

## Gating of population flow in resonant multiphoton excitation

Bruce W. Shore\*

Lawrence Livermore National Laboratory, University of California, P.O. Box 808,  
Livermore, California 94550

(Received 17 January 1983; revised manuscript received 27 September 1983)

I discuss the flow of excitation probability along a multistate chain of resonant excitations as driven by coherent photoexcitation, in particular the gated interruption of this probability flow caused by a strongly coupled side branch. If the side branch is itself a chain, not a single transition, then flow inhibition occurs if the number of branch transitions is odd, but no inhibition occurs if the number is even. The inhibition provides a limiting bound on the intensity of amplified stimulated emission.

### INTRODUCTION

The behavior of  $N$ -state systems excited coherently by near-monochromatic radiation has been studied for several years;<sup>1</sup> the relevant dynamics obtains from solutions of the time-dependent Schrödinger equation in the rotating wave approximation (RWA).

The application of Schrödinger dynamics to multistep excitation in isotope separation is by now well known, but realistic modeling of an actual atom including hyperfine structure, magnetic-sublevel degeneracy, Doppler shifts, inhomogeneities, and excitation pulse characteristics often inject such complexity that it is difficult to interpret either the results of realistic modeling or of experiment; i.e., it is not always easy to foresee the result of parameter changes upon excitation dynamics. For this reason it is useful to have available a corpus of results for elementary models (e.g., monochromatic illumination of stationary nondegenerate multilevel atoms) to serve as touchstones for interpreting more realistic models. These models of time-dependent dynamics, often admitting exact analytic solution, are the analogs of the hydrogen atom as an element of molecular structure. By examining and understanding simplified models we gain insights which aid the interpretation of complicated time dependence. The present Comment examines some examples of such simple models. Just as the coherence of the three-level lambda system<sup>2</sup> finds applications as an element of more complicated linkages, so too are the chains described here observable as elements in general excitation schemes.

It is now generally recognized that the coherent flow of probability along a chain of coupled near-resonant transition proceeds most efficiently when the successive Rabi frequencies appropriate to the transitions are roughly equal.

Einwohner, Wong, and Garrison (EWG) have pointed out<sup>3</sup> that the regular flow of probability along a chain can be severely curtailed to levels beyond the point of insertion by the presence of a strong resonant branching transition, and they present analytical expressions for the special case  $n = 2$  to support this observation.

It is instructive to consider this branched system from the viewpoint of dressed states;<sup>4</sup> we then observe that the hindered flow of probability is a consequence of the familiar ac Stark effect. We can also see consequences not noted in the EWG analysis. In particular, we observe a rather striking dependence of excitation upon the number of links to the side chain, i.e., whether this number is even or odd.

### DISCUSSION

Consider the chain of excitation transitions shown in Fig. 1(a): a sequence of states (nondegenerate energy levels) linked by near-resonant excitation. Population flows along this chain beginning, at time  $t = 0$ , from an initial concentration in state 1. One may imagine monitoring an ionization or fluorescence signal to register time-dependent arrival of population into the terminus of the chain, state  $N$ . We assume near-resonant relaxation-free excitation: at each step the cumulative detuning is much less than the Rabi frequency for the step. At some point in the chain, state  $X$  say, a competing transition (or chain of transitions) provides an alternative excitation route, say to state  $Y$ .

For this system the rotating wave approximation time-dependent Schrödinger equations for probability amplitudes  $A_m$  of the main chain have the structure

$$\begin{aligned} i\frac{d}{dt}A_{x+1} &= \frac{1}{2}\Omega_c A_{x+2} + \frac{1}{2}\Omega_c A_x, \\ i\frac{d}{dt}A_x &= \frac{1}{2}\Omega_c A_{x-1} + \frac{1}{2}\Omega_c A_{x+1} + \frac{1}{2}\Omega_b A_Y, \\ i\frac{d}{dt}A_{x-1} &= \frac{1}{2}\Omega_c A_x + \frac{1}{2}\Omega_c A_{x-2}. \end{aligned} \quad (1)$$

