Coherent Population Transfer via the Continuum

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We present a remarkable analytic result which suggests that a continuum can be used as an intermediary for a significant transfer of population from one discrete state to another discrete state in a stimulated Raman transition. The population transfer is accomplished by employing two laser pulses that overlap in time, arranged in the so-called counterintuitive order. That a continuum can be used for population transfer is shown here for the first time. It promises to open up more channels for selective coherent excitation of atoms or molecules.

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A new method of coherent population transfer in a stimulated Raman transition with the use of two spatially overlapping laser beams arranged in the so-called counterintuitive order [1,2] has received a great deal of attention recently [3–8], following the first and striking experimental demonstration of the procedure in an effective excitation of sodium molecules to selected high vibrational states, by Bergmann and co-workers [9]. Spatially overlapping laser beams are used, so that each atom or molecule sees two laser pulses that overlap in time. The two pulses are said to be in the counterintuitive order when the transition which connects the initially populated state 1 to an unpopulated intermediate state 3 is strongly driven *after* the transition which connects the initially unpopulated intermediate state 3 to the initially unpopulated final state 2 is strongly driven. The procedure not only effectively transfers population from state 1 to state 2, it also has the striking features that (1) the population transfer is accomplished without the intermediate state being significantly populated at any time even though the two lasers used may be at one-photon resonance with the 1-3 and 3-2 transition frequencies and (2) the high efficiency of population transfer is not sensitively dependent on the pulse shape, intensity, or frequency modulation of the lasers. These attractive features give the counterintuitive procedure a distinct advantage over more conventional methods not only for selective population transfer, but also, as has been recently suggested [7], for atomic beam splitters and mirrors in atomic interferometry.

Knowing the above, here we ask: What happens if the intermediate state 3 is replaced by a continuum?

In this Letter, we present an analytic calculation for a simple model with a continuum of intermediate states. We believe that our result captures the essential features of the excitation process. It suggests that a continuum can be used as an intermediary for a significant transfer of population from one discrete state to another and again shows the great advantage of the counterintuitive pulse order over the more conventional or intuitive procedure in which the lasers are arranged in the order of laser 1 preceding laser 2.

Consider an atom or molecule driven by two laser beams in a stimulated Raman transition from state 1 to state 2, through an intermediate continuum, which we model by closely spaced discrete states $3,4,\ldots,N$, where N is large, as shown in Fig. 1. Transitions from state 1 to intermediate states to state 2 are driven by two classical oscillating external fields which represent two laser pulses that overlap in time. We use the rotating-wave approximation and assume that each laser drives only its own transitions. This amounts to neglect of intermediate states that are far off resonance, and it suggests that we should neglect energy-dependent properties of the continuum. Since states of very large detuning have a negligible effect on the physical process, we use a topless and bottomless sequence of states; this is our model of a featureless continuum. Furthermore, we neglect spontaneous emission of photons and electrons from the continuum states, which is justified if the continuum is lightly populated for a short time, and we neglect the continuum-continuum transitions. We set $\hbar=1$ and use the time-dependent Schrödinger equation as the equation of motion. The transformation of Einwohner, Wong, and Garrison [10] is used to eliminate optical-frequency terms from the Hamiltonian and wave function. The transformed Hamiltonian is represented by a matrix whose nonzero elements are

$$H_{kk}=-\Delta_k,\quad H_{mj}(t)=H_{jm}(t)=-\tfrac{1}{2}\,\Omega_{mj}(t)\,, \qquad (1)$$

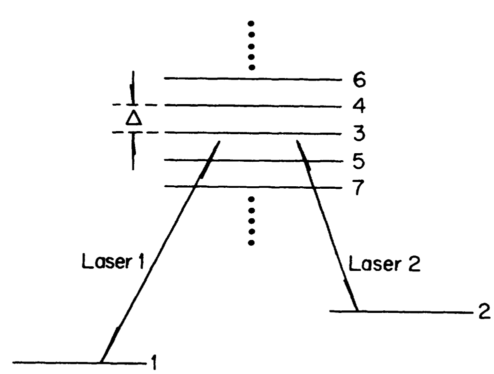

FIG. 1. Model for the continuum that connects state 1 and state 2.

