2001

Optimally shaped terahertz pulses for phase retrieval in a Rydberg-atom data register

Chitra Rangan
University of Windsor

P.H. Bucksbaum

PHYSICAL REVIEW A, VOLUME 64, 033417

Optimally shaped terahertz pulses for phase retrieval in a Rydberg-atom data register

C. Rangan and P. H. Bucksbaum
Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1120
(Received 30 March 2001; published 17 August 2001)

We employ optimal control theory to discover an efficient information-retrieval algorithm that can be performed on a Rydberg-atom data register using a shaped terahertz pulse. The register is a Rydberg wave packet with one consituent orbital phase reversed from the others (the ''marked bit''). The terahertz pulse that performs the decoding algorithm does so by driving electron probability density into the marked orbital. Its shape is calculated by modifying the target of an optimal-control problem so that it represents the direct product of all correct solutions to the algorithm.

DOI: 10.1103/PhysRevA.64.033417 PACS number(s): 32.80.Qk, 32.80.Rm, 03.67.−a, 42.30.Rx

I. INTRODUCTION

The interaction of terahertz-frequency electromagnetic pulses with Rydberg atoms has produced many insights into the dynamical properties of atomic systems [1]. The comparable time scales of terahertz pulses with those of Rydberg-state lifetimes make it possible to envision schemes of quantum control. In this paper, we propose a method of controlling Rydberg wave packets using shaped terahertz pulses, and theoretically show how these pulses can be designed to execute a quantum algorithm on a Rydberg-atom data register.

It has been shown that information can be stored in the phases of the constituent orbitals of a Rydberg wave packet [2,3]. Recently, a terahertz half-cycle pulse was used to decode the information stored in a Rydberg-atom data register [3]. This half-cycle pulse decodes the phase structure by retaining the population only in the orbital that was initially 180° out of phase with respect to the other orbitals, i.e., the marked bit. However, this guess (unshaped) pulse does not decode all marked bits of the register with the same efficiency. In this paper, we aim to find the shaped THz pulse that will optimally transfer most of the population to any marked bit of the quantum data register.

II. OPTIMAL CONTROL THEORY

To design the terahertz pulse, we use a method that has been used extensively in mathematical and engineering applications — optimal control theory (OCT) [4,5]. This theory has also been applied to the control of quantum systems [6–12] with some success in experimental implementation [13]. We use OCT to design a terahertz frequency pulse that can be used to achieve a desired target state. We then modify the OCT target state to make it possible to discover not a single target, but an optimized quantum algorithm. The wave function of the Rydberg electron is the state variable and the electric field of the terahertz pulse is the control parameter. A functional J is defined, whose extremum must be calculated. The functional consists of two parts, representing the desired target and the cost. Our aim is to maximize the fraction of the electron probability density in a target orbital $|a_k\rangle$ at a time T (after the end of the terahertz pulse). That is, the target functional $\langle P_k(T)\rangle=\langle\psi(T)|a_k\rangle\langle a_k|\psi(T)\rangle$ must be a maximum. The cost functional represents the constraint on the control parameter, the terahertz field $E(t)$. The integrated energy of the pulse must be kept low, therefore the cost functional is defined as $Y(T)=\int_0^T dt\,\ell(t)|E(t)|^2$. Here ℓ is a penalty parameter, in general time dependent, that controls the cost functional and hence the peak terahertz field. The functional J written as

$$J=\langle\psi(T)|P_k|\psi(T)\rangle-Y(T) \tag{1}$$

must be maximized.

The aim is to find an optimal control function $E(t)$ that maximizes J. The wave-packet evolution is governed by the Schrödinger equation. In atomic units, $e=m_e=\hbar=1$,

$$|\dot{\psi}(t)\rangle=-iH(t)|\psi(t)\rangle, \tag{2}$$

where $H(t)=H_0+E(t)z$. The equation of motion acts as a constraint on the evolution of the state and, in a manner similar to that used in variational calculus, we introduce a Lagrange multiplier $|\lambda(t)\rangle$ [14]. The unconstrained functional that must be optimized is written as

$$\overline{J}=J-\int_0^T dt[\langle\lambda(t)|\dot{\psi}(t)\rangle+\langle\lambda(t)|iH|\psi(t)\rangle+\langle\psi(t)|\lambda(t)\rangle - i\langle\dot{\psi}(t)|H|\lambda(t)\rangle] \tag{3}$$

$$=\langle\psi(T)|P_k|\psi(T)\rangle-\int_0^T dt\,\ell(t)|E(t)|^2 - 2\,\mathrm{Re}\langle\lambda(t)|\psi(t)\rangle|_0^T + \int_0^T dt\,2\,\mathrm{Re}\langle\lambda(t)|\psi(t)\rangle - \int_0^T dt\,2\,\mathrm{Re}\langle\lambda(t)|iH|\psi(t)\rangle. \tag{4}$$