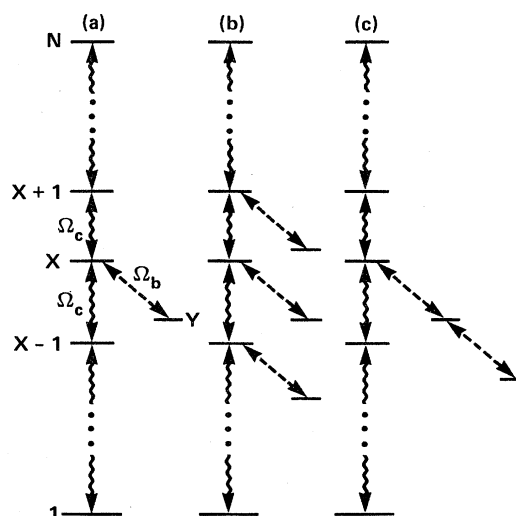


FIG. 1. Linkage patterns for branched multistep excitation chains. Wavy arrows designate chain of  $N$  states; dashed lines indicate branches [(a) and (b)] or branched chains (c).

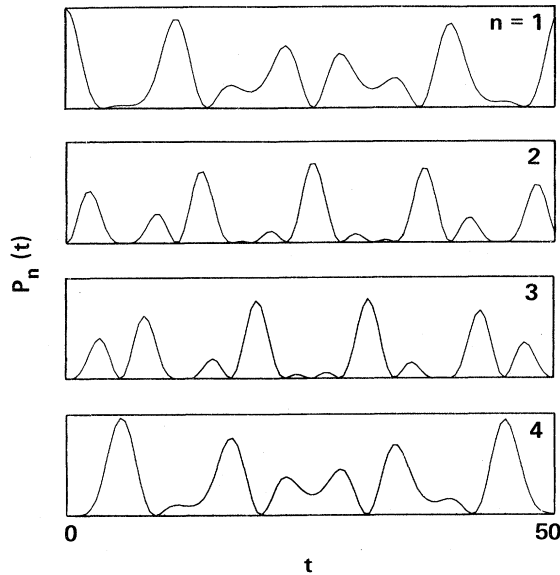


FIG. 2. Population histories  $P_n(t)$  for unbranched  $N=4$  state chain (resonantly tuned) chain. All Rabi frequencies are set equal to unity, thereby defining the time scale.

Suitable additional linked equations, such as

$$i \frac{d}{dt} A_Y = \frac{1}{2} \Omega_b A_X \quad (2)$$

for the system of Fig. 1(a), describe the side branch. For weak coupling,  $\Omega_b \ll \Omega_c$ , the dynamics are basically governed by Eq. (1) with the neglect of  $A_Y$ .

If all the main-sequence transition (denoted by solid-line linkages) have comparable Rabi frequencies (i.e., coupling strengths) and the branched transition (dashed line) has a much smaller Rabi frequency, then the excitation dynamics of the  $N$ -state chain is essentially unaffected by the branch. Population flows out of (and subsequently returns into) the initially populated level 1, arriving at (and departing from) terminal state  $N$  in a manner appropriate to  $N$  strongly coupled (or dressed) states.

Figure 2 exhibits the population probability histories

$$P_n(t) = |A_n(t)| \quad (3)$$

for such a lossless 4-state chain ( $N=4$ ). Each state receives population during the course of time, and the time-averaged population  $\bar{P}_n$  of the terminal level equals that of the ground state,  $\bar{P}_4 = \bar{P}_1$ .

When the Rabi frequency  $\Omega_b$  of the branch becomes comparable with the Rabi frequency  $\Omega_c$  of the chain then one can no longer regard the branch linkage to level  $Y$  as a negligible perturbation: one must explicitly diagonalize the Hamiltonian of the  $(N+1)$ -state system.

A particularly interesting situation occurs when the branching Rabi frequency  $\Omega_b$  far exceeds the adjacent chain Rabi frequency  $\Omega_c$ , for under this condition almost no population at all flows into states  $X, X+1, \dots, N$ . The strong transition  $\Omega_b$  has acted as a gating switch to turn off population flow into states above the linkage.

