Piecewise Adiabatic Passage with a Series of Femtosecond Pulses

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We develop a method of executing complete population transfers between quantum states in a piecewise manner using a series of femtosecond laser pulses. The method can be applied to a large class of problems as it benefits from the high peak powers and large spectral bandwidths afforded by femtosecond pulses. The degree of population transfer is robust to a wide variation in the absolute and relative intensities, durations, and time ordering of the pulses. The method is studied in detail for atomic sodium where piecewise adiabatic population transfer, as well as the induction of Ramsey-type interferences, is demonstrated.

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In the past decade two strategies have emerged as being most useful for achieving quantum control. These are coherent control (CC) with broad and narrow-band laser pulses [1] and "adiabatic passage" [2]. Control with broadband pulses relies on the possibility to shape the pulses both in the time and frequency domains [3]. This enables realization of various CC schemes [1], which utilize coherent laser radiation to induce interferences between indistinguishable quantum pathways. One can enforce the desired evolution of a quantum system with high accuracy due to the ability to control quantum interferences. Though applicable to a wide range of problems, control with ultrashort pulses is often highly sensitive to the durations and exact shapes and intensities of the laser fields used.

Adiabatic passage (AP) is a complementary control strategy that enables the execution of complete population transfer between quantum states, using pulses that are long on the time scale of the system's evolution. The AP approach, when exercised in the context of bound-bound [2,4] and bound-free [5] transitions can bring about high yields, often approaching unity. In contrast to control with shaped pulses, the AP method is very robust with respect to variations in the durations and amplitudes of the laser pulses used [2,6]. Unfortunately, the long pulse lengths can be detrimental in certain applications, such as quantum computation, where decoherence is to be strictly avoided. Moreover, the adiabaticity condition requires slowly changing field envelopes which are hard to shape due to their narrow spectral bandwidth. This limits the set of quantum control tools and the range of achievable goals of AP.

In this work we propose a way of merging the strategies of adiabatic passage and the control with shaped pulses (for a review of ways of combining AP with CC see Ref. [7]). We do so by performing the population transfer in a piecewise manner using a series of femtosecond pulses. Our technique combines the robustness of AP with the flexibility afforded by the use of broadband light sources. The present proposal extends the concepts of "coherent accumulation" [8] and the Ramsey experiment [9], where a slow process is implemented in a piecewise manner using a train of short, mutually coherent, laser pulses. As our primary example, we consider below the stimulated Raman adiabatic passage (STIRAP) [2] scheme. In its simplest version, population from an initial quantum state $|1\rangle$ is transferred to a target state $|3\rangle$ via an intermediate state $|2\rangle$ by means of two narrow-band laser pulses, the "pump" $E_p(t) \equiv \mathrm{Re}\{\varepsilon_p(t)e^{-i\omega_p t}\}$, tuned on resonance with the $\epsilon_2 - \epsilon_1$ transition and the "dump" $E_d(t) \equiv \mathrm{Re}\{\varepsilon_d(t)e^{-i\omega_d t}\}$, tuned on resonance with the $\epsilon_3 - \epsilon_2$ transition. The system is described under the rotating wave approximation in the interaction representation [1,2]; when both laser fields are turned off, the state amplitudes do not evolve. In the presence of the laser fields, the eigenstate corresponding to the zero eigenvalue ("null vector") is

$$\Psi_0 = \sin\theta|1\rangle - \cos\theta|3\rangle, \qquad \tan\theta = \Omega_d/\Omega_p^*, \quad (1)$$

where $\Omega_p(t) \equiv \mu_{2,1}\varepsilon_p(t)$ and $\Omega_d(t) \equiv \mu_{3,2}\varepsilon_d(t)$, and μ_{ij} being the dipole matrix elements. If at the beginning of the process we set $|\Omega_d| > 0$ and $\Omega_p = 0$ and at the end we let $|\Omega_p| > 0$ and $\Omega_d = 0$ (the "counterintuitive" pulse ordering [2]), the null vector will evolve from state $|1\rangle$ to state $|3\rangle$ without ever populating the intermediate state $|2\rangle$. The wave function $\Psi(t)$ will follow the null vector, making an adiabatic passage from $|1\rangle$ to $|3\rangle$. The particular values of the Rabi frequencies do not matter as long as we meet the adiabaticity condition [2]