Several iterative techniques for determining the optimal solution have been developed [15–19]. Following the scheme for an iterative solution proposed in Ref. [19], the functional $\overline{J}$ is written as the sum of a terminal part and an integral

$$\overline{J}=G+\int_0^T dt\, R, \tag{5}$$

where

$$G=\langle\psi(T)|P_k|\psi(T)\rangle-2\mathrm{Re}\langle\lambda(t)|\psi(t)\rangle|_0^T, \tag{6}$$

$$R=-\ell(t)|E(t)|^2+2\mathrm{Re}[\langle\dot{\lambda}(t)|\psi(t)\rangle-i\langle\lambda(t)|H|\psi(t)\rangle]. \tag{7}$$

The maximum of both G and R is sufficient to ensure the maximum of $\overline{J}$. Our objective is to iteratively determine the optimal function $E(t)$ that maximizes $\overline{J}$. The functional $\overline{J}$ at the kth and $(k+1)$th iteration is defined by the changes in the wave function and the field. That is,

$$|\psi(t)^{k+1}\rangle\equiv|\psi(t)^k\rangle+|\Delta\psi(t)\rangle,$$

$$E^{k+1}\equiv E^k+\Delta E. \tag{8}$$

The difference in $\overline{J}$ between two successive iterations is written as

$$\overline{J}^{k+1}-\overline{J}^k=\Delta_1+\Delta_2+\Delta_3, \tag{9}$$

where

$$\Delta_1\equiv G(\psi^{k+1}(T))-G(\psi^k(T))=2\mathrm{Re}[\langle\psi^k(T)|P_k|\Delta\psi(T)\rangle-\langle\lambda(T)|\Delta\psi(T)\rangle]+\langle\Delta\psi(T)|P_k|\Delta\psi(T)\rangle, \tag{10}$$

$$\begin{aligned}\Delta_2&\equiv\int_0^T dt[R(t,\psi^{k+1},E^{k+1})-R(t,\psi^{k+1},E^k)]\\&=-2\mathrm{Re}\int_0^T dt\,\ell(t)E^{*k}(t)\Delta E(t)\\&\quad-\int_0^T dt\,\ell(t)|\Delta E(t)|^2\\&\quad-2\mathrm{Re}\int_0^T dt[i\langle\lambda^k(t)|z\Delta E(t)|\psi^k(t)\rangle\\&\quad+i\langle\lambda^k(t)|z\Delta E(t)|\Delta\psi(t)\rangle],\end{aligned} \tag{11}$$

$$\begin{aligned}\Delta_3&\equiv\int_0^T dt[R(t,\psi^{k+1},E^k)-R(t,\psi^k,E^k)]\\&=2\mathrm{Re}\left[\int_0^T dt\langle\lambda(t)|\Delta\psi(t)\rangle-i\int_0^T dt\langle\lambda(t)|H^k|\Delta\psi(t)\rangle\right].\end{aligned} \tag{12}$$

Choosing

$$|\lambda(T)\rangle=P_k|\psi^k(T)\rangle \tag{13}$$

and

$$|\dot{\lambda}(t)\rangle=-iH^k|\lambda(t)\rangle, \tag{14}$$

we find

$$\Delta_1=\langle\Delta\psi(T)|P_k|\Delta\psi(T)\rangle, \tag{15}$$

$$\Delta_2=-2\mathrm{Re}\int_0^T dt\left[\ell(t)E^{*k}(t)\Delta E(t)+\frac{1}{2}\ell(t)|\Delta E(t)|^2+i\langle\lambda^k(t)|z\Delta E(t)|\psi^k(t)\rangle+i\langle\lambda^k(t)|z\Delta E(t)|\Delta\psi(t)\rangle\right], \tag{16}$$

$$\Delta_3=0. \tag{17}$$

The solution improves when $\overline{J}$ increases or stays the same at each iteration. P_k is a positive semidefinite operator; therefore, Δ_1 is greater than or equal to zero. The change in the control parameter ΔE is chosen to maximize Δ_2. The expression for Δ_2 suggests the following change in the field at the (k+1)th iteration [19]:

$$\Delta E(t)=\frac{-i}{\ell(t)}\langle\lambda^k(t)|z|\psi^{k+1}(t)\rangle. \tag{18}$$

The appearance of $|\psi^{k+1}(t)\rangle$ in the above expression implies that the overlap of $|\psi(t)\rangle$ and $|\lambda(t)\rangle$ is fed back immediately to find the field at the next time step.