Figure 3 shows such a linkage pattern: a strong branching transition  $\Omega_b$  connects a fifth state to state  $X=3$  of a 4-

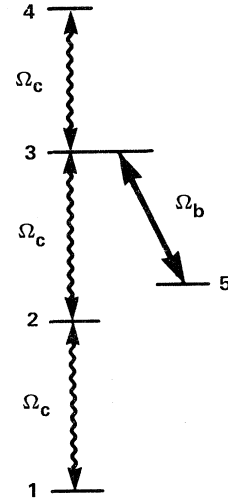


FIG. 3. Linkage pattern for  $N=4$  state chain with branch from state  $x=3$ .

state chain. Figure 4 shows the resulting population histories. We recognize that the branching transition has essentially excluded population from states beyond and including state  $X$ : the system has become very nearly a 2-state system.

The extent to which the branch suppresses population flow depends upon the strength of the branch transition  $\Omega_b$  relative to the values  $\Omega_c$  for the chain, as Fig. 5 shows: we see that the ratio  $\Omega_b/\Omega_c=2$  suffices to suppress excitation probabilities by an order of magnitude. Note that when

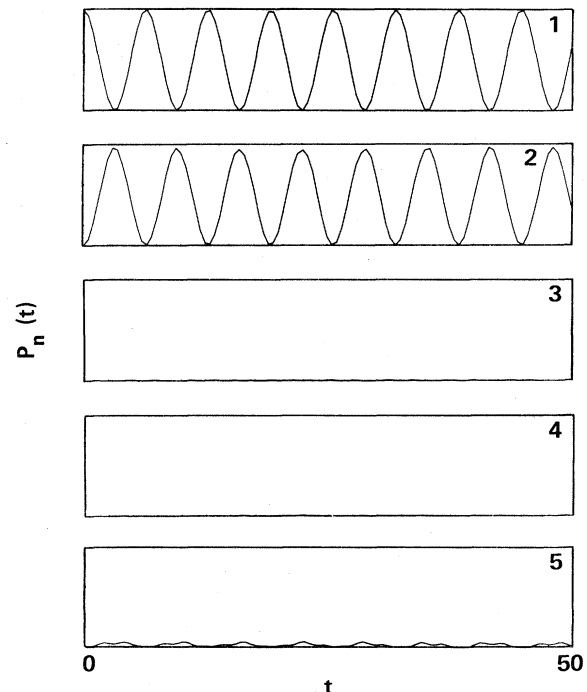


FIG. 4. Population histories for  $N=4$  state chain, having Rabi frequencies  $\Omega_c=1$ , with strong branch transition  $\Omega_b=5$ .

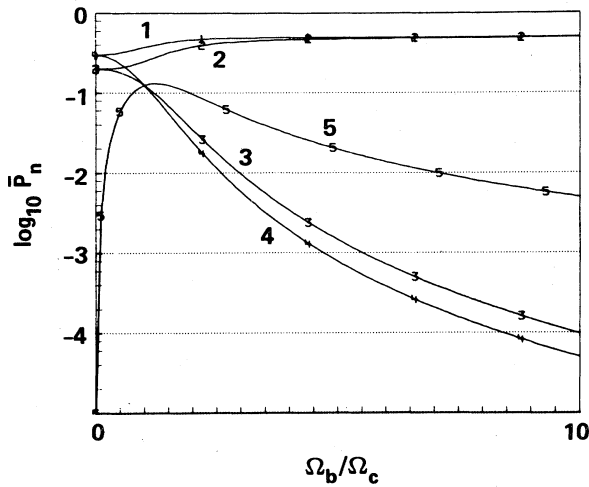


FIG. 5. Plots of time-averaged population  $P_n$  in the system of Fig. 3 vs the ratio  $\Omega_b/\Omega_c$  of branch-to-chain Rabi frequencies.

branch and chain have equal Rabi frequencies,  $\Omega_b/\Omega_c=1$ , populations equalize in states 3, 4, and 5.

