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Population transfer through the continuum

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We show that complete population transfer is not in general possible through continuum intermediate states. We present a formal theoretical argument and supporting numerical results. In addition, the behavior of the system is compared with the well-known Λ system.

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It is well established by now that a counterintuitive laser pulse sequence [1–7] in a three-level system can transfer population most efficiently between the end levels of a Λ or ladder system. As long as all three levels are bound [Fig. 1(a)], the theoretical argument is well established and rigorous even when the intermediate state decays. Basically, the effect is due to the adiabatic evolution of one linear combination of the states not involving the intermediate state. For related experiments, see Refs. [2–6]. The possibility of extending the technique to the case of a transition via a continuum is of considerable interest as it would add flexibility and generality. The question has been addressed in a paper by Carroll and Hioe [8] based on a discretized model of the continuum. It is our purpose in this paper to show that, although correct within the confines of their model, their conclusions are valid only under very special circumstances which do not represent the real situation in the continuum of an atom or molecule.

The question is how efficiently the population can be transferred from state $|0\rangle$ to a state $|1\rangle$ of the same parity, by a sequence of two pulses, when the transition takes place through the continuum, as depicted in Fig. 1(b). The continuum, which may represent ionization or dissociation, replaces the intermediate bound state in the usual Λ system studied in the context of a counterintuitive pulse sequence. The most general expression for the wave function must here be written as a linear combination of the two bound states and the continuum with time-dependent amplitudes. Substituting in the Schrödinger equation and taking the Laplace transforms of the resulting differential equations for the amplitudes, one can eliminate the continuum by substituting the solution for its amplitude in the other two equations [9]. The summation over continuum states leads to a real and an imaginary part representing a decay (pole) into the continuum (ionization) and an adiabatic coupling (via the principal-value part) between the two bound states. The role of the continuum is thus contained in certain parameters introduced and discussed below, which are completely valid for intensities up to at least 10^{13} W/cm^2 or until above-threshold ionization (ATI) begins becoming significant.

As a result of that elimination, the wave function can be written as $\Psi(t) \equiv c_0(t)|0\rangle + c_1(t)|1\rangle$ with the initial condition $\Psi(t=-\infty)=|0\rangle$. The equations governing the time development of the amplitudes $c_0(t)$ and $c_1(t)$ can then be written as

$$\frac{d}{dt}\begin{pmatrix} c_0 \\ c_1 \end{pmatrix} = (-i)\begin{pmatrix} -\frac{i}{2}\Gamma_0 & -\frac{1}{2}\Gamma(q+i) \\ -\frac{1}{2}\Gamma(q+i) & -\frac{i}{2}\Gamma_i + D \end{pmatrix}\begin{pmatrix} c_0 \\ c_1 \end{pmatrix} \equiv -iH\begin{pmatrix} c_0 \\ c_1 \end{pmatrix}, \tag{1}$$

where Γ_0 and Γ_1 are the ionization widths of $|0\rangle$ and $|1\rangle$ due to lasers 1 and 2, respectively. They are given by the photoionization cross section of the state multiplied by the respective photon flux, which implies that they are proportional to the intensity of the laser. Γ is defined by $\Gamma \equiv \sqrt{\Gamma_0 \Gamma_1}$ and the detuning $D \equiv (\tilde{\omega}_1 - \tilde{\omega}_2) - (\omega_1 - \omega_0)$, with ω_j being the energy of an atomic state $|j\rangle$ ($j=0$ or 1) and $\tilde{\omega}_k$ ($k=1,2$) the frequencies of the lasers. q is an atomic parameter defined by

$$q \equiv \frac{\displaystyle\sum_l \frac{\mu_{1l}\mu_{l0}}{-\tilde{\omega}_1 + \omega_{1l}} + \mathrm{P}\sum_l \frac{\mu_{1l}\mu_{l0}}{\tilde{\omega}_2 + \omega_{1l}}}{\frac{1}{2}\sqrt{\Gamma_0 \Gamma_1 / I_1 I_2}} \tag{2}$$

and independent of the laser intensities I_1 and I_2, being determined solely by the states and the photon frequencies, which indirectly determine the strength of the coupling of each state to the continuum by fixing the energy in the continuum at which the bound-free matrix element is calculated.

