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Pulsed Adiabatic Structure and Complete Population Transfer

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Pulsed Adiabatic Structure

The title of my talk mentions pulsed adiabatic structure and population transfer. By the term "pulsed" I have in mind excitation induced by laser pulses, although I shall mention also an example of collisions.

The term "adiabatic" refers, in the present context, to a slowly varying Hamiltonian.

By "structure" I refer to the instantaneous eigenstate structure of this time varying Hamiltonian. These are known variously as dressed states or adiabatic states, to distinguish them from the original unperturbed or undressed or diabatic states that originally occur, before the Hamiltonian begins to change slowly.

This talk is intended as a review of some of the theoretical developments of the last few years.

Adiabatic Population Transfer

Population can be transferred between atomic or molecular energy states in a variety of ways. The basic idea of adiabatic transfer, discussed in many textbooks, is as follows.

We begin with an atom that is in some single energy state (an eigenstate of an initial Hamiltonian). This energy state is one of many possible states, known variously as the unperturbed states or basis states or diabatic states.

Next we begin to change the Hamiltonian very slowly. The changes may occur in either the diagonal elements (the basis state energies) or in the off-diagonal elements (interactions between basis states). If there are off-diagonal elements then the Hamiltonian will no longer commute with the original one. Because the Hamiltonian is no longer the one that was used to define the original basis states, it will cause these states to become mixed.

However, if the change is sufficiently slow, the system can remain in a single eigenstate of the changing Hamiltonian -- an adiabatic state, composed of a combination of basis states.

Finally, at some later time, we examine the system once again in the original basis. We find that the population has undergone a change, and now resides in a different unperturbed state. We have produced population transfer.

The elements of this procedure, idealized to two states, were suggested originally by Landau, Zener and Stueckelberg, and so one often sees references to this as an LZS or Landau-Zener curve crossing. I shall examine this simple model briefly.

Adiabatic Coordinates

We must imagine two coordinate systems for statevectors: that of the original basis states, and that of the slowly changing adiabatic states, defined as eigenstates of the instantaneous Hamiltonian. The coordinate axes represent the complex-valued probabililty amplitudes, whose absolute squares are the populations.

To achieve complete population transfer by adiabatic means, the two coordinate systems must coincide initially. At the end of the process there must again be a simple connection between to two coordinate systems, but now the alignment of the two coordinate systems is different. The result is interpreted as a transfer of population, meaning population referenced to the original basis states.

Examples of Adiabatic Passage

There are many illustrative examples of adiabatic passage, both theory and experiment. I shall mention briefly two common examples, inelasitc collisions between atoms, and the static Stark effect in Rydberg atoms, before continuing with the main objective, a discussion of adiabatic passage induced by laser pulses.

Inelastic Collisions

The essential idea of an inelastic collision, here greatly simplified, is that there are two distinct types of atoms that collide. One of these might be an ion, the other a neutral, or the atoms might differ in some internal structure such as electronic excitation or spin. Initially we have atom A colliding with atom B. As a result of the collision each of these can be altered, say by transferring an electron or by transferring internal excitation. As a result of this rearrangement we can have atom C and atom D. Each of these collision pairs has an interaction energy -- this is the energy that repels the two atoms and prevents them from coalescing. The energies of the two types of pairs, as a function of internuclear separation, form the diabatic energy curves. If there were no possibilty of an inelastic collision, these two curves would be separate. But there is some chance of collision induced change. There is an interaction that mixes these two unperturbed states. It has the greatest effect when the unperturbed energies are equal, that is, when the diabatic curves cross. When we take this interaction into account we have the adiabatic Hamiltonian. The eigenvalues of this Hamiltonian, as a function of separation, form the adiabatic curves. These curves do not cross. They exhibit an avoided crossing.

We can use the forms of these curves to understand what happens during a collision. If the relative velocity of the two atoms is large as the separation nears the crossing point, then the system tends to remain in the original diabatic states, and to follow the diabatic curves. However, if the velocity is sufficiently slow, then the system follows an adiabatic curve.

After the atoms have passed through the curve crossing, the lowest energy curve is now associated with a different combination of basis states, and so a transition has occurred.

In a collision there is a crossing as the atoms approach and another as the atoms move apart. The overall probabillity for a reaction is the product of the two probabilities. In actual atomic collisions there is a distribution of velocities, over which one must average. And there are usually several curve crossings to consider. Thus the theory of adiabatic passage, when applied to inelastic collisions, attempts to predict collision rates that require averages over many conditions. One does not control the time evolution, one accepts what nature provides.

Static Stark shifts in Rydberg atoms

Another example of adiabatic passage occurs when one applies a static electric field to an atom in a Rydberg state. The static field splits the degeneracy of the Rydberg level into various sublevels. By properly tuning a laser a transition can be produced that places population into a particular sublevel. Then the field can be steadily increased. The Stark shift causes the energies of sublevels that originated with different principal quantum numbers to cross. As the field steadily increases a particular sublevel will cross a number of other sublevels. These appear as avoided crossings when one evaluates the eigenvalues of the full Hamiltonian, including the effect of the electric field. Population placed into one sublevel will therefore undergo a succession of crossings or avoided crossings until the field becomes sufficiently intense that the atom ionizes.

Unlike the situation with inelastic collisions, the experimenter can control the details of the time evolution produced by Stark shifts. By carefully controlling the rate of field increase one can guide the population through the various crossings and produce a predictable final state.

Examples, adiabatic Passage via Lasers

Today I want to discuss a number of examples of adiabatic passage induced by laser radiation.

The simplest of these examples, a two state atom acted on by a pulse of laser radiation, can be very much like the LZS model or the collision model, except that one can control the time variation of both the diagonal and the off-diagonal elements of the Hamiltonian. The diagonal elements vary with the frequency detuning of the laser away from the atomic Bohr frequency, while the off-diagonal elements depend on the instantaneous pulse intensity.

Another class of examples, similar to what happens with the Stark shift in Rydberg atoms, occurs when one sweeps the frequency of a laser across a band of energy states, each of which has an allowed dipole transition to the originally populated state.

Perhaps the most interesting class of excitation problems occur when one has a chain of energy levels. The three-state case is the simplest of these, but more elaborate chains have been considered. One may sweep the frequency in various ways, or one may combine frequency variation with pulse variation. As I shall point out, a particularly interesting class of excitation events become possible when we can have two or more distinct but overlapping pulses, one for each transition. Some very interesting effects then become possible.

Coherent Excitation Equations (1)

Before proceeding, it is useful to present a few of the basic equations. I shall be consemed with coherent excitation, so that either the Schrödinger equation for a statevector or the Liouville equation for a density matrix may be used. I shall follow the former approach and shall assume that the initial state is a single quantum state. The Liouville equation is simpler when one wishes to consider initial conditions that involve incoherent mixtures of states.

When one wishes to include homogeneous incoherence, such as occurs with spontaneous emission, then it is neccessary to work with a density matrix and to generalize the Liouville equation. Such treatments have been made. But to emphasise the essential physics of adiabatic passage is is a little simpler to deal with statevectors than with density matrices.

Coherent Excitation Equations (2)

Given the equation of motion, in our case the Schrodinger equation, we choose some set of basis states and express the statevector in terms of these states. These states, the basis states, should be such that in the absense of the laser field they are eigenstates of the atomic Hamiltonian. We wish to determine probabilities for making a transition between these states as a result of applying the laser radiation.

Solving the Equations

We are faced with the need to solve a set of simultaneous ordinary differential equations. The number of equations is the number of basis states that are important. The coefficients are the matrix elements of the Hamiltonian for the atom acted on by the field, and so these generally depend on time.

There are several practical ways to solve these equations. Naturally it is always possible to find numerical solutions. Numerous excellent methods exist for producing solutions, and these can be implemented for any case of practical interest.

For certain special choices of pulse shapes and frequency variation it is possible to obtain analytic solutions, that is, solutions expressed in terms of various special functions of 19th century mathematicians. Often these solutions can be manipulated to produce asymptotic values that are not readily obtainable by numerical methods -- for example, limits of infinitely long time durations.

Lastly, there are various approximate methods that may be applied when conditions warrent them. I shall be considering one of these, the adiabatic approximation, applicable when the Hamiltonian varies slowly. Another complementary regime occurs when the Hamiltonian changes abruptly and thereafter remains constant.

Conventional Adiabatic Approximation

The essence of the adiabatic approximation is as follows. At each instant of time we consider the instantaneous Hamiltonian. We construct the eigenstates and eigenvalues for this Hamiltonian. This is very easy if there are only two states, but it can also be done in a number of other cases. These eigenstates do not generally coincide with the actual statevector, because the Hamiltonian at one time does not commute with that at another time. Nevertheless, under certain circumstances the statevector may be very closely approximated by one of these states. At any rate, we can always express the actual statevector as a linear combination of the adiabatic states. In general the coefficients in this expansion will change with time. However, if the Hamiltonian is sufficiently slowly varying, then these coefficients remain nearly constant. This is the adiabatic regime. All time dependence of the statevector then occurs in the time dependence of the adiabatic states.

The condition for the adiabatic approximation to hold is that the change in the adiabatic states should be slow compared with the differences of the adiabatic eigenvalues.

Adiabatic Following

Adiabatic following occurs when the following conditions hold. First, we should start in a single quantum state. Then the Hamiltonian should change sufficiently slowly that the adiabatic condition holds. We then follow the time evolution until the adiabatic state is again a single basis state.

LZS Curve crossing

Before discussing more complicated, and more realistic, cases, it is instructive to review the simple LZS model of adiabatic passage. The model assumes that there are only two energy levels of importance, that the off-diagonal elements of the Hamiltonian in the unperturbed basis are fixed, and that the diagonal elements (the diabatic energies) vary linearly with time. Time varies from minus infinity to plus infinity. The diabatic energies, plotted as a function of time, appear as two straight lines that cross at time zero.

The LZS Transition Probability

Analytic solutions to this two state model are known; they involve parabolic cylinder functions. From asymptotic forms for these functions, or by various other analytical techniques, one can deduce the desired transition probabilities, that is, the probability that the system will be found in a particular basis state in the distant future, given that it was in a particular state in the remote past.

From the LZS formula we can determine the conditions that are required for complete population transfer. They are that the interaction be strong or that the sweep be slow. These are the conditions for adiabatic passage.

It should be recognized that thes LZS formula refers to an infinite time duration. For finite times, near to the crossing time, the probabilities typically exhibit oscillations. The oscillations die away, and the probabilities approach the LZS values as time increases.

Pulsed Two-State Atom

The simplest example of coherent atomic excitation is a two-state atom excited by a pulse of nearly monochromatic radiation whose carrier freqeuncy is nearly resonant with the Bohr frequency of the atom.

After making the traditional rotating wave approximation, which is valid when the radiation is not too intense and is close to resonance, we obtain a 2 x 2 Hamiltonian matrix characterized by two functions of time: the detuning (or difference between carrier and Bohr frequency) and the Rabi frequency (the product of dipole transition moment and instantaneous electric field).

Two-State Solutions

A variety of analytic solutions are known for various analytic forms for pulse shapes. In addition to the simple LZS model of a linearly changing detuning and constant Rabi frequency, the known solutions include the case of a hyperbolic secant resonant pulse, pulses with changing detuning, and asymmetric pulses.

Numerical solutions are, of course, always possible. One must sometimes take care in comparing numerical with analytic solutions, which may treat infinite times.

When the pulse is slowly varying, then it may be possible to use adiabatic eigenstates todescribe the time evolution.

Adiabatic States

The construction of adiabatic states for a two-level system is quite simple. The eigenvalues are the two roots of a quadratic equation, and from these one can construct the two eigenvectors. The components of these vectors can be expressed as sine and cosine of a mixing angle.

The traditional adiabatic passage occurs when one sweeps the detuning across resonance, either by changing the laser frequency or by shifting the Bohr frequency. As the detuning runs between a large negative value and a large positive value, the mixing angle changes by π and the adiabatic state changes from identity with one basis state to identity with the other. That is, population transfer occurs.
Pulsed Two-State Atom

Examples of two-state pulses

When the field amplitude remains constant, at least during the vicinity of the curve crossing, and the detuning varies linearly, then we have the traditional LZS curve crossing. Plots of eigenvalues are useful in two extremes: very fast chirp, or very slow chirp.

The LZS theory provides a simple expression for the probabililty of population transfer for any value of Rabi frequency or detuning rate, whether or not the transition is adiabatic. Complete population transfer will occur if the pulse is sufficiently slow or sufficiently strong.

Two levels, pulse without chirp

When detuning remains fixed, say at resonance, and the Rabi frequency has the time dependence of a pulse, the populations undergo Rabi oscillations. The population transfer depends on the pulse area: if the area is π, 3π, 5π, etc. then complete transfer occurs, whereas if the area is 2π, 4π, etc. then the population returns, at the end of the pulse, to its original location. Plots of adiabatic eigenvalues do not assist in determining the final population in this case.

Two levels, pulse with chirp

When both frequency variation and amplitude modulation occur in a pulse, the behavior may be either adiabatic or diabatic.The changing pulse amplitude distorts the population histories from the pattern they have with the LZS adiabatic transfer. There is a tendency for the populations to equilibrate during the interval when detuning is small and amplitude is large.

