Quantum control of multilevel atoms with rotational degeneracy using short laser pulses

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We study the quantum control of multilevel atoms with rotationally degenerate levels using short laser pulses. Various control schemes are considered, ones using π pulses, frequency-chirped pulses, two consecutive pulses, or two pulses that overlap each other partially. We study the possibilities of controlling the quantum state of an ensemble of atoms distributed randomly over one or more rotationally degenerate levels initially. For the sake of concreteness we use the hyperfine level scheme of the ^{85}Rb D line, but the results can easily be generalized for any of the alkali-metal atoms used in cooling and trapping experiments. We find that even though a number of difficulties arise, such as unequal coupling constants between rotational sublevels or dephasing between different hyperfine levels during the interaction, control schemes using simple or multiphoton adiabatic passage can be used to control the internal states of the atoms effectively as well as the center-of-mass motion. Furthermore, it is shown that in some cases it is possible to exploit the inequality of the coupling constants to entangle the rotational substates with specific distinct translational quantum states and hence separate these substates in momentum space.

I. INTRODUCTION

The quantum control of the states of atomic or molecular systems using laser pulses is a diverse and fertile field of study with numerous applications. In particular, adiabatic passage in various forms is a versatile and useful tool that has been studied extensively [1–4] with applications in selective population transfer in atoms [5], molecules [6], in Rydberg atoms [7], or for preparing atoms in various coherent superpositions of quantum states [8], or their coherent acceleration [9,10].

Most schemes of quantum control are applicable when the atomic or molecular system can be reduced to a few (two, three, four) well-defined working levels with individually controllable couplings between them. This is, of course, a drastic simplification of real physical systems—atoms and molecules most often have rotationally degenerate quantum states. Furthermore, the electronic levels often come in almost degenerate groups (such as the hyperfine levels of atomic states) that are so close to each other that one cannot simply consider a single one of these levels when trying to apply quantum control schemes. A simplified system with a "few" quantum states is usually the combined result of specific narrow-band laser frequencies, selection rules in conjunction with laser polarization and often also the careful preparation of the initial state. Thus the application of a specific quantum control scheme designed with a few working levels in mind may be problematic in a situation where the atomic states we start out with cannot be controlled precisely. For this reason, control schemes which are "robust" with respect to initial conditions of the atomic or molecular state are of great interest. "Robust" in this sense can have two meanings. Either the outcome of the control scheme is not sensitive at all with respect to the initial atomic state within certain bounds [the effect of the interaction is the same for a set of possible initial states (e.g., rotationally degenerate substates of

a certain level)] or, to the contrary, the interaction is perfectly selective (i.e., it is capable of "picking out" the atoms with suitable initial conditions and leaving all others more or less unchanged). Either one of these two extremes can be very useful. Quantum control schemes that involve adiabatic population transfer have been studied extensively precisely because they are known to be robust with respect to the change of interaction parameters such as laser intensities or detunings. The two most prominent examples of these is stimulated Raman adiabatic passage [1] (STIRAP) and adiabatic passage (AP) using frequency-chirped pulses [11]. As it turns out, this kind of robustness can be very useful also when dealing with rotationally degenerate states of atoms.

In this paper we study various schemes of quantum control that are usually discussed for the manipulation of atoms with a few working levels, and consider their application to atoms with a more complicated level scheme. In particular, we study the effect of rotational degeneracy of the working levels, in a setting where the atoms cannot be assumed to be in just one of these sublevels. As a case study we use the explicit level scheme of the D lines of ^{85}Rb which is often used in cooling and trapping experiments. The full hyperfine structure of the transitions and full rotational degeneracy of each hyperfine level is thereby taken into account. The results we derive, however, are quite generic, our considerations can easily be carried over to any of the alkali-metal atoms and also to atoms with a more complicated level scheme.

We will mostly study schemes that use AP by frequency-chirped laser pulses and compare them with π-pulse schemes. We show that several schemes that implement quantum control via AP may work also if we assume a random initial condition over rotational substates. We discuss the necessary conditions as well as the advantages of AP in this situation. We investigate cases when pulse bandwidth cannot be assumed to select a single hyperfine level to target and also the case of overlapping pulses that induce multiphoton adiabatic passage [12–14]. We will not discuss STIRAP, as this scheme has been studied at great length also for degenerate or nearly degenerate states

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[15,16]. We will consider a situation, however, when the inequality of the coupling constants between rotational substates can be turned to an advantage and used to entangle rotational substates with distinct translational quantum states of the atom.

II. FORMAL DESCRIPTION

A. Internal atomic state

Figure 1 depicts the structure of the relevant levels of ^{85}Rb atoms. The equations that describe the coherent interaction between the atoms and laser fields resonant with the $5^2S_{1/2} \rightarrow 5^2P_{1/2}$ transition (D_1 line) or the $5^2S_{1/2} \rightarrow 5^2P_{3/2}$ transition (D_2 line) may be obtained from the usual description of the Schrödinger equation using the Hamiltonian

$$\hat{H} = \hat{H}_A + \hat{H}_{AF}, \quad (1)$$

where the atomic Hamiltonian $\hat{H}_A$ may be written, in a frame rotating with the resonance frequency ω_0, as

$$\hat{H}_A = \sum_{F,m} \hbar \delta_F |F,m\rangle\langle F,m| + \sum_{F',m'} \hbar \delta_{F'} |F',m'\rangle\langle F',m'|. \quad (2)$$

