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Laser-induced population transfer in multistate systems: A comparative study

B. W. Shore,* K. Bergmann, A. Kuhn, S. Schiemann, and J. Oreg†
Fachbereich Physik der Universität, Postfach 3049, 6750 Kaiserslautern, Germany

J. H. Eberly
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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We examine similarities and differences in three schemes that are capable of producing complete population transfer in multistate systems: generalized π pulses, adiabatic passage by pulse chirping, and counterintuitive pulses or stimulated Raman adiabatic passage. We use the picture of adiabatic following through avoided crossings of instantaneous eigenvalues to exhibit the essential differences between the latter two procedures.

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I. INTRODUCTION

For many purposes it is desirable to produce samples of atoms or molecules whose population resides almost entirely in a particular excited state. For complete population transfer, from the initially populated state into this desired final state, the excitation must be coherent, i.e., it must be described by the Schrödinger equation rather than by rate equations [1]. Within this category there exist a variety of schemes that can, in principle, produce complete population inversion of a two-state system or, more generally, produce complete population transfer in a multistate system. The present paper offers comments on the differences and similarities of several of these schemes.

II. TWO-STATE POPULATION TRANSFER

The problem is simplest when only two nondegenerate levels (quantum states) are involved. Two well-known procedures can, in principle, produce complete population transfer in an ensemble of two-state atoms or molecules.

A. π pulse

The first procedure makes use of the Rabi population oscillations that characterize coherent excitation [1]. By employing nearly monochromatic light tuned to the resonance (Bohr) frequency of two levels one can induce complete but momentary population inversion. The radiation frequency remains constant during the pulse, and the pulse envelope may have any convenient time dependence. One adjusts the laser intensity and pulse duration so that the time integral of the Rabi frequency (the pulse area) has the value π (or any odd-integer multiple of π) [2]. A longer pulse, with area 2π, will return the population to the initial state.

A single π pulse can, in principle, produce complete population transfer between two states. In turn, a succession of separate π pulses, with different carrier frequencies, can produce a sequence of population transfers and thereby move population eventually into any desired state. With such a multiple-step scheme the population resides in a succession of intermediate states, each of which may undergo spontaneous emission with consequent loss of transfer efficiency.

Alternatively, one may expose the atom to the simultaneous influence of two or more pulses, each tuned to an appropriate Bohr frequency, so that the atom undergoes multilevel Rabi oscillations. By suitably controlling the relative intensity and duration of the concurrent pulses one may produce complete population transfer between specified states (e.g., Cook-Shore pulses [3]).

A π-pulse procedure will induce complete population transfer between any two levels that couple via a one-photon transition. The population transfer can also proceed by multiphoton transitions (two-photon ones in the case of three states). Such a transition requires that single-photon steps be detuned from resonance. A multiphoton transition moment is inversely proportional to the product of one or more detunings, and so it is typically much smaller than a single-photon transition moment. Therefore a multiphoton π pulse will require higher intensity than will a single-photon π pulse.

The main disadvantage of the π-pulse method and its multistate generalizations is the requirement for resonant light (needed for complete population transfer) and the need to control very precisely the pulse area. The presence of Doppler shifts diminishes the achievable population transfer unless the intensity is so large that the Rabi frequency greatly exceeds the Doppler bandwidth. The presence of magnetic sublevel degeneracy introduces further difficulties: in an ensemble of dipole moments the pulse area can be π for only one of the transition moments.

B. Adiabatic passage

An alternative mechanism for population transfer between two states is based on sweeping the pulse frequency through a resonance [4,5]. If the sweep (chirp) is sufficiently slow, then it is possible to produce complete population transfer between the two states that are connected by the resonance. The mechanism is often explained by plotting the two dressed-atom eigenvalues (i.e., the energies of the atom and fields together, including the radiative interaction) as a function of the carrier frequency $\omega(t)$ of the radiation pulse [4]. In the absence of radiative interaction, these curves differ by the detuning between Bohr frequency and carrier frequency. Thus for the simple case of a linear chirp the curves are two straight lines. These lines, often termed diabatic curves, cross when $\omega(t)$ equals the Bohr frequency. The radiative interaction causes the curve crossing to become an avoided crossing; the resulting curves are termed adiabatic.

To understand adiabatic passage with the aid of such a diagram, one considers an initial situation in which the frequency is far from resonance, so that it is possible to identify unambiguously the initially populated state with one of the dressed states (i.e., an eigenstate of the full Hamiltonian for the atom together with its interaction with radiation). If the frequency can be swept sufficiently slowly, then population will remain in this dressed state and at the conclusion of the chirp the state will again be unambiguously identifiable as the excited state. In this way slow passage through the region of avoided crossing will induce population transfer (sometimes termed a Landau-Zener transition).