Sublevels, single pulse

The elementary LZS theory generalizes in several ways. One class of extensions occurs when the initially populated level, the ground level, has dipole linkages to a band of closely spaced levels.

The Hamiltonian matrix for this case is a bordered matrix (in contrast to a banded tridiagonal matrix that occurs for ladder-linkage excitation)

Curve Crossings

When the dipole moments are weak, the pattern of adiabatic energies appears as a succession of independent curve crossings. The populations oscillate in time, but approach limiting values predicted by the LZS formula.

When the evolution is not completely adiabatic, population is left in the various sublevels as the chirp passes through the several resonances.

Sublevels, Adiabatic Chirp

When the evolution is adiabatic, then population is transferred to the first of the sublevels whose Bohr frequency matches the laser. The remaining sublevels do not participate in the evolution.

In modelling this evolution it is neccessary to employ a pulse that turns on smoothly while the detuning is still large. If the pulse is taken as a square pulse, the abrupt initiation introduces oscillations in the population history.

Sublevels, Pulsed Adiabatic Chirp

When the pulse has appreciable variation during the interval of population transfer, the population histories vary appreciably from those of the LZS type chirp. There is a tendency of the populations to equilibrate during the most intense portion of the pulse, although ultimately the population transfer is to a single sublevel.

Population transfer by pulses without chirp

It has been suggested by Peterson and Cantrell that population might be transferred into a quasicontinuum of sublevels by means of pulses that maintain constant frequency, tuned off resonance. For such a scheme to work the first portion of the pulse must rise very slowly, producing various curve crossings, and then the pulse is abrubptly terminated.

Although the adiabatic states produced in this way could, in principle, transfer population, it is not easy to achieve the required slow pulse growth.

Three Level Systems

Let us turn next to the simplest of the excitation chains, a three0level atom excited by at most two laser beams. There are three possible configurations of a coherently excited three-level system, often termed the vee, the lambda and the ladder. In both the vee and the ladder population starts in one end of the chain, so these two linkages are equivalent. In the vee system population starts in the center level of the chain, so it can exhibit interesting interference effects.

For definiteness let us consider the ladder system. We wish to transfer population from the initial state 1 to the terminal state of the ladder, state 3.

Although the systems differ in initial conditions, they have the same Hamiltonian, a matrix characterized by two Rabi frequencies and two detunings. There are various ways to produce population inversions. Some involve simple frequency chirps, generalizations of the LZS curve crossings. The more interesting cases are those in which there are two distinct pulses.

3 Levels, Single chirp

Consider the case of constant amplitude Rabi frequencies and time varying detunings. There are actually three detunings to consider. These involve two single-photon resonances and one two-photon resonance. We can choose the ordering in which we sweep through these several resonances.

Intuitive Order

The intuitive order is that in which we first bring the 1-2 transition into resonance and then the 2-3 transition into resonance. If each transition is adiabatic we would first transfer all population into level 2 and thensfer this population into level 3.

This ordering can succesfully transfer population from level 1 to level 3.

Counterintuitive Order

An alternative possibility is to pass through the two-photon resonance 1-3. In this case we first encounter the 2-3 resonance (though there is no population in level 2 to be affected) and then we encounter the 1-3 resonance.

This ordering can also transfer population. Indeed, as Oreg, Hazek and Eberly showed, this ordering can be a more efficient mechanism to transfer population than is the intuitive ordering.

Delayed Pulses

These chirping schemes make use of frequency variation at constant Rabi frequency to produce population transfer. What if we allow pulse variation of the two Rabi frequencies?

Let us consider the case in which the two pulses are each resonant with their respective transitions. We allow the pulses to be independent: either one may come first or they may occur simultaneously. for definiteness we consider a lambda system, in which the middle level lies highest, although the ladder dynamics are identical. Our system is that of stimulated Raman scattering.

Intuitively one might expect that it would be preferable to apply first the pulse that produces the 1-2 excitation (the pump pulse), followed by the pulse that produces the 2-3 excitation (the Stokes pulse). Rather surprisingly, this is not the case.

STIRAP

Experiments show, and theory supports, the conclusion that it is preferable to apply the pulses in a counterintuitive sequence. It is better to apply the Stokes pulse first.

Three-Level Adiabatic States

One way to understand this behavior is by an examination of the adiabatic states of the 3 level system. For simplicity, we assume that all transitions are resonant. Then one eigenvalue is null, and the other two are roots of a quadratic equation. The eigenstates are quite simple. The null-eigenvalue state is expressible as a combination of states 1 and 3, weighted by the values of Rabi frequencies. The other two eigenstates both involve all three basis states.

The null-value eigenstate has very interesting properties. When the Stokes pulse dominates, this state approaches the basis state 1. When the pump pulse dominates, this state approaches the basis state 3. At no time does this state contain any component of state 2. The consequences of this construction are the following.

The STIRAP Process

We start by applying the Stokes field while the atom is in basis state 1. Then this state is also, very nearly, the adiabatic null-eigenvalue state.

We maintain the adiabatic condition while diminishing the Stokes field and increasing the pump field. The atom stays in the same adiabatic state.

Finally we turn off the pump field. The atom remains in the same adiabatic state, which now coincides with basis state 3. We have produced adiabatic passage from state 1 to state 3. This is an example of stimulated Raman adiabatic passage, or STIRAP.

Extensions of STIRAP

The original concept of counterintuitive pulses was worked out for the rather simple case of a nondegenerate three level atom. Quite naturally, one wonders whether the various corrections needed to make this a more realistic model, particularly sources of relaxation and incoherence, will destroy the ability to move population. Several subsequent studies have shown that modifications need not disrupt the adiabatic passage.

A first concern is for spontaneous emission from the intermediate state. Because there is very little population in this state such decay has little effect.

Noise in the pulse (appearing as a broad bandwidth) will introduce incoherence that can prevent coherent population transfer. However, if the bandwidth remains narrow the process continues to work.

Connections between the essential STIRAP levels and other levels need not disrupt the process.

The intermediate level of the simple model can be generalized to a band of levels. It has been suggested by Hioe and Carrol that the process will succeed even when the intermediate states are a continuum.

5-Level STIRAP

It is possible to extend the STIRAP concept to excitation along a chain of levels, by means of multiple pulses.

A particularly simple analytic expression becomes possible when the number of levels is an odd integer. Consider, as an example, the 5-level system. We can construct, in all of these systems, STIRAP solutions that require only two pulse shapes, The pulses are applied in counterintuitive order. It is important to maintain resonance conditions for 2-photon and 4-photon transitions, but the other detunings are arbitrary.

This model has simple analytic solutions that predict complete population transfer for adiabatic behavior.

4-Level STIRAP

The model discussed for the 5 level system will not work for an even number of levels. However, it is possible to find relatively simple solutions for the 4 level system too. What is required is that there be nonzero detunings for 1-photon and 2-photon transitions, while the 3-photon transition must be resonant. It is possible to achieve population transfer with either 3 or two pulse shapes. Two of these must be applied in the counterintuitive order, while the middle pulse need only be present during the overlap of these two.

This model also permits complete population transfer. Like the 5-level system, it does place a nonzero population into the intermediate levels.

Adiabatic Passage and the Density Matrix

Although I have described coherent excitation by means of the Schrodinger equation and a statevector, much interesting insight can be obtained by examining the density matrix. By considering a lossless system, so that probability is conserved, the number of independent elements of the density matrix for an N level system is reduced from N^2 to $N^2 - 1$. The Lie group associated with this matrix is SU(N).

When the elements of the density matrix are suitably organized, they can be presented as the real-valued components of a vector in a space of dimension $N^2 - 1$. For the two level atom this organization gives the three components of the Bloch vector.

The generalization of the Bloch vector to multilevel atoms has been pointed out by Elgin and by Hioe and Eberly.

When one has characterized the density matrix as a vector, one also obtains a corresponding characterization of the Hamiltonian matrix as a vector. Adiabatic passage can be viewd as time evolution in which these two vectors remain parallell. (Or, more accurately, in which they both remain in the same subspace of the full space.)

Conclusion

There are a variety of ways in which adiabatic passage can be used to transfer population in multilevel systems.

Plots of adiabatic energies and curve crossings offer guidance in some of these instances. What is really required, however, is an appreciation of the structure of the adiabatic states, ie. how their basis-state components change as pulsed excitation proceeds.

Pulsed Adiabatic Structure

and

Complete Population Transfer

$\hbar$

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Pulsed Adiabatic Structure

Pulsed: Laser induced

Adiabatic: Slowly varying $H(t)$

Structure: Instantaneous eigenstates of $H(t)$

Adiabatic or Dressed states

Adiabatic Passage

Basic idea:

- Start in a single energy state

  (Unperturbed or basis or diabatic)

- Slowly change the Hamiltonian

  (Diagonal or off diagonal elements)

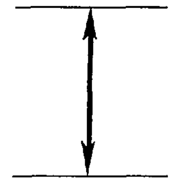
- Later will be in a different unperturbed state

Examples, Adiabatic Passage

Two States

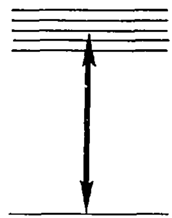
Chirp (LZS)

Pulse



Sublevels

Chirp (multi LZS)

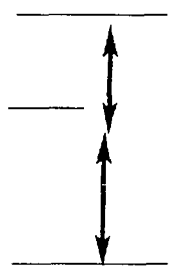


Ladder

Chirp

Single pulse

Multiple Pulses



Coherent Excitation Equations

Schrödinger

$$i\hbar\frac{\partial}{\partial t}\Psi(t) = H(t)\ \Psi(t),$$

Liouville

$$i\hbar\frac{\partial}{\partial t}\rho(t) = [H(t),\ \rho(t)] \qquad [1]$$

Completely coherent

Facilitate partial coherent

Extension to incoherent

Coherent Excitation Equations

Introduce Basis States

$$\Psi(t) = \sum_{n=1}^{N} c_n(t)\ \psi_n \qquad 1 = \sum_{n=1}^{N} | c_n(t) |^2 \qquad [2]$$

$$P_n(t) = | \langle \Psi(t) | \psi_n \rangle |^2 = | c_n(t) |^2 = \rho_{nn}(t) \qquad [3]$$

Obtain coupled linear ODEs

$$i\hbar \frac{d}{dt} c_n(t) = \sum_{m=1}^{N} H_{nm}(t)\ c_m(t) \qquad [4]$$

Transition probabilities

$$P_1(-\infty) = 1 \qquad P(1 \rightarrow m) = P_m(+\infty)$$

Solving the Equations

Solving coupled linear ODEs

$$i\hbar \frac{d}{dt} c_n(t) = \sum_{m=1}^{N} H_{nm}(t)\ c_m(t) \qquad [5]$$

Numerical

Any case

Analytical

Special pulses

Approximations

Abrupt change, then constant

Slow variation: **Adiabatic**

Conventional Adiabatic Approximation

Adiabatic Eigenstates

$$H(t)\ \Phi_n(t) = \hbar\ \omega_n(t)\ \Phi_n(t), \qquad [6]$$

$$\Psi(t) = \sum_{n=1}^{N} a_n(t)\ \Phi_n(t) \exp\left[-i \int_0^t dt'\ \omega_n(t) \right]. \qquad [7]$$

$$1 = \sum_{n=1}^{N} |\ a_n(t)\ |^2 \qquad [8]$$

Adiabatic Condition (remains in same adiabatic state)

$$\left| \left\langle \Phi_h \left| \frac{d}{dt} \Phi_n \right. \right\rangle \right| << |\ \omega_h - \omega_n\ | \qquad [9]$$

(Slow change wrt.. Eigenvalue differences)

Adiabatic Following

Require

- Initially single basis state
- Initial basis state = single adiabatic state
- Adiabatic condition thereafter

LZS Curve Crossing

Landau-Zener Stueckleberg (LZS) Model

Constant interaction (off diagonal H elements)

Linear varying energies (diagonal H elements)

Two-state Hamiltonian

$$H(t) = \begin{bmatrix} E_0 & V \\ V & E_0 - st \end{bmatrix} . \quad [10]$$

Analytic solutions = parabolic cylinder functions

Asymptotic values $\Rightarrow$ transition probabilities

Adiabatic LZS solutions

Adiabatic Hamiltonian

$$H^{ad}(t) = \hbar \begin{bmatrix} \omega_+(t) & 0 \\ 0 & \omega_-(t) \end{bmatrix} \qquad [11].$$