Here the states $|F,m\rangle$ denote the $5^2S_{1/2}$ ground-state hyperfine levels, with their rotational substates. The energies relative to the line center are denoted by $\hbar \delta_F$ and are defined by the relations $\sum_F \delta_F(2F+1) = 0$ and $\delta_3 - \delta_2 = 2\pi \times 3035.7$ MHz [17]. Similarly, the states $|F',m'\rangle$ denote the $5^2P_{1/2}$ excited-
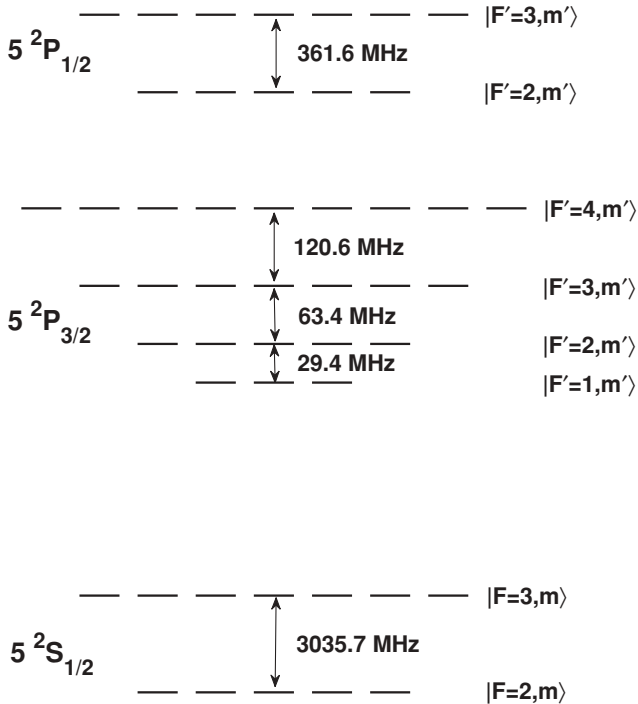
state hyperfine levels (D_1 line, $F' \in \{2,3\}$) or the $5^2P_{3/2}$ excited-state hyperfine levels (D_2 line, $F' \in \{1,2,3,4\}$) and their rotational substates. The energies $\hbar \delta_{F'}$ are defined by the relations $\sum_{F'} \delta_{F'}(2F'+1) = 0$ together with $\delta_{3'} - \delta_{2'} = 2\pi \times 361.6$ MHz (D_1 line) or $\delta_{4'} - \delta_{3'} = 2\pi \times 120.6$ MHz, $\delta_{3'} - \delta_{2'} = 2\pi \times 63.4$ MHz, and $\delta_{2'} - \delta_{1'} = 2\pi \times 29.4$ MHz (D_2 line).

The interaction with a classical laser field **E** is considered in the dipole approximation. Decomposing the interaction Hamiltonian $\hat{H}_{AF}$ in spherical components

$$\hat{H}_{AF} = -\hat{\mathbf{d}}\mathbf{E} = -\sum_q (-1)^q \hat{d}_q E_{-q}, \quad (3)$$

using the frame rotating with ω_0, and dropping terms counter-rotating with approximately twice this frequency, we write the interaction Hamiltonian as [18]

$$\hat{H}_{AF} = \frac{\hbar}{2} \sum_{F,m;F',m'} (\Omega_{F,m;F',m'} |F',m'\rangle\langle F,m| + \text{H.c.}). \quad (4)$$

The Rabi frequency for the $|F,m\rangle \rightarrow |F',m'\rangle$ transition is given by

$$\Omega_{F,m;F',m'} = \mathcal{D}_{F,m;F',m'} \Omega_{m-m'} \quad (5)$$

with the $q = m - m'$, $q \in \{-1,0,1\}$ spherical component of the vector Rabi frequency defined as

$$\Omega_q = -\frac{2}{\hbar} \langle J \| \hat{d} \| J' \rangle E_q^{(+)}. \quad (6)$$

Here $E_q^{(+)}$ is the qth component of the electric field's positive-frequency part and $\langle J \| \hat{d} \| J' \rangle$ is the reduced matrix element for the D_1 or D_2 line. The "geometrical" coefficients $\mathcal{D}_{F,m;F',m'}$ can be expressed via Clebsch-Gordan coefficients and Wigner $6j$ symbols as

$$\mathcal{D}_{F,m;F',m'} = (-1)^{F'+J+1+I} \sqrt{(2F'+1)(2J+1)}$$
$$\times \langle Fm|F'm';1q\rangle \begin{Bmatrix} J' & J & 1 \\ F & F' & I \end{Bmatrix}, \quad (7)$$

where $I = 5/2$ is the nuclear spin of ^{85}Rb while $J = 1/2$, and $J' = 1/2$ or $J' = 3/2$ for the D_1 or the D_2 line, respectively. The $\mathcal{D}_{F,m;F',m'}$ coefficients for ^{85}Rb have been tabulated in [17]. Writing the state of the atom as

$$|\psi\rangle = \sum_{F,m} \alpha_{F,m} |F,m\rangle + \sum_{F',m'} \beta_{F',m'} |F',m'\rangle \quad (8)$$

with the ground- and excited-state probability amplitudes $\alpha_{F,m}$ and $\beta_{F',m'}$, the Schrödinger equation yields

$$i\partial_t \alpha_{F,m} = \delta_F \alpha_{F,m} + \frac{1}{2} \sum_{F',q} \mathcal{D}_{F,m;F',m-q} \Omega_q^* \beta_{F',m-q},$$
$$\qquad\qquad\qquad\qquad\qquad (9)$$
$$i\partial_t \beta_{F',m'} = \delta_{F'} \beta_{F',m'} + \frac{1}{2} \sum_{F,q} \mathcal{D}_{F,m'+q;F',m'} \Omega_q \alpha_{F,m'+q}.$$

Note that we have used $q = m - m'$ to rewrite the sum over rotational substates and that the σ^+ (σ^-) transition from $|F,m\rangle$ to $|F',m+1\rangle$ ($|F',m-1\rangle$) belongs to the spherical index $q = -1$ ($q = 1$). This form of the equation is still very general, any constant detuning relative to the line center ω_0, or any

FIG. 1. The level scheme of ^{85}Rb showing the two hyperfine levels of the $5^2S_{1/2}$ ground state, the four hyperfine levels of the $5^2P_{3/2}$ state (D_2 line) with the frequency splittings between them, and the two hyperfine levels of the $5^2P_{1/2}$ state (D_1 line) with their frequency splittings (not drawn to scale). The rotational degeneracy $2F+1$ of each hyperfine level is indicated schematically by the number of lines representing the level.

slowly varying frequency modulation can be inserted as a time varying phase of the complex envelope functions $\Omega_q(t)$. If there are two or more fields interacting with the atoms at the same time, the envelope function is a sum for all fields. The response of the atom to any combination of pulsed fields may thus be calculated as long as the interaction time is short enough for spontaneous emission to be neglected and the knowledge of the internal state of the atom is sufficient.

The question how a statistical ensemble of atoms would evolve during the interaction with some predefined set of pulses can be investigated conveniently by solving Eqs. (9) numerically for an orthogonal set of relevant basis states (e.g., the $\{|F,m\rangle\}$ set) and then using the set of time-dependent vectors to construct the unitary time-evolution operator $\hat{U}(t,t_0)$ for which

$$|\psi(t)\rangle = \hat{U}(t,t_0)|\psi_0\rangle. \tag{10}$$

Using the operator $\hat{U}(t,t_0)$ it is then simple and fast to obtain the time evolution of the atomic state for any initial condition or ensemble of initial conditions.