The optimal control algorithm consists of the following steps.

(1) Starting from the initial wave packet $|\psi^{(0)}(0)\rangle=|\psi(0)\rangle$ and a first guess for the terahertz field $E^{(0)}(t)$, the wave packet is propagated according to Eq. (2) to find $|\psi^{(0)}(T)\rangle$.

(2) Using Eq. (13) to find $|\lambda(T)\rangle$, Eq. (14) is iterated backward to time $t=0$ and $|\lambda(t)\rangle$ is found at every time step.

(3) With $|\psi^1(0)\rangle=|\psi(0)\rangle$, Eq. (18) is then used to find a new value of the control field $E^1(t)$ and Eq. (2) is used to propagate the wave-packet forward in time.

The second two steps are repeated until the target yield converges to within the desired accuracy.

III. OPTIMAL PULSE FOR A SINGLE TARGET STATE

The first step in our approach is to find the optimal terahertz field required to decode a single flipped state. The initial state of the Rydberg data register is a wave packet made of the $24p$ through $29p$ orbitals of equal amplitudes and the phase of the $26p$ (the marked bit) orbital opposite to that of the others. The initial guess terahertz pulse is a half-cycle pulse [20] of pulse width 1 ps. The desired initial phase structure occurs at the peak of the half-cycle pulse (i.e., at 0.5 ps). We find the terahertz pulse that will optimally transfer most of the population to the marked bit. The best value of the penalty parameter ℓ that controls the peak field of the terahertz pulse to a reasonable value, and at the same time produces the desired final state, was found to be roughly

10^{10}. One feature of several optimal fields obtained theoretically [7,11] is that the fields do not go to zero smoothly at the times $t=0$ and $t=T$. In the present system, the correct evolution of the wave packet depends very sensitively on the fields at the end points. The addition of a smooth switch on and switch off of the calculated optimal field drastically changes the evolution of the wave packet. Therefore, the condition that the terahertz field continuously goes to zero before and after the time interval of choice must be built into the algorithm. To ensure that the terahertz field goes smoothly to zero at times $t=0$ and $t=T$, the penalty parameter $\ell(t)$ is made a smoothly varying time-dependent function. The penalty on the pulse fluence is a thousand times more at the end points than at the rest of the pulse duration. The smoothness of the penalty function ensures a smooth switching on and switching off of the terahertz pulse.

This OCT implementation is very successful in describing the terahertz control of a Rydberg wave packet. The method takes macrosteps in the control field at every iteration and convergence is swift. The computational complexity is of the same order as that of the wave-packet propagation. Therefore, we use a split-operator method in a restricted basis of essential states [21,3]. The energy eigenstates of cesium are calculated using a pseudopotential method on a nonlinear radial grid [22]. A time step of 10 fs ensures the accuracy of the propagator, which is correct to the second order in the time step. The numerical implementation of the local iterative algorithm is extremely sensitive to numerical error, and Δ_3 must be maintained equal to zero to very high precision [19]. The unitary nature of the symmetrized-product propagator maintains this condition. The restricted basis consists of 195 energy eigenstates with principal quantum number n ranging between 21 and 31, and angular momentum quantum number $\ell<17$. Absorbers ensure that population does not get reflected from the $n=21$, $n=31$, and $\ell=16$ "boundaries." Using the selected state basis also imposes the condition that the spectrum of the terahertz pulse is decided by the energy range of the selected state basis. In this implementation of optimal control theory, we have chosen a fixed pulse length T of roughly 8 ps. In other formalisms, this time T may also be varied as a parameter.

The terahertz field that optimizes the population in the marked 26p state is shown in Fig. 1(a). The initial population in the 26p state is 16.7%. The optimal pulse will decode the information stored as phase by transferring most of the population into the 26p state. With the initial guess pulse, the population is 29.5%. After 50 iterations, the target yield is increased to 52.8%. The spectrum and Husimi distribution [23] of this optimal pulse are shown in Fig. 1(b) and Fig. 1(c), respectively. Notably, the strong peaks in the spectrum and the Husimi distribution do not correspond to any resonance between the energy levels of the selected state basis. The optimal terahertz pulse does not drive the system to any particular resonant condition. Instead, it alters the phases of the constituent orbitals of the wave packet so that they interfere to produce the desired probability distribution.