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for $k=1,2,\ldots,N$, $m=1,2$, and $j=3,4,\ldots,N$. Here Δ_k is essentially the detuning of state k from the resonance, and $\Omega_{mj}(t)$, the Rabi frequency, is proportional to the amplitude of the mth laser beam, and is also proportional to the dipole matrix element for the transition. Assuming that the two lasers are equally detuned from any one intermediate state, we set $\Delta_1=\Delta_2$. The work of Gottlieb [11], Pegg [12], and the present authors [5,13] suggests the use of two overlapping laser pulses which last from $t=0$ to $t=T$ and have the forms

$$\Omega_{1j}(t)=M\cos\theta(t),\quad \Omega_{2j}(t)=M\sin\theta(t) \tag{2}$$

for $j=3,4,\ldots,N$, and $\Omega_{1j}(t)=\Omega_{2j}(t)=0$ for $t<0$ and for $t>T$. M is the maximum value of each Rabi frequency. We assume that $\theta(t)$ is a linear function of t that varies from 0 to $\pi/2$ or from $\pi/2$ to 0. This gives the simple pulse shapes shown in Fig. 2; $d\theta/dt=\pm\pi/2T$ correspond to the intuitive and counterintuitive pulse orders, respectively. The transformation

$$B_1=A_1\cos\theta(t)+A_2\sin\theta(t)\,, \tag{3a}$$

$$B_2=-A_1\sin\theta(t)+A_2\cos\theta(t)\,, \tag{3b}$$

gives an equation of evolution for $B_1,B_2,A_3,\ldots,A_N$ that has the form of the Schrödinger equation, with the Hamiltonian H', and can be solved. The diagonal matrix elements of H' are given by $H'_{kk}=-\Delta_k$, and the remaining matrix elements in the first row and column are given by

$$H'_{12}=H'^{*}_{21}=i\,d\theta/dt,\quad H'_{1j}=H'_{j1}=-\tfrac{1}{2}M\,, \tag{4}$$

for $j=3,4,\ldots,N$. The other matrix elements are zero. The states we use to represent the continuum are uniformly spaced in energy:

$$\begin{aligned}&\Delta_3=0,\ \Delta_4=\Delta,\ \Delta_5=-\Delta\,,\\ &\Delta_6=2\Delta,\ \Delta_7=-2\Delta,\ \ldots,\end{aligned} \tag{5}$$

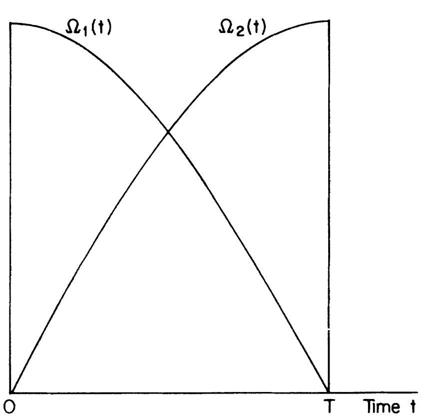

FIG. 2. Pulse shapes given by Eq. (2) in the intuitive order. For the counterintuitive order, the labels Ω_1 and Ω_2 are interchanged.

where Δ is the energy difference between adjacent states (Fig. 1).

The Hamiltonian H' is now represented by a time-independent $N\times N$ matrix, because $d\theta/dt$ is time independent. Matrix elements of $\exp(-iH't)$ are needed to compute the occupation probabilities at time t, and we are especially interested in $t=T$, the end of the two overlapping pulses. These matrix elements can be evaluated by the Laplace-transform technique of Stey and Gibberd [14]. Simple results are obtained in the limit as $N\to\infty$ if $\Delta<2\pi/T$ holds. Since $\Delta\to 0$ is needed for a continuum, we may assume this. We solve one quadratic equation to get the Laplace transforms, rather than a secular equation of degree N.