B. Center-of-mass motion, multiphoton transitions

Often the knowledge of internal atomic dynamics is not enough [e.g., when the center-of-mass (c.m.) motion is also to be controlled through photon absorption and emission]. Sometimes there are several running light fields present at once (e.g., a standing wave or a pair of counterpropagating pulsed fields $\Omega_q(t) = \Omega_q^+(t) + \Omega_q^-(t)$ with the upper $\pm$ index showing the direction of propagation) and it is important to know the direction of momentum transfer that accompanies the change of internal state. Using Eqs. (9) to determine the internal dynamics is insufficient for this. One way to proceed is to generalize the description by including the center-of-mass coordinates of the atom in the description and derive equations for the momentum-space probability amplitudes $\alpha_{F,m}(p,t), \beta_{F',m'}(p,t)$. While this description requires considerably longer computational time, it gives us all necessary information and can also be used to study multiphoton transitions where N photons are exchanged between the two counterpropagating pulses. Alternatively, when there are two laser fields of constant-frequency difference present (or at least very slowly varying frequency difference), one may use the Floquet formalism [3,19] to obtain the same information—at a comparable cost of computational complexity.

A reasonable compromise is to consider $\hat{H}_{AF}$ to depend on the (classical) c.m. coordinate and use the evolution of the atomic state obtained from (9) to calculate the quasiclassical dipole force on the atom

$$\hat{F} = -\nabla \hat{H}_{AF}. \tag{11}$$

Clearly, we will not obtain information on the momentum-space dispersion of coherent wave packets this way, but for our purpose this method will suffice. In what follows, we write the fields as a product of slowly varying envelope functions and plane waves propagating along the z direction: $\Omega_q^\pm(z,t)\exp(\pm ikz)$. If the pulses are long compared to the wavelength (picosecond to nanoseconds pulses) the spatial derivative of the envelope functions can be neglected compared to the phase derivatives, so the dipole force becomes

$$\hat{F} = -\frac{\hbar}{2} \sum_{F,m;F',m'} [|F,m\rangle\langle F',m'|(ik\Omega_{F,m;F',m'}^{-*} - ik\Omega_{F,m;F',m'}^{+*})$$
$$+ |F',m'\rangle\langle F,m|(ik\Omega_{F,m;F',m'}^{+} - ik\Omega_{F,m;F',m'}^{-})] \tag{12}$$

and the change of the atoms' mean center-of-mass momentum along the z coordinate can be obtained from

$$\Delta p = \int \langle\psi|\hat{F}|\psi\rangle dt. \tag{13}$$

The average force on a statistical ensemble can also be calculated efficiently, as it is given by the forces acting on each of the basis states used in calculating $\hat{U}(t,t_0)$ averaged with their statistical weight in the ensemble.

C. Quantum control of atoms with random orientation

There are several difficulties in applying quantum control schemes to an atom with a complex level structure. The first one is already evident in Eqs. (9): the coupling coefficients between the various rotational substates $\mathcal{D}_{F,m;F',m'}$ can be very different, and are fixed. Given any specific light field, we are unable to control the relative interaction strengths of different transitions. As an illustration, the numerical values for the coefficients $\mathcal{D}_{3,m;4,m'}$ have been plotted in Fig. 2 for all three polarizations—the full list can be calculated from Eq. (7) or found in [17]. There are well-known cases when specific sum rules of the $\mathcal{D}_{F,m;F',m'}$ coefficients can be used to simplify the equations (such as the case for isotropic light or linear polarization) but these are not applicable in all situations. Second, the excited-state hyperfine levels $|F',m'\rangle$ lie fairly close to each other. Quantum control schemes often use laser pulses that are shorter than the spontaneous lifetime of the excited state, whose bandwidth is larger than the hyperfine-level separation, interacting with several (all) levels at once. For the ^{85}Rb D_1 or D_2 line this is always the case as $\tau_{\rm spont} \approx 27$ ns [17]. Third, a complex, possibly random distribution of the initial states in the ensemble is the generic case. Unless the atoms have been carefully prepared using optical pumping, all rotational substates are populated (e.g., in
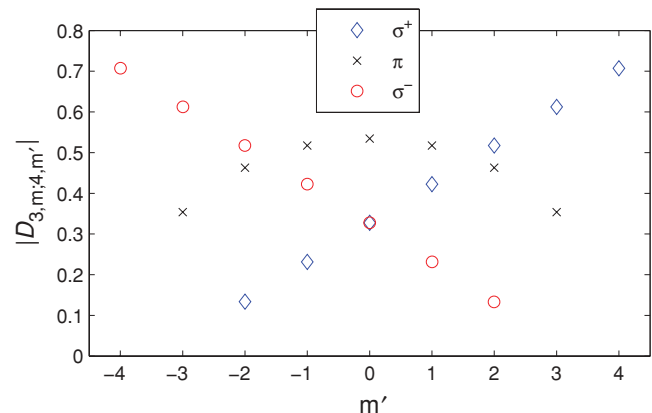

FIG. 2. (Color online) The magnitude of the coupling coefficients $\mathcal{D}_{F,m;F',m'}$ for the particular case of $F = 3, F' = 4$ on the D_2 line of ^{85}Rb.

the typical situations where a cloud of cold atoms are prepared in a magneto-optical trap or optical molasses by several lasers with various polarizations). Thus the question raised is what kind of quantum control mechanisms are actually applicable when the atom is in some unknown (and possibly incoherent) mixture of different rotational ground-state sublevels?

In what follows, we will always assume that initially we have a random ensemble of atoms with a uniform distribution over the rotational substates of the hyperfine levels that are populated (i.e., a uniform distribution over one or both of the $F = 2$ and $F = 3$ levels). For the pulsed fields propagating in either direction along the z coordinate we will assume a Gaussian envelope of the form $\Omega_q^{\pm}(t) = \Omega_{q0}^{\pm}\exp[-(t - T_d^{\pm})^2/2\tau^2]/2$, where a delay parameter has been included to order, and possibly separate the pulses in time. Multiple polarization components may be superposed to yield linearly polarized waves. For the phases of the pulses we use $\phi_q^{\pm}(t) = \delta_q^{\pm}(t - T_d^{\pm}) + \beta_q^{\pm}/2(t - T_d^{\pm})^2 + \phi_0^{\pm}$ which includes a constant detuning $\delta_q^{\pm}$ and a possible linear frequency chirp of the form $\beta_q^{\pm} t$. In most cases $\delta_q^{\pm}$ and $\beta_q^{\pm}$ will be assumed to be the same for both pulses. Also, without constraining generality, we will assume $T_d^+ = 0, T_d^- = T_d$ and $\phi_0^+ = 0, \phi_0^- = \phi_0$. These choices are appropriate for a pair of pulses whose second member has been produced simply by retroreflection of the first, which is convenient and appears in several quantum control schemes [13,20].