Eigenvalues

$$\hbar\omega_\pm(t) = (st/2) \pm \sqrt{(st/2)^2 + V^2} \qquad [12]$$

Adiabatic States

$$\Phi_+(t) = \begin{bmatrix} \cos\vartheta(t) \\ \sin\theta(t) \end{bmatrix}, \qquad \Phi_-(t) = \begin{bmatrix} -\sin\theta(t) \\ \cos\theta(t) \end{bmatrix} \qquad [13]$$

$$\tan 2\theta(t) = -2V / st \qquad [14]$$

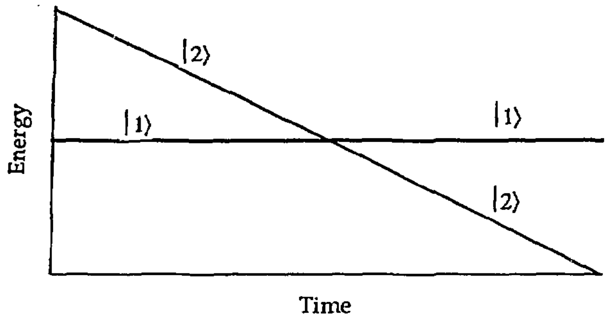
Time evolution

$$-\infty \rightarrow t \rightarrow \infty$$

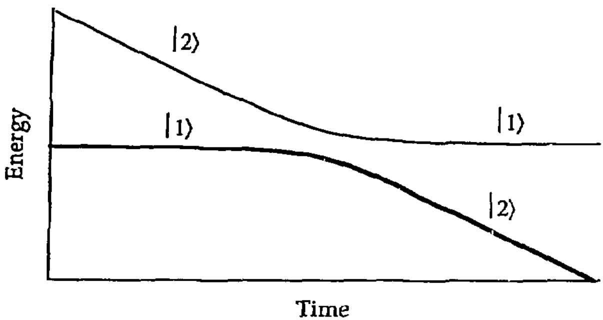
$$0 \rightarrow 2\theta \rightarrow 180^o$$

$0 \rightarrow \theta \rightarrow 90^o$ *population inversion*

Diabatic Energies



Adiabatic Energies



LZS Evolution

Rapid (Diabatic) Evolution: no transition

$$\Psi(t) = \begin{cases} \psi_1 & \text{for } t \to -\infty \text{ (past)} \\ \psi_1 & \text{for } t \to +\infty \text{ (future)} \end{cases} \qquad [15]$$

Slow (Adiabatic) Evolution: transition

$$\Psi(t) = \begin{cases} \Phi_+ = \psi_1 & \text{for } t \to -\infty \\ \Phi_+ = \psi_2 & \text{for } t \to +\infty \end{cases} \qquad [16]$$

The LZS Transition Probability

Probability over infinite time, $-\infty \rightarrow t \rightarrow \infty$

$$P(1\rightarrow 1) = \exp\left[-\frac{2\pi V^2}{\hbar s} \right] \rightarrow \begin{cases} 0 & \text{if adiabatic (large } V \text{ or small } s) \\ 1 & \text{if diabatic (small } V \text{ or large } s) \end{cases}$$

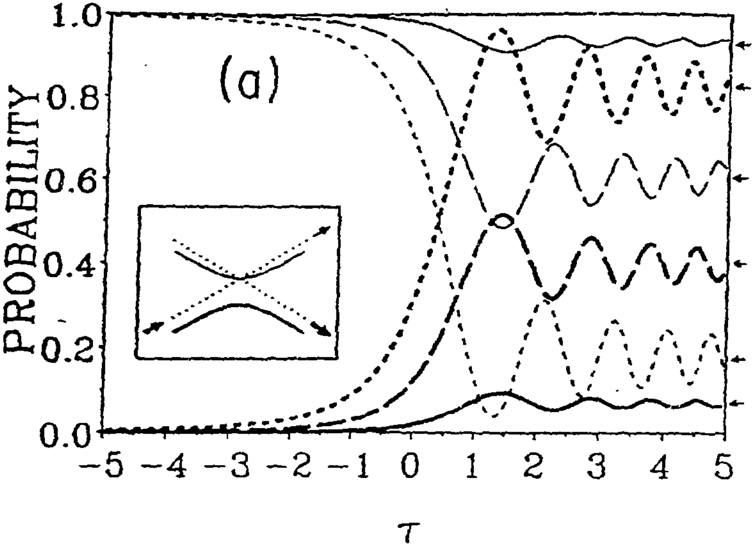
$$P(1\rightarrow 2) = 1 - P(1\rightarrow 1) \rightarrow \begin{cases} 1 & \text{if adiabatic} \\ 0 & \text{if diabatic} \end{cases}$$

Complete Transfer = Adiabatic Passage requires

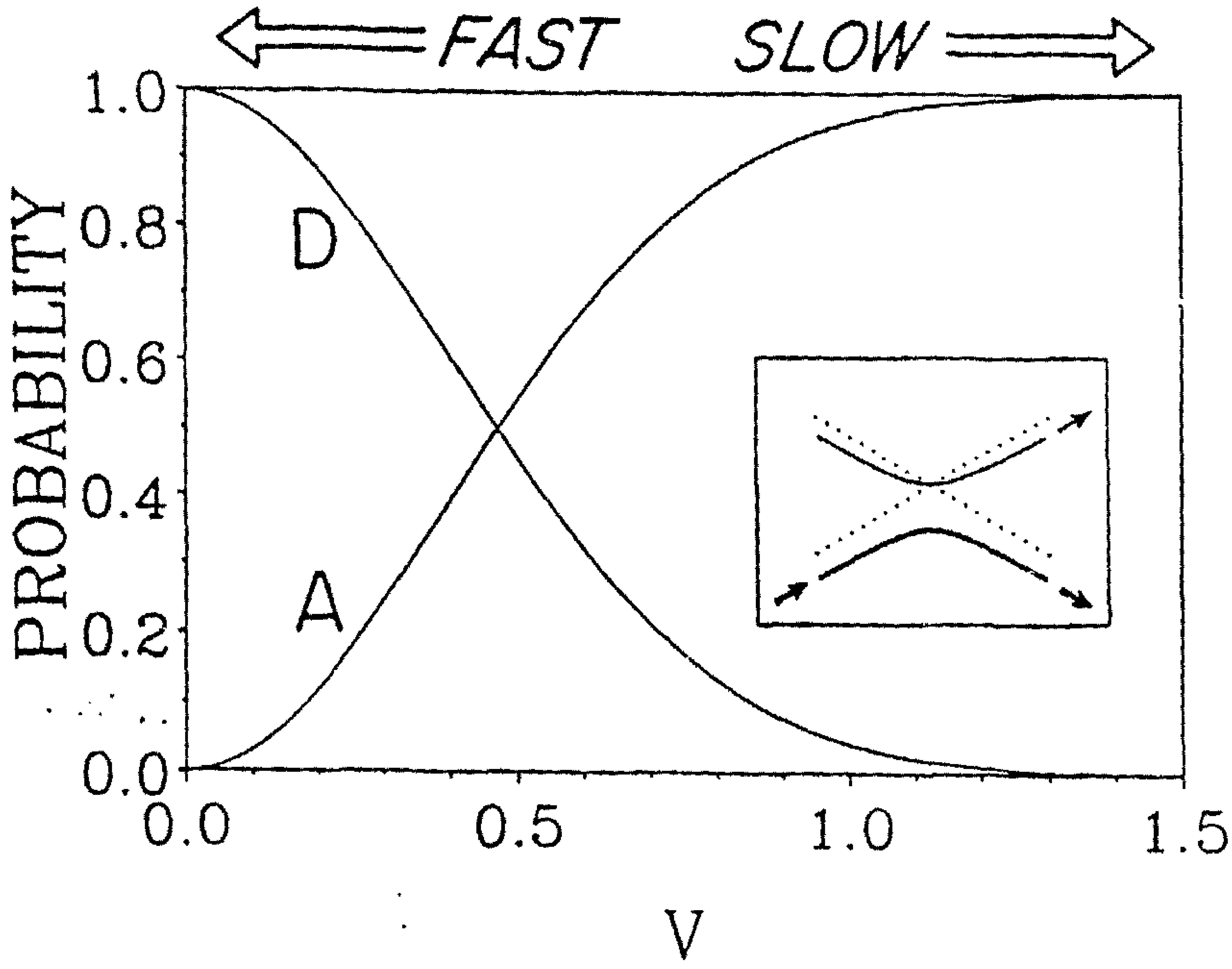
$$2\pi V^2 >> |\hbar s| \quad \text{(strong or slow)}$$

More generally

$$2\pi |H_{12}|^2 >> \left| \hbar \frac{\partial}{\partial t}(H_{11} - H_{22}) \right|$$



Harmin 91



Harmin 91

LZS Shortcomings

- Real system is not constant V for $-\infty < t < \infty$
- Must turn on V well before crossing
- Turn on must not be abrupt

Inelastic Collisions

Two states, A + B and C + D

$$H(R) = \begin{bmatrix} E_{AB}(R) & V(R) \\ V(R) & E_{CD}(R) \end{bmatrix}. \quad [17]$$

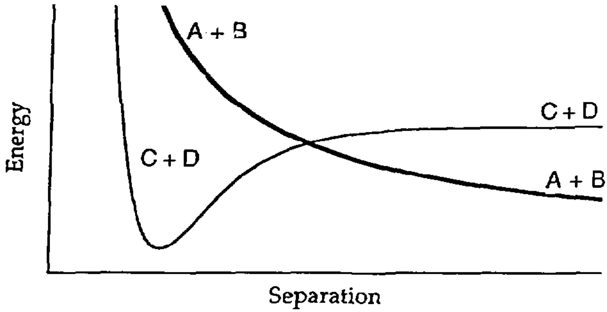
Adiabatic States $\Phi_+ \rightarrow \psi_{AB}, \quad \Phi_- \rightarrow \psi_{CD}.$

$$H^a(R) = \begin{bmatrix} E_+^a(R) & 0 \\ 0 & E_-^a(R) \end{bmatrix}. \quad [18]$$

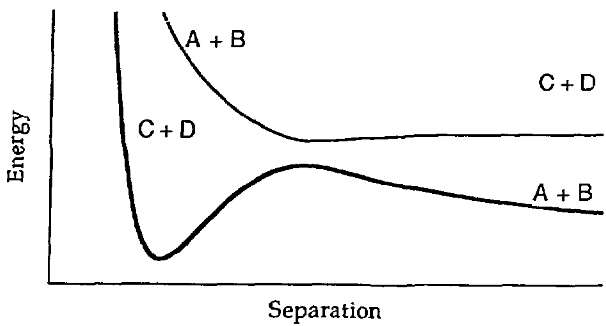
Adiabatic Condition (complete transfer)

$$2\pi | H_{12} |^2 >> | \hbar \frac{dR}{dt} \frac{\partial}{\partial R}(H_{11}-H_{22}) | \quad [19]$$

