = \hat{\rho}_T$, even for mixed states. In both weak- and strong-field domains we obtained an achievement of $A = 0.999 \pm 0.001$, indicating that the target state was obtained with extremely high fidelity in both cases.

Fields that generate other distributions with more complicated phase-space structure can also be designed. For our second test case, we used Eq. (8) to design a field that generates a Rydberg "cat" state in the upper manifold. This state corresponds to a coherent superposition of two classically distinguishable quasicoherent states of the same shape at the inner and outer turning points. Here, $b_{N+j}(\tau) = \{\Delta[(\bar{n} - N)^j \exp(N - \bar{n})/j!]\}^{1/2}[(-1)^j + 1]$, and, as before, $j = 0, 1, \ldots, 25$; $N = 75$ and $\bar{n} = 85$. The field that generates such distribution is shown in Fig. 1b for both weak and strong excitation regimes. As one would expect, two pulses are necessary to produce this state [10–13]. The second pulse arrives approximately a time $T/2$ after the first pulse and creates a second wave packet, of identical configuration space structure to the first. This second wave packet interferes with the first one, canceling the population in the even numbered levels. Again, the differences between pulse shapes in the small and large depletion cases can

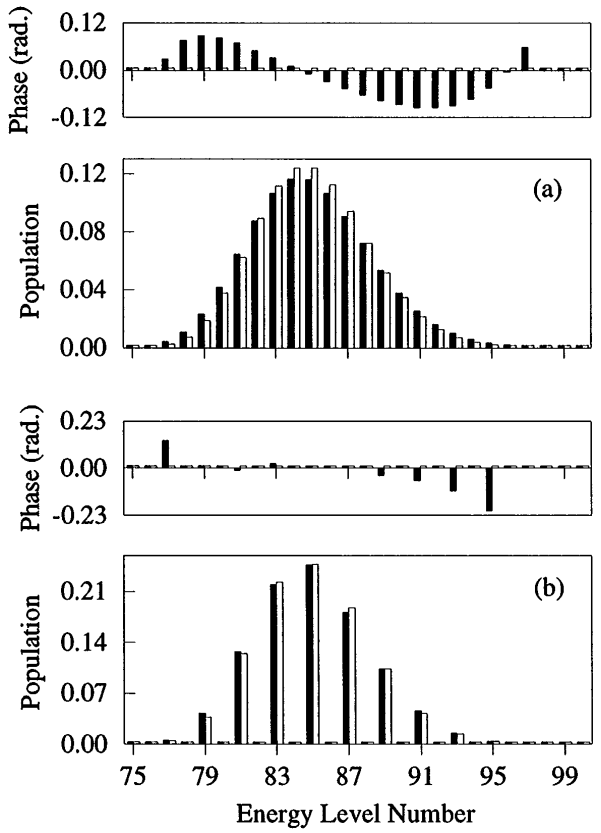

FIG. 2. Solutions to Schrödinger's equation using the field of Eq. (8) for a target wave packet that is (a) a quasicoherent state and (b) a "cat" state in the strong-field regime. Black and white columns correspond to actual and target distribution, respectively. The target phases are zero, and we show only the actual phases for levels for which $|b_n|^2$ is greater than 1% of the maximum target probability.

be explained by the smaller absorption seen by the second pulse, and the rapid depletion of the ground state during each pulse. Figure 2b shows the actual and target populations and phases, for the strong-field case. Here, $A = 0.999 \pm 0.001$ as well.

Thus we have shown that the idea of restricting the driving force to a short enough duration that the discreteness of the system's level structure is not operative leads to a great simplification of the nonlinear quantum control problem. In contrast to other approaches to quantum control such as optimal control theory (OCT), this restriction allows one to derive a simple, approximate analytic solution for the driving field, directly from the quantum state amplitudes in the upper manifold, even in the limit of large population transfer. The main approximation used in finding an analytic solution was that of a rapid electronic response function. This approximation was tested by substituting the driving field from Eq. (8) in Eqs. (2) and (3), which were then numerically integrated. As shown in Figs. 2a and 2b, the approximation seems to hold well even in the strong regime.

Even though more complex models of an atom, or of a molecule, may be more tractable under OCT, due to its greater flexibility, the driving field of Eq. (8) can be used as the initial guess for the control field required by that method. The output of OCT would then probably yield better achievements and, correspondingly, a field closer to the globally optimal solution.

L. d. A. acknowledges the financial support of CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico, Brazil). This work was supported in part by the NSF and the Army Research Office.

[1] J. I. Cirac and P. Zoller, Phys. Rev. Lett. **74**, 4091 (1995); L. K. Grover, Phys. Rev. Lett. **79**, 325 (1997).
[2] A. Bard *et al.,* J. Vac. Sci. Technol. B **15**, 1805 (1997); R. Younkin *et al.,* Appl. Phys. Lett. **71**, 1261 (1997).
[3] J. Somloi, V. A. Kazakov, and D. J. Tannor, Chem. Phys. **172**, 85 (1993).
[4] L. Shen, S. Shi, and H. Rabitz, J. Phys. Chem. **97**, 12 114 (1993).
[5] M. Shapiro and P. Brumer, J. Chem. Phys. **97**, 6259 (1992).
[6] J. L. Krause, M. Messina, Y. Yan, and K. R. Wilson, J. Phys. Chem. **99**, 13 736 (1995).
[7] K. G. Kim and M. D. Girardeau, Phys. Rev. A **52**, 891 (1995).
[8] J. Botina, N. Rahman, and H. Rabitz, J. Chem. Phys. **104**, 4031 (1996).
[9] M. Demiralp and H. Rabitz, Phys. Rev. A **47**, 809 (1993).
[10] M. W. Noel and C. R. Stroud, Jr., Phys. Rev. Lett. **77**, 1913 (1996).
[11] I. A. Walmsley and M. G. Raymer, Phys. Rev. A **52**, 681 (1995).
[12] J. Janszky, A. Vinogradov, T. Kobayashi, and Z. Kis, Phys. Rev. A **50**, 1777 (1994).
[13] N. Scherer *et al.,* J. Chem. Phys. **95**, 1487 (1991).