III. QUANTUM CONTROL WITH π PULSES

A. Population transfer with π pulses

It is instructive to consider concisely the well-known results of one of the earliest schemes that may be termed quantum control: the coherent excitation by a resonant laser pulse with a well-defined pulse area, the π pulse [11]. Since the coupling constants for any given light polarization for different sublevels are different, a given laser pulse will not in general be a π pulse for all transitions simultaneously. Furthermore, since $\tau_{\text{spont}} \approx 27$ ns, for coherent excitation the pulse duration can be 1–2 ns at the most, so pulse bandwidth will be larger than the separation of excited-state hyperfine levels.

One specific case where pure excitation may be possible regardless of the initial atomic distribution over m, however, is when we have linearly polarized light. An important sum rule for the coupling coefficients states that for any $|F,m\rangle$ state, $\sum_{F'} |\mathcal{D}_{F,m;F',m}|^2 = 1/3$ and $\sum_{F'} |\mathcal{D}_{F,m;F',m-1}|^2 + |\mathcal{D}_{F,m;F',m+1}|^2 = 2/3$ [18] so any linearly polarized field can be used to craft a π pulse if it interacts with all excited-state hyperfine levels "equally" (i.e., $\tau^{-1} \gg \delta_{F'}$, the pulse bandwidth is larger than the separation of the hyperfine levels). Depending on the bandwidth of the exciting pulse and the detuning, one can either transfer the populations of one particular F level to the excited state and leave the other intact if $\delta_F \gg \tau^{-1} \gg \delta_{F'}$ or transfer the atomic population from both F levels if the pulse bandwidth permits (i.e., $\tau^{-1} \gg \delta_F, \delta_{F'}$). In the particular case of ^{85}Rb a Gaussian pulse with $\tau = 0.1$–0.2 ns half-width and tuned from the $F = 3$ ($F = 2$) hyperfine level to the $5^{1/2}P_{3/2}$ excited-state line center will move the population of this level to the excited state, while leaving the population of the $F = 2$ (or $F = 3$)

level intact. On the other hand, a $\tau = 5$–10 ps pulse (or shorter) will transfer the atom from the ground state to the excited state regardless of the initial distribution over the two hyperfine levels. This means that using pulses of suitable length and area, we can empty either one of the ground-state hyperfine levels selectively, or both of them together, even if we have a random initial distribution over rotational substates.

Some further comments: (i) Whether the requirement $\delta_F \gg \tau^{-1} \gg \delta_{F'}$ for the selective excitation from a single ground-state hyperfine level can be fulfilled depends on both the choice of initial and final levels. Most obviously, the D_1 line is less of a candidate because the splitting between the $5^{1/2}P_{1/2}$ hyperfine levels is greater. Less obvious is that the success of selectivity also depends on the F level chosen. Since the splittings of the $F' = 2,3,4$ levels that $F = 3$ is coupled to is greater than the splittings of the $F' = 1,2,3$ levels that $F = 2$ is coupled to, the cleanest selective population transfer can be expected when exciting the $F = 2$ level on the D_2 line. (ii) Selectivity with respect to the excited state to which we transfer the population is ruled out by the requirement that we must interact with all F' levels "equally" to be able to construct the π pulse.

B. Acceleration of atoms and dephasing on the excited-state manifold

The next question to be considered is the whether one can implement quantum control by a series of π pulses, either identical in frequency and polarization or different. A series of identical π pulses that propagate in alternating directions can confine a cloud of atoms, or accelerate them efficiently [21–23]. The essence of this scheme is the absorption of a photon from one beam and emission into the counterpropagating one with the $2\hbar k$ momentum transfer that accompanies the process. Clearly, the second pulse that deexcites the atoms should follow fast enough for spontaneous emission to be negligible.

There is one other important point, however. The unitary time evolution operator $\hat{U}(t_{\max},t_0)$ that describes the coherent interaction with the laser pulse maps any subspace of the full state space to another subspace of equal dimensionality. Thus the subspace spanned by the $\{|F = 2,m\rangle\}$ set is mapped to a five-dimensional subspace of the 15-dimensional $F' = 1,2,3$ manifold, and the subspace spanned by the $\{|F = 3,m\rangle\}$ set to a seven-dimensional subspace of the 21-dimensional $F' = 2,3,4$ manifold. These subspaces, in turn, are not invariant under the atomic Hamiltonian $\hat{H}_A$, as different F' components have slightly different energies and hence gradually dephase in time. If the delay of the deexciting pulse is too large, some of the atoms will have already left this subspace (i.e., they will not be coupled to the initial ground-state level by the second pulse). This dephasing can limit our ability to transfer all of the excited-state population back to the ground state even if we neglect spontaneous emission altogether.

The dephasing effect is illustrated on Fig. 3 which depicts the ability of the second π pulse of a pair to return the atom to the $F = 3$ level after the (identical) first pulse has excited it completely. The plot shows overall population of the $F = 3$ level $P_g = \sum_m |\alpha_{3,m}|^2$ and the overall population of the excited levels $P_e = \sum_{F',m'} |\beta_{F',m'}|^2$ after the interaction, and the average momentum transferred to the atoms Δp. The
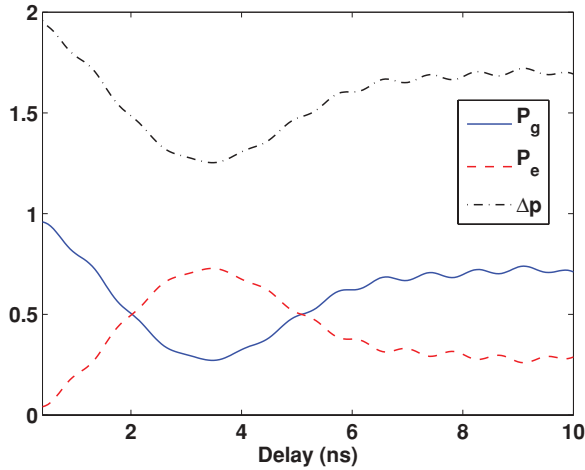
FIG. 3. (Color online) Final population of the $F = 3$ level (solid blue line), excited-state population (dashed red line), and average momentum transferred (dash-dotted black line, in units of $\hbar k$) of a ^{85}Rb atom after the interaction with a pair of counterpropagating $\tau = 0.1$ ns π pulses as a function of the delay between the two pulses.