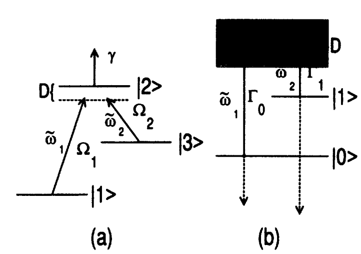

FIG. 1. (a) Well-known Λ system, which is considered for comparisons in our study. (b) Schematic diagram of the systems studied. Two bound states are coupled by a two-photon Raman-type process through the continuum as well as the coupling below the state $|0\rangle$ depicted by the dashed line.

The summations Σ_l in the numerator of Eq. (2) imply complete summation over the whole spectrum of the atom, with integration over the continuum part. Since the second term has a pole in the continuum, the principal-value part of the integral (indicated by P) must be taken. The matrix elements of μ represent electric dipole moments between the respective states. Note that under pulsed excitation, the quantities Γ_0 and Γ_1 are time dependent, while of course q is not.

The equations set up above are equivalent to those employed in the description of LICS (laser-induced continuum structure), which has received renewed attention both experimentally [10,11] and theoretically [9,12,13] during the last three years; except that we have cast these equations here in terms of the wave function instead of the density matrix, in order to conform to the usual practice followed in the literature on counterintuitive pulses. The parameter q is also referred to as the asymmetry parameter (in the context of LICS), as it determines whether the line shape of ionization is symmetric or not. As we show below it is a pivotal parameter in this context as well.

The characteristic equation of the Hamiltonian in Eq. (1) is

$$\lambda^2+\left[\frac{i}{2}(\Gamma_0+\Gamma_1)-D\right]\lambda+\frac{i}{2}\Gamma_0\left(\frac{i}{2}\Gamma_1-D\right)-\left[\frac{\Gamma}{2}(q+i)\right]^2=0. \tag{3}$$

Let us note first that, if $D=0$ and $q=0$, one of the roots of this equation is $\lambda=0$. The eigenfunction corresponding to this eigenvalue is

$$u_0=\frac{1}{\sqrt{\Gamma_0+\Gamma_1}}\begin{pmatrix}\sqrt{\Gamma_1}\\ -\sqrt{\Gamma_0}\end{pmatrix}. \tag{4}$$

In that case, if I_2 precedes I_1, we have $\Gamma_0 \ll \Gamma_1$ as $t\rightarrow-\infty$ and $\Gamma_0 \gg \Gamma_1$ as $t\rightarrow\infty$, which leads to $|u_0\rangle \sim |0\rangle$at $t\rightarrow-\infty$ and $|u_0\rangle \sim -|1\rangle$ at $t\rightarrow\infty$. This implies complete population transfer.

We seek now a condition for complete population transfer under the less restrictive situation in which $q\neq0$. For this to be possible, it is necessary and sufficient that the characteristic equation have one real solution. This leads to the two conditions,

$$\lambda^2-D\lambda-\tfrac{1}{4}\Gamma_0\Gamma_1-\tfrac{1}{4}\Gamma^2(q^2-1)=0 \tag{5}$$

and

$$\tfrac{1}{2}(\Gamma_0+\Gamma_1)\lambda-\tfrac{1}{2}\Gamma_0 D-\tfrac{1}{4}\Gamma^2 2q=0, \tag{6}$$

which require that $D=q/2(\Gamma_0-\Gamma_1)$.

This condition is the same as that obtained by Knight, Lauder, and Dalton [13], who examined population trapping in connection with LICS, under the assumption of square pulses, i.e., constant intensity. But this condition cannot be satisfied during the entire laser pulse for sequential pulses unless $q=0$. If $q\neq0$ it can only be satisfied in the special case $D=0$ and $\Gamma_0(t)=\Gamma_1(t)$ for all t, which is possible only for completely overlapping pulses.