Diabatic Energies

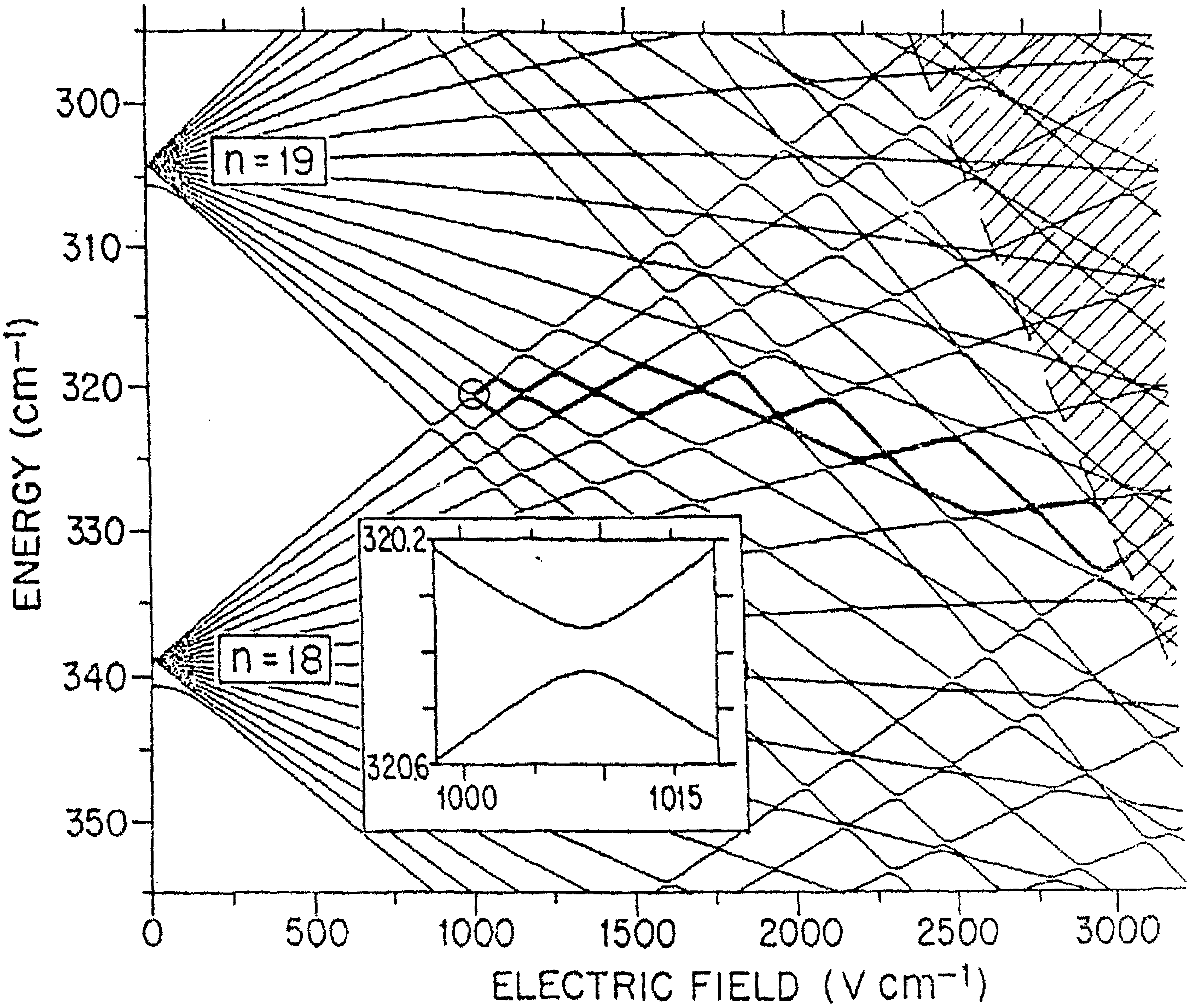


Adiabatic Energies

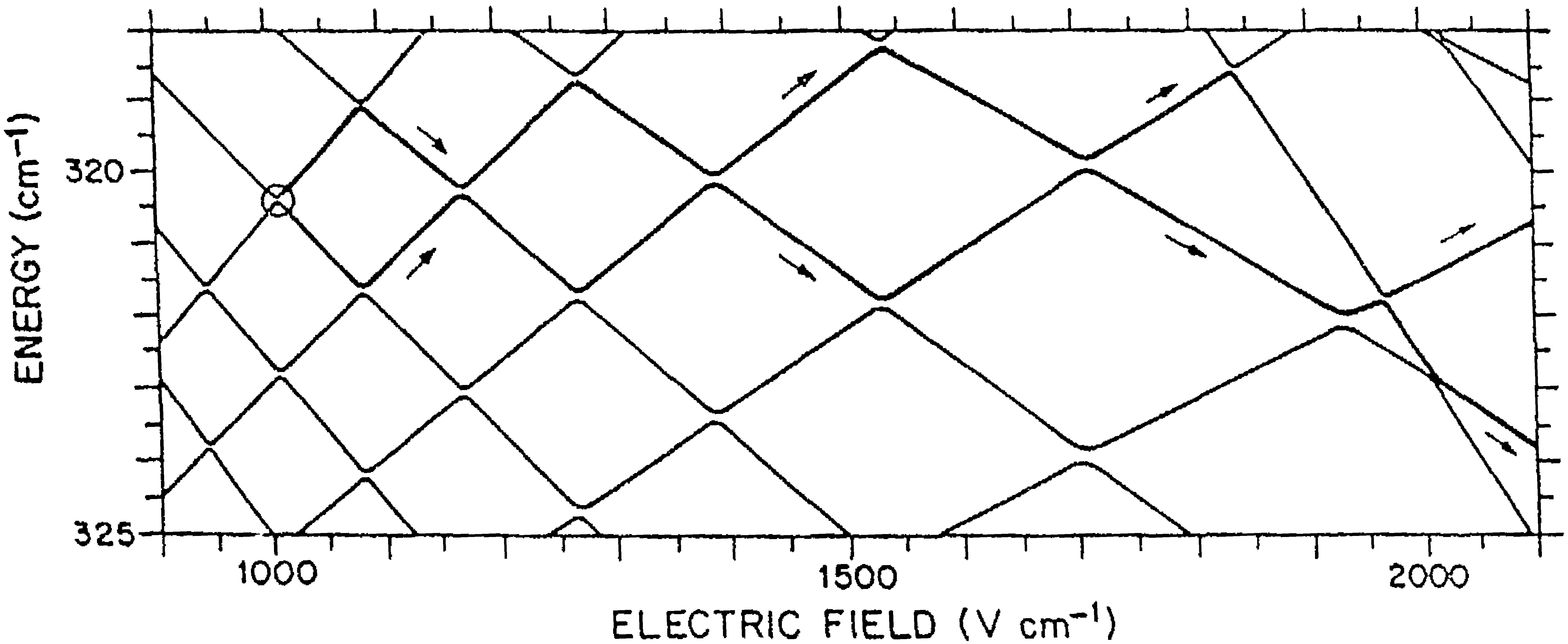


Rydberg Atoms

- Stark splittings of degenerate levels
- Multiple LZS crossings
- Population transfer at crossings



n = 19
n = 18
300
310
320
330
340
350
ENERGY (cm^{-1})
0
500
1000
1500
2000
2500
3000
ELECTRIC FIELD (V cm^{-1})
320.2
320.6
1000
1015



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Pulsed Two-State Atom

Two-state Hamiltonian

$$H(t) = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega(t) \\ \Omega(t) & 2\Delta(t) \end{bmatrix}. \qquad [20]$$

Adiabatic states

$$\Phi_+(t) = \begin{bmatrix} \cos\theta(t) \\ \sin\theta(t) \end{bmatrix}, \qquad \Phi_-(t) = \begin{bmatrix} -\sin\theta(t) \\ \cos\theta(t) \end{bmatrix} \qquad [21]$$

$$\tan 2\theta(t) = -\Omega(t) / \Delta(t). \qquad [22]$$

Eigenvalues

$$\omega_\pm(t) = \Delta(t) \pm \sqrt{\Delta(t)^2 + \Omega(t)^2} \qquad [23]$$

Population inversion: for change in sign of $\Delta(t)$

Examples of Two-State Pulses

Analytic solutions

Special cases (with and without detuning)

Numerical solutions

- LZS: Constant Amplitude, Linear Chirp

  Can have Adiabatic passage

- Pulsed Amplitude, Constant Frequency

  No population transfer

- Pulsed Amplitude and Linear Chirp

  Can have Adiabatic passage

Examples, Adiabatic Passage

- Pulsed, constant tuning
- Chirped, constant amplitude
- Pulsed with chirp

2 Levels, Pulse without chirp

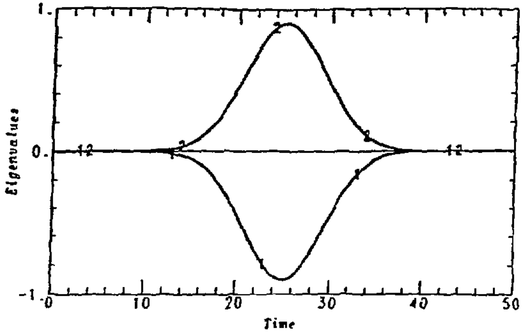
Resonant excitation

Population transfer depends on pulse area

complete for π, 3π, 5π, ...

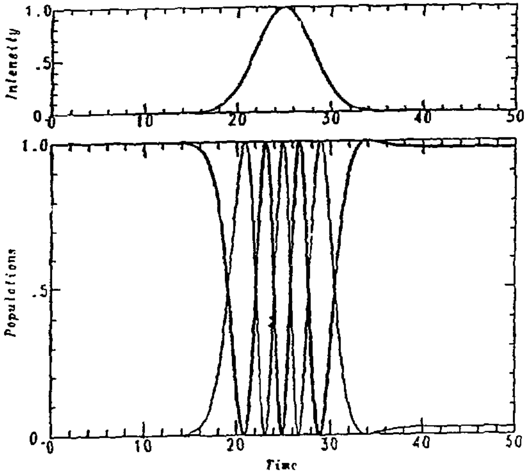
null for 0, 2π, 4π, ...

Pulse



Pulse

N = 2

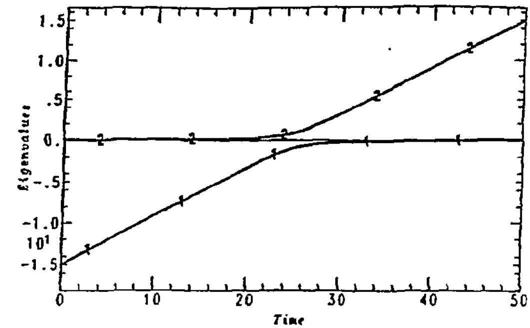


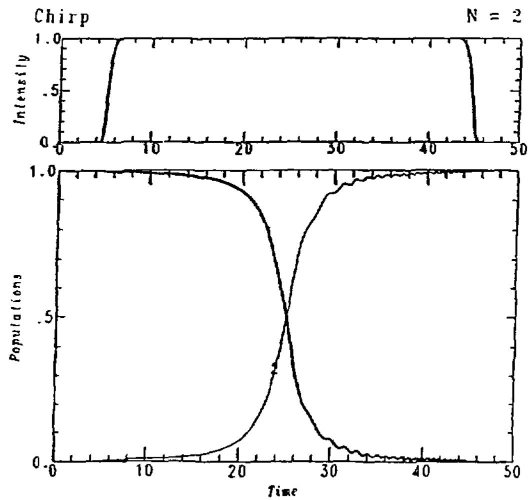
frame 4 12:06.05 01-29-92 y

2 Levels, Chirp (LZS)

- Numerical time window is finite
- Need to turn on interaction in past
- Complete population transfer if LZS predicts