Figure 2 shows the evolution of the wave packet as a function of time while the optimal pulse is on. During the terahertz pulse, probability can leak into other states not in the register (the other states in the essential basis). At the end of the pulse, a large fraction of the electron probability density lies in the flipped orbital (marked bit) of the data register. This can be thought of as using the other states of the data register as working qubits, which are used during the computation, but are not measured for any useful retrieval of information.

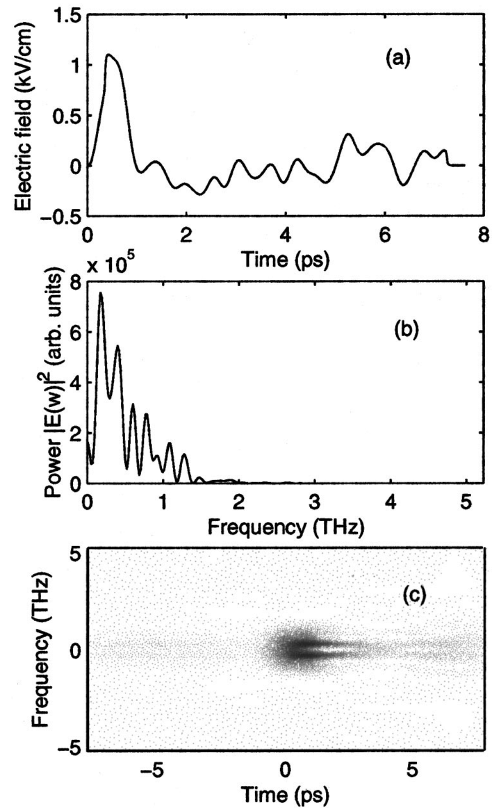

FIG. 1. Optimal terahertz pulse that maximizes the population in the marked 26p orbital. (a) Electric field of the pulse as a function of time shows a strong peak around 0.5 ps when the marked phase structure occurs. (b) The Fourier transform of the pulse does not indicate any atomic resonance. (c) The Husimi distribution of the terahertz pulse.

One interesting feature of this optimal pulse is that the peak field of roughly 1 kV/cm lasts for roughly 0.5 ps. For a $\bar{n}=26$ wave packet, this field, which is beyond the field-ionization limit, lasts for more than half the Kepler period ($\sim 2\pi n^3$). Yet, 99% of the population remains in the selected state basis. This feature is an example of interferometric stabilization [24], seen in other atomic systems.

IV. OPTIMAL PULSE FOR A QUANTUM ALGORITHM

This terahertz pulse is optimal only for decoding the flipped 26p orbital. That is, if the phase of a different state were flipped, this pulse will not decode it. We wish to design a universal terahertz pulse that will optimally decode any flipped orbital of the wave-packet register. Therefore, we redefine the optimal-control problem by considering an initial state that is a product state of independent wave packets with singly flipped orbitals,

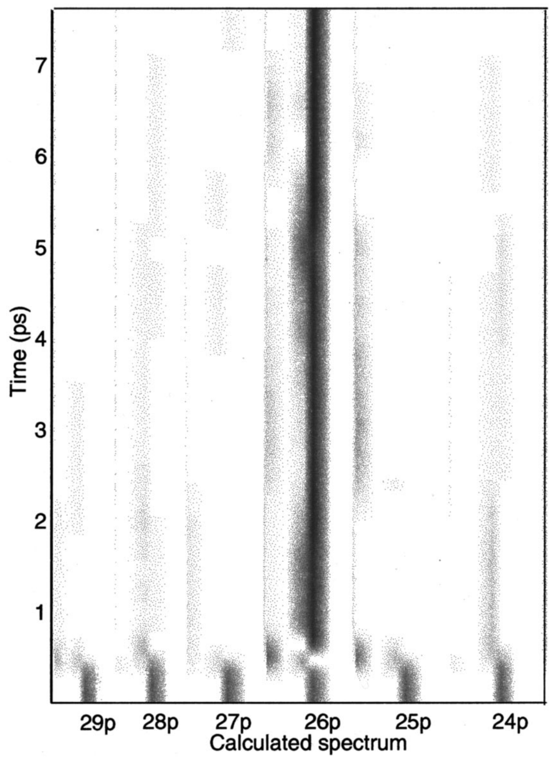

FIG. 2. Calculated spectrum of the atomic wave packet as a function of time while under the influence of the optimal terahertz pulse. The wave packet is redistributed such that a significant portion of the population lies in the marked orbital at the end of the pulse.