In their paper Carroll and Hioe [8] have modeled the continuum by an infinity of equally spaced discrete levels extending from $-\infty$ to ∞ in energy. They have in addition assumed that the matrix elements μ_{l0} and μ_{1l} of the dipole operator, connecting states $|0\rangle$ and $|1\rangle$ to the states of the model continuum labeled by l, obey the relation $\varepsilon_1\mu_{l0}=\varepsilon_2\mu_{1l}$ (for all l), with ε_1 and ε_2 being the electric field amplitudes of lasers 1 and 2, respectively. The above special assumptions about the continuum automatically lead to $q=0$, because due to its infinite extent from $-\infty$ to ∞, it is completely symmetric with respect to any position of the resonance energy in the continuum. It is straightforward to verify that this symmetry causes each of the two terms in the numerator of Eq. (2) to vanish, thus making q identically zero. This, as we demonstrated above, leads to complete population transfer. Unfortunately, continua of electrons bound in atoms or molecules do not have this desirable property. It should be stressed here that the above special model implicitly models the whole spectrum of the system and not just the continuum. It pushes the ground state to $-\infty$ and makes no distinction between bound and continuum spectrum. It is known on the other hand that, in a typical atom or molecule, the bound spectrum carries a large part of the oscillator strength, which prevents q from vanishing except in accidental situations. As a result, the two terms in the numerator of Eq. (2) are in general unequal.

We discuss now in some detail what we can expect in a realistic situation. It is useful to use the standard Λ system with three bound states as a reference basis. With the notation as shown in Fig. 1(a), the evolution of that system is governed by

$$\frac{d}{dt}\begin{pmatrix}c_1\\ c_2\\ c_3\end{pmatrix}=(-i)\begin{pmatrix}0 & -\Omega_1 & 0\\ -\Omega_1 & D-\frac{i}{2}\gamma & -\Omega_2\\ 0 & -\Omega_2 & 0\end{pmatrix}\begin{pmatrix}c_1\\ c_2\\ c_3\end{pmatrix}\equiv-iH\begin{pmatrix}c_1\\ c_2\\ c_3\end{pmatrix}, \tag{7}$$

where the complete wave function is assumed to have the form $\Psi(t)=\Sigma_{j=1}^{3}c_j(t)|j\rangle$, Ω_1 and Ω_2 are the Rabi frequencies between $|1\rangle$ and $|2\rangle$, and $|2\rangle$ and $|3\rangle$, respectively, D is the detuning [as shown in Fig. 1(a)], and γ the rate of decay of $|2\rangle$ out of the Λ system. Two-photon resonance, i.e., $\omega_1+\tilde{\omega}_1=\omega_3+\tilde{\omega}_2$, is assumed. We define as before the adiabatic state $|u_0(t)\rangle=1/\sqrt{\Omega_1^2+\Omega_2^2}\,(\Omega_2|1\rangle-\Omega_1|3\rangle)$. We also define (what we shall call "purity") $f(t)\equiv|\langle u_0(t)|\Psi(t)\rangle|^2$. Initially the system is in the eigenstate $u_0(t=-\infty)$, which is the same as $|1\rangle$. The degree to which the system evolves adiabatically is characterized by how close to 1 is the purity $f(t)$.

In order to make a quantitative comparison with a realistic continuum, we have chosen the states $|0\rangle=|3s\rangle$ and $|1\rangle=|5s\rangle$ of a simple atom like Na. We consider the coupling of these two states by a two-photon process through the continuum [as in Fig. 1(b)] assuming two pulsed lasers of frequencies $\tilde{\omega}_1$ and $\tilde{\omega}_2$ and pulse durations $5ns$ (full width at