A scheme based on adiabatic passage has the advantage that it is insensitive to pulse area (and to pulse shape) and to the precise location of the resonance. Thus it is useful for producing population transfer in an ensemble of atoms that have different Doppler shifts and different dipole moments. In principle, if the chirp is sufficiently slow, the entire ensemble can undergo complete population transfer.

The difficulty with two-state adiabatic passage arises from the need to employ a slow chirp and hence a long pulse, and from the requirement that the frequency should sweep a range that is large compared with the peak Rabi frequency. The requirement for slow population transfer often conflicts with the need to move population more rapidly than spontaneous emission depletes population.

Like the π-pulse procedure, the adiabatic-passage scheme has possibilities for extension to multiphoton transitions, thereby enabling population transfer between two levels that have no allowed single-photon transition moment. However, the weakness of the multiphoton transition requires that the chirp rate be appropriately slower than for a single-photon transition.

III. THREE-STATE SYSTEMS

Both these techniques, the π pulse (with constant frequency) and the adiabatic passage (with swept frequency), require pulsed coherent excitation, but they proceed in very different ways. Each permits generalization to multiphoton transitions or to multistep excitation. The multistep schemes suffer the same disadvantages of the two-state schemes.

By considering three (or more) states, however, a third procedure becomes possible. It too is based on pulsed coherent excitation, by resonant light, but the pulses are applied in a "counterintuitive" sequence [5]. That is, the second-step excitation pulse is applied before the first-step pulse.

As an example of a typical situation, we consider an ensemble of nondegenerate three-state molecules. We assume that the states 1, 2, and 3 are connected in a chain 1-2-3 by electric dipole transitions, and that the population initially resides entirely in state 1, which we take to be the state having lowest energy. For definiteness we take state 2 to have the highest energy, so that the excitation is analogous to a Raman process: the pump transition links states 1 and 2, while the Stokes transition links states 2 and 3. An "intuitive" pulse sequence would apply the pump pulse prior to applying the Stokes pulse.

Stimulated Raman adiabatic passage

In the third scheme [5,6], typically accomplished with resonant pump and Stokes pulses, the carrier frequencies are kept constant and only the pulse amplitude varies. The pulses are applied to the atom in the "counterintuitive" order: first the Stokes transition (coupling the unpopulated states 2 and 3) and then the pump transition (coupling the initial state 1 with state 2). This method, stimulated Raman adiabatic passage (STIRAP), has been demonstrated [7] to produce complete population transfer between states 1 and 3, without ever placing appreciable population into state 2.

This method requires that the temporal pulse area be much larger than π (typically the area should be larger than around 3π), but it does not require careful control over pulse area, and so it is applicable to atoms or molecules with magnetic sublevels. This insensitivity to pulse area makes possible efficient use of laser beams whose intensity exhibits a Gaussian spatial variation. STIRAP does not require single-photon resonance, although it works most efficiently when both pulses maintain such tuning. However, successful population transfer does require that the two frequencies maintain the condition of two-photon resonance.

IV. QUESTIONS

In view of the success of STIRAP, it is natural to ask whether STIRAP can be considered as a kind of chirp-induced adiabatic passage. The immediate answer is no: adiabatic passage requires a frequency chirp, whereas STIRAP, as hitherto developed, requires constant frequency. Undaunted, one might wonder whether the needed counterintuitive pulse sequence might, in some way, be provided by a frequency chirp of a single pulse, such that the carrier frequency passes successively through Stokes and then pump resonance frequencies (in the sequence that is used for STIRAP). To answer this question, we proceed as follows.

V. FORMULATION

Let the laser P, with time varying frequency $\omega_p(t)$, connect states 1 and 2. Let the laser S, with frequency $w_S(t)$, connect states 2 and 3. We permit, but do not require, two distinct frequency variations. Upon introducing the traditional rotating-wave approximation (RWA) together with suitable choices of phases we can write the first two diagonal elements of the three-state RWA Hamiltonian as

$$\hbar W_{11}(t) = +\hbar\omega_p(t) - (E_2 - E_1) \ , \quad \hbar W_{22}(t) = 0 \ . \quad (1)$$