The occupation probabilities are $|A_k(t)|^2$, for $k=1,2,3,\ldots$. For $j\geq 3$, $|A_j(t)|^2$ depends on the pulse strength and on

$$\delta=T(\Delta_j-\Delta_1)\,, \tag{6}$$

the dimensionless detuning parameter. This parameter should replace the index j when we take the continuum limit.

In the continuum limit, each dipole matrix element for a transition vanishes, because the continuum wave functions are normalized to unity in a large box [15]. This means that each Rabi frequency vanishes in the continuum limit. Although both M and Δ vanish in this limit, M^2/Δ approaches a nonzero limit. Indeed, use of the so-called "golden rule" [15], which is applicable only for weak pulses, would give a transition rate proportional to M^2/Δ and a probability of transition to the continuum proportional to M^2T/Δ. The results of our model show that all the final occupation probabilities depend on

$$x=\pi M^2T/8\Delta\,, \tag{7}$$

the dimensionless pulse strength, for strong as well as for weak pulses. The occupation probability of a small interval in the continuum is

$$P_\delta(t)d\delta=\{|A_j(t)|^2/\Delta T\}d\delta\,, \tag{8}$$

where $d\delta$ is the small change in (6). As the occupation probabilities should sum to unity, we check that

$$|A_1(t)|^2+|A_2(t)|^2+\int_{-\infty}^{\infty}P_\delta(t)d\delta=1\,.$$

Two of the final occupation probabilities are

$$|A_1(T)|^2=[\tfrac{1}{2}\pi S]^2\,, \tag{9}$$

$$|A_2(T)|^2=[C\mp xS]^2\,, \tag{10}$$

where the upper and lower signs correspond to $d\theta/dt=\pm\pi/2T$, meaning intuitive and counterintuitive pulse order. Here the functions C and S, which depend on x, denote

$$C=e^{-x}\cosh\phi,\quad S=e^{-x}\sinh\phi/\phi\,, \tag{11}$$

where $\phi=(x^2-\pi^2/4)^{1/2}$. Although ϕ may be positive, zero, or imaginary, C and S are real when x is real.

The density of final occupation probability in the continuum is

$$P_\delta(T) = 2x\{\delta^2(\cos\delta - C + xS)^2 + (\delta\sin\delta - \pi^2 S/4)^2\}/D\pi \tag{12}$$

for the intuitive pulse order, and

$$P_\delta(T) = \pi x\{(C + xS - \cos\delta)^2 + (\delta S - \sin\delta)^2\}/2D \tag{13}$$

for the counterintuitive pulse order. Here, $D = (\delta^2 - \pi^2/4)^2 + 4x^2\delta^2$.

Let us first examine the behaviors of the final occupation probabilities $|A_1(T)|^2$ of state 1, which has all the population initially, and $|A_2(T)|^2$ of state 2, which the population is to be transferred to, for the two different laser pulse orders, as functions of x. The behavior of $|A_1(T)|^2$ is the same for both pulse sequences, namely,

$$|A_1(T)|^2 \approx \begin{cases} 1 - 2x \to 1 \quad \text{as } x \to 0, & \text{(14a)} \\ \{\pi/4x\}^2 \to 0 \quad \text{as } x \to \infty. & \text{(14b)} \end{cases}$$

The behaviors of $|A_2(T)|^2$ are also similar for both pulse orders for the case of small x and they are given by

$$|A_2(T)|^2 \approx (2x/\pi)^2 \quad \text{as } x \to 0. \tag{15a}$$

On the other hand, the behaviors of $|A_2(T)|^2$ become dramatically different as x becomes large for the two different pulse orders. They are given by

$$|A_2(T)|^2 \approx \begin{cases} \{\pi^2/16x^2\}^2 \to 0 \quad \text{as } x \to \infty, & \text{(15b)} \\ 1 - \pi^2/4x \to 1 \quad \text{as } x \to \infty & \text{(15c)} \end{cases}$$