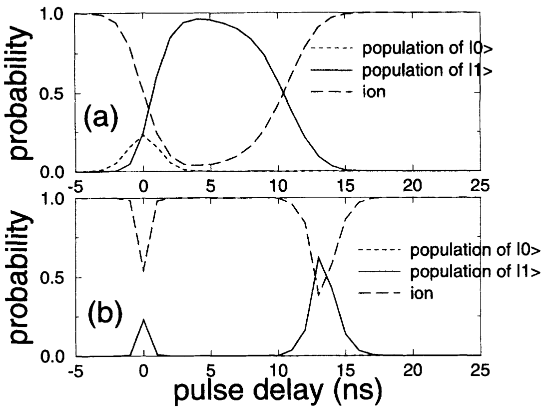

FIG. 2. Population and ionization after the laser pulses are turned off as a function of the laser pulse delay time. A pulse delay of I_1 is taken with respect to the time when I_2 reaches its maximum. Negative delay indicates that the $\tilde{\omega}_1$ laser precedes the $\tilde{\omega}_2$ laser. A 5-ns Gaussian pulse is employed for both lasers. In (a), the q value is set to 0 artificially while the original q value is used in (b). In both cases, at the peak intensities $\Gamma_0 = 2.55\times10^{10}$ s^{-1} and $\Gamma_1 = 2.37\times10^{10}$ s^{-1}. Detuning D is set to 0. Note that the short dashed line in (b) coincides with the bottom horizontal axis for all pulse delays except around 0, where it coincides with the population of state $|1\rangle$.

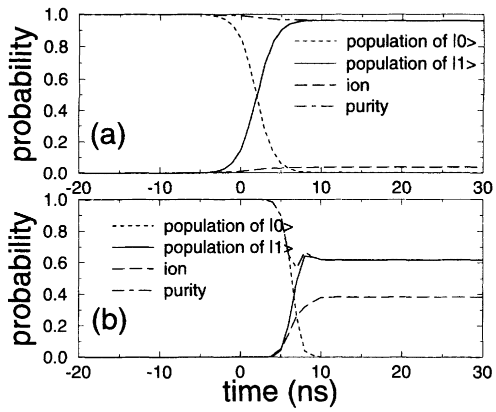

FIG. 3. Time evolution of the population and ionization. I_2 reaches the peak value at 0 ns in this time scale. (a) $q=0$ and time delay=4 ns (i.e., I_1 is at peak at time 4 ns). (b) Original q and time delay=13 ns. All of the parameters employed here are the same as those in Fig. 2. The purity $f(t)$ is defined in the text.

half maximum of Gaussian temporal pulse shapes). The values of the parameters needed in the description of this system are $q=-4.0$, $\Gamma_0 = 0.128 I_1(t)$ s^{-1}, and $\Gamma_1 = 0.395 I_2(t)$ s^{-1} (I_1 and I_2 in units of W/cm^2) at photon energies $\tilde{\omega}_1 = 43\,498$ cm^{-1} and $\tilde{\omega}_2 = 10\,398$ cm^{-1}.

To assess how much the value of q affects the efficiency of population transfer, we have calculated two cases. The results are shown in Figs. 2(a) and 2(b), in which the populations of $|0\rangle$ and $|1\rangle$, as well as ionization at the end of the delayed pulse, are calculated as a function of pulse delay. Positive (negative) delay means that the $\tilde{\omega}_2$ ($\tilde{\omega}_1$) laser precedes the $\tilde{\omega}_1$ ($\tilde{\omega}_2$) laser. Thus positive delay stands for counterintuitive pulse order. The first graph [Fig. 2(a)] is obtained by setting $q=0$ artificially. With the laser intensities we have chosen, about 95% of the population is transferred at a pulse delay of 4 ns. By increasing the laser intensity further, we have checked that almost a 100% population transfer occurs, which we do not show here. In Fig. 2(a), nearly 23% and 25% of the populations are left in $|0\rangle$ and $|1\rangle$, respectively, with a total ionization of 52% after the pulses, at zero pulse delay. Note that the laser intensities and pulse shape of the two lasers we have chosen almost satisfy the population trapping condition $D = q/2(\Gamma_0 - \Gamma_1)$ at delay time 0. Under the above perfect population trapping condition, 50% of the atoms ionize, while the rest are distributed equally between states $|0\rangle$ and $|1\rangle$. A further increase or decrease of delay time causes 100% ionization. This is due to the fact that the $\tilde{\omega}_1$ laser alone is sufficiently intense to ionize all atoms during the 5-ns pulse duration. Having examined the fact that complete population transfer works for the $q=0$ case, we performed another calculation with the original q value $q=-4.0$ [Fig. 2(b)]. The maximum population transfer of 61% is obtained with the pulse delay of 13 ns. Although the pulse duration is 5 ns (full width at half maximum), there is sufficient overlap of the two pulses in the time domain (5–10 ns) where the population transfer occurs. As a next step, we have calculated the time evolution of the populations of $|0\rangle$ and $|1\rangle$, of ionization, and of $f(t)$, with a 4-ns delay for $q=0$ and a 13-ns delay for the original $q(=-4.0)$. The results are shown in Figs. 3(a) and 3(b). In Fig. 3(a), the system follows quite closely the eigenstate $u_0(t)$. Once the system begins to deviate from $u_0(t)$, it never comes back. For comparison, we plot the time evolution of the Λ system with $\gamma=0$ [Fig. 4(a)]. It is interesting to see that when the time evolution of the system is close to that of $u_0(t)$, it will recover, even if it begins to deviate from $u_0(t)$. If $\gamma \neq 0$, however, the system does not completely recover, if it begins to deviate from $u_0(t)$ [Fig. 4(b)]. These results indicate that