It is always possible to add a constant to all diagonal elements of the Hamiltonian, thereby defining a convenient zero-point of energy. Here we choose to make the middle element W_{22} zero. The remaining element is, for a ladder configuration [$E_3 > E_2$, see Fig. 1(a)],

$$\hbar W_{33}(t) = -\hbar\omega_S(t) - (E_2 - E_3) \ , \quad (2a)$$

while for a Λ configuration [$E_3 < E_2$, see Fig. 1(b)] it is

$$\hbar W_{33}(t) = +\hbar\omega_S(t) - (E_2 - E_3) \ . \quad (2b)$$

We shall proceed by assuming that the time dependence of these matrix elements comes, as shown here explicitly, by controlled variation of the carrier frequencies. It is also possible to vary the Bohr frequencies, say by causing the atoms to experience a varying static Stark effect. (We comment below on dynamic Stark shifts.) If there are two lasers, then we have two separate functions of time. Otherwise we have just one frequency. The complete RWA Hamiltonian includes nonzero elements

$$\hbar W_{12}(t) = \hbar[W_{21}(t)]^* \ , \quad \hbar W_{23}(t) = \hbar[W_{32}(t)]^* \ . \quad (3)$$

Let us regard an arbitrary pulse sequence by considering the instantaneous eigenvalues of the RWA Hamiltonian $W(t)$. These can be regarded as the three diagonal elements of $W(t)$, modified by repulsion at (avoided) crossings. The extent of the repulsion increases as the instantaneous Rabi frequency increases. We therefore require a plot of three curves versus frequency. We then follow a path, starting with W_{11} initially, and proceeding through avoided crossings until we find the final state. If the chirp is slow, then the population follows the avoided crossing, resulting in population transfer. If the chirp is fast, then no population transfer occurs at the crossing.

The present challenge, to produce population transfer from state 1 to state 3, can evidently be met in two ways.

(i) Indirect. Obviously, we can proceed in two separate steps: first a single-photon resonance crossing that moves population from state 1 to state 2 (the pump transition), and then a second crossing (the Stokes transition) that moves population from state 2 to state 3. Because state 2 is a necessary intermediary in this scheme, and is therefore populated, this excitation procedure is evidently not equivalent to STIRAP. This method of pulsed excitation is sometimes termed "intuitive," in contrast to the "counterintuitive" pulse sequence of STIRAP.

The general scheme of this process, viewed as a succession of curve crossings, must be topologically equivalent to the representation in Fig. 2(a). Under the assumption that with increasing time the frequency variation goes from left to right, the pump frequency sweeps through resonance from below whereas the Stokes frequency sweeps through resonance from above. The (avoided) crossing of states 1 and 3 does not affect this scheme; it is only necessary that we adjust the separate crossings of 1-2 and 2-3. In principle this can be accomplished with a single laser that sweeps across the two resonances.

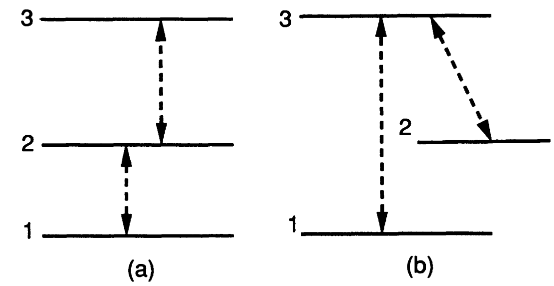

FIG. 1. (a) Ladder configuration and (b) Λ configuration.

(ii) Direct. Alternatively, we can proceed to transfer population directly between state 1 and state 3 by a single crossing of W_{11} with W_{33}. The diagram of diabatic curves must have the topological form of Fig. 2(b) (again reading from left to right). The diagram is drawn for a case in which only the Stokes frequency changes, by sweeping through the two-photon resonance. It is immaterial whether curve 1 moves or crosses curve 2, so long as the 1-3 crossing takes place prior to any 1-2 crossing. Similarly, it is immaterial whether E_3 lies above E_2 (a ladder, as shown) or below E_2 (a Λ configuration).

It should be noted that this direct adiabatic process requires two separate pulse frequencies when it is applied to a Raman type pair of transitions (a Λ configuration): it is not possible to sweep through two-photon resonance with a single frequency. To see this constraint most clearly it is useful to redefine the phases such that the first two diagonal elements of the RWA Hamiltonian become

$$\hbar W_{11}(t) = 0 \ , \quad \hbar W_{22}(t) = (E_2 - E_1) - \hbar\omega(t) \ . \quad (4)$$

The remaining element is, for a Λ configuration ($E_3 < E_2$), the constant value

$$\hbar W_{33}(t) = (E_3 - E_2) \ . \quad (5a)$$

From the constancy of this matrix element we recognize that it is not possible to produce a 1-3 curve crossing in the Λ configuration by means of frequency chirp with a single laser, unless the states 1 and 3 are degenerate. (Pulses may then be distinguished by their polarization.)