The nature of this gating action can be understood easily if we regard states  $X$  and  $Y$ , the two strongly coupled states of Figs. 1(a) and 3, as forming a dressed 2-state system upon which the remaining chain acts as a perturbation. Equation (2), together with the approximation

$$i\frac{d}{dt}A_x \approx \frac{1}{2}\Omega_b A_Y, \quad (4)$$

expresses this coupling. In this view the (relatively) weak linkage  $\Omega_c$  serves as a probe of the dressed  $X$ - $Y$  system. The absorption rate as a function of probe frequency will show the well-known double-peaked Autler-Townes or ac-Stark doublet structure of two peaks separated by the Rabi frequency  $\Omega_b$ . Thus, if the chain transitions remain tuned to the resonant Bohr frequency when the branch transition is turned on, we shall find that the tuning falls midway between the Autler-Townes doublet, and hence the effective excitation rate is quite small.

Figure 6(a) shows the detuning needed to reveal this effect: each main-chain transition remains tuned to the appropriate Bohr frequency and each additional branch transition similarly remains tuned to its Bohr frequency, but chain and first branch are detuned by  $D$ . Figure 7 displays the time-averaged populations for this case [Fig. 6(a)]. The resonant choice  $D=0$  expresses the situation of Fig. 4: populations in states 3 and 4 are then more than two orders of magnitude below those of states 1 and 2.

The branch transition  $XY$  thus acts as a gate preventing population flow along the chain. There need never be any actual population in state  $Y$ : the dressed  $XY$  states occur even in the absence of population. The blockage occurs when we impose a branch Rabi frequency appreciably exceeding the adjacent chain Rabi frequency.

The gating action does not occur instantaneously, of course: roughly one  $XY$  Rabi cycle must be completed to define the Autler-Townes doublet.

Although population does not reach states further along the main chain than the branch joint  $X$ , some population

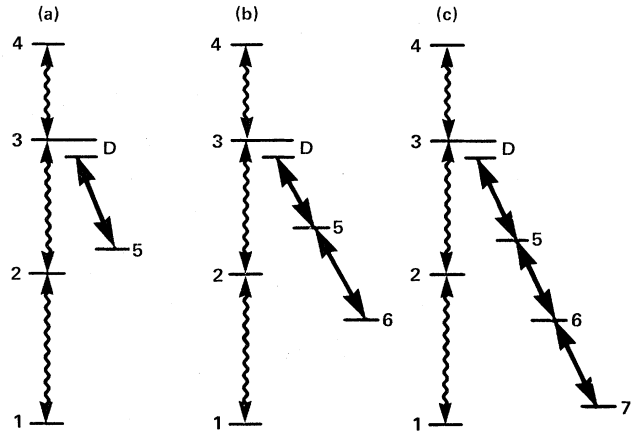


FIG. 6. Linkage patterns for  $N=4$  state chain linked to 1-state branch (a), 2-state branch (b), and 3-state branch (c), showing definition of detuning  $D$ .

does in time leak, by two-photon transition, into the branch terminus (state 5).

It might be thought that the dressing of state  $X$  by the branch is equivalent to making state  $X$  an off-resonant virtual state on the main chain, and that a two-photon transition between states  $X-1$  and  $X+1$  could thereby bypass this bottleneck. However, Figs. 5 and 7 show that such transitions do not occur.

Obviously, the gating action of a single branch extends to multiple branches as in Fig. 1(b). Here any one of the branches, if strongly driven can exclude population from states further along the chain.

Interestingly, the presence of a second strong resonantly tuned transition in the side chain [see Fig. 1(c)] negates the gating action and dramatically alters the excitation dynamics. Figure 8 shows population-history plots for this case, a strong 2-state side branch joined to a 4-state main chain at state  $X=3$ . As in the case for Fig. 4, the branch Rabi fre-