$$|\Psi(0)\rangle = |\psi_{25p}^{(1)}(0)\rangle \otimes \psi_{26p}^{(2)}(0)\rangle \otimes \psi_{27p}^{(3)}(0)\rangle \otimes \cdots . \quad (19)$$

The terahertz pulse acts simultaneously but independently on all these wave packets. The desired final state is also a product state of independent wave packets with the flipped bit correctly decoded,

$$|\Psi(T)\rangle = |25p^{(1)}\rangle \otimes |26p^{(2)}\rangle \otimes |27p^{(3)}\rangle \otimes \cdots . \quad (20)$$

The counterparts of Eqs. (13) and (14) are straightforward. At every time step, the updated terahertz field is found by using a modified version of Eq. (18), with the matrix element of z replaced by a sum of matrix elements of z, one from each independent "subspace,"

$$\Delta E(t) = \frac{-1}{\mathscr{E}(t)} \sum_{i=1}^{N} \langle \lambda_{(i)}^{k}(t) | iz | \psi_{(i)}^{k+1}(t) \rangle . \quad (21)$$

Using this method, we find the terahertz pulse that detects any flipped orbital of the N-bit data register. The advantage of this refinement is that the computational resources needed increase only by a factor of the number of constituent states in the wave-packet register.

We now find the optimal terahertz pulse that will decode any flipped state in a six-state Rydberg data register. The register consists of np states of cesium, with n from 24 to 29.

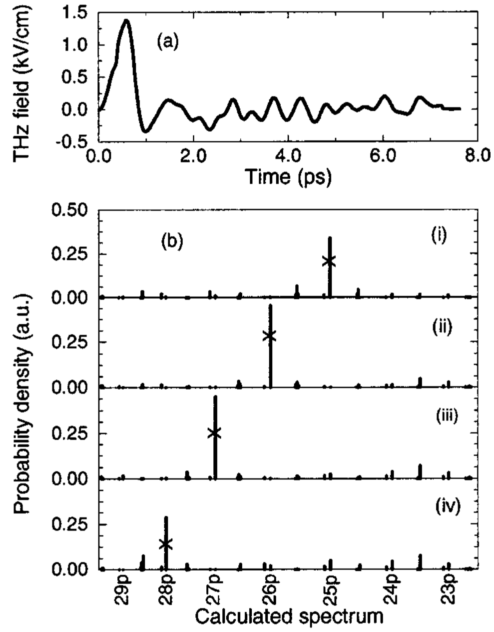

FIG. 3. The terahertz pulse designed to optimally retrieve any marked orbital of the Rydberg-atom data register. (a) The electric field as a function of time. (b) Calculated spectra of the Rydberg wave packet after the terahertz pulse for initial states with different marked orbitals (i) $25p$, (ii) $26p$, (iii) $27p$, and (iv) $28p$. The crosses indicate the population of the marked orbital when the guess HCP is used to retrieve the phase information.

Population in the flipped orbital is amplified by the diffusion of probability density from the adjacent states. This is an example of the implementation of Grover's search algorithm, where information is stored in states with differing phases, and a marked bit is amplified by "quantum diffusion" [25]. The outer states $n=24$ and $n=29$ are therefore not included in the optimization. The universal decoding pulse and its effect on a wave packet with different marked bits is shown in Fig. 3. After the pulse, the wave-packet population is distributed so that the flipped state is clearly amplified.

V. CONCLUSIONS

In conclusion, we have designed a terahertz pulse to implement a search algorithm on a quantum data register. Phase information stored in a Rydberg wave packet was optimally retrieved through the interaction with the pulse. Careful attention was paid to the smooth switch on and switch off of the terahertz pulse. We also show that it is possible to design an optimal pulse that can achieve not only a desired target state of an atom, but also implement a desired algorithm.

To our knowledge, this is the first time that optimal control theory has been applied to the terahertz control of a quantum system. This theoretical study motivates the experimental design and control of broadband terahertz-frequency pulses. Beyond quantum control, these results point to the possibilities of using Rydberg atoms as quantum computers and terahertz pulses to implement quantum algorithms.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge discussions with and help from Professor D. J. Tannor, Professor A. M. Bloch, Professor V. F. Krotov, and Dr. V. Malinovsky. C.R. acknowledges support from the Center for Ultrafast Optical Science at the University of Michigan. This research was supported by the National Science Foundation through Grant No. 9987916.