data were obtained by solving Eqs. (9) repeatedly for a pair of $\tau = 0.1$ ns Gaussian π pulses to construct $\hat{U}(t,t_0)$ and averaging over a set of random initial states on $F = 3$. It has been noted before that it is possible to excite selectively one of the ground-state hyperfine levels in ^{85}Rb by a $\tau = 0.1$ ns pulse. From the figure one can now see that the selective acceleration of atoms residing in $F = 3$ cannot be achieved because the population returned to the initial level degrades too fast with the delay between the pulses. The maximum occurs at a delay of 0.4 ns and it is about 95%, which is not good enough if the interaction has to be repeated many times. For delays smaller than 0.4 ns the process degrades because the two pulses overlap each other appreciably. One can also see that the effectiveness of the second pulse does not decrease monotonically but shows partial collapses and revivals as can be expected in case of a coherent dephasing process.

In general, the time scale on which this dephasing becomes a problem and how it compares to τ_{spont} depends on the hyperfine splittings of the atomic levels chosen. In the particular case of ^{85}Rb the time scale for dephasing τ_{deph} is about an order of magnitude shorter than τ_{spont} if excitation of the $F = 3$ level is considered on the D_2 line. Should we consider the excitation (and coherent acceleration) of the $F = 2$ level, τ_{deph} becomes somewhat longer because the separation of the $F' = 1,2,3$ levels to which the atom is excited is about half of the separation of the $F' = 2,3,4$ levels. This is still shorter than τ_{spont}, however. Excitation on the D_1 line yields an even shorter τ_{deph}. Of course using 5–10 ps pulses does make it possible to accelerate the atoms by repeated action of π pulses similar to [21,23]—at the expense of ground-state selectivity. The important conclusion here is that when quantum control of atoms with multiple excited-state levels is attempted, limiting time scales may appear that are shorter than τ_{spont}, even in the absence of any collisions or inhomogeneous line-broadening mechanisms.

IV. QUANTUM CONTROL USING FREQUENCY-CHIRPED PULSES

A. Selective population transfer between distinct hyperfine levels

The use of frequency-chirped pulses in the adiabatic passage (AP) regime instead of π pulses for the quantum control of atoms has a long history. It has been proposed and applied for the coherent excitation and acceleration of two-level atoms [9,24,25] as well as population transfer between stable states of a Λ atom [26]. The most often emphasized advantage that AP has over π pulses is that it is robust with respect to a moderate change of experimental parameters, so imperfections of exact resonance conditions or pulse amplitude can be tolerated. For an atomic state distributed randomly over rotational substates which have unequal coupling constants to an excited state, the latter property is very attractive. Because interaction with all possible hyperfine levels for a complete population transfer is no longer necessary, selective population transfer between two hyperfine levels becomes possible. One immediate advantage is that when a single excited-state hyperfine level is populated, there is no dephasing. Any further repetition of the pulses is constrained only by τ_{spont}. This way a selective manipulation and acceleration of atoms residing within a single ground-state hyperfine level can be performed.

For a successful population transfer between selected hyperfine levels, several conditions must be fulfilled. First, the bandwidth of the exciting pulse must be narrow enough to achieve selectivity. It is clearly sufficient if the full bandwidth (including chirp) is smaller than the distance to the nearest level, but it is not always necessary. In some cases, AP can be realized between selected levels if only the transform-limited bandwidth $1/\tau$ is smaller than the frequency distance to neighboring levels, but the direction of the chirp can be chosen to realize resonance with the targeted level first [27]. The second condition is that the degeneracy of the targeted level cannot be smaller than that of the initial state. As noted before, the coherent process driven by any laser pulse defines a mapping [via the unitary $\hat{U}(t,t_0)$] between state spaces of equal dimensionality, so a transfer from $F = N$ to $F' = N - 1$ will always leave the population of a two-dimensional "dark" subspace behind on the initial level. In the case of $F' = F$, the completeness of population transfer depends on light polarization—a purely circularly polarized light or a purely linearly polarized light will necessarily leave the population of a dark state behind. (This will be the $|F, m = F\rangle$ state in the case of σ^+ polarization, or the $|F, m = 0\rangle$ state in the case of π polarization.)

Figure 4 shows the effect of a pair of consecutive linearly polarized, chirped pulses with varying length τ on an ensemble of atoms residing on the $F = 2$ level initially. The central frequency of the pulses was chosen to be resonant with the $F = 2 \rightarrow F' = 3$ transition on the D_1 line, and the effect of the pulses is plotted as a function of the pulse length. The pulse amplitude is $\Omega_{-1}^{\pm} = \Omega_{+1}^{\pm} = 10\pi/\tau$ (scaled so that the pulse maintains its area as its temporal length changes) and the chirp parameter is $\beta = \pm 4/\tau^2$ (scaled so that the frequency sweep remains the same order of magnitude as the maximum Rabi frequency). The delay between the first and second pulses is $T_d = 8\tau$, which is sufficient for the pulses to interact
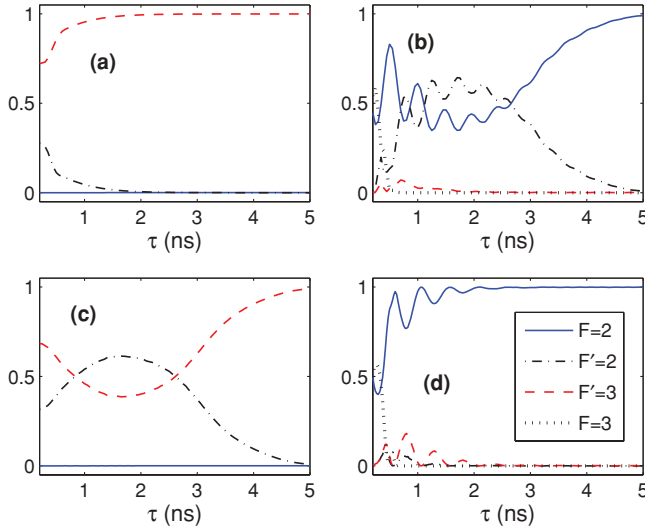
FIG. 4. (Color online) Population transfer between the $F = 2$ and $F' = 3$ hyperfine levels of the D_1 line with a pair of frequency-chirped pulses. The solid blue, dashed red, dot-dashed black, and dotted black lines depict the populations of the $F = 2$, $F' = 3$, $F' = 2$, and $F = 3$ levels, respectively. The populations have been plotted as a function of the pulse length τ.