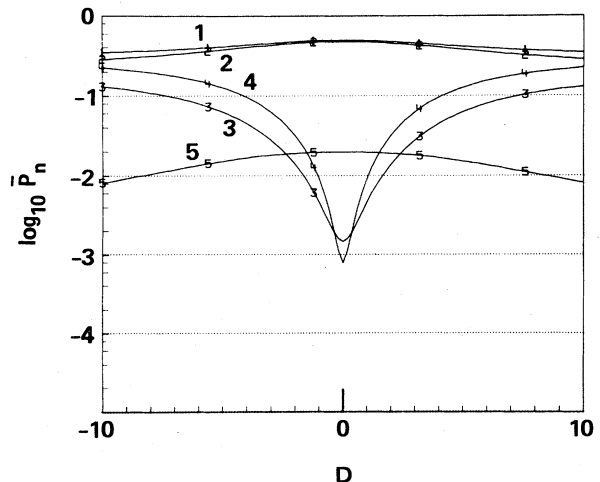


FIG. 7. Plot of time-averaged populations  $P_n$  vs branch detuning  $D$  for 1-state branch ( $\Omega_c=1$ ,  $\Omega_b=5$ ) of Fig. 7(a).

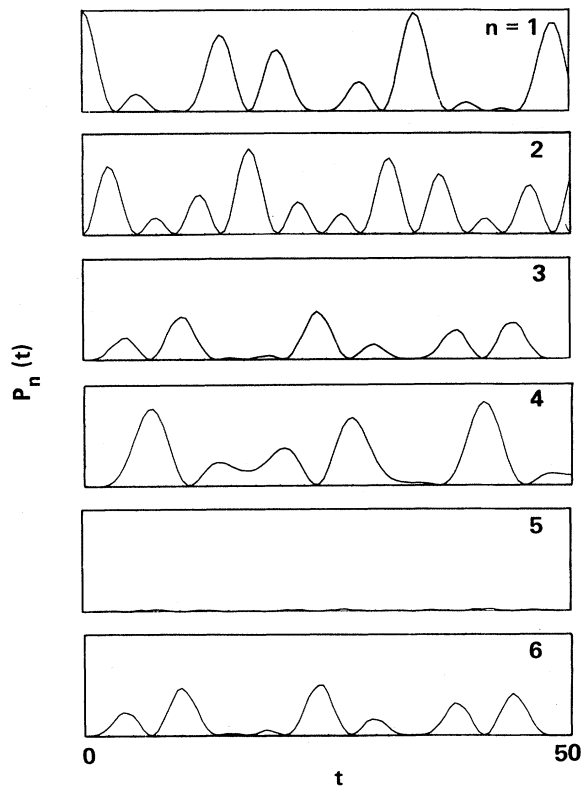


FIG. 8. Population histories for  $N=4$  state chain linked at state  $X=3$  to a 2-state branch [pattern of Figs. 1(c) or 7(b)]. Chain Rabi frequencies are all  $\Omega_c=1$ ; branch Rabi frequencies are all  $\Omega_b=5$ .

quency is  $\Omega_b=5\Omega_c$ . We see that population regularly reaches the main-chain terminus, state 4, as well as the branch terminus, state 6, and no population blockage occurs.

A plot of time-averaged populations versus detuning for this case, Fig. 9, shows that the resonant case,  $D=0$ , yields maximum population in state 4, in sharp contrast to the minimum population which occurs (Fig. 7) with a single

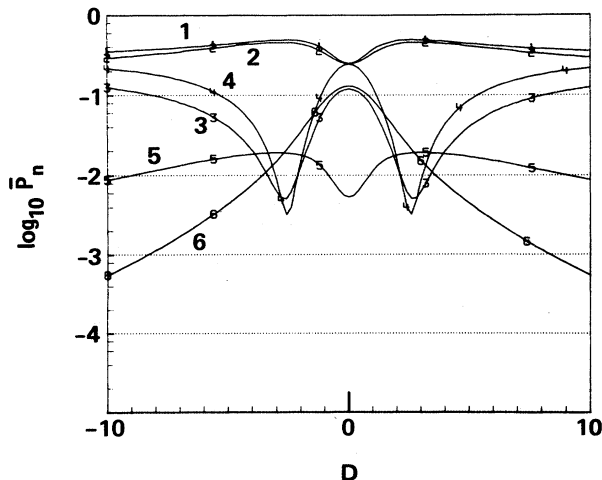


FIG. 9. As in Fig. 8 for 2-state branch of Fig. 7(b).