[1] R.R. Jones, D. You, and P.H. Bucksbaum, Phys. Rev. Lett. **70**, 1236 (1993); G.M. Lankhuijzen and L.D. Noordam, *ibid.* **74**, 355 (1995); C. Raman, C.W.S. Conover, C.I. Sukenik, and P.H. Bucksbaum, *ibid.* **76**, 2436 (1996); R.R. Jones, *ibid.* **76**, 3927 (1996); M.T. Frey, F.B. Dunning, C.O. Reinhold, S. Yoshida, and J. Burgdörfer, Phys. Rev. A **59**, 1434 (1999).

[2] J. Ahn, T.C. Weinacht, and P.H. Bucksbaum, Science **287**, 463 (2000).

[3] J. Ahn, D.N. Hutchinson, C. Rangan, and P.H. Bucksbaum, Phys. Rev. Lett. **86**, 1179 (2001).

[4] V.F. Krotov, *Global Methods in Optimal Control Theory*, Pure and Applied Mathematics Vol. 195 (Monographs and Textbooks, New York, 1996).

[5] A.M. Bloch, J. Baillieul, P. Crouch, and J. Marsden (unpublished).

[6] A.P. Pierce, M.A. Dahleh, and H. Rabitz, Phys. Rev. A **37**, 4950 (1988).

[7] S. Shi and H. Rabitz, J. Chem. Phys. **92**, 364 (1990).

[8] R. Kosloff, S.A. Rice, P. Gaspard, S. Tersigni, and D.J. Tannor, Chem. Phys. **139**, 201 (1989).

[9] W. Jacubetz, J. Manz, and H.-J. Schreier, Chem. Phys. Lett. **165**, 100 (1990).

[10] J. Somloi, V.A. Kazakov, and D.J. Tannor, Phys. Rev. A **60**, 3081 (1999).

[11] K.G. Kim and M.D. Girardeau, Phys. Rev. A **52**, R891 (1995).

[12] Y.J. Yang, J. Che, and J.L. Krause, Chem. Phys. **217**, 297 (1997).

[13] R.S. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992); M. Shapiro and P. Brumer, Chem. Phys. Lett. **208**, 193 (1993).

[14] E. Gerjuoy, A.R.P. Rau, and L. Spruch, Rev. Mod. Phys. **55**, 725 (1983).

[15] S. Shi and H. Rabitz, Comput. Phys. Commun. **63**, 71 (1991).

[16] V.F. Krotov and I.N. Fel'dman, Eng. Cybernetics **21**, 123 (1983).

[17] V.A. Kazakov and V.F. Krotov, Autom. Remote Control (Engl. Transl.) **5**, 430 (1987).

[18] W. Zhu, J. Botina, and H. Rabitz, J. Phys. Chem. **108**, 1953 (1998).

[19] D.J. Tannor, V.A. Kazakov, and V. Orlov, in *Time Dependent Quantum Molecular Dynamics*, edited by Broeckhove and Lathouwers (Plenum, New York, 1992), p. 347.

[20] A. Bugacov, B. Piraux, M. Pont, and R. Shakeshaft, Phys. Rev. A **51**, 4877 (1995).

[21] M.D. Feit, J. Comput. Phys. **47**, 412 (1982); M.R. Hermann and J.R. Fleck, Phys. Rev. A **38**, 6000 (1988).

[22] J. N. Bardsley, in *Case Studies in Atomic Physics*, edited by E. W. McDaniel and M.R.C. McDowell (North-Holland, Amsterdam, 1974), Vol. 4, p. 302; C. Rangan, K.J. Schafer, and A.R.P. Rau, Phys. Rev. A **61**, 053410 (2000).

[23] H.-W. Lee, Phys. Rep. **259**, 147 (1995).

[24] M.V. Fedorov and A.M. Movesian, J. Phys. B **21**, L155 (1988); M.V. Fedorov and O.V. Tikhonova, Phys. Rev. A **58**, 1322 (1998); O.V. Tikhonova, E.A. Volkova, A.M. Popov, and F.M. Fedorov, *ibid.* **60**, R749 (1999).

[25] L.K. Grover, Phys. Rev. Lett. **79**, 325 (1997); Science **280**, 228 (1998).