for the intuitive and counterintuitive pulse orders, respectively. Figure 3 shows how these final occupation probabilities vary as x increases. We thus have the remarkable result that the counterintuitive laser pulse order at appropriately high intensities can be used to transfer a significant proportion of population from one discrete state to another with a continuum of states as an intermediary, while the intuitive laser pulse order would simply disperse the population into the continuum. Compare this result with our previous result for a three-state model [5] using laser pulses of the same shapes where we found that the counterintuitive pulse order is generally effective for population transfer so long as the laser intensities are sufficiently high, while the intuitive pulse order can be effective for population transfer but only for laser pulses of certain areas.

Figure 3 not only shows how effectively the counterintuitive pulse order can be used for population transfer via a continuum, but also shows how ineffective the result would be if the intuitive pulse order were used, as is evidenced by the exceedingly small maximum value of 0.02644 for $|A_2(T)|^2$ occurring at $x \approx 0.7285$ for this case.

The next question of interest is how the population

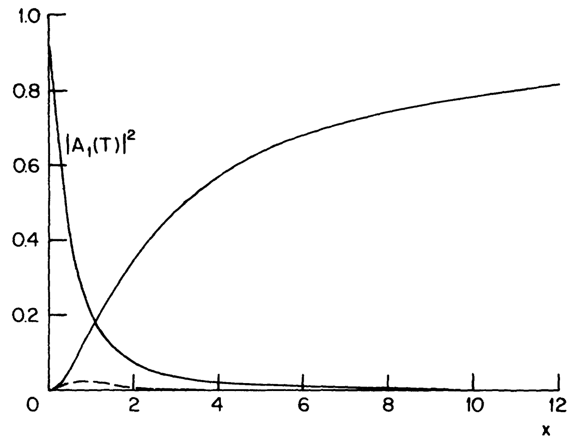

FIG. 3. The final occupation probabilities $|A_1(T)|^2$ of state 1 for both pulse sequences, and $|A_2(T)|^2$ of state 2 for the intuitive (dashed line) and the counterintuitive (solid lines) pulse sequences, as functions of x.

which gets transferred into the continuum is distributed among the continuum states. That is, what is the shape of $P_\delta(T)$ as a function of δ? For the intuitive pulse order, we find that as x increases from 0, the distribution changes from one that has a single maximum at $\delta = 0$ to one that has two maxima located symmetrically about $\delta = 0$, with the maximum at $\delta = 0$ becoming a minimum. As x increases further, the distribution changes into one that has three maxima, and as x becomes large, the distribution becomes very broad, as the states in the continuum take up essentially all the population from state 1. On the other hand, for the counterintuitive pulse order, the distribution of $P_\delta(T)$ always has a central maximum at $\delta = 0$ for all values of x, and it never becomes very broad. The total final population in the continuum initially increases as x increases from zero, reaches a maximum of 0.6311 at $x \approx 1.0992$, and then decreases as x further increases. Moreover, we find that the population in the continuum at any time between $t = 0$ and $t = T$ is never greater than that at the final time for the counterintuitive pulse order. Thus, for sufficiently high pulse strengths, the continuum is never significantly populated *at any time* in the process.

These results are all quite unexpected and are ones which could not have been easily foreseen. For the intuitive pulse order, some detailed features of $P_\delta(T)$ may depend on our use of the specific pulse shapes. For the counterintuitive pulse order, on the other hand, experiments [9] and our analytic calculation [5] in the case of three states suggest that the principal features of $|A_2(T)|^2$ and $P_\delta(T)$, especially in the limits of large x, are most probably quite independent of the laser pulse shapes.

In summary, we have presented a remarkable analytic result that shows that a continuum can be used as an intermediary for population transfer from one discrete state to another when two overlapping laser pulses in the counterintuitive order are used. This result is shown to be in dramatic contrast to one when the two overlapping laser pulses are in the intuitive order. Experimental verification of these results will open up opportunities for many applications, in selective excitation as well as in atomic interferometry.

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