completely separately with the atoms. The overall populations of the four hyperfine levels $F = 2,3 F' = 2,3$ are depicted on the figures: (a) after the first pulse, with $\beta_1 < 0$ (blue to red chirp); (b) after the second pulse with $\beta_1,\beta_2 < 0$; (c) after the first pulse with $\beta_1 > 0$ (red to blue chirp), and finally (d) after the second pulse with $\beta_1 < 0, \beta_2 > 0$. It is clear that acceptable population transfer by the first pulse persists for much smaller pulse lengths and thus much larger pulse bandwidths compare Figs. 4(a) and 4(c)—when the resonance condition with the $F = 2 \rightarrow F' = 3$ transition is fulfilled first. In the opposite case, a sizable portion of the atomic population is transferred to the $F' = 2$ level [(black dash-dotted curve on Fig. 4(c)]. Figures 4(b) and 4(d) show the level populations after the second pulse for both signs of the chirp β_2. It can be seen that the best case is for $\beta_1 < 0, \beta_2 > 0$, when the pulses transfer the population there and back effectively until about $\tau = 2$ ns. As the $F = 3$ level is not perturbed by the pulses significantly, a selective acceleration of the $F = 2$ ground-state level may be attempted using pulses of opposite chirp. While generating two consecutive pulses with opposite chirp does not seem very easy, using a method of chopping pulses from cw lasers with harmonically modulated frequencies does yield a series of pulses with alternating chirp directions [28]. The oscillations in the effectivity of the second pulse visible in Fig. 4(d) for $\tau < 2$ ns is caused by the dephasing effect on the excited-state manifold, as there is a slight population transfer to $F' = 2$. Complete population transfer on the $F = 2 \rightarrow F' = 2$, $F = 3 \rightarrow F' = 2$, or $F = 3 \rightarrow F' = 3$ transitions is not possible, however, because of subspace dimension problems mentioned earlier. Also, selective excitation of any of the $5^2P_{3/2}$ state levels within a time $\ll \tau_{\text{spont}}$ is ruled out by the fact that they are much closer to each other then the levels of the $5^2P_{1/2}$ state.

B. Multiphoton transitions induced by overlapping pulses

Another interesting possibility for implementing quantum control of atomic systems is by using two partially overlapping pulses, the second of which arrives with a distinct time delay. Apart from the well known STIRAP scheme and its variants, several others have also been proposed, such retroreflection induced bichromatic adiabatic passage [20] or multiphoton adiabatic passage (MAP) in Rydberg atoms or molecules [29,30]. Two pulses with a constant frequency difference have been shown to be able to induce MAP in two-level atoms [12–14] where the internal state of the atom returns to its initial one by the end of the interaction, but with several photons exchanged between the driving fields. This leads to a considerable amplification of the force acting on the atoms if the pulses counterpropagate with respect to each other, as the momentum change that accompanies the process is $2N\hbar k$, where N is a small integer, the order of the transition [14]. Such overlapping pulse schemes are preferable to two consecutive separate pulses not only because the force is amplified, but also because the time spent in the excited state is much shorter, lessening the chance of spontaneous emission [10,14].

In accordance with Sec. II B, we have used formulas (12) and (13) to calculate the integral of the quasiclassical force on atoms that interact with a pair of counterpropagating, linearly polarized, chirped pulses that overlap each other. Figure 5(a) shows the average momentum transferred to atoms (solid blue line) as well as the final population of the excited state after the
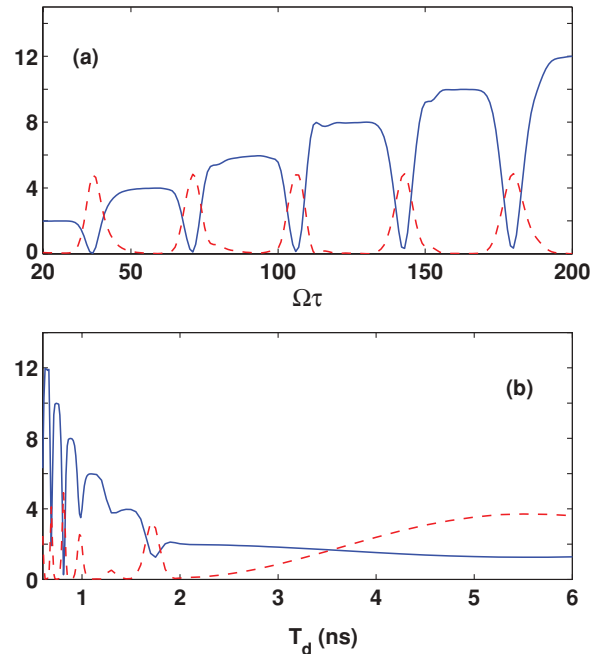

FIG. 5. (Color online) Regions of multiphoton adiabatic passage induced by a pair of overlapping, linearly polarized, chirped laser pulses as function of (a) pulse amplitude and (b) second pulse delay. Solid blue lines depict the momentum transferred to the atoms during the interaction (in units of $\hbar k$), while dashed red lines depict the final population of the excited state multiplied by five. The parameters of the calculation were $\tau = 0.5$ ns, $\beta = -20/\tau^2$, and (a) $T_d = 1.5\tau$ and (b) $\Omega_{-1}^{\pm} = \Omega_{+1}^{\pm} = 160/\tau$. The central frequency was tuned to the $F = 2 \rightarrow F' = 3$ transition on the D_1 line.

interaction (multiplied by five to be very visible—dashed red line) as a function of pulse amplitude. The atoms were assumed to populate the $F = 2$ level initially and the central frequency of the pulses was assumed to be resonant with the $F = 2 \rightarrow F' = 3$ transition on the D_1 line. Other parameters used for the calculation were $\tau = 0.5$ ns, $\beta = -20/\tau^2$, and $T_d = 1.5\tau$. The curves clearly indicate that multiphoton adiabatic passage of increasing order N takes place as the pulse amplitude increases, similarly to the two-level atom case [14] despite the fact that we have an ensemble of randomly oriented initial states. It is also visible that the atoms are returned cleanly to the ground state at the end of the interaction in the plateau regions, which is an important property for the repeatability of the interaction. Figure 5(b) shows the the same two curves (momentum transferred to the atoms—solid blue line; final population of the excited state—dashed red line) as a function of the delay between the two pulses for a fixed amplitude of $\Omega_{-1}^{\pm} = \Omega_{+1}^{\pm} = 160/\tau$. Again, the curves are similar to the ones found for two-level atoms [13].