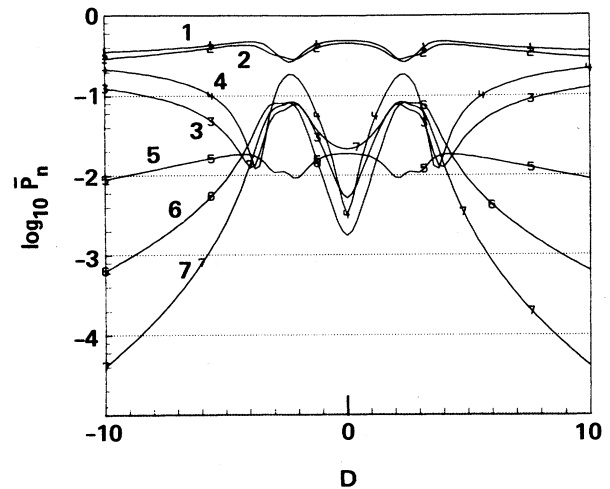


FIG. 10. As in Fig. 8 for 3-state branch of Fig. 7(c).

transition branch. Indeed, with an odd number of steps in the side chain, gating occurs, whereas an even number of steps induces no strong gating effect.

The explanation of this curious odd-even effect, noted earlier in a slightly different context,<sup>5</sup> appears quite natural if we regard the side chain as a strongly coupled  $M$ -state system. When we sweep the detuning then we will observe an  $M$ -peaked spectrum. When  $M$  is an odd integer one of these peaks occurs at zero detuning, and hence the major chain remains "on resonance" at the unperturbed Bohr frequency, even in the presence of the side chain. Figure 10 illustrates this effect for the 3-state branch of Fig. 6(c). Of course, in this situation, with an odd-state chain, the population flow does not simply flow without impediment along the original chain.

As the dressed-atom picture makes clear, the gating action of a side chain only holds if the main-chain excitation frequencies remain tuned to the Bohr frequencies, as is appropriate to excitation in the absence of a side chain. If we instead retune the excitation frequency to the relevant Autler-Townes multiplet, then population can flow upward (as well as into the side chain).

We note in passing that the Autler-Townes splitting of the branch transition provides a limiting bound on the intensity of amplified stimulated emission into any branch of the excitation chain: if the branch Rabi frequency were to exceed the chain Rabi frequency, then the branch transition no longer satisfies the resonant condition necessary for continued stimulated emission of population out of the chain sequence.

Although gated probability flow of the sort described here is an inevitable consequence of coherent multistep excitation it would be of interest to demonstrate experimentally the behavior, particularly the striking odd-even dependence upon branch length.

#### ACKNOWLEDGMENT

This work was performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W7405-Eng-48.

\*Address until June 1984: Spectroscopy Group, Imperial College, London SW7 2AZ, England.

<sup>1</sup>T. H. Einwohner, J. Wong, and J. C. Garrison, *Phys. Rev. A* 14, 1452 (1976); J. H. Eberly, B. W. Shore, Z. Bialynicka-Birula, and J. Bialynicki-Birula, *ibid.* 16, 2038 (1977).

<sup>2</sup>H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., *Opt. Lett.* 3, 218 (1978); G. Orriols, *Nuovo Cimento* B53, 1 (1979); B. J. Dalton and P. L. Knight, *Opt. Commun.* 42, 411 (1982); also, J.

*Phys. B* 15, 3997 (1982).

<sup>3</sup>T. H. Einwohner, J. Wong, and J. C. Garrison, *Phys. Rev. A* 20, 940 (1979).

<sup>4</sup>C. D. Cantrell, V. S. Letokhov, and A. A. Markarov, in *Coherent Nonlinear Optics, Recent Advances*, edited by M. S. Feld and V. S. Letokhov (Springer, New York, 1980), p. 185.

<sup>5</sup>B. W. Shore and R. J. Cook, *Phys. Rev.* 20, 1958 (1979).