$$|\dot{\theta}| \ll \sqrt{|\Omega_p|^2 + |\Omega_d|^2}. \quad (2)$$

Figures 1(a) and 1(b) show a computation for an ordinary ("reference") AP for $|1\rangle = 3s$, $|2\rangle = 3p$, and $|3\rangle = 3d$ electronic states of the Na atom, using the empirically derived $\mu_{i,j}$ matrix elements [10]. In Figs. 1(a) and 1(b) we use two circularly polarized laser pulses with sine-squared $\varepsilon_{p(d)}$ field envelopes and temporal duration of 0.75 ps (FWHM in amplitude). The pulses are applied with a

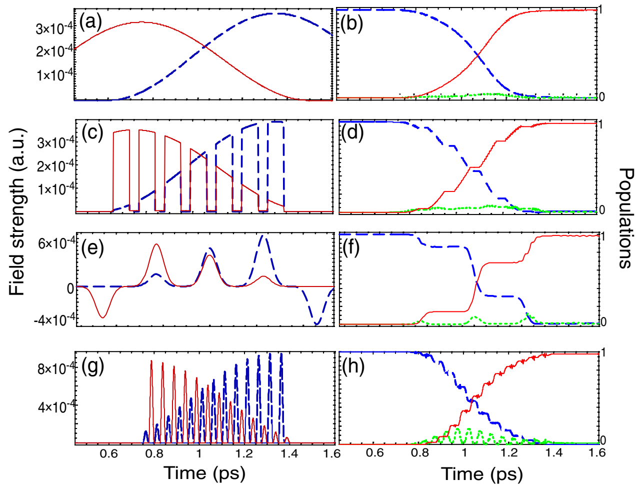

FIG. 1 (color online). Four types of PAP processes: (a),(c),(e),(g)—the pulse sequences; dashed blue—the pump, firm red—the dump. (b),(d),(f),(h)—the populations ($|b_i(t)|^2$); dashed blue—$i = 1$, dotted green—$i = 2$, firm red—$i = 3$. (a,b) The reference AP; (c),(d) PAP with sharp turn-ons and turn-offs; (e),(f) and (g),(h) PAP with smooth overlapping and nonoverlapping pulses.

0.6 ps delay and have central wavelengths $\lambda_p = 2\pi c/\omega_p = 589$ nm and $\lambda_d = 2\pi c/\omega_d = 819$ nm. Each pulse is capable of driving about three Rabi oscillations within the time pump-dump overlap. We observe a complete transfer of population from the state $|1\rangle$ to the state $|3\rangle$, while never significantly populating the state $|2\rangle$. Note that in the counterintuitive ordering, only the overlapping region of the pulses is important, since the population of the states does not change when only one of the pulses is present.

We now modify the above scenario by letting the material system evolve according to the above scheme until time t_1, at which point both laser fields are abruptly turned off. At some later time t_2, we abruptly turn both fields back on, making sure that the absolute phase of each field is retained, i.e., the relative phase of the carrier oscillations after t_2 and before t_1 is zero. Since the coefficients $b_i(t)$ do not evolve during the zero-field period, $b_i(t > t_2)$ continues to follow the adiabatic passage with $b_i(t_1) = b_i(t_2)$ serving as a new starting point. Repeating the turn-off–turn-on procedure many times allows us to execute the AP process in a controlled piecewise manner.

Figures 1(c) and 1(d) demonstrate the action of a pulse sequence obtained by turning off the original reference STIRAP fields at periodic intervals, while scaling up $\varepsilon_{p(d)}(t)$ during the "on" periods, as described in detail below. As shown, the resulting pulse sequence closely reproduces the effect of an ordinary AP. Moreover, we find that this "piecewise adiabatic passage" (PAP) procedure is insensitive to the particular shape and strength of the pulses, as well as to the durations of the "off" periods.