One interesting thing to notice in Fig. 5(b) is that as the delay increases, the $N = 1$ (i.e., $\Delta p = 2\hbar k$) plateau degrades with the atoms being left partly in the excited state and the momentum transfer decreases. The reason again is that in this case the pulse bandwidth is larger than the excited-state hyperfine separation, so multiple excited-state levels are populated and dephasing occurs when we have two separate pulses. It is important to note, however, that for the $N \geqslant 2$ (i.e., when the two pulses interact with the atoms together), there is no dephasing even though it would take place without pulse overlap on this time scale. (Compare Fig. 5(b) where the $N = 2$ plateau exists for $T_d = 1.5$ ns and Fig. 3 where the second pulse leaves a sizable excited-state population behind after a delay of $T_d = 0.4$ ns.) Thus the overlapping pulse scheme enjoys a measure of immunity from the dephasing problem and it is advantageous if the atoms are driven to and from the excited state in a single interaction without a pause.

It must be mentioned that since the application of multiphoton adiabatic schemes requires higher pulse intensities than AP induced by a single field, power broadening will prevent us from applying it to a single hyperfine ground-state level selectively for ^{85}Rb. Thus we cannot accelerate atoms residing in just one of the ground-state hyperfine levels separately. Also, we would get practically identical figures if the atoms are assumed to populate both ground-state hyperfine levels initially. For the same reason, the direction of the chirp is unimportant in this case.

V. ENTANGLING ROTATIONAL AND TRANSLATIONAL QUANTUM STATES

It has been mentioned in the Introduction that in a setting when we initially have atoms distributed randomly over rotational substates of a level, only certain types of quantum control schemes may be applied. One type (which has been discussed in the preceding sections) is when all rotational substates of the level are affected in exactly the same way. Another is when only one rotational substate is affected at all. A third type, which can be considered to be the generalization of the second one, would be a control scheme in which all

rotational substates are affected in a distinctly different way. In this section we provide an example for such a control scheme.

To start, we first note that for the $F = 3 \rightarrow F' = 4$ transition on the D_2 line, the plot of $\mathcal{D}_{F,m;F',m+1}$ as a function of m looks like a straight line—see Fig. 2. This observation can be made more quantitative by considering the $F' = F + 1$ case and σ^+ polarization (i.e., $q = -1$, $m' = m + 1$). The only dependence of $\mathcal{D}_{F,m;F+1,m+1}$ on m is through $\langle Fm|F'm'; 1q \rangle$, which, using the standard formulas for Clebsch-Gordan coefficients and expanding in powers of m around $m = 0$, can be written as

$$\langle Fm|F + 1m + 1; 1 - 1\rangle$$
$$= \mathcal{G}(F)\left(\sqrt{2 + 3F + F^2} + \frac{3 + 2F}{2\sqrt{2 + 3F + F^2}}m \right.$$
$$\left. - \frac{1}{[8(2 + 3F + F^2)]^{3/2}}m^2 + O(m^3) \right), \quad (14)$$

where $\mathcal{G}(F)$ is a coefficient that depends only on F. It is easy to see that the linear term in m is always $O(1) \times m$, while the quadratic term for $m = \pm F$ is $O(10^{-2})$. Thus the magnitude of $\mathcal{D}_{F,m;F+1,m+1}$ coefficients can be considered to be a linear function of m to a very good approximation. Clearly, by virtue of symmetry the $\mathcal{D}_{F,m;F+1,m-1}$ coefficient of the $q = 1$, $m' = m - 1$ case (a σ^- transition) would also possess this property. Furthermore, this relation also holds for atoms or isotopes without a hyperfine structure (no nuclear spin), the only difference being that F must be replaced by J in the formulas.

Next we consider a pair of circularly (say σ^+) polarized, frequency-chirped pulses that overlap each other partially. Assuming that the central frequency and pulse bandwidth is such that only the $F' = F + 1$ transition is excited, the state space is decomposed into a series of independent two-level systems. By virtue of the arguments of the previous paragraph, for any given pulse amplitude the coupling of the individual $|F,m\rangle \Rightarrow |F + 1, m + 1\rangle$ transitions $\Omega_{-1}\mathcal{D}_{F,m;F+1,m+1}$ is (approximately) a linear function of m. On the other hand, the order N of the MAP transition of a two-level system also increases linearly with the coupling. This is a general property that can be best seen from a Floquet analysis [12,14], but is also visible from Fig. 5(a). Thus it is not difficult to deduce that using a pair of pulses with suitable parameters ($\Omega_q^{\pm}$, β, and T_d) it is possible to tune the interaction such that each distinct two-level $|F,m\rangle \Rightarrow |F + 1, m + 1\rangle$ transition goes through a multiphoton adiabatic process with a different N. This happens when the pulse amplitudes are such that each $\Omega_{-1}^{\pm}\mathcal{D}_{F,m;F+1,m+1}$ is on a different plateau of Fig. 5(a).

For a specific example, with a pulse pair of length $\tau = 50$ ns, $\Omega_{-1}^{\pm} = 224/\tau$, $\beta = 20/\tau^2$, central frequency tuned to the $F = 2 \rightarrow F' = 3$ transition on the D_1 line, and a delay of $T_d = 1.5\tau$ [apart from a temporal scaling the same parameters as those used to produce Fig. 5(a)], we obtain a situation where the $m = -2, -1, 0, 1, 2$ sublevels of $F = 2$ go through MAP of order $N = 2, 3, 4, 5, 6$, respectively. This means that if the two pulses of the pair counterpropagate, atoms residing initially in the $m = -2, -1, 0, 1, 2$ ground-state sublevels experience a state-dependent force and acquire $\Delta p = 4\hbar k, 6\hbar k, 8\hbar k, 10\hbar k, 12\hbar k$ momentum by the end of the interaction, respectively. This property of the interaction holds

irrespective of whether the initial state is some pure quantum state (i.e., a coherent superposition of various ground-state sublevels) or a statistical mixture of pure states. The scheme can thus be used to separate (in momentum space and thus eventually in coordinate space) atoms in various ground-state sublevels or, in other words, to entangle the the atoms' internal quantum states with specific translational states.