Chirp


Eigenvalues
Time
1.5
1.0
.5
0.
-.5
-1.0
-1.5
0
10
20
30
40
50


Chirp
N = 2
Intensity
Populations
Time
1.0
.5
0
0
10
20
30
40
50

2 Levels, Pulse with chirp

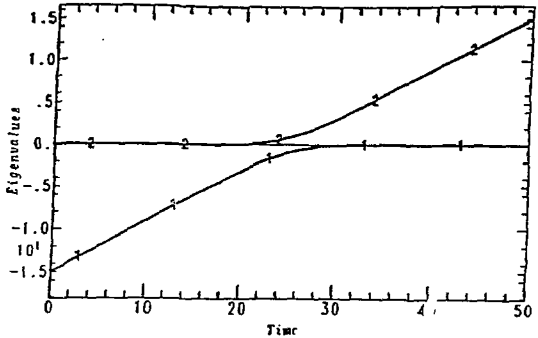
Weak pulse

complete, like LZS result

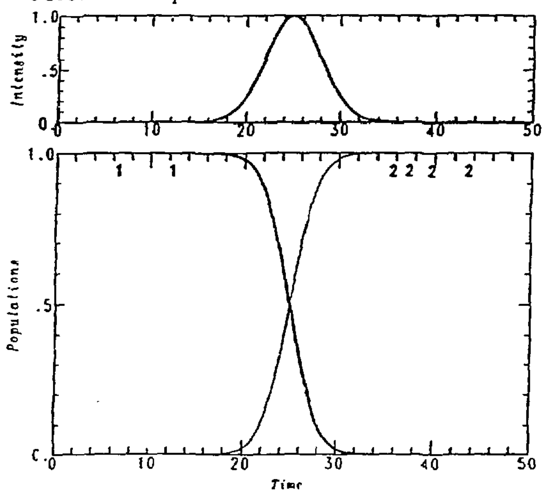
Strong pulse

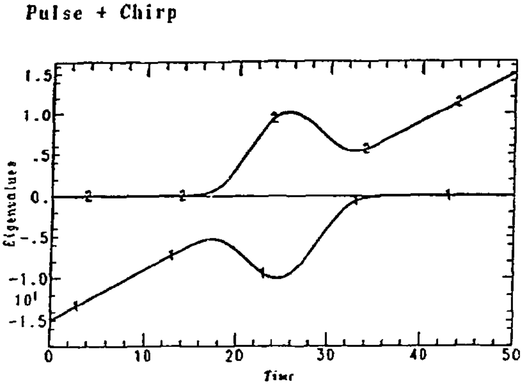
complete, intermediate equilibration

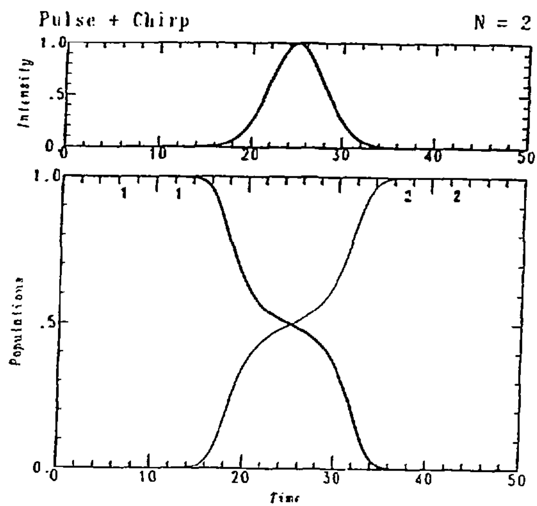
Pulse + Chirp



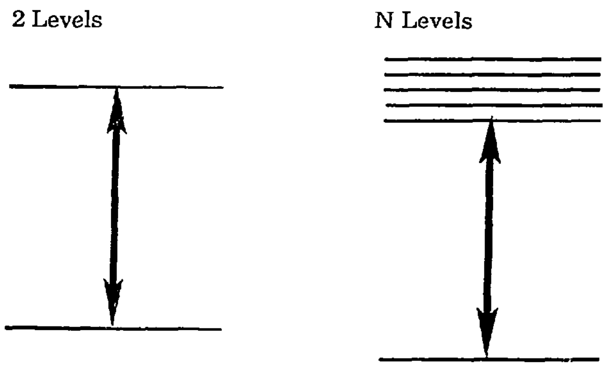
Pulse + Chirp N = 2




Pulse + Chirp
Eigenvalues
Time
1.5
1.0
.5
0.
-.5
-1.0
10¹
-1.5
0
10
20
30
40
50


Pulse + Chirp
N = 2
Intensity
Populations
Time
1.0
.5
0
0.
0
10
20
30
40
50

Sublevels, Single Pulse

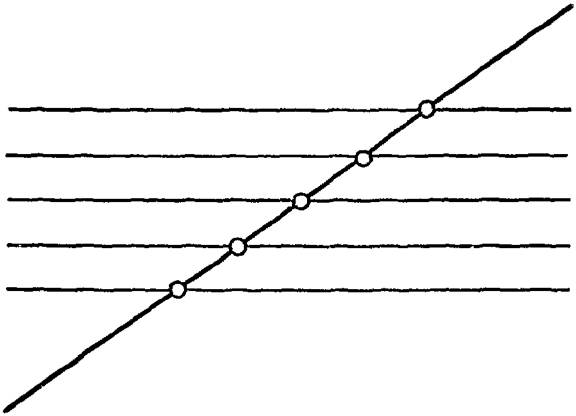


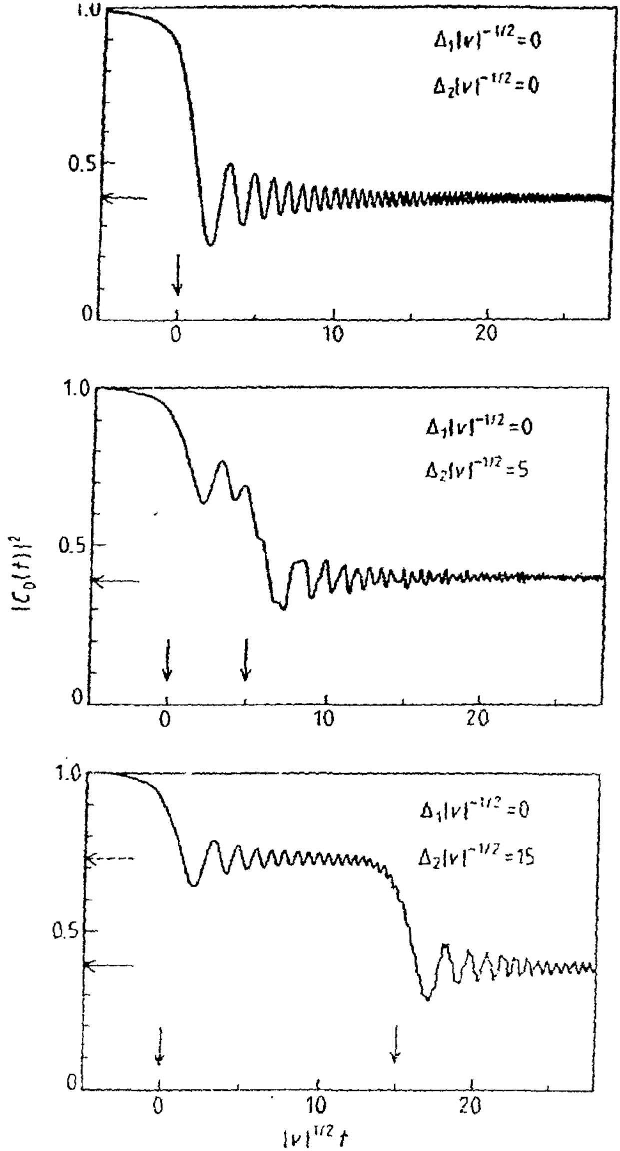
Sublevel Hamiltonian

Single level, Multiple crossings

$$H = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_1 & \Omega_2 & \Omega_3 & \cdots \\ \Omega_1 & 2\Delta_1 & 0 & 0 & \cdots \\ \Omega_2 & 0 & 2\Delta_2 & 0 & \cdots \\ \Omega_3 & 0 & 0 & 2\Delta_3 & \cdots \\ \vdots & 0 & 0 & 0 & \ddots \end{bmatrix}$$

Curve Crossings Multilevel, single chirp





1.0
0.5
0
0
10
20
Δ1|v|^-1/2=0
Δ2|v|^-1/2=0
Δ1|v|^-1/2=0
Δ2|v|^-1/2=5
|C0(t)|^2
Δ1|v|^-1/2=0
Δ2|v|^-1/2=15
|v|^1/2 t

'ayanrira... 85

Multilevel Adiabatic Passage via Pulse

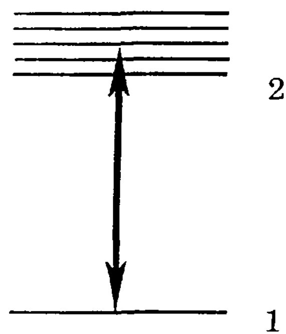
Transfer with constant frequency

Only possible for N > 2 Levels

Consider quasicontinuum,

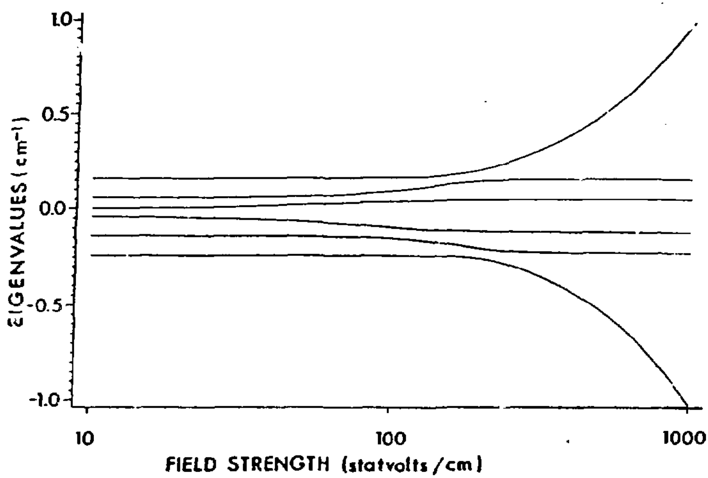
Evenly spaced

Not Resonant $1 \longleftrightarrow 2$



EIGENVALUES OF A (1,5) SYSTEM

ν = 947.96 cm^{-1}



Peterson, Cantrell 84

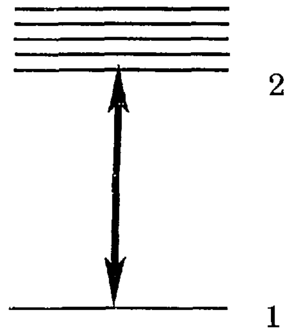
Multilevel Adiabatic Passage via Chirp

Consider quasicontinuum,

Evenly spaced

Equal Rabi

Resonant $1 \longleftrightarrow 2$

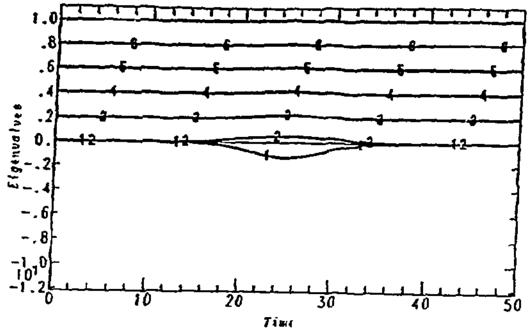


Sublevels, Weak Pulse

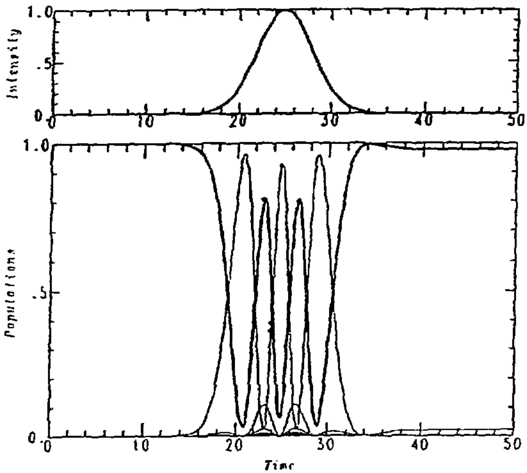
Only two levels involved (resonant)

Population transfer depends on pulse area

Pulse

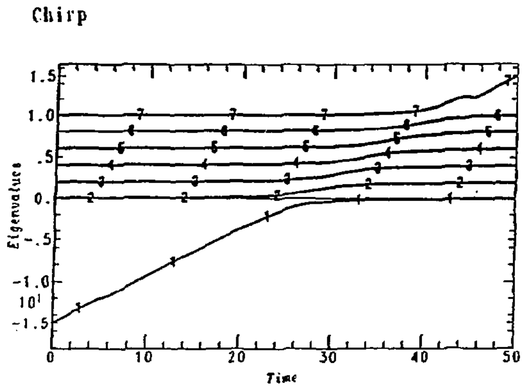


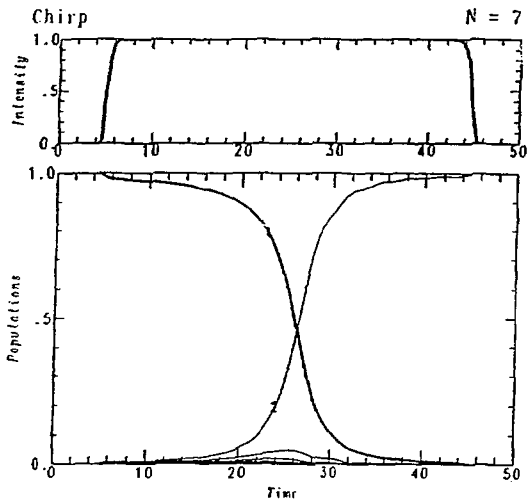
Pulse N = 7



Sublevels, Chirp

Can get complete population transfer to first crossing


Chirp
Eigenvalues
Time
10^1


Chirp
N = 7
Intensity
Populations
Time

Sublevels, Pulsed Chirp

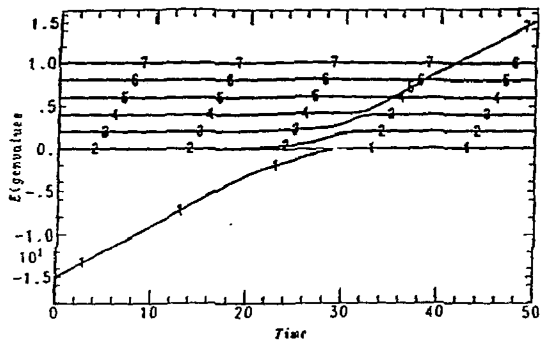
Weak pulse

- Complete transfer, first crossing
- Like LZS

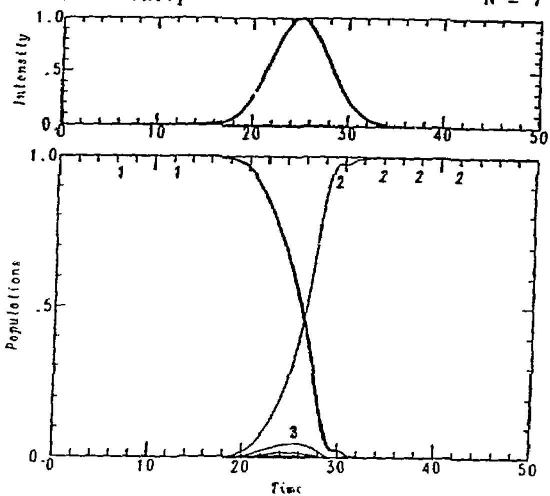
Strong pulse

- complete transfer, first crossing
- Intermediate equilibration

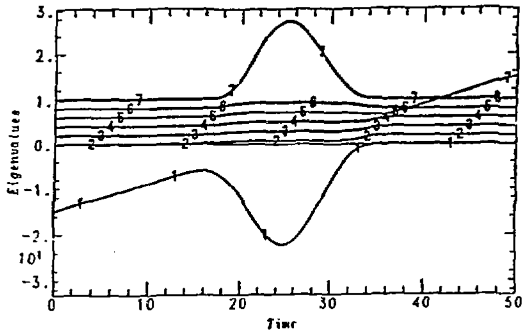
Pulse + Chirp



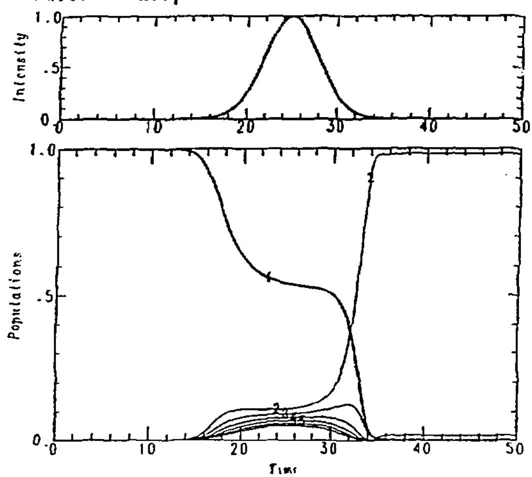
Pulse + Chirp N = 7



Pulse + Chirp

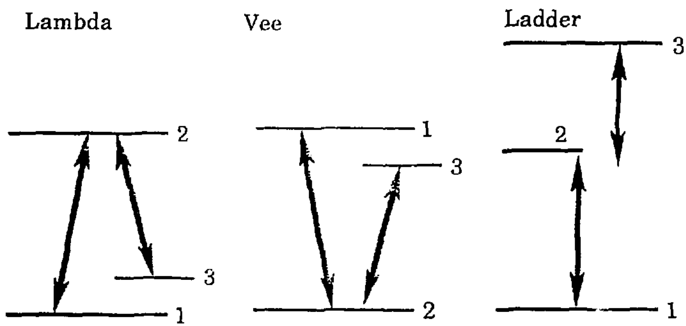


Pulse + Chirp N = 7



frame 2 11:15:54 04-25-93 g

Two Pulses, 3 Levels



Sequencing of pulses is important

Three Levels

The ladder, vee or lambda system

$$H(t) = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_1(t) & 0 \\ \Omega_1(t) & 2\Delta_2(t) & \Omega_2(t) \\ 0 & \Omega_2(t) & 2\Delta_3(t) \end{bmatrix}$$