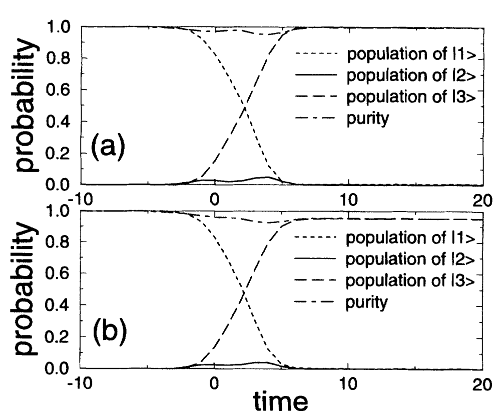

FIG. 4. Time evolution of the population for the Λ system. The pulse duration $T=5$ for both lasers. Detuning is 0. I_2 is at peak at time 0. The pulse delay is $0.8T$ (I_1 is at a peak at time $4=0.8T$). $\Omega_1 = 5T^{-1}$ and $\Omega_2 = 5T^{-1}$ at the peak. (a) $\gamma=0$. (b) $\gamma=T^{-1}$. (All parameters are dimensionless here.) The purity $f(t)$ is defined in the text.

when the adiabatic conditions are not 100% satisfied, the system loses adiabatic evolution in time. It may be worth pointing out that, contrary to the correct perception that adiabatic following does not require any strict pulse shape or intensities for a Λ system with $\gamma=0$, it does require more restrictive conditions for the two-level system coupled through the continuum, as well as the Λ system with $\gamma \neq 0$. The Λ system with $\gamma=0$ has the ability to recover, even if the adiabatic conditions are not strictly satisfied. In other words, the adiabatic following for the Λ system with $\gamma \neq 0$ and a two-level system coupled through the continuum are more fragile than that for a Λ system with $\gamma=0$. Figure 3(b) shows the time evolution of the system with the original $q(=-4.0)$. As the intensity of the laser with $\tilde{\omega}_1$ increases, $f(t)$ decreases to about 0.62. This verifies our argument that if q is not 0, complete population transfer does not occur at any intensity and pulse delay. Having established this limitation, in general, the exact amount of population transfer will depend on the value of q. The amount of 61% obtained above simply represents one example and can be much smaller for larger q.

In conclusion, we have demonstrated that complete population transfer in a Λ-like arrangement through the continuum is possible only under very special conditions which, given two levels and two frequencies, can be satisfied only accidentally. Unfortunately the situation is much worse. In order to have a direct comparison with the model of Carroll and Hioe [8], we neglected the incoherent channels of ionization; namely, ionization of level $|1\rangle$ by laser $\tilde{\omega}_1$ and depending on the position of level $|0\rangle$, ionization of level $|0\rangle$ by laser $\tilde{\omega}_2$. At least one of these channels is always present and inevitably leads to irreversible decay into the continuum, thus reducing the population. This aspect has been discussed in detail elsewhere [9] and has been shown to play a decisive role in LICS [10–12]. It will obviously have a deleterious effect on population transfer as well, reducing, for example, the 61% of the case above to 0.01%.

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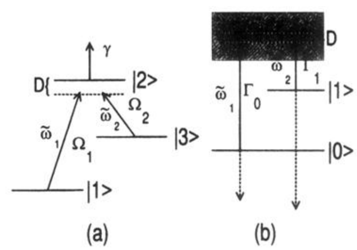

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