The restriction against a single laser does not apply to

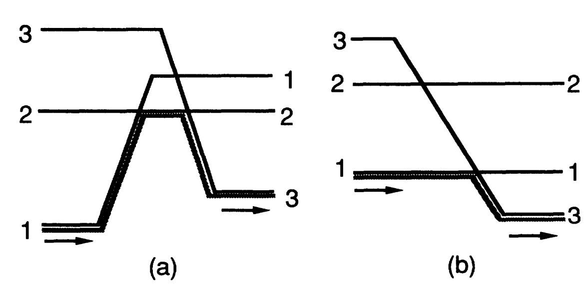

FIG. 2. (a) Frequency variation of RWA detunings needed to produce indirect population transfer 1-2-3. (b) Frequency variation of RWA detunings needed to produce direct population transfer 1-3. Population follows shaded path if excitation is adiabatic.

a ladder excitation, for which $E_1 < E_2 < E_3$. In that case the RWA Hamiltonian has the element

$$\hbar W_{33}(t) = (E_3 - E_1) - 2\hbar\omega(t) . \qquad (5b)$$

We see that it is possible to produce a curve crossing in the ladder configuration by sweeping a single laser through a two-photon resonance.

This indirect scheme places no population into state 2. It therefore accomplishes just what STIRAP does. However, the transfer is by means of a two-photon resonance, in which some combination of pump or Stokes frequency sweeps through resonance with the 1-3 transition. Population will be excluded from state 2 only if the pump and Stokes pulses are not resonant (at least during the population transfer) with their respective transitions. We must chirp the combined pump and Stokes frequencies through the two-photon resonance. The chirp must be slow compared with the two-photon transition rate. This rate is much smaller than a resonant single-photon transition rate, and so the chirp must be correspondingly slower. This is a disadvantage of any process that relies on a two-photon transition.

VI. STIRAP AS A CHIRP

Although STIRAP cannot be produced by chirping a pulse carrier frequency, there is a sense in which the two-photon adiabatic passage of nonresonant STIRAP can be considered as a chirp [8]: the process accompanies a detuning between a fixed laser frequency and a changing dressed-state eigenvalue (i.e., a dynamic Stark shift). To see this we adiabatically eliminate state 2 from the three-state sequence. The resulting effective two-state Hamiltonian has a detuning which is [8]

$$\Delta_{\text{eff}} = (|W_{32}|^2 - |W_{21}|^2)/W_{22} . \qquad (6)$$

This effective detuning, involving the difference of two pulse intensities, acts to produce adiabatic passage in exactly the same way as does the conventional frequency chirp of a true two-state system. However, this chirp involves fixed laser frequencies and variable (Stark shifted) Bohr frequencies.

VII. CONCLUSION

Population transfer by frequency chirped adiabatic passage can move population directly between states 1 and 3, bypassing the intermediate state 2. However, the desire for minimal population in the intermediate state requires a two-photon transition in which this state is far from resonance. This condition means that the two-photon Rabi frequency will be weak. In contrast, the STIRAP process modulates the amplitudes alone (rather than the carrier frequencies) of two fields. It can use carrier frequencies that are on resonance, and so it can proceed rapidly.

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*Permanent address: Lawrence Livermore National Laboratory, Livermore, CA 94550.

†Permanent address: Nuclear Research Center of the Negev, Beer-Sheva, Israel.

[1] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990).

[2] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1987), Sec. 3.2; see also B. W. Shore, *The Theory of Coherent Atomic Excitation* (Ref. [1]), Sec. 5.1.

[3] R. J. Cook and B. W. Shore, Phys. Rev. A **20**, 539 (1979).

[4] E. E. Nikitin, in *Physical Chemistry. An Advanced Treatise, Vol. VIA,* edited by W. Jost (Academic, New York, 1974), pp. 216–222; J. P. C. Kroon, H. A. J. Senhorst, H. C. W. Beijerinck, B. J. Verhaar, and N. F. Verster, Phys. Rev. A **31**, 3724 (1985).

[5] J. Oreg, G. Hazak, and J. H. Eberly, Phys. Rev. A **32**, 2776 (1985); J. Oreg, F. T. Hioe, and J. H. Eberly, *ibid.* **29**, 690 (1984).

[6] C. E. Carrol and F. T. Hioe, J. Math. Phys. **29**, 487 (1988); J. Opt. Soc. Am. B **5**, 1335 (1988); J. R. Kuklinski, U. Gaubatz, F. T. Hioe, and K. Bergmann, Phys. Rev. A **40**, 6741 (1989).

[7] U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988); U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, J. Chem. Phys. **92**, 5363 (1990); H.-G. Rubahn, E. Konz, S. Schiemann, and K. Bergmann, Z. Phys. D **22**, 401 (1991).

[8] D. Grischkowsky and M. M. T. Loy, Phys. Rev. A **12**, 1117 (1975).