A soft turn-on–turn-off version of PAP is shown in Figs. 1(e)–1(h). In panels (e),(f) the pump and the dump laser fields are obtained by the spectral phase shaping of 75 fs pulses:

$$\varepsilon^{\rm shaped}_{p(d)}(\omega) = \varepsilon^{\rm unshaped}_{p(d)}(\omega) F_{p(d)}(\omega), \tag{3}$$

where $F_{p(d)}(\omega) = \exp\{iA_{p(d)} \sin[\Delta T(\omega - \omega_{p(d)})]\}$ with $A_{p(d)} = \mp 1.2$, $\Delta T = 229$ fs. The resulting pulse sequence, shown in Fig. 1(e), is comprised of copies of the original transform-limited femtosecond pulse of duration τ separated in time by ΔT, and with smoothly changing amplitudes defined by the value of A [11]. We see that the soft turn-on–turn-off PAP is as successful in bringing about the desired population transfer as the abrupt turn-on/turn-off version discussed above. The negative field envelopes (opposite sign of the field) in Fig. 1(e) do not influence the population transfer, and are due to the sinusoidal phase modulation of the spectrum [11].

It is instructive to consider each pump-dump segment of the combined pulse train as a deviation from the smoothly varying reference AP field [see Fig. 1(a)]. We do so by writing the interaction Hamiltonian as $H(t) = H_0(t) + H_1(t)$, where $H_0(t) \equiv -\mu E_0(t)$, and $E_{0p(d)}(t) = \mathrm{Re}\{\varepsilon_{0p(d)}(t) \exp(-i\omega_{0p(d)} t)\}$ is the field from the reference Hamiltonian. The evolution operator in PAP will be equal to that in the reference AP, if

$$\begin{aligned} S_n &= \mathcal{T} \exp\left[-i \int_{t_n-\tau/2}^{t_n+\tau/2} H_0(t)dt - i \int_{t_n-\tau/2}^{t_n+\tau/2} H_1(t)dt\right] \\ &= \mathcal{T} \exp\left[-i \int_{t_n-\tau/2}^{t_n+\tau/2} H_0(t)dt\right] \equiv S_{0n}, \end{aligned} \tag{4}$$

where $\mathcal{T}$ denotes the time ordering operator and S_{0n} is the evolution operator of the reference AP during the nth segment of duration τ. By requiring $\int_\tau H dt = \int_\tau H_0 dt$, condition (4) can be satisfied within the first order of the shake-off perturbation theory [12] with respect to $\varpi\tau$, where $\varpi \sim \Omega_{p,d}$ is the typical frequency in the dressed Hamiltonian. Therefore, in each segment we can replace the reference Hamiltonian H_0 by a piecewise H of the same area, as long as each of the fields drives no more than a fraction of a Rabi cycle in that segment. Within the same level of accuracy, the actions S_n and S_{0n} are composed of independent actions of the pump and dump fields. In order to enforce condition (4), we require that $\int_\tau \varepsilon_{p(d)} dt = \int_\tau \varepsilon_{0p(d)} dt \simeq \varepsilon_{0p(d)}(t_n)\tau$. Constructed this way, the field of the pulse train is only defined by the coarse-grained $\varepsilon_{0p(d)}(t_n)$ profile, and is insensitive to its short-time behavior. At the same time, since the pulse area of the reference AP satisfies the adiabaticity condition, so does the area of the PAP pulse train. Thus constructed PAP is as robust to the particular choice of $\varepsilon_{0p(d)}(t_n)$ as the reference AP. Effective coarse graining of the evolution [Eq. (4)] explains the observed robustness of the transfer both to the pulse shapes, and to additional noise in the pulse envelopes (for an analysis of noise effects in a conventional STIRAP, see Ref. [13]). The discretized coarse-grained description is complementary to the search of the generalized Floquet states of Ref. [14], and can help in deriving conditions of complete population transfer without relying on the particular field spectrum (see below).

The phases of the pump and dump pulses can be used in PAP as additional control knobs. Similar to the Ramsey interference effect [9], if either one or both fields are detuned off the exact resonance, the probability transfer in PAP becomes sensitive to the duration of the turn-off periods. In Fig. 2(a) we show PAP performed with just two pairs of (75 fs) pump and dump pulses inducing one-photon transitions between the levels $3s$, $3p$, and $3d$ of Na. Panel (b) depicts the final population of the state $3d$ as a function of the duration of the turn-off periods δt. Figures 2(c) and 2(d) show the same results for the pulse trains composed of 100 segments with linearly increasing (decreasing) pump (dump) field strengths. The dump wavelength was set to ~818 nm, 1 nm away from the exact $3p$-$3d$ resonance.

