Laser-driven population transfer in four-level atoms: Consequences of non-Abelian geometrical adiabatic phase factors

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(Received 4 August 1998)

We analyze the influence of three pulsed laser fields interacting with a four-state atomic system for which there occur two degenerate population-trapping dressed states. We present a simple expression for the geometrical phase acquired by such an atom during a stimulated Raman adiabatic passage (STIRAP) process using a delay between Stokes and pump pulses, during which the two Rabi frequencies adiabatically traverse a closed path in the parameter space. We describe techniques that can produce (geometrical) state-vector phases that are independent of the longitudial velocity with which atoms move across a laser beam, and that are insensitive to radiative decay from the intermediate excited state. The geometrical phase can be changed by changing the relative delay of the pulses. The geometrical phase can be measured by observing the population in atomic states. We show that when the pump and Stokes pulses are properly timed, high population transfer from the initial state to the target state is achieved. In the adiabatic limit the robustness of the population transfer is equivalent to that of STIRAP. [S1050-2947(99)05104-5]

PACS number(s): 42.50.Hz, 03.65.Bz

I. INTRODUCTION

A. Phase in quantum systems

One of the significant differences between a classical statistical system, described by a probability distribution, and a quantum system, described by probability amplitudes, is the presence of a phase factor with each probability amplitude. These make possible a variety of interference effects characteristic of wavelike behavior, of which the familiar two-slit diffraction pattern is perhaps the simplest.

In elementary cases, each probability amplitude acquires a scalar phase with passing time. Such scalar objects commute — they are Abelian. More interesting are the cases, discussed here, involving matrices of phases — these are non-Abelian.

We will describe techniques that can produce state-vector phases which are independent of the duration of pulses; such phases are thus independent of the velocity with which atoms move across a laser beam. We will note possibilities for applying the general theory in atomic beams of helium and neon.

The work reported here is an extension to some of the methods developed in recent years to produce atomic or molecular population transfer in multilevel systems by means of pulsed coherent excitation [1]. Section II considers the method of stimulated Raman adiabatic passage [2]. A femtosecond variant of this technique, adiabatic passage by laser-induced potentials, forces molecular vibrational motion into a desired target state [3].

B. Schrödinger equation

We consider the effect on an atom of classical pulses of coherent radiation, i.e., fields free of stochastic variation. We assume the pulses that are sufficiently short so that the excitation dynamics is coherent, and can be described by the time-dependent Schrödinger equation

$$\hbar \frac{d}{dt}\Psi(t) = -iH(t)\Psi(t), \tag{1}$$

where $H(t)$ is the time-dependent Hamiltonian operator for the atom in the presence of a specified field. Because probability is conserved, the norm of the state vector remains fixed, $|\Psi(t)|^2 = 1$.

The initial conditions on a state vector, and the analysis of its evolution in time, are presented most conveniently by introducing a set of atomic basis states, ψ_n, associated in the absence of pulsed radiation with energies E_n. Typically one assumes, as do we, that the atom is known to be in state ψ_1 initially (at time $t \to -\infty$). Of interest are the probabilities $P_n(t) = |\langle \psi_n | \Psi(t)|^2$ for finding the atom at a later time t in state ψ_n.

A simple approach to this problem, now quite standard, is to express the state vector $\Psi(t)$ as a superposition of the basis states, in the form [1]

$$\Psi(t) = \sum_n C_n(t)\exp[-i\zeta_n(t)]\psi_n \equiv \sum_n C_n'(t)\psi_n \tag{2}$$

where the functions $\zeta_n(t)$ are chosen *a priori* and the complex variables $C_n(t)$ are to be determined so as to satisfy the Schrödinger equation and the initial conditions. For any choice of the phase functions $\zeta_n(t)$, the functions $C_n(t)$ are probability amplitudes: the associated probabilities are $P_n(t) = |C_n(t)|^2$. Following this usual approach, one obtains, in place of the original operator equation for $\Psi(t)$, a set of coupled ordinary differential equations for the probability amplitudes. Upon listing these amplitudes $C_n(t)$ as components of a vector $\mathbf{C}(t)$, one obtains the Schrödinger equation in the form

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$$\frac{d}{dt}\mathbf{C}(t) = -i\mathsf{W}(t)\mathbf{C}(t), \tag{3}$$

where $\mathsf{W}(t)$ is a time-varying matrix, whose elements depend explicitly upon the choice of phase functions $\zeta_n(t)$ made in defining the probability amplitudes. It is usually desirable to choose these phases such that the off-diagonal elements of $\mathsf{W}(t)$ are slowly varying. This is possible in the cases we shall consider.

C. Adiabatic states and the RWA

One of the important situations of coherent atomic excitation occurs when the time variation of the Hamiltonian, as presented in the matrix $\mathsf{W}(t)$, is very slow. It then becomes useful to introduce a set of adiabatic (or dressed) states $\Phi_k(t)$ defined as instantaneous eigenstates of $\mathsf{W}(t)$,

$$\mathsf{W}(t)\Phi_k(t) = \lambda_k(t)\Phi_k(t) \tag{4}$$

and to express the state vector in terms of these,

$$\Psi(t) = \sum_a B_a(t)\Phi_a(t). \tag{5}$$

Such an expansion is always possible.

Our concern is with excitation of an atom by near-resonant laser fields. Specifically we consider the customary case when laser frequencies are nearly resonant with atomic transitions, and when each transition can be uniquely assigned a single near-resonant laser field. Under these well-studied conditions we deal with the rotating wave approximation (RWA) Hamiltonian matrix $\hbar\mathsf{W}(t)$, whose off-diagonal elements $\hbar\Omega_{nm}(t)$ express the interaction energy of a transition dipole moment in a pulsed electromagnetic field, and whose diagonal elements are detunings expressing cumulative differences between Bohr frequencies and laser carrier frequencies. We will assume that the various laser frequencies are resonant with the appropriate transitions, so that the diagonal elements $W_{nn}(t)$ of $\mathsf{W}(t)$ vanish. We also assume that the pulse areas $\int dt W_{nm}(t)$ are large, so that the adiabatic approximation [4] can be used to solve the RWA equations (3).

D. Dynamical, geometric, and non-Abelian phases

In special circumstances the initial state coincides with one of the adiabatic states, say $\Phi_a(t)$,

$$\Psi_a(-\infty) = \Phi_a(-\infty),$$

where the label on $\Psi_a(t)$ identifies the initial condition. If the system evolves adiabatically thereafter, then it remains at all times in a combination of adiabatic states having a common eigenvalue. When the adiabatic states are not degenerate, then only the state $\Phi_a(t)$ can contribute to $\Psi_a(t)$, and we can write

$$\Psi_a(t) = B_{aa}(t)\Phi_a(t) \equiv \exp[i\phi_a(t)]\Phi_a(t),$$

thereby defining a (scalar) phase $\phi_a(t)$. Thus adiabatic evolution, which returns externally controlled parameters to their initial values, will return the system to the initial state, but with the acquisition of some phase ϕ_a.