Rabi frequencies:

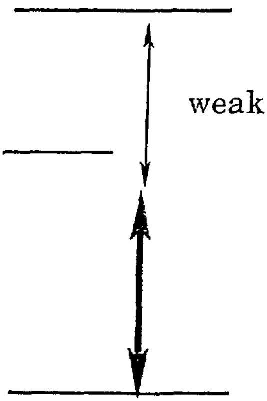
$\Omega_1 = 1 \longleftrightarrow 2$ (pump) $\Omega_2 = 2 \longleftrightarrow 3$ (Stokes)

Cumulative detunings:

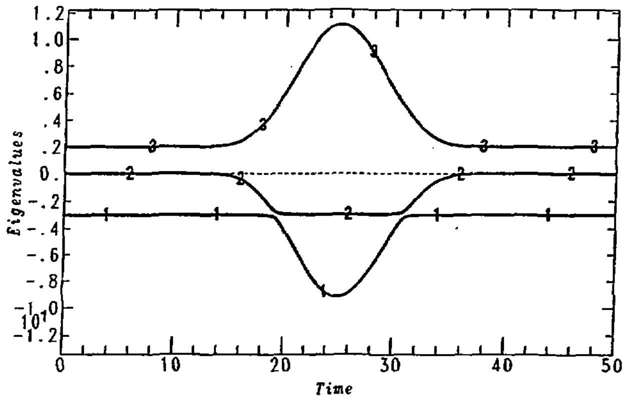
$\Delta_2 = 1$ photon $\Delta_3 = 2$ photon

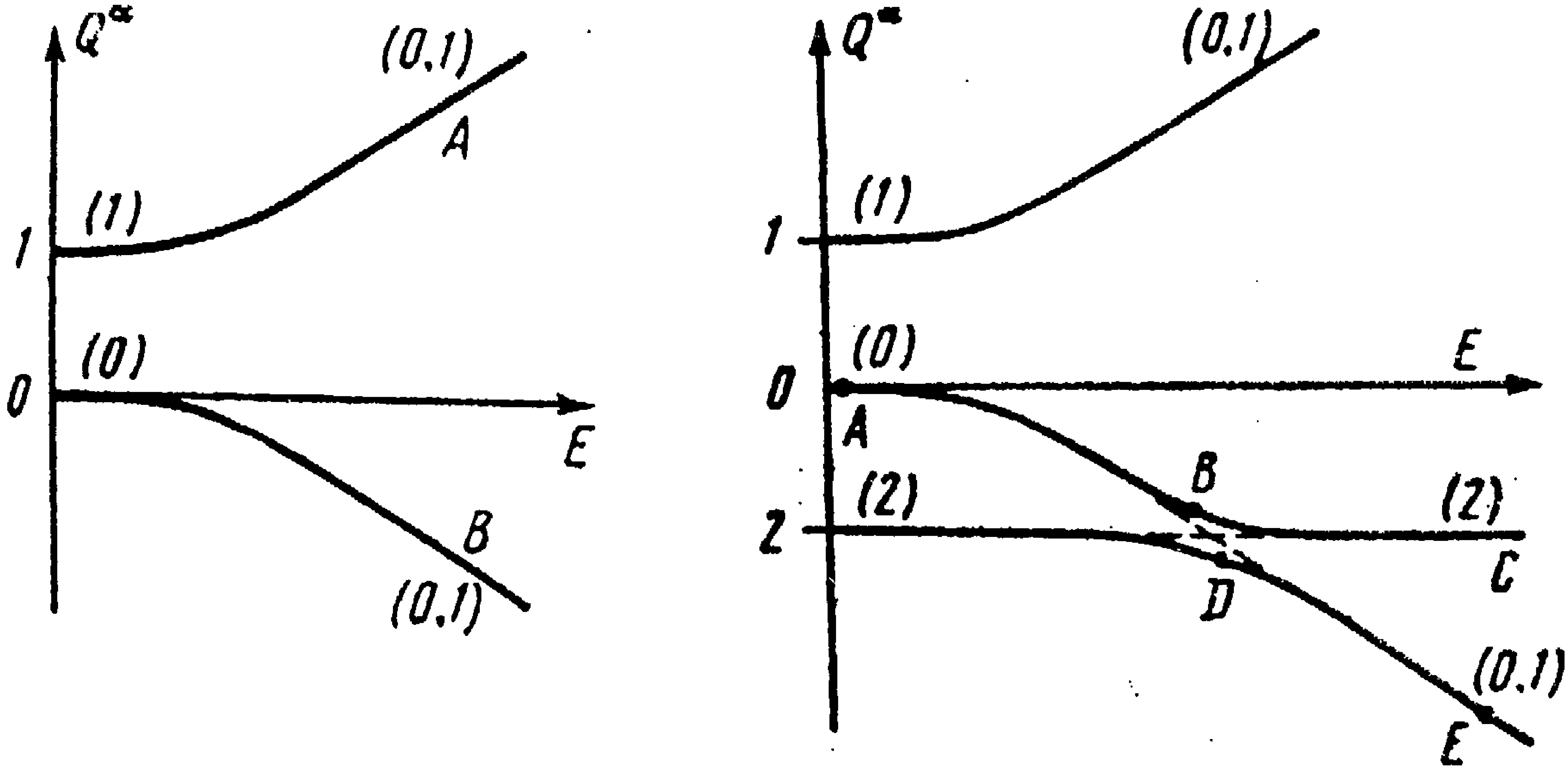
3 Levels, Single Pulse

Pulse with constant frequency, then abrupt stop

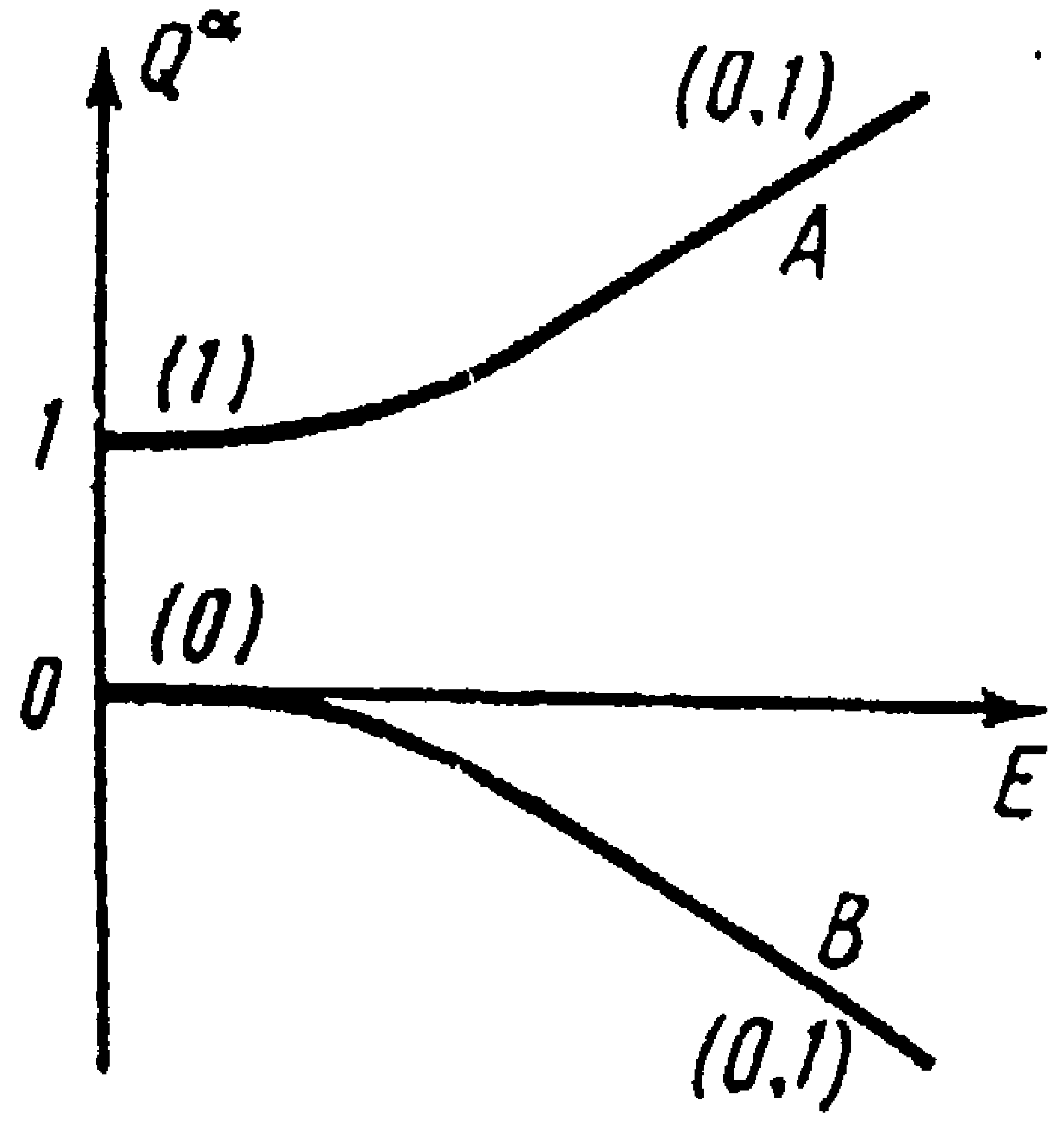


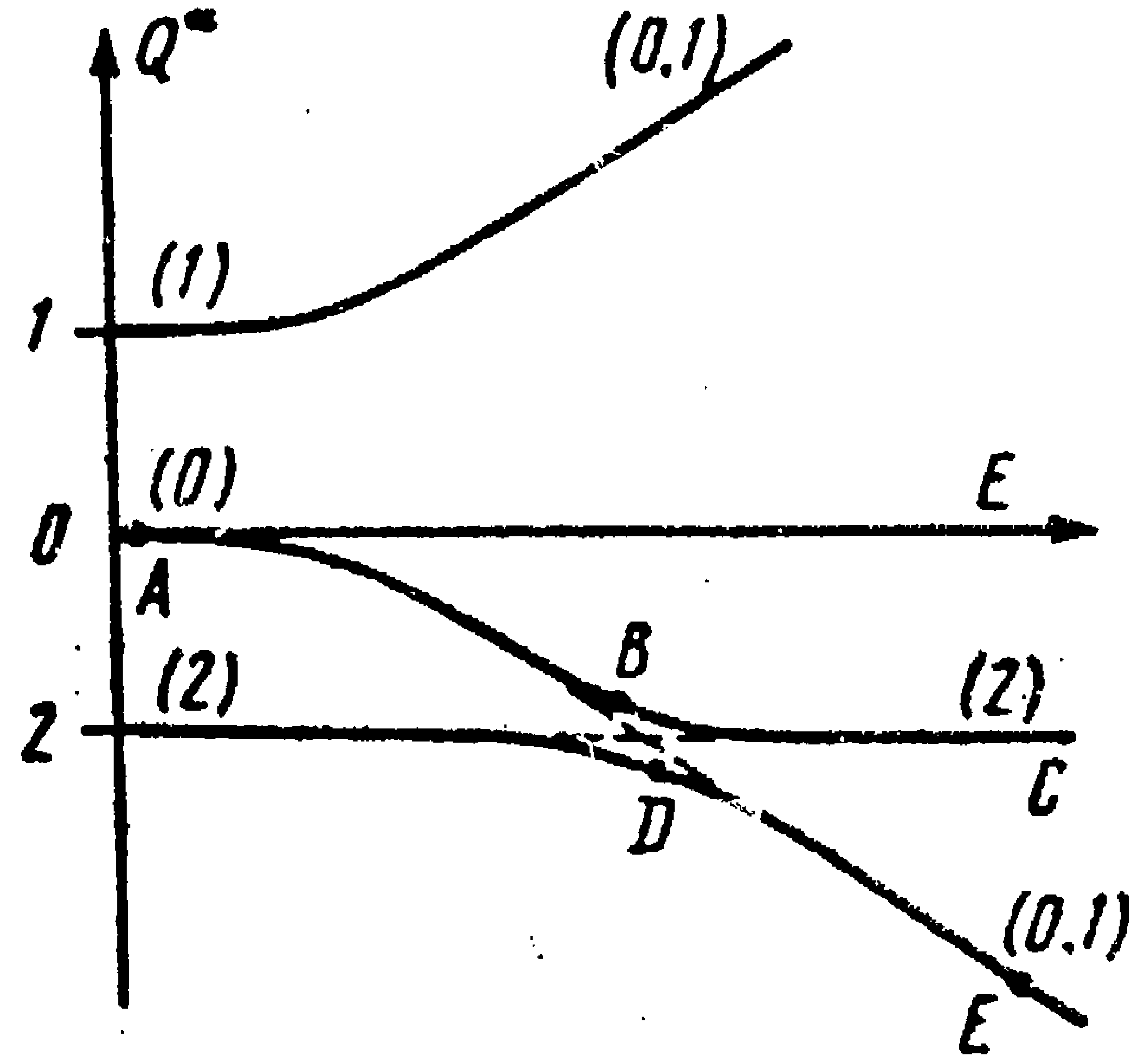
Pulse, weak 2-3 N = 3





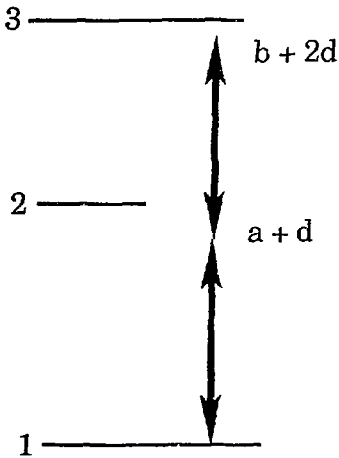
Kuz'min ... 80





Kuz'min ... 80

3 Levels, Single Chirp



Resonances:

One photon

Two photon

Results depend on sequencing of resonances

Three Levels: Two Photon Transition

For large intermediate detuning $\Delta_2(t)$

$$H(t) = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_1(t) & 0 \\ \Omega_1(t) & 2\Delta_2(t) & \Omega_2(t) \\ 0 & \Omega_2(t) & 2\Delta_3(t) \end{bmatrix} \Rightarrow \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega(t) \\ \Omega(t) & 2\Delta(t) \end{bmatrix}.$$

Effective detuning and Rabi Frequency

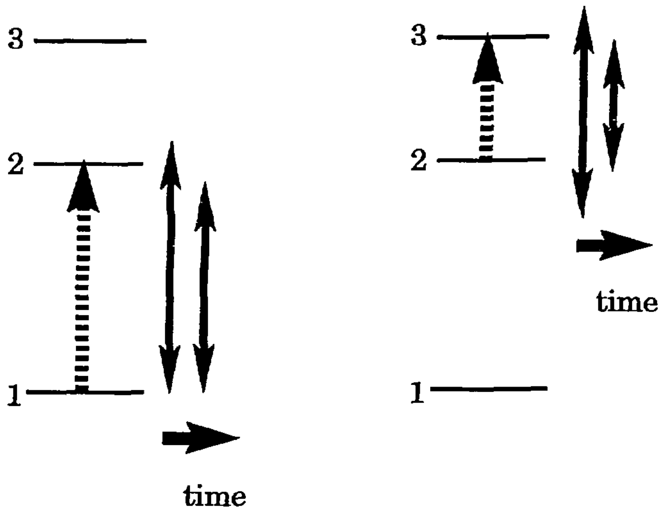
$$\Omega(t) = \frac{\Omega_1(t)\Omega_2(t)}{2\Delta_2(t)},$$

$$\Delta(t) = \Delta_3(t) + \frac{|\Omega_1(t)|^2 - |\Omega_2(t)|^2}{4\Delta_2(t)}.$$