We first note that even as few as two pairs of pulses suffice to reproduce the effect of conventional AP. It can be also seen in Figs. 2(b) and 2(d), that there is a strong dependence of the final state population on the duration of the turn-off period(s). The sharp peaks seen in the latter case are analogous to the Ramsey fringes; the longer the pulse trains the narrower the fringes. This suggests that applying PAP, implemented with a frequency-comb source [15], to a metastable target state might present a useful alternative to the precision measurements of atomic transitions and frequency standards.

There is an underlying connection between PAP and the coherent accumulation schemes studied in Ref. [8]. Consider the example shown in Figs. 1(g) and 1(h) in which the pump and dump pulses do not overlap. Within each non-overlapping pulse pair, the pump first drives the population into the intermediate level, from which it gets subsequently dumped into the target state. We have ascertained numerically that the accumulating action of this coherent non-overlapping scheme is insensitive to the intensity of the pump and dump trains, as long as their envelope mimics that of the reference AP. Unlike the preceding examples, the population is transferred, in small portions, via the intermediate level. Yet, similarly to PAP with overlapping pulses, the transfer is complete and robust due to the effective coarse graining.

A very informative, though, in general, incomplete, description of the system is given by following the trajectory of the amplitudes vector **a** shown in Fig. 3(a) [2,16]. If Ω_p and Ω_d are real, and if at $t=0$ $b_1=1$, $b_2=b_3=0$, then $\mathbf{a}_x=\mathrm{Re}(b_1)$, $\mathbf{a}_y=-\mathrm{Im}(b_2)$, and $\mathbf{a}_z=\mathrm{Re}(b_3)$. Each pump pulse, transferring populations between states $|1\rangle$ and $|2\rangle$, rotates **a** by a small angle $\alpha_P=\int_\tau \Omega_P(t)dt$ about the z axis. Likewise, each dump pulse rotates **a** about the x axis by an angle $\alpha_D=\int_\tau \Omega_D(t)dt$. These two rotations result in an overall rotation of **a** by an angle α_0 about an axis defined by the (θ_0, ϕ_0) polar and azimuthal angles, given in the lowest-order expansion of α_P, α_D as,

$$\alpha_0=\sqrt{(\alpha_P^2+\alpha_D^2)/2}, \tag{5}$$

$$\phi_0=-\alpha_P/2, \qquad \tan\theta_0=\alpha_D/\alpha_P. \tag{6}$$

As $\Omega_{p(d)}(t)$ evolves, the otherwise stable (θ_0, ϕ_0) vector, which coincides with the null vector of our reference AP in the limit of small pulse areas [(see Eq. (1)], moves slowly from being aligned along the x axis to being aligned along the z axis. As shown in Fig. 2, as long as the individual α_P and α_D are small, the **a** vector will faithfully follow the (θ_0, ϕ_0) null vector. A trajectory of the vector **a** on the sphere of states, composed of piecewise rotations around the moving center (θ_0, ϕ_0), resembles a cycloid. Coarse-grained adiabaticity can be shown to be guaranteed as long as

$$\Delta\theta_0 \ll \sqrt{(\alpha_P^2+\alpha_D^2)/2}. \tag{7}$$

In the limit of small rotations, this condition reduces, up to an insignificant numerical factor, to the adiabaticity condition, Eq. (2), of conventional AP.

We thus expect the overall evolution to be robust as long as Eq. (7) is satisfied and the α_P, α_D angles at each step are small. These expectations were verified numerically for various sequences of alternating rotations with slowly varying amplitudes. Figure 3(b) depicts two such trajecto-

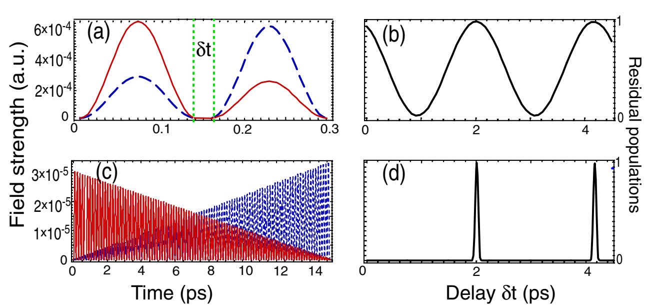

FIG. 2 (color online). Ramsey interference in PAP. (a),(c) The field envelopes of the pump (dashed blue) and dump (red) pulse sequences. The green dotted lines in panel (a) mark the onset of the turn-off period(s). (b),(d) The final $|b_3|^2$ population as a function of the duration of the turn-off period(s).