To see this more clearly, let us consider an initial atomic state of the form

$$|\psi_0\rangle = \sum_{m=-2}^{2} \int_{-\infty}^{+\infty} f_m(p)|J,m,p\rangle dp, \quad (15)$$

where the complex momentum-space probability amplitudes $f_m(p)$ for each m describe a narrow momentum distribution peaked around $p = 0$ and the states $|J,m,p\rangle$ denote a simultaneous eigenstate of the angular momentum and translational momentum. The effect of a pair of counterpropagating pulses with the above parameters will then transform this wave function in the adiabatic regime into

$$|\psi_1\rangle = \sum_{m=-2}^{2} \int_{-\infty}^{\infty} f_m(p - [m+4]2\hbar k)e^{i\phi_m}|J,m,p\rangle dp, \quad (16)$$

where $e^{i\phi_m(p)}$ is a phase that arises from the time integral of the adiabatic eigenvalues. Thus the wave function components for different m are shifted by a different amount, but the shapes of the momentum-space distributions do not change. If this cycle is repeated several times to separate the different wave-function components in momentum space, we will have a new wave function in which the internal rotational states are entangled with distinct translational states. The process is illustrated on Fig. 6 where the overall momentum-space distribution of the atom has been plotted after each cycle of interaction with a pair of overlapping chirped pulses. The initial state has been chosen to have $f_m(p) \sim \exp(-p^2/2\sigma^2)$
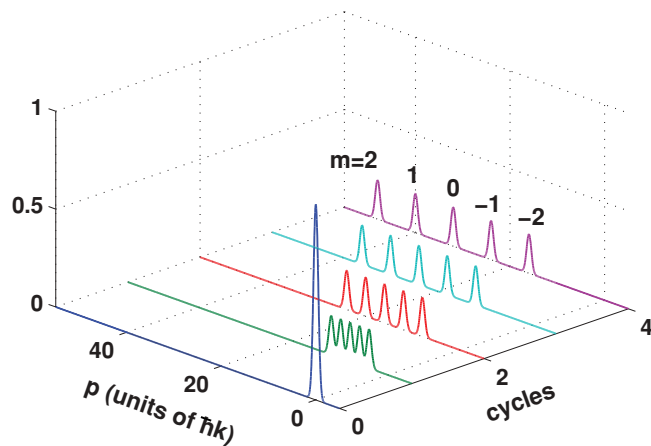
FIG. 6. (Color online) Entangling rotational substates with translational states. A pair of counterpropagating, frequency-chirped pulses transform the initial-wave function by shifting each m component of the momentum-space wave function by a different amount. The overall momentum-space distribution is plotted on the figure after each cycle. After a few cycles, several distinct peaks in momentum space are visible with each peak corresponding to a different m indicated above the peaks after four cycles.

the same shape and amplitude for each m. After a few cycles the initial-state transforms into one in which distinct momentum-space peaks correspond to distinct rotational states (the peaks are labeled with the corresponding m after four cycles).

Note that since this scheme relies on the interaction with a single excited-state hyperfine level, the pulse length had to be chosen far too long for the scheme to really work—larger than the spontaneous lifetime of ^{85}Rb. However, in other cases such a scheme may well be feasible either if hyperfine levels other than the targeted one are distant enough, or when there is no hyperfine structure at all (atoms or isotopes with zero nuclear spin). Also, it has to be stressed that while the linear dependence of the coupling on m is only approximate, the momentum shift of each component is exactly $2N\hbar k$, because adiabatic passage is fault tolerant with respect to the precise value of the coupling parameters. Thus as long as the conditions for adiabaticity are fulfilled and the coupling for a certain m places us somewhere on the plateau for a specific N, the mth component of the wave function will be shifted exactly by $2N\hbar k$.

VI. SUMMARY

We have investigated the quantum control of atoms with rotationally degenerate hyperfine levels by short laser pulses. As a case study, we have used the hyperfine-level scheme of the ^{85}Rb D_1 and D_2 lines and assumed that we have an ensemble of atoms that are distributed evenly on the rotational substates of the ground-state hyperfine levels initially. Solving Schrödinger's equation for the interaction, we tested the applicability of several quantum control schemes usually discussed for two-level atoms, such as population transfer by π pulses, acceleration of atoms using counterpropagating π pulses, adiabatic population transfer using frequency-chirped pulses, acceleration of atoms using counterpropagating chirped pulses, and multiphoton adiabatic passage using pairs of overlapping chirped pulses.

We found that due to the unequal coupling constants between different rotational state sublevels, π pulses have limited applicability when an ensemble of randomly oriented atoms is to be excited by a laser pulse, or accelerated coherently using counterpropagating pulses. The main obstacles are that pulse bandwidth should be much larger than the separation of the excited-state hyperfine levels and that dephasing between excited-state hyperfine levels degrades performance faster than does spontaneous emission, even in the absence of any line-broadening mechanisms.

AP induced by chirped pulses on, the other hand, can be used to transfer atomic population between distinct hyperfine levels, regardless of the differences of coupling constants and randomly oriented initial states. While this may be a valuable property in itself, it can also be advantageous when performing quantum control with multiple pulses or accelerating atoms with a series of pulses, because the dephasing between excited-state hyperfine levels can be reduced or eliminated.

Multiphoton adiabatic passage by overlapping pulses found for two-level atoms can also be made to work when we have rotationally degenerate substates and multiple hyperfine levels. Thus schemes which transfer $2N\hbar k$ momentum to the atoms by a single pulse pair are available. Overlapping pulses have also

been found to be advantageous because performing quantum control in a single interaction instead of two consecutive pulses eliminates the dephasing problem even if multiple hyperfine levels are excited.

Moreover, in certain cases it may be possible to turn the difference of coupling constants to an advantage. Such a scheme has been demonstrated where the order of multiphoton adiabatic passage depends on the rotational substate. This scheme can be used to entangle the rotational substates of the atom with distinct translational quantum states. This entanglement works irrespective of whether the initial state is a pure state (a coherent superposition of rotational substates) or an incoherent statistical ensemble.

Overall, we have shown that the functioning of a quantum control scheme in the presence of multiple hyperfine levels with rotational degeneracy depends on several factors which must all be considered carefully. The most trivial are, of course, pulse length versus spontaneous lifetime and pulse bandwidth versus distance to neighboring levels. (Transform-limited bandwidth and full bandwidth in the case of chirped pulses may count separately.) Less obvious but equally important are the direction of chirp for frequency-modulated pulses, the degeneracy of the levels that are to be coupled by the interaction (i.e., the dimensionality of the ground- and excited-state manifolds that are coupled) and the possible dephasing effects due to excitation to multiple hyperfine levels. It has also been shown that in numerous cases, the robustness of adiabatic passage with respect to pulse parameters can be made to translate into a robustness with respect to randomly oriented initial states of the atoms.

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