As first noted by Berry [5], one can distinguish two contributions to the phase acquired after completed time evolution: a dynamical part and a geometrical part. Usually the major contribution to the phase depends explicitly upon the time variation of the Hamiltonian; it is a *dynamical* phase. Berry [5] showed that when the Hamiltonian of the system depends on a set of parameters which evolve along a closed curve in the parameter space, then the state vector corresponding to a simple *nondegenerate* eigenvalue develops a phase which depends only on the curve in the parameter space. This is the *geometrical* phase (or Berry phase). Unlike the dynamical phase, the geometrical phase does not depend on the duration of the interaction. It is therefore, for instance, independent of the speed with which an atom moves through an interaction region.

Berry's original identification of a geometric phase assumed that the eigenstates of the Hamiltonian were nondegenerate, and that the adiabatic theory of Born and Fock [4] could be applied. The notion of Berry's phase was generalized to the case of degenerate levels by Wilczek and Zee [6]. Aharonov and Anandan [7] went beyond the assumption of adiabatic evolution to further generalize Berry's results.

When the evolution is adiabatic but the adiabatic eigenvalues are degenerate, then we can write the state vector which evolves from $\Psi_a(-\infty)$ as

$$\Psi_a(t) = \sum_b^{\mathcal{N}} B_{ab}(t)\Phi_b(t),$$

where $\mathcal{N}$ is the number of adiabatic states which have a common eigenvalue. The coefficients here, $B_{ab}(t)$, form a unitary matrix; they can be regarded as the elements of a generalized non-Abelian phase matrix $[\exp(i\hat{\phi})]_{ab}$. Thus when degenerate eigenvalues are considered the Berry phase needs to be replaced by a unitary transformation among the degenerate eigenfunctions. The transformation has the properties of a non-Abelian gauge field [6]. The non-Abelian phase has been invoked, for example, in discussions of atoms in external collinear electric and magnetic fields [8], and for nuclear quadrupole resonance [9].

Usually the geometrical phase is much smaller than the dynamical phase. This makes measurement of the geometric phase difficult — it is a small effect on a large effect. For example, the variation of dynamical phase due to variations of the atom residence time in a laser beam places stringent constraints on allowable variation of atomic velocities — atomic beams must be nearly monoenergetic.

Here we present a scheme to measure the geometric phase. The proposal is based on using null-eigenvalue trapped states [1] to describe the state vector at all times. When a system evolves as a trapped state, the state vector does not acquire a dynamical phase; only the geometrical phase, if nonzero, is present. We show how the geometrical phase can be mapped into atomic excitation. The excitation can, in turn, be detected by standard techniques (laser-induced fluorescence or photoionisation) or as deflection of an atomic beam. Thus there are numerous possibilities for using degenerate dressed states to observe the geometric phase.

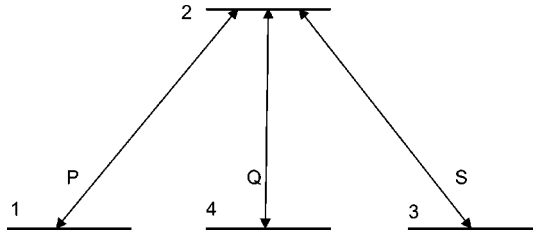

FIG. 1. Energy levels and linkages. States 1 and 2 are coupled by the pump pulse $P(t)$, states 2 and 3 by the Stokes pulse $S(t)$, and states 2 and 4 by the pulse $Q(t)$. The system is initially in state 1.

II. NON-ABELIAN PHASE IN A FOUR-STATE SYSTEM

The extension from three states to four states brings with it numerous interesting possibilities for interesting effects, much as the earlier extension of the two-state atom to a three-state system brought new classes of phenomena under examination. In this paper, we discuss geometrical-phase effects in a four-state atomic system which interacts with three pulsed laser fields. The system has similarities with the three-state system of two pulses acting to produce stimulated Raman adiabatical passage (STIRAP) [2]. The STIRAP mechanism offers, in principle, a simple scheme for transferring all atomic population from a single populated initial state to a target final state via a sequence of two partially overlapping pulses applied in counterintuitive order: first a Stokes pulse and then a pump pulse. During the course of such a pulse sequence the adiabatic parameters (the two Rabi frequencies) traverse a closed-loop path in parameter space. Reference [10] presented a concept for measurement of the Berry phase for atomic interferometry based on two cycles of STIRAP.

Because there occurs only a single population-trapping state for the three-state STIRAP, the adiabatic evolution involves an Abelian transformation (process). Such a process can only change the phase of the quantum state, and so any observation of such a change requires the observation of interference between two states. However, a *non-Abelian* transformation, possible for a four- (or more) state system, can change the expectation value of a physical observable (for example atomic state population). Therefore, it is relatively easy to detect its consequences experimentally.

We study a four-state atomic system interacting with three external laser fields, whose carrier frequencies are ω_p, ω_s, and ω_d (see Fig.1). The frequencies need not be different if the laser polarizations give unique associations of pulses and transitions. The pulses are delayed in time, and obey the condition of exact one-photon resonance.

A version of such a system has been investigated previously [11]. Recently we suggested [12] a method to create any preselected coherent superposition of the atomic states 1 and 3, in a controlled and robust way, by using a sequence of three pulses in the four-state system of Fig. 1. We have shown that by changing the delay of the control pulse (the $2\leftrightarrow 4$ transition) with respect to the pump and Stokes pulses (which are themselves simultaneous) it is possible to control the final populations in states 1 and 3. A time-reversed version of the technique makes possible a determination of the phase occurring in a superposition of two atomic states.

In this present paper we provide further details of the underlying principles, and of the consequences of the non-Abelian geometric phase. We discuss various pulse sequences which produce a complete removal of population from the initial state 1. In these cases the final state is a superposition of states 3 and 4; the relative amounts of these two possibilities depend only on the geometric phase, and hence a measurement of population in state 4 (say) provides a simple determination of this phase. We discuss both intuitively and counterintuitively ordered pulse sequences.

It is important to note that in this atomic system the intermediate state may decay but we can choose the ordering of the laser pulses such that the population of state 2 will be negligibly small during the interaction time. Thus one can investigate population transfer between atomic bare states and the consequences of a non-Abelian geometric effect in a purely adiabatic situation when the interaction time is large. This is analogous to the situation in ordinary STIRAP. In this part of the paper we will assume the single-photon detunings vanish for all transitions to state 2. We remark, in section IV A, on the effects of nonzero detuning.

We solve the time-dependent Schrödinger equation (3) for a four-state atom subject to an excitation scheme in which three of the states are connected to a single exited state [a tripod linkage (see Fig. 1)]. The RWA Hamiltonian matrix of this system has the form

$$\hbar \mathsf{W}(t)=\frac{\hbar}{2}\begin{pmatrix} 0 & P(t) & 0 & 0 \\ P(t) & 0 & S(t) & Q(t) \\ 0 & S(t) & 0 & 0 \\ 0 & Q(t) & 0 & 0 \end{pmatrix}. \tag{6}$$

Here the Rabi frequencies $P(t)$, $S(t)$ and $Q(t)$ are real-valued functions of time. Although we do not account here for spontaneous emission, the usual application of the STIRAP procedure is to a system in which the intermediate state ψ_2 undergoes spontaneous emission not only to states ψ_1, ψ_3, and ψ_4 but to other unrecorded states (a loss of population from the four-state system). Thus it is usually desirable that population be kept from this state at all times.

A. Adiabatic states for the tripod linkage

It is easy to verify that $\mathsf{W}(t)$ of Eq. (6) has the following eigenvalues, two of which are degenerate:

$$\begin{gathered} \lambda_1(t)=\lambda_2(t)=0, \\ \lambda_3(t)=+\Omega_0(t)/2, \quad \lambda_4(t)=-\Omega_0(t)/2. \end{gathered} \tag{7}$$

Here

$$\Omega_0(t)\equiv\sqrt{Q(t)^2+S(t)^2+P(t)^2}.$$

The corresponding eigenvectors (the dressed states) are expressible in terms of two time-dependent angles $\vartheta(t)$ and $\varphi(t)$:

$$\tan\vartheta(t)=\frac{P(t)}{S(t)}, \quad \tan\varphi(t)=\frac{Q(t)}{\sqrt{P(t)^2+S(t)^2}}. \tag{8}$$

The angle $\vartheta(t)$ is the mixing angle used in standard STIRAP, and $\varphi(t)$ is an additional mixing angle related to the additional pulse. The two degenerate null-eigenvalue eigenvectors are

$$\Phi_1(t)=\begin{bmatrix}\cos\vartheta(t)\\0\\-\sin\vartheta(t)\\0\end{bmatrix},\qquad \Phi_2(t)=\begin{bmatrix}\sin\varphi(t)\sin\vartheta(t)\\0\\\sin\varphi(t)\cos\vartheta(t)\\-\cos\varphi(t)\end{bmatrix}, \tag{9}$$

while the remaining eigenvectors are

$$\Phi_3(t)=\frac{1}{\sqrt{2}}\begin{bmatrix}\cos\varphi(t)\sin\vartheta(t)\\1\\\cos\varphi(t)\cos\vartheta(t)\\\sin\varphi(t)\end{bmatrix},\qquad \Phi_4(t)=\frac{1}{\sqrt{2}}\begin{bmatrix}\cos\varphi(t)\sin\vartheta(t)\\-1\\\cos\varphi(t)\cos\vartheta(t)\\\sin\varphi(t)\end{bmatrix}. \tag{10}$$

When the $Q(t)$ pulse is absent we have the usual three-state atomic system and the adiabatic states turn into the well-known adiabatic states for STIRAP [2]. However, the occurrence of two degenerate null-eigenvalue states here adds complications, and flexibility, not present with three-state STIRAP.

In the adiabatic limit, which we assume to be applicable, the time derivative of the mixing angles $\vartheta(t)$ and $\varphi(t)$ is small compared to the splitting of eigenvalues, given by $\Omega_0(t)$. Under this condition there is negligible nonadiabatic coupling of dressed states $\Phi_1(t)$ or $\Phi_2(t)$ to the states $\Phi_3(t)$ or $\Phi_4(t)$. Therefore, in the adiabatic limit we must take into account only transitions between the degenerate dressed states $\Phi_1(t)$ and $\Phi_2(t)$.

The systems of interest for the present discussion are those for which the atomic states ψ_1, ψ_3, and ψ_4 are stable states. Spontaneous emission occurs only, if at all, from state ψ_2. The two degenerate adiabatic states $\Phi_1(t)$ and $\Phi_2(t)$ receive no contribution from state ψ_2 — these states are known as trapped states — and hence there is no difficulty in considering long pulses, as needed to ensure adiabatic evolution.

B. Non-Abelian adiabatic potential

The assumption that $\mathsf{W}(t)$ varies adiabatically implies that only the two degenerate adiabatic states $\Phi_1(t)$ and $\Phi_2(t)$ are needed for the construction of any state vector $\Psi(t)$. Let us consider the possibilities that initially $\Psi(t)$ is either one of these, by defining the state vector $\Psi_a(t)$ to be the state vector which evolves from the initial condition

$$\Psi_a(-\infty)=\Phi_a(-\infty). \tag{11}$$

At a later time we can write

$$\Psi_a(t)=\sum_b B_{ab}(t)\Phi_b(t). \tag{12}$$

Substituting this expansion into the Schrödinger equation, taking the scalar product with adiabatic states, and using the fact that the adiabatic states [Eq. (9)] are orthonormal, we find immediately that for null-eigenvalue adiabatic states (trapped states) the elements of $\mathsf{B}(t)$ obey the equation

$$\frac{d}{dt}B_{ba}(t)=-\sum_c A_{bc}(t)B_{ca}(t)\quad (a,b,c=1,2), \tag{13}$$

where

$$A_{bc}(t)\equiv\left\langle \Phi_b(t)\left|\frac{d}{dt}\right|\Phi_c(t)\right\rangle \tag{14}$$

form elements of a matrix $\mathsf{A}(t)$.

A matrix of formal solutions to Eq. (13) can be obtained by direct integration, as

$$\mathsf{B}(t)=\mathcal{P}\exp\left[-\int_{-\infty}^{t}\mathsf{A}(t')dt'\right], \tag{15}$$

where $\mathcal{P}$ indicates a time-ordered product. The exponentiated matrix $\mathsf{A}(t)$, often termed a potential, is geometrical because it depends on the Hilbert-space structure, i.e., the choice of bases. If we choose a different basis

$$\Phi'_a(t)=\sum_b U_{ab}(t)\Phi_b(t), \tag{16}$$

where $\mathsf{U}(t)$ is a unitary matrix, then $\mathsf{A}(t)$ will transform as components of a non-Abelian gauge potential

$$\mathsf{A}'(t)=\mathsf{U}(t)\mathsf{A}(t)\mathsf{U}(t)^{-1}+\dot{\mathsf{U}}(t)\mathsf{U}^{-1}(t). \tag{17}$$

It proves useful to follow the common approach of regarding the Hamiltonian as a functional of parameters S, P, and Q and to define a non-Abelian potential [6] A_ν comprised of elements

$$A_{ab\nu}=\left\langle \Phi_a\left|\frac{\partial}{\partial x^\nu}\right|\Phi_b\right\rangle,\quad \nu=1,2,3, \tag{18}$$

where x^ν is one of the three coordinates in the space S, P, or Q. A complete pulse sequence, from $t=-\infty$ to $t=+\infty$, returns all pulse amplitudes $P(t)$, $S(t)$ and $Q(t)$ to their initial values (zero). For a closed curve in parameter space (i.e., completed pulses) one obtains the result

$$\mathsf{B}(\infty)=\mathcal{P}\exp\left[-\sum_\nu \oint \mathsf{A}_\nu dx^\nu\right]. \tag{19}$$

Integral (19) transforms as Eq. (17) but without the inhomogeneous term $\dot{U}U^{-1}$. This means that the eigenvalues of the matrix $\mathsf{B}(t)$ are gauge invariant [6].

C. Coupling of degenerate dressed states

The computation of integral (19) is difficult in the general case, because a given segment in the path-ordered exponential integral does not commute with the next one. However, for our case, we have a two-state system, and after some algebra we find the solution of Eq. (13). For the $\mathsf{B}(t)$ matrix we have the simple formula

$$\mathsf{B}(t)=\begin{bmatrix} \cos\gamma(t) & \sin\gamma(t) \\ -\sin\gamma(t) & \cos\gamma(t) \end{bmatrix}, \tag{20}$$

where

$$\gamma(t)\equiv\int_{-\infty}^{t}\frac{d\vartheta(t')}{dt'}\sin\varphi(t')dt'. \tag{21}$$

After some time T the parameters return to their original value. This means that after the interaction we have the matrix

$$\mathsf{B}(\infty)=\begin{bmatrix} \cos\gamma_f & \sin\gamma_f \\ -\sin\gamma_f & \cos\gamma_f \end{bmatrix}, \tag{22}$$

where

$$\gamma_f=\oint_{\mathcal{C}}\frac{Q}{(P^2+S^2)\sqrt{Q^2+P^2+S^2}}(SdP-PdS), \tag{23}$$

and $\mathcal{C}$ is the closed path in the parameter space. The eigenvalues of matrix (22) are $\exp(\pm i\gamma_f)$. Thus γ_f, which is only defined modulo 2π, is gauge invariant, i.e., it does not depend on the choice of the adiabatic basis.

The geometric phase γ_f can be evaluated using Stokes' theorem. Specifically, to evaluate γ_f we must find the integral of a vector $\mathbf{V}\equiv\mathbf{R}/R^3$ through an area element $d\mathbf{S}$ in this pulse-parameter space:

$$\gamma_f=\oint_{\mathcal{S}}\mathbf{V}\cdot d\mathbf{S}. \tag{24}$$

Equation (24) expresses the phase γ_f as the flux through $d\mathbf{S}$ of a field $\mathbf{V}$ of a monopole with unit strength located at the point of degeneracy ($P=0$, $S=0$, $Q=0$). Therefore, $\gamma_f=0$ if $\mathcal{C}$ is in the radial plane; otherwise the geometric phase is generally nonzero.

III. POPULATION TRANSFER

For a quantitative analysis of the effects of the control pulse we assume Gaussian pulses of forms

$$\begin{aligned} S(t)&=\Omega_S e^{-(t+\tau)^2/T^2}, \\ Q(t)&=\Omega_Q e^{-t^2/T^2}, \\ P(t)&=\Omega_P e^{-(t-\tau)^2/T^2}. \end{aligned} \tag{25}$$

Here T and τ are the pulse lengths and the delay between the Stokes and pump pulses, respectively. In keeping with the terminology of STIRAP, we refer to the pulse ordering as *intuitive* when the pump pulse precedes the Stokes pulse, and *counterintuitive* when the Stokes pulse arrives before the pump pulse. In all cases we assume that initially all population resides in atomic state 1, and that we wish to transfer this population into state 3.

There are two possible pulse sequences which will achieve this goal with adiabatic evolution. These two sequences are associated with the two trapped adiabatic states $\Phi_1(t)$ and $\Phi_2(t)$, and they are distinguished by the choice of pulse ordering.

A. Counterintuitively ordered pulses

When the Stokes pulse precedes the pump pulse (counterintuitively ordered pulses), the angle ϑ is initially ($t\rightarrow-\infty$) zero. The trapped states then have the components

$$\Phi_1(-\infty)=\begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \Phi_2(-\infty)=\begin{bmatrix} 0 \\ 0 \\ \sin\varphi(-\infty) \\ -\cos\varphi(-\infty) \end{bmatrix}. \tag{26}$$

The initial condition (all population in atomic state 1) is fulfilled by associating the initial state vector $\Psi(-\infty)$ with the dressed state $\Phi_1(t)$, because $|\langle 1|\Phi_1(-\infty)\rangle|^2=1$ regardless of the order in which the Q pulse occurs. This is just the conventional STIRAP trapped state; after the conclusion of the final (pump) pulse (when $|\varphi|=\pi/2$) it coincides, apart from a phase, with the target state ψ_3.

The Q pulse does not affect $\Phi_1(t)$ directly, but because the two trapped states are degenerate, transitions may occur between them. Such transitions will alter the population transfer, and will place population into a coherent superposition of states ψ_1, ψ_3, and ψ_4.

From Eqs. (12) and (22) for the counterintuitive ordering of pulses, we obtain the probability $N_i=|\langle\Phi_i(\infty)|\Psi\rangle|^2$ for the adiabatic state $\Phi_i(t)$ after the interactions

$$\begin{aligned} N_1&=\cos^2\gamma_f, \quad N_2=\sin^2\gamma_f, \\ N_3&\approx 0, \quad N_4\approx 0. \end{aligned} \tag{27}$$

When there is no control pulse, $Q(t)\equiv 0$, then $\gamma_f=0$ and consequently $N_1=1$, $N_2=0$. This means that all the population remains in the trapped state Φ_1, as in STIRAP.

For counterintuitively ordered pulses we have the initial values $\varphi(-\infty)=0$ and $\vartheta(-\infty)=0$. Therefore, the desired initial condition (all population in atomic state 1) is fulfilled by associating the initial state vector $\Psi(-\infty)$ with the dressed state Φ_1 [see Eq. (9)]. At the end of the interaction, the following connection exists between the adiabatic states and the atomic states:

$$\Phi_1(\infty)\rightarrow-\psi_3, \quad \Phi_2(\infty)\rightarrow-\psi_4.$$

From Eqs. (9) and (27), we find that the atomic (bare state) populations are

$$P_3(\infty)=\cos^2\gamma_f, \quad P_1(\infty)\approx 0, \tag{28}$$

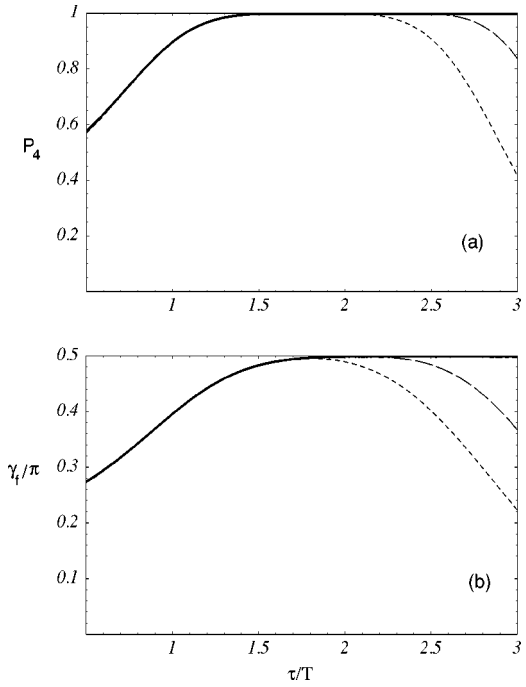

FIG. 2. Population of state 4 (a) and the geometrical phase (b) in units of π as function of the delay between S and P pulses. The results shown are obtained from numerical solutions to the Schrödinger equation for the Gaussian pulses given by Eq. (25); the dashed curve is for $\Omega_S T=\Omega_P T=\Omega_Q T=A$ and $A=100$. The short-dashed curve is for $A=50$. Analytical results are shown as a solid curve.

$$P_4(\infty)=\sin^2\gamma_f, \quad P_2(\infty)\approx 0.$$

Thus the population of state 4 is determined completely by the geometrical phase γ_f. In turn, the geometrical phase γ_f can be determined directly by measuring the population of state 4.

B. Numerical calculations

To illustrate this point, Fig. 2 shows the population of state 4, and the geometrical phase as a function of the delay between the Stokes and pump pulses. These results were obtained from the numerical solution of the Schrödinger equation for the Gaussian pulses [Eq. (25)], when $\Omega_Q T=\Omega_S T=\Omega_P T=100$ (dashed line) and $\Omega_Q T=\Omega_S T=\Omega_P T=50$ (short dash). The solid line gives the analytical solution [Eqs. (28) and (21)]. This figure confirms the good agreement of the numerical and analytical results in the adiabatic limit (large pulse area). For large delay between Stokes and pump pulses we have good population transfer from the state 1 to state 4, and the geometrical phase is equal to $\pi/2$.

The geometrical phase γ_f reaches its maximum value, $\pi/2$, when the delay τ is large. There is a nice geometrical interpretation for this value for the phase, as we see from the simple arguments of Sec. III C.

It is interesting to note that we can understand the behavior of the population of state 4 for large τ as an example of the usual STIRAP. In fact, for large delay we can neglect the influence of the Stokes pulse on the population transfer, i.e., one can consider a four-state atomic system as a three-state system whose counterintuitive interactions Q and P involve the states 1, 2, and 4. Thus we see that there is a mapping of the geometrical phase onto atomic population distributions: we can deduce the geometrical phase by measuring the population of the atomic states.

C. Geometrical interpretation

In the STIRAP process there occurs only a single population-trapping adiabatic state. The composition of this state can be described by a single angle, expressing the relative contribution of two atomic states to the null-eigenvalue adiabatic state. With passing time the Hamiltonian is completely described by the motion of a point in the two-dimensional parameter space spanned by the pump and Stokes Rabi frequencies (as x and y axes). As the pulses vary with time, this Hamiltonian point moves, and the state vector changes its angle in a corresponding plane.

Our considered extension involves two degenerate adiabatic states, which contain portions of three atomic states but exclude one of the atomic states. To describe this composition we require two angles; the subspace of the dressed states is a sphere rather than the plane of the simpler STIRAP case, and the position of the state vector in this subspace depends on three Rabi frequencies rather than the two of STIRAP. As the three pulses evolve in time, expressed by the changing location of the Hamiltonian point in a three-dimensional parameter space (Fig. 3), they force motion of the state vector on a sphere. The phase acquired by the state vector from a pulse sequence is the integral of a phase along a path on this sphere; it has the interpretation of a solid angle. Equations (23) and (24) provide alternative expressions for this total geometric phase.

The geometrical interpretation of the parameter space provides a simple explanation of predicted phases. For example, the pump and Stokes pulses alone, taken as nonnegative functions of time, force the statevector to move within one quarter of a circle in the plane defined by these two parameters. As long as there is no control pulse, the state-vector motion is planar and the enclosed solid angle is zero. The addition of a nonnegative control pulse moves the state vector away from this plane but, given the quarter-circle constraint of pump and Stokes pulses, allows motion only within one-eighth of the full sphere. The solid angle subtended by this motion is at most one eighth of the full 4π solid angle: the maximum geometrical phase is $\pi/2$.

The geometrical picture provides a simple understanding of the effect of changing the ordering of pump and Stokes pulses: reversing this order reverses the motion of the Hamiltonian point in the pump-Stokes plane of parameter space, and therefore reverses the motion of the state vector on the sphere, meaning that there is a change in sign of the geometrical phase.

D. Generalizations of counterintuitive pulses

Unlike the three-state case, there are many possible generalizations of the counterintuitive pulse order for four states. Let us consider a control pulse Q whose duration exceeds that of the combined S and P pulses. For simplicity we consider the choice $Q(t)=\text{const}$. Then we have late- or early-time $\vartheta=\pi/2$ and $\varphi=\pi/2$ [see Eq. (8)]. Therefore we have

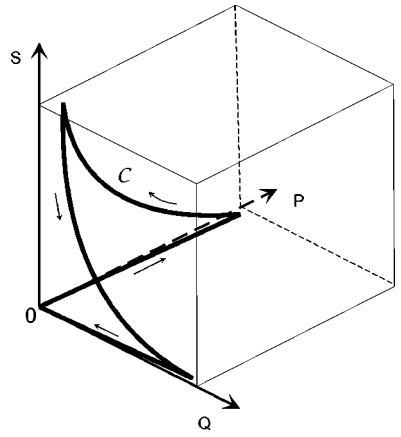

FIG. 3. Contour followed in the S-P-Q parameter space for the laser pulses given by Eq. (25). The parameters are $\Omega_S T=\Omega_P T=\Omega_Q T$. The delay between the Stokes and pump pulser is $\tau=T$.

the following connection between the adiabatic states and the atomic states at the end of the interaction [see Eq. (9)]

$$\Phi_1(\infty)\rightarrow-\psi_3, \quad \Phi_2(\infty)\rightarrow\psi_1.$$

From Eqs. (9) and (27) we find the atomic (bare state) populations to be

$$\begin{aligned} P_1(\infty)&=\sin^2\gamma_f, \quad P_2(\infty)\approx 0,\\ P_3(\infty)&=\cos^2\gamma_f, \quad P_4(\infty)\approx 0. \end{aligned} \tag{29}$$

The geometric phase γ_f is independent of pulse areas, such as $\Omega_P T$, $\Omega_S T$ and $\Omega_Q T$. It depends on the ratios τ/T, Ω_P/Ω_Q, and Ω_S/Ω_Q.

When $Q(t)\approx\Omega_Q=\text{const}$ is very large compared with $P(t)$ and $S(t)$ the atomic system will return to the state ψ_1 because $\varphi\approx\pi/2$ and thus [see Eq. (21)]

$$\gamma_f\approx\int_{-\infty}^{+\infty}\frac{d\vartheta}{dt}dt=\vartheta(+\infty)-\vartheta(-\infty)=\frac{\pi}{2}.$$

We can explain this in the following way. For large $Q(t)$ states ψ_2 and ψ_4 are not populated, and one can eliminate these states from the initial equations for the amplitudes [Eq. (3)]. After this procedure we have an effective two-state atom. The population transfer from the atomic state 1 to atomic state 3 can proceed in two ways, as expressed by the amplitudes PS/Q and $-PS/Q$. These two amplitudes interfere destructively, so that no net transfer occurs. Thus the atomic population will return to the state 1.

When the amplitudes of the pump and Stokes pulses large compared with the control pulse, and $\Omega_Q T\gg 1$, then the geometrical phase is small. We then find from Eq. (29) that complete population transfer occurs from the initial atomic state 1 to the target atomic state 3. The evolution of the atomic populations is plotted in Fig. 4. The lower frame of this figure shows the pulses. The parameters are $\tau=0.7T$, $\Omega_P T=\Omega_S T=500$, and $\Omega_Q T=35$. The upper frame shows the resulting populations. Because the time evolution is adiabatic the population of the excited state ψ_2 remains small. The geometrical phase, from Eq. (23), is $\gamma_f=0.126$. With Eqs. (29) this gives a value $P_3=0.984$ for the population of

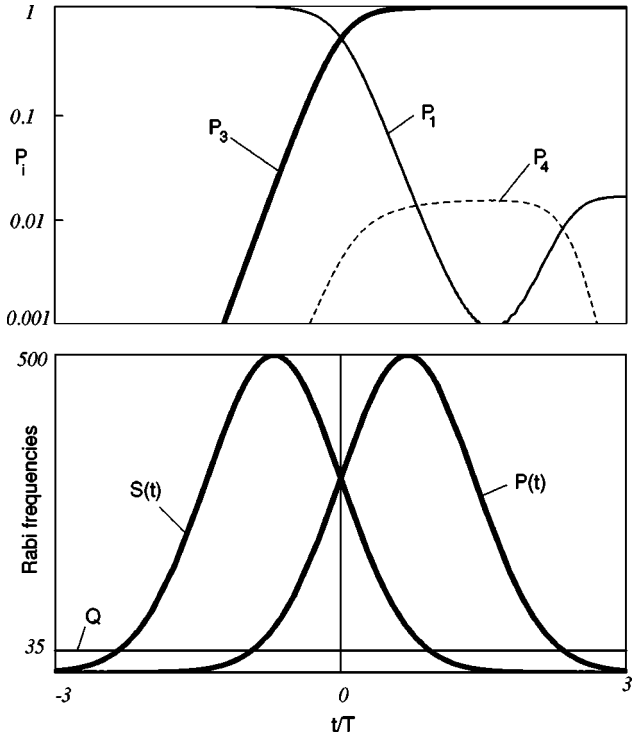

FIG. 4. Top: time evolution of atomic populations for counterintuitively ordered pulses. The population of the intermediate state 2 remains zero. Bottom: pulses producing this excitation; the amplitudes are given in units of T^{-1}. Parameters are $\Omega_P T=\Omega_S T=500$, $\Omega_P T=35$, and $\tau=0.7T$.

state 3 after the interaction. The numerical solution of Eq. (3) gives the same numerical value. It is interesting to note that at the end of the Stokes interaction there is population in 4. The Q and P pulses together transfer this population into state 1 via adiabatic evolution, as in STIRAP. The transfer into state 4 involves three simultaneous pulses, and so it has no such simple explanation.

The variation of the final population of state 3 with the areas of pump and Stokes pulses is shown in Fig. 5, for $\tau=0.7T$, with $\Omega_Q T=35$ and $\Omega_Q T=0$ (STIRAP).

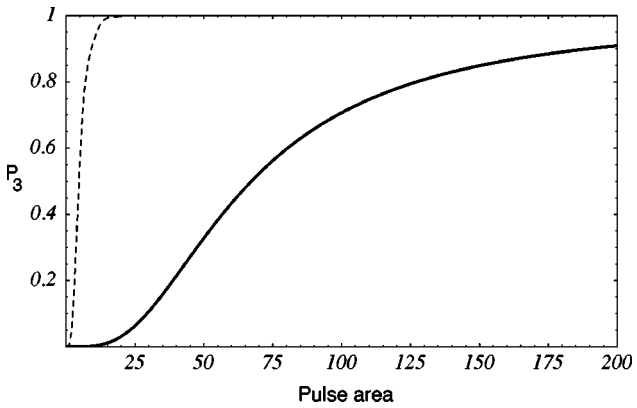

FIG. 5. Population of state 3 as a function of the area of Stokes and pump pulses (counterintuitive sequence with delay $\tau=0.7T$). Solid curve, no control pulse ($\Omega_Q T=0$); dashed curve, control pulse $\Omega_Q T=35$. Results were obtained from a numerical solution of the Schrödinger equation for the Gaussian pulses of Fig. 2.

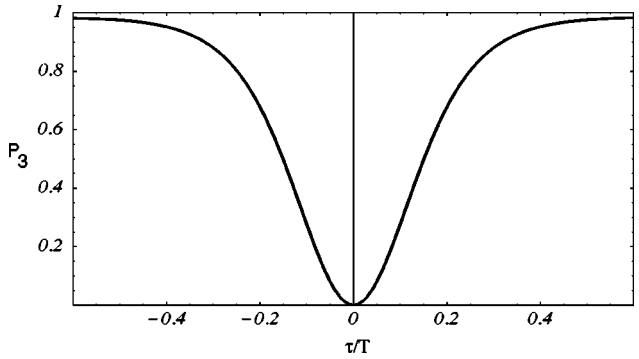

FIG. 6. Population of state 3 as a function of the delay between pump and Stokes pulses. The negative delay $\tau<0$ is a counterintuitive sequence, and the positive delay $\tau>0$ is an intuitive pulse sequence. The curve is symmetrical about $\tau=0$, and agrees with Eq. (29). Parameters are $\Omega_S T=\Omega_P T=500$ and $\Omega_Q T=35$.

As can be seen, the evolution differs significantly in these two case. This is not surprising. For any adiabatic process involving only nondegenerate adiabatic states, the transition between adiabatic states is proportional to $e^{-\Omega T}$, where ΩT is the effective pulse area [13]. The dependence of the final population on the area was considered in Ref. [14] for the STIRAP process. They found breakdown of the Dykhne-Davis-Pechukas [13] exponential dependence of the nonadiabatic transition probability. As can be see from Fig. 5, population growth does not follow the simple pattern exp $(-\Omega T)$ when the control pulse is present. The coupling between degenerate trapped states Φ_1 and Φ_2 changes the population dynamics dramatically.

In Fig. 6 we show the dependence of the population transfer from state 1 to state 3 on the delay of the pump and Stokes interactions when $\Omega_P T=\Omega_S T=500$ and $\Omega_Q T=35$ for Gaussian pulses [Eq. (25)] and $Q(t)=$const. A negative delay means counterintuitive pulses and positive delay means intuitively ordered pulses. Thus when $\Omega_Q T\gg 1$ the variation of the transfer probability with delay τ is symmetric with respect to $\tau=0$; both counterintutive and intuitive pulse sequences produce the same results. Let us consider the intuitively ordered pulses in more detail.

E. Intuitively ordered pulses

When the pump pulse precedes the Stokes pulse, then we have initially $|\vartheta|=\pi/2$, and state $\Phi_1(t)$ does not fit the initial conditions (because then $|\langle 1|\Phi_1(-\infty)\rangle|=0$). The state vector coincides initially with state $\Phi_2(t)$ if we require that the Q pulse extends earlier and later than the pump pulse and the Stokes pulse, so that initially and finally $|\varphi|=\pi/2$. For $\varphi=\pi/2$ the trapped states are

$$\Phi_1(t)=\begin{bmatrix}\cos\vartheta(t)\\0\\-\sin\vartheta(t)\\0\end{bmatrix},\quad \Phi_2(t)=\begin{bmatrix}\sin\vartheta(t)\\0\\\cos\vartheta(t)\\0\end{bmatrix}. \tag{30}$$

When the pump pulse precedes the Stokes pulse, then the initial trapped states are

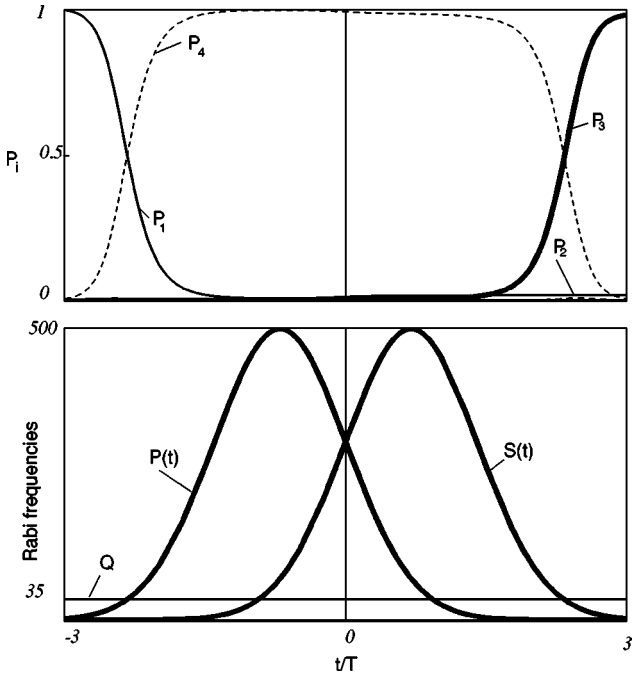

FIG. 7. Top: time evolution of the atomic populations for intuitively orderd pulses, for parameters the same as those in Fig. 4. Bottom: the pulses.

$$\Phi_1(-\infty)=\begin{bmatrix}0\\0\\-1\\0\end{bmatrix},\quad \Phi_2(-\infty)=\begin{bmatrix}1\\0\\0\\0\end{bmatrix}, \tag{31}$$

and the state vector will begin as the adiabatic state $\Phi_2(-\infty)$. If, in addition, the pump pulse ends at $t=t_P$, prior to the Stokes pulse, we have

$$\Phi_2(t_P)=\begin{bmatrix}0\\0\\1\\0\end{bmatrix}, \tag{32}$$

i.e., the dressed state $\Phi_2(t_P)$ coincides with the target state ψ_3 at the end of the pump-Stokes pulse sequence. Thus when the Q pulse is present at the start and at the termination of the pump-Stokes pulses, then it is possible to achieve complete adiabatic population transfer with intuitively ordered pulses. However, unlike the case of counterintuitively ordered pulses, this transfer places population into state ψ_4 during the time evolution; see Fig. 7 [the parameters are the same as in the counterintuitive case (Fig. 4)].

Although the first step of the population transfer from state 1 to state 4 is a STIRAP process, in which the interaction Q acts as the counterintuitively ordered Stokes pulse, the transition from state 4 to the final atomic state 3 is not. In this step the interaction Q occurs in the intuitive ordering compared to the transition between states 2 and 3.

The transition from atomic state 1 to atomic state 3 takes place via the trapping state $\Phi_2(t)$. This can be understood by examining the dressed state $\Phi_2(t)$ when, at intermedia-

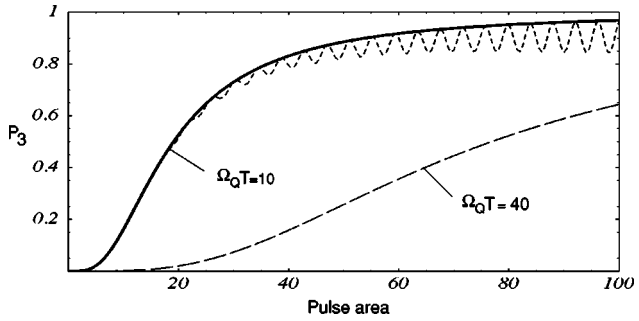

FIG. 8. Variation of the final population of state 3 with pulse area. The solid curve refers to the counterintuitive sequence, with $\Omega_Q T=10$; the short dash corresponds to the intuitive sequence, again with $\Omega_Q T=10$; the dashed curve is for both sequences; and the large area $\Omega_Q T=40$ (intuitive and counterintuitive sequences are indistinguishable). This figure demonstrates how the adiabatic limit is approached, as predicted by the analytical results [Eq. (29)], when the area of the Q pulse is increased.

tetimes, S and P are not small compared to Ω_Q, so that we have $\varphi\approx 0$, and the dark state is

$$\Phi_2(t_P)=\begin{bmatrix} 0 \\ 0 \\ 0 \\ -1 \end{bmatrix}. \tag{33}$$

The final populations of the atomic states in the adiabatic limit are the same as given by Eq. (29). After the completed population transfer the atoms are in state 3.

As shown in Ref. [15] for the intuitive case when $\Omega_Q T=0$, the final population of state 1 is zero but the final populations of the states 2 and 3 oscillate with the effective Rabi frequency of the two pulses. However, the situation changes qualitatively for large $\Omega_Q T$; one then has a monotonic dependence of the population in state 3 on the area of the Stokes and pump pulses.

It is interesting to note that the population of level 2 is small during the adiabatic transfer between the initial and final states. Thus in the adiabatic limit we have the same final population in the states 1 and 3, in accordance with Eq. (29). In order to show this, in Fig. 8 we plot the final population of state 3 as a function of $\Omega_Q T$, for counterintuitive and intuitive pulse sequences. For large $\Omega_Q T=40$ the population of state 3 as a function of the area of the Stokes and pump pulses is the same for the both cases. However, for $\Omega_Q T=10$ the intuitive and counterintuitive sequences give different results. For the intuitive sequence the evolution is not adiabatic. Equations (29) and (28) are valid only in the adiabatic limit, i.e.,

$$\left|\frac{d\vartheta(t)}{dt}\right| \ll \Omega_0(t), \quad \left|\frac{d\varphi(t)}{dt}\right| \ll \Omega_0(t), \tag{34}$$

or, for pulse duration T,

$$\Omega_0 T \gg 1.$$

In fact, in atomic beam experiments (see for example, Refs. [16], [17]) pulse areas as large as $\Omega_0 T\sim(100-500)$ have been used.

IV. COMMENTS

A. Effects of Detuning

For application to experiments involving atomic beams moving transversely across laser beams, a consequence of the velocity distribution is to change the interaction time of the individual atoms with the laser beams. This amounts to a rescaling of the time unit τ/T, but the integral [Eq. (23)], is not changed. Thus the geometrical phase is insensitive to the longitudinal velocity distribution of an atomic beam.

The present results were derived with the simplifying assumption that the carrier frequencies of the three pulses P, S, and Q are each tuned to resonance with the respective transition. However, the results also apply when the pulses have the same nonzero detuning. Then there still exists a pair of null-eigenvalue adiabatic states Φ_1 and Φ_2. The common nonzero detuning alters only the structure and eigenvalues of the two adiabatic states Φ_3 and Φ_4. These states are not involved in the adiabatic evolution.

B. A Comment on null eigenvalues

In the simplest coherent excitation by resonantly tuned fixed-frequency pulses, population transfer occurs between two states having common unperturbed energy eigenvalues in the rotating-wave approximation — the atomic states combined with field states are degenerate before and after the interaction which couples them. It is convenient, and always possible, to take this common energy to be the (arbitrary) zero point for expressing other energies; these two states then share a null eigenvalue of the unperturbed RWA Hamiltonian. During the course of pulsed interaction the adiabatic eigenvalues of the two-state single-pulse system differ from zero.

In the STIRAP procedure, based on three states and two pulses, there occurs a single eigenvalue of the full adiabatic Hamiltonian which remains at all times equal to the original degenerate energy (zero, by convention). In the tripod linkage considered here, with four states and three pulses, there are two null adiabatic eigenvalues during the interaction: a pair of dressed states remain degenerate at all times. As we show, the location of the state vector within the restricted subspace of the two strongly coupled adiabatic states is not only a phase angle (the geometric phase), but it has a direct association with observable population transfer.

More elaborate systems, with more pulses, may involve population transfer between any number of states sharing a null eigenvalue of the adiabatic Hamiltonian. Such cases offer opportunities to examine couplings between more than two closely coupled null-eigenvalue adiabatic states [18].

C. Experimental realizations

There are many ways to realized a tripod linkage pattern within atomic systems examined in earlier studies of STIRAP, such as metastable neon (Ne*) [16] and metastable helium (He*) [7].

For the Ne* state ψ_1, we can take the metastable state $(2p^5 3s)\,^3P_0(M=0)$, which can be coupled to the state $(2p^5 3p)\,^3P_1(M=0)$ (state ψ_2) by linearly polarized radiation of wavelength 616 nm. The state $^3P_1(M=0)$ can be coupled with $(2p^5 3s)\,^3P_2(M=1)$ (state ψ_4) and $^3P_2(M=-1)$ (state ψ_3) by a laser ($\lambda=588$ nm) with $\sigma_\pm$ polarization [16].

For He*, the state 2^3S_1 of helium is metastable, and can be coupled to the level $^3P_0$ by three laser beams of the same wavelength and with different polarizations. The state $^3P_0(M=0)$ (state ψ_2) can be coupled with $^3S_1(M=1)$ (state ψ_1), $^3S_1(M=-1)$ (state ψ_3), and $^3S_1(M=0)$ (state ψ_4) by a laser ($\lambda=1083$ nm) with $\sigma_\pm$ and π polarization, for example, the pump pulse $P(t)$ with σ_+ polarization, the Stokes pulse $S(t)$ with counterpropagating σ_- polarization, and the control pulse $Q(t)$ with π polarization. In this case we can derive all of the pulses from a single laser. Alternatively, we can use $\lambda=389$ nm to couple the metastable state to the next-higher-lying level, giving the option of easily detecting decay from there.

Note that not only the control laser, but also the pump and Stokes lasers, may be at a given wavelength, or that the pump and Stokes laser are at one wavelength, while the control laser is at the other. The choice depends on availability, and needs to be judged eventually based on other considerations. The power of suitable cw lasers (say 50W/cm^2) is adequate to satisfy the adiabaticity conditions (see Ref. [8]).

V. CONCLUSIONS

In this paper we have shown that, in the adiabatic limit and with resonant excitation, the nature of the transition between four atomic states, linked in a tripod configuration, involves only a geometrical phase. Consequently the phase does not depend on the exact value of the Rabi frequencies as long as the conditions for adiabatic following are satisfied.

The stimulated Raman adiabatic passage (STIRAP) process (i.e. a delay between Stokes and pump pulses), during which the Rabi frequencies traverses a closed path in parameter space and the state vector evolves adiabatically, provides a remarkable possibility of experimental determination of a geometrical phase.

Even when the lasers are each taken to be resonant with the associated Bohr frequencies, the excited state 2 is never populated during the adiabatic evolution. The absence of spontaneous emission preserves the coherence of the atomic wave function. At all times the atom is in a trapped state, which suppresses the evolution of a nonzero dynamical phase and permits the observation of the geometrical phase, even if small.

We have shown how to measure this phase, say in an atomic beam experiment, by observing the population in atomic bare states. When pump and Stokes pulses are applied in either counterintuitive or intuitive orders, one can obtain high population transfer from the initial state to the target state. In the adiabatic limit the robustness of this transfer (i.e. insensitivity to details such as pulse area) is equivalent to that of the STIRAP process.

ACKNOWLEDGMENTS

The work of R.U. was supported by the Alexander von Humboldt Foundation. The work of B.W.S. was supported in part under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48, and by the Alexander von Humboldt Foundation. We gratefully acknowledge a NATO Collaborative Research grant, support by the Deutsche Forschungsgemeinschaft, and partial support by the EU network ''Laser Controlled Dynamics of Molecular Processes and Applications'' (Network No. ERB-CH3-XCT-94-0603). We are indebted to M. Fleischhauer and S. Guérin for stimulating discussions.

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