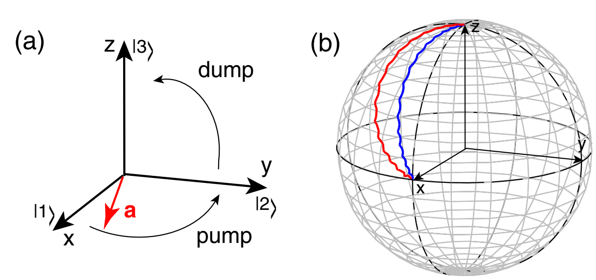

FIG. 3 (color online). (a) Depiction of the state vector **a**. (b) Two sample trajectories of the **a** vector during the PAP process. Both the pump and the dump sequences of small rotations rotate the vector by an overall angle of $10\times 2\pi$. The left (right) trajectory corresponds to 50 (100) pulses in each sequence.

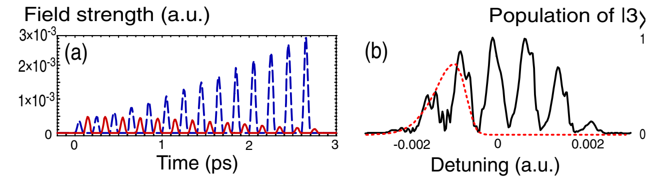

FIG. 4 (color online). Field envelopes of the (2 + 1) PAP, and the final population of the target state as a function of the dump detuning in PAP (black), and in the reference conventional STIHRAP (dashed red).

ries. Each of them starts at the state $|1\rangle$ and follows the AP route slightly outside the zeroth meridian (i.e., somewhat populating state $|2\rangle$) to coincide finally with state $|3\rangle$.

The ability to alternate between the pump and dump segments can be useful in minimizing deleterious effects of the Stark shifting of levels. This is particularly the case in the two-photon + one-photon ("2 + 1") Stimulated hyper-Raman adiabatic passage (STIHRAP [17,18]) whose implementation with short laser pulses is considered very difficult due to the strong time-dependent ac Stark shifts of the levels. Although one can transfer the population into the target level by detuning both the pump and the dump lasers from the exact resonance [17,18], the robustness of the method is strongly reduced since the conditions connecting the field-induced Stark shifts with the required frequency offsets are stringent.

We have considered the action of strong pump pulses which drive a two-photon transition from the ground state of Na ($3s$) to an intermediate state $4s$, while the dump pulses cause a one-photon down-transition from $4s$ to the target state $3p$. Figure 4(a) shows the pulse sequence with nonoverlapping pump and dump segments which drives the (2 + 1) hyper-Raman PAP. When the relatively weak dump pulses transfer populations to the target state, the strong pump field is turned off. As a result, the levels are not Stark-shifted away from the dump resonance. Stark shifts are still present during each pump segment, introducing an additional relative phase to the state amplitudes.

We set the center wavelength of the pump field to 796 nm (instead of the resonant wavelength of 777 nm) to compensate for the average shift of the levels during the pump segments. An offset in the dump frequency is only needed to compensate for the relative phase between the states $|2\rangle$ and $|3\rangle$ acquired during one pump pulse. Thus the probability of the successful transfer is a periodic function of the dump detuning with the period $2\pi/T$, where T is the delay between the individual pulses. This can be seen in Fig. 4(b) where the final population of the target state is shown as a function of the dump detuning. For comparison, we show the same dependance for the reference STIHRAP with smooth pulses [19]. One can see that the conditions of successful implementation of a (2 + 1) PAP are drastically different from those for a conventional (2 + 1) STIHRAP: instead of carefully matching the detuning with the Stark shifts [18], the field has to satisfy the Ramsey-type condition within the bandwidth of a single pulse. Further studies are under way in order to find out whether (2 + 1) PAP can prove to be a practical tool for robust hyper-Raman transfer. The possibility to cancel an arbitrary Stark shift by a combination of the pump and dump detunings with additional delays [20] are of particular interest.

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