Conditions for validity: large $\Delta_2(t)$

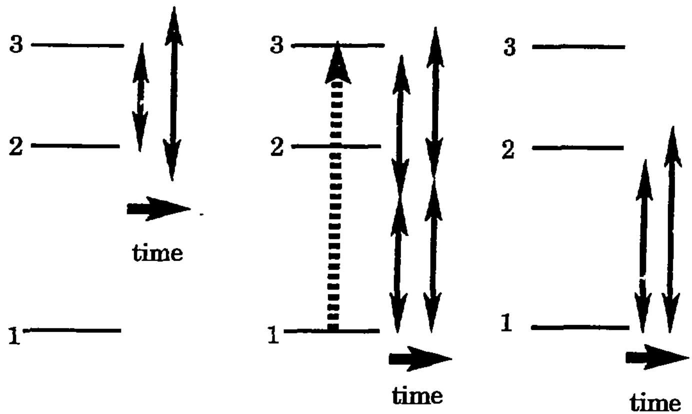
Intuitive Order

Pass through two 1-photon resonances



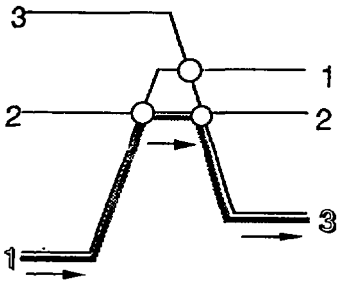
Counterintuitive Order

Pass through one 2-photon resonance

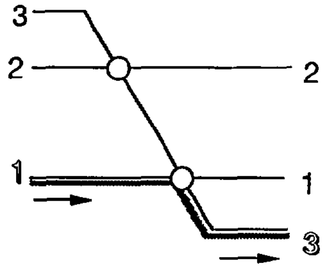


Curve Crossings

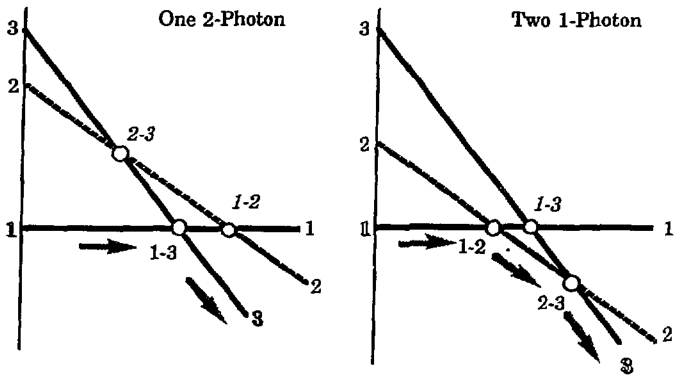
Two 1-photon crossings



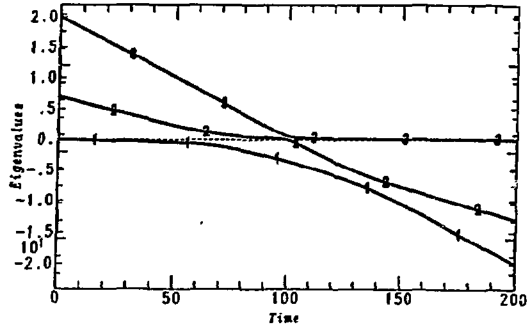
One 2-photon crossing



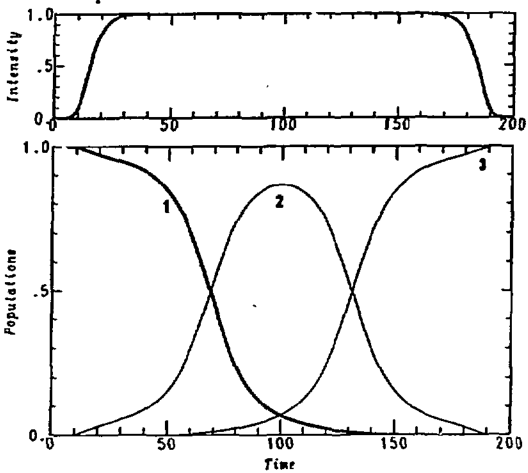
Curve Crossings 3 Level, single chirp



Two 1-photon

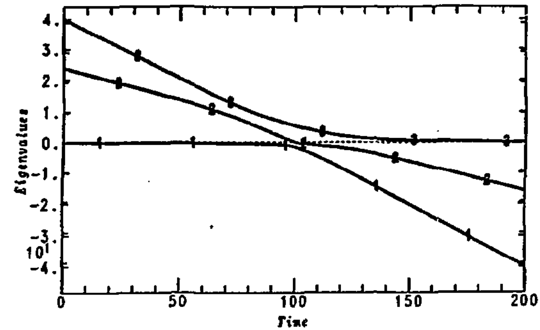


Two 1-photon

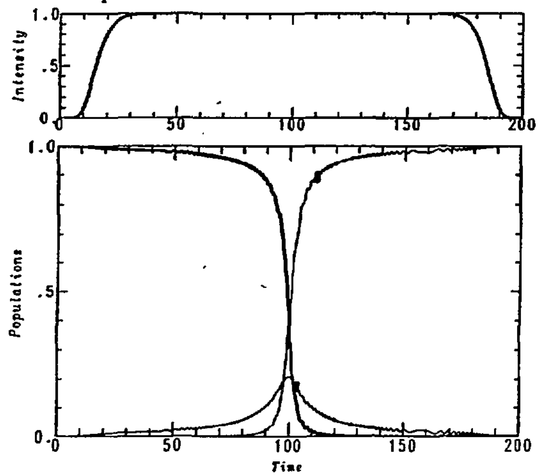


frame 1 12:05:52 03-07-92 y

One 2-photon

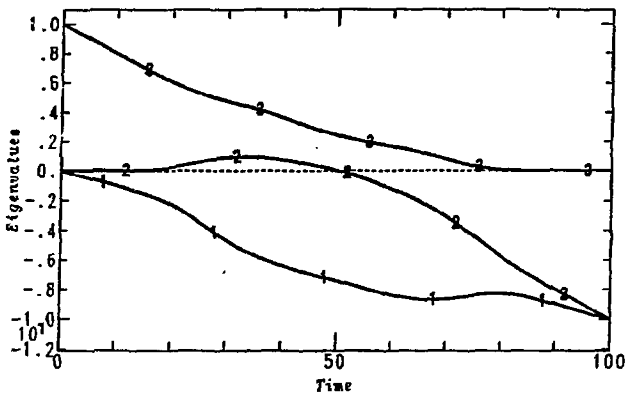


One 2-photon

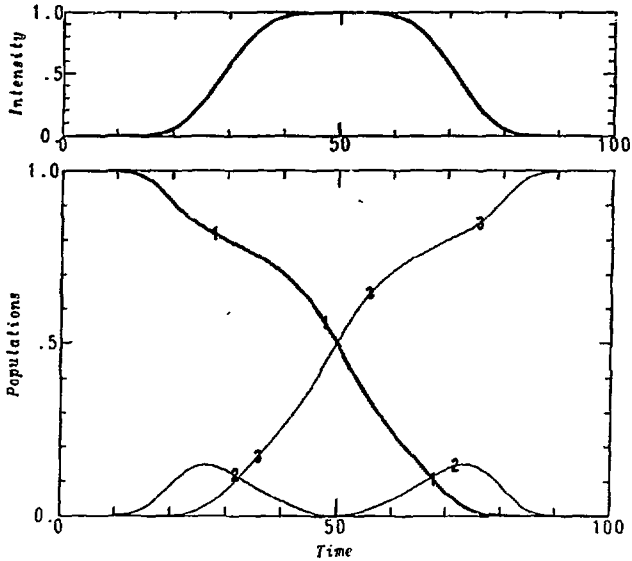


frame 4 12:06:50 03-07-82 y

Intuitive

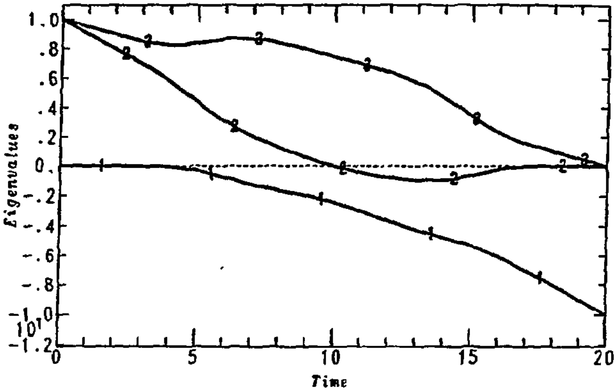


Intuitive

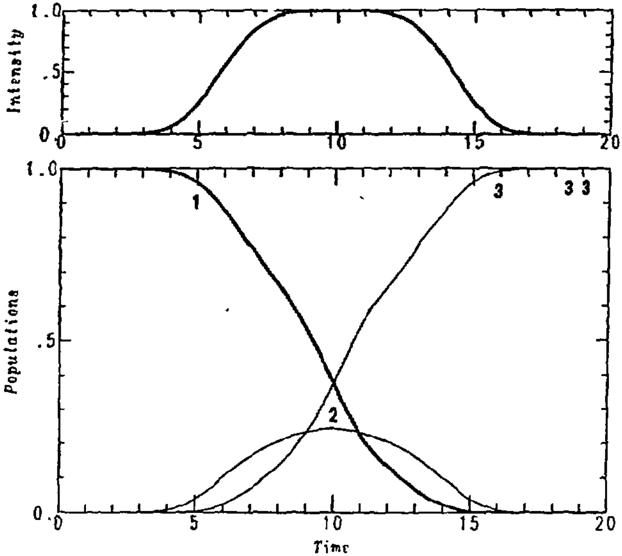


frame 4 11:46:16 09-03-92 y

Counterintuitive



Counterintuitive



frant t 11:44:09 09-09-92 y

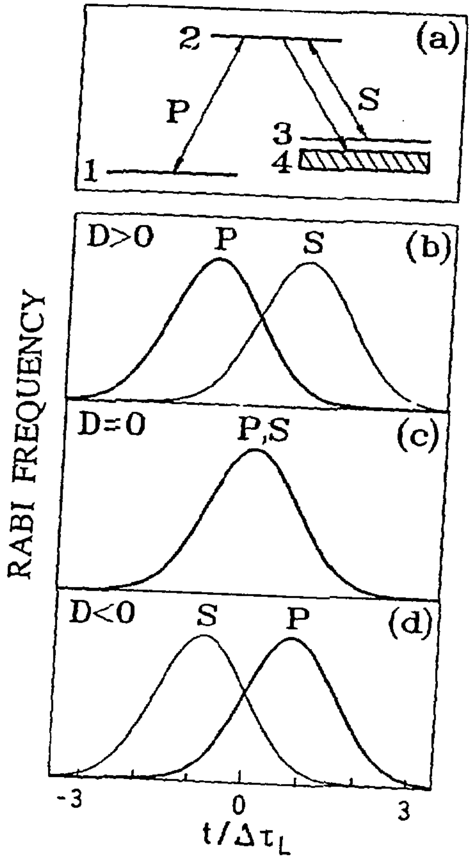
Delayed Pulses

Completely resonant case

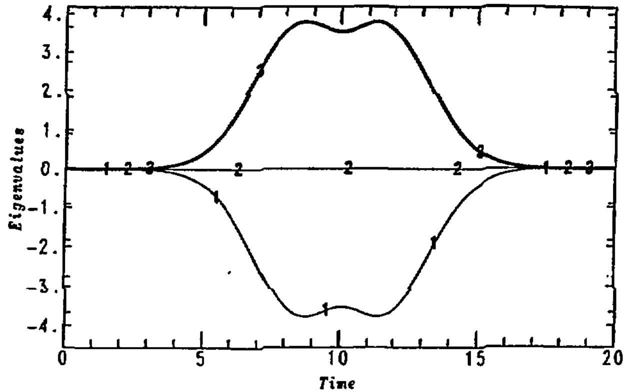
$$i\frac{d}{dt}\begin{bmatrix} C_1(t) \\ C_2(t) \\ C_3(t) \end{bmatrix} = -\frac{1}{2}\begin{bmatrix} 0 & \Omega_1(t) & 0 \\ \Omega_1(t) & 0 & \Omega_2(t) \\ 0 & \Omega_2(t) & 0 \end{bmatrix}\begin{bmatrix} C_1(t) \\ C_2(t) \\ C_3(t) \end{bmatrix} \qquad [24]$$

Possible pulse sequencing:

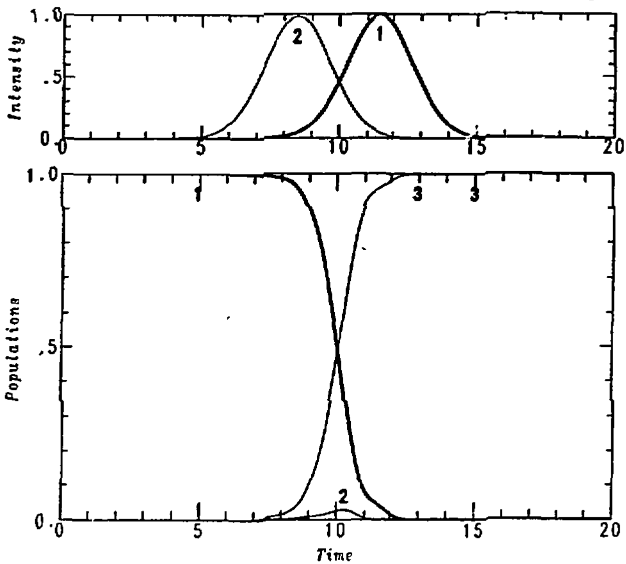
- **Intuitive:** first Ω_1 then Ω_2 (pump, then Stokes)
- **Simultaneous**: both Ω_1 and Ω_2
- **Counterintuitive**: first Ω_2 then Ω_1 (Stokes, then pump)



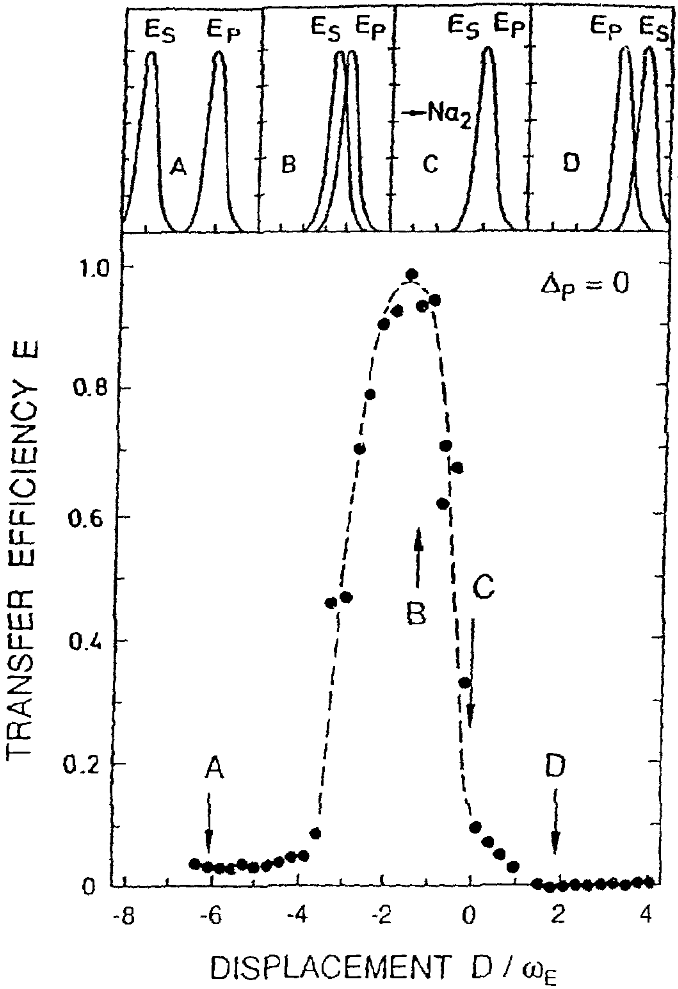
He... 30



Counterintuitive 10 pi



frame 1 16:32:36 09-09-92 y



Es
Ep
A
B
C
D
→Na₂
1.0
0.8
0.6
0.4
0.2
0
Δp = 0
TRANSFER EFFICIENCY E
-8
-6
-4
-2
0
2
4
DISPLACEMENT D / ωE

Three-Level Adiabatic States

Define

$$\Phi_0(t) = \cos\Theta\ \psi_1 \quad - \sin\Theta\ \psi_3 \qquad \omega_0 = 0$$

$$\Phi_+(t) = \frac{1}{\sqrt{2}}[\sin\Theta\ \psi_1 + \psi_2 + \cos\Theta\ \psi_3] \qquad \omega_+ = \overline{\Omega}(t) \qquad [25]$$

$$\Phi_-(t) = \frac{1}{\sqrt{2}}[\sin\Theta\ \psi_1 - \psi_2 + \cos\Theta\ \psi_3] \qquad \omega_- = -\overline{\Omega}(t)$$

where

$$\overline{\Omega}(\mathrm{t}) = \sqrt{(\Omega_1)^2 + (\Omega_2)^2} \qquad [26]$$

$$\tan\Theta = \Omega_1(t) / \Omega_2(t). \qquad [27]$$

Then

$$\Psi(t) = a_+(t)\ \Phi_+(t) + a_0(t)\ \Phi_0(t) + a_-(t)\ \Phi_-(t) \qquad [28]$$

$$1 = |\ a_+(t)\ |^2 + |\ a_0(t)\ |^2 + |\ a_-(t)\ |^2 \qquad [29]$$

Adiabatic condition (constant a_n)

$$\left|\ \frac{d\Theta}{dt}\ \right| << |\ \omega_0 - \omega_\pm\ | = |\ \overline{\Omega}(t)\ |. \qquad [30]$$

Important:

...$\Phi_0(t)$ has no component of ψ_2 ..

STIRAP

Stimulated Raman Adiabatic Passage

Initial:

$\Psi(-\infty) = \psi_1.$

$\Omega_1 << \Omega_2, \quad \cos\Theta \approx 1, \quad \Psi \approx \Phi_0 \approx \psi_1$

Intermediate:

Maintain adiabatic conditions

(Slow change wrt.. Eigenvalue differences)

Final:

$\Omega_1 >> \Omega_2, \quad \cos\Theta \approx 0, \quad \Psi \approx \Phi_0 \approx \psi_3$

$\Psi(+\infty) = \psi_2.$

STIRAP Properties

Advantages:

Complete population transfer

No population in intermediate level

Works on or off intermediate resonance

Insensitive to pulse area (need > 5π)

Constraint:

Must maintain 2-photon resonance

Extensions of STIRAP

- Multiple intermediate levels
- Continuum of intermediate Levels
- Noisy pulses
- Extraneous Levels
- 4-Level (3 pulse) STIRAP
- N-Level STIRAP (N = 3, 5, 7, 9, ...)

The Bloch Equations

Atom variables

$$\begin{aligned} u &= \rho_{12} + \rho_{21} && \textit{(coherence)} \\ v &= -i(\rho_{12} - \rho_{21}) && \\ w &= \rho_{22} - \rho_{11} && \textit{(inversion)} \end{aligned} \qquad [31]$$

Hamiltonian variables

$$\begin{aligned} \Lambda_1 &= H_{12} + H_{21} && = 2\,Re\,V_{12} \\ \Lambda_2 &= -i(H_{12} - H_{21}) && = 2\,Im\,V_{12} \\ \Lambda_3 &= H_{22} - H_{11} && = \Delta \end{aligned} \qquad [32]$$

Equation of motion

$$\frac{d}{dt}\begin{bmatrix} u \\ v \\ w \end{bmatrix} = \begin{bmatrix} 0 & -\Lambda_3 & \Lambda_2 \\ \Lambda_3 & 0 & \Lambda_1 \\ -\Lambda_2 & -\Lambda_1 & 0 \end{bmatrix}\begin{bmatrix} u \\ v \\ w \end{bmatrix}. \qquad [33]$$

Adiabatic Following

$$0 = \begin{bmatrix} 0 & -\Lambda_3 & \Lambda_2 \\ \Lambda_3 & 0 & \Lambda_1 \\ -\Lambda_2 & -\Lambda_1 & 0 \end{bmatrix}\begin{bmatrix} u \\ v \\ w \end{bmatrix}. \qquad [34]$$

Bloch Equations as Rotation

Bloch vector

$$r = [u, v, w]^T \qquad [35]$$

Angular velocity (torque) vector

$$\Lambda = [\Lambda_1\ \Lambda_2, \Lambda_3]^T \qquad [36]$$

Bloch eqn = Rotation (torque) equation

$$\frac{d}{dt}r(t) = \Lambda(t) \times r(t) \quad \text{or} \quad \frac{d}{dt}r_j(t) = \sum_{jk} \varepsilon_{ijk}\, \Lambda_j(t)\, r_k(t). \qquad [37]$$

Adiabatic following

$$\Lambda(t) \times s = 0 \quad \text{or} \quad \begin{bmatrix} 0 & -\Lambda_3 & \Lambda_2 \\ \Lambda_3 & 0 & \Lambda_1 \\ -\Lambda_2 & -\Lambda_1 & 0 \end{bmatrix} \begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix} = 0. \qquad [38]$$

Require r parallel to Λ

Multilevel Bloch Equations

Define coherences

$$u = [u_1,...,u_M]^T = [u_{12},u_{23},...,u_{13}, ...,u_{14}, ... ,u_{1N}]^T \quad [39]$$

$$v = [v_1,...,v_M]^T = [v_{12},v_{23},...,v_{13},...,v_{14},...,v_{1N}]^T \quad [40]$$

where

$$u_{jk} = \rho_{jk} + \rho_{kj} \quad for \ 1 \le j < k \le N \quad [41]$$

$$v_{jk} = -i(\rho_{jk} - \rho_{kj}) \quad for \ 1 \le j < k \le N \quad [42]$$

Define inversions

$$w_j = \sqrt{\frac{2}{j(j+1)}} \left[j\rho_{j+1,j+1} - \sum_{k=1}^{j} \rho_{kk} \right] \quad for \ 1 \le j \le N-1 \quad [43]$$

Generalized Bloch (or coherence) vector

$$r = [u_1,....,v_1,...,w_1,...w_{N-1}]^T \quad [44]$$

Equation is a rotation

$$\frac{d}{dt} r_j(t) = \sum_{jk} \varepsilon_{ijk} \Lambda_j(t) \ r_k(t) \quad [45]$$

Conclusion

- Adiabatic passage can produce complete population transfer
- Interesting pulsed schemes in multilevel systems
- Theory deals with